CHARACTERIZATION OF ICE CRYSTALS AND WATER DROPLETS WITH THE ICE OPTICAL DETECTOR DEVICE (IODE) USING DEPOLARIZATION MEASUREMENTS

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« En essayant continuellement, on finit par réussir. 
Donc plus ça rate, plus on a de chances que ça marche »

*Devise Shadok*
ABSTRACT

In order to determine the efficiency of aerosol particles of several types to nucleate ice, an ice optical depolarization detector device (IODE) was developed to distinguish between water droplets and ice crystals in cloud chambers. In this thesis, results of simulated depolarization ratios ($\delta$) of single ice particles in fixed orientations are presented in order to determine if discrimination between non-spherical ice crystals (causing depolarization) and spherical water droplets (inducing no depolarization) can be made. A T-matrix method is used to compute $\delta$ over a range of particle diameters from 0.13 to 4 $\mu$m and aspect ratios $\Gamma = d/h$ ($d$ is the diameter and $h$ the height of the particle) from 0.3 to 3, where ice crystals are assumed to have a circular cylindrical shape. The depolarization ratio is primarily dependent on the orientation of the particle. Some orientations return no depolarization whereas others generate values reaching almost $\delta = 1$. Considering the depolarization averaged over all orientations, a dependence of $\delta$ with the particle size is observed where values close to 0.25 are reached. No strong influence of the aspect ratio on the depolarization for a given particle size of 2 $\mu$m is evident, as values remain in a range between 0.2 and 0.3.

The second part of this work will show the design of the detector and experimental results. This system operates at the bottom of a continuous flow diffusion chamber (ZINC chamber) and works in the following way: A laser beam polarized linearly (power: 50 mW, wavelength: 407 nm) is directed through the ZINC chamber. The scattered light intensity from particles is measured at a scattering angle of $\Theta = 175^\circ$ in both polarization components (parallel and perpendicular). The ratio between the perpendicular intensity over the total one gives the depolarization ratio $\delta$. Single particle detection is possible, using a peak detection algorithm. For high particle concentrations, a real-time signal averaging method can also be run simultaneously. To rule out depolarization ratios and intensities for water droplets, the perpendicular and parallel components have to be considered at the same time. In presence of ice crystals, peaks were detected in both channels, generating depolarization signals.

Experiments are first made in the laboratory with spherical particles and water droplets in order to see if they are detectable and if they do not depolarize light. Average intensities from 7.8 pW to 25.3 pW were obtained in the parallel channel. No significant peaks were recorded in the perpendicular channel. For 75-$\mu$m diameter water droplets, the average intensities were around 106 pW. Experiments with the ZINC chamber were then led, using montmorillonite as ice nuclei. Ice crystals concentrations reaching 1 cm$^{-3}$ were detected, giving mean depolarization ratios of 0.124 and 0.112. Finally the IODE detector was used during the ICIS 2007 workshop where ice nucleation experiments
were performed with several aerosol types. Mean values of $\delta$ ranged from 0.24 to 0.37.
The peak intensities obtained range from 5 pW (perpendicular channel) up to 1000 pW
in some rare peaks (parallel channel) and mean depolarization values between 0.09 and
0.2 were found in water breakthrough cases. Ice crystals can still be detected and
distinguished within water drops.
Dans le but de déterminer l'efficacité que possèdent les aérosols pour former des cristaux de glace par nucléation, un détecteur optique (IODE) utilisant la propriété de dépolarisation a été construit dans le but de pouvoir distinguer des gouttelettes d'eau des particules de glace dans des chambres de diffusion. Dans cette thèse, des résultats de coefficients de dépolarisation (δ) obtenus par simulations numériques et concernant des particules de glace unitaires dans une orientation fixe seront exposés. Ceci dans le but de déterminer si la distinction entre cristaux de glace non-sphériques (dépolarisant la lumière) et gouttelettes d'eau supposées sphériques (ne générant aucune dépolarisation) peut être réalisée. La méthode « T-matrix » est utilisée pour modéliser δ pour des particules de diamètre entre 0.13 et 4 µm et des coefficients de forme Γ = d/h (d étant le diamètre et h la hauteur) entre 0.3 et 3, où les cristaux sont considérés étant cylindriques. Le coefficient de dépolarisation dépend avant tout de l'orientation de la particule. Certaines orientations spécifiques ne créent aucune dépolarisation tandis que d'autres peuvent générer des valeurs de δ atteignant quasiment 1. En moyennant δ sur toutes les orientations possibles, une dépendance au niveau de la taille de la particule est constatée où des valeurs proches de 0.25 sont mises en évidence. En revanche, le coefficient de taille Γ ne possède pas d'influence notable pour une particule de 2 µm de diamètre par exemple, δ restant compris entre 0.2 et 0.3.

La seconde partie de cette thèse concerne l'élaboration du détecteur, ainsi que les résultats expérimentaux obtenus avec ce dernier. Le système est utilisé au bas d'une chambre de diffusion à flux continu (chambre ZINC) et fonctionne de la manière suivante : Un rayon laser polarisé linéairement (puissance: 50 mW, longueur d'onde: 407 nm) est dirigé transversalement dans la chambre ZINC. L'intensité de la lumière rétrodiffusée est mesurée à un angle de diffusion de Θ = 175° dans les deux composantes de polarisation (parallèle et perpendiculaire). Le rapport entre l'intensité du canal perpendiculaire et l'intensité totale donne le coefficient de dépolarisation δ. La détection de particules isolées est possible, moyennant l'utilisation d'un algorithme de détection de pics. Dans les cas de hautes concentrations en particules, une méthode de moyennage du signal brut peut également être utilisée simultanément. Pour obtenir les coefficients de dépolarisation, les intensités subséquentes des signaux des deux canaux doivent être prises en compte au même moment. En présence de cristaux de glace, des pics sont obtenus dans les deux canaux, générant des signaux de dépolarisation.

Dans un premier temps, des expériences ont été réalisées au laboratoire avec des particules sphériques en polystyrène et des gouttelettes d'eau. Le but est de voir si d'une part si ces éléments sont détectables, et d'autre part s'ils ne génèrent aucune
dépolarisation. Des intensités moyennes de l’ordre de 7.8 pW à 25.3 pW ont été obtenues dans le canal parallèle. Aucun pic significatif n’a été enregistré par le canal perpendiculaire. Pour des gouttelettes d’eau de 75 µm de diamètre, les intensités moyennes étaient de l’ordre de 106 pW. Des expériences avec la chambre ZINC ont ensuite été menées en utilisant de la montmorillonite comme aérosol. Des concentrations de cristaux de glace de l’ordre de 1 cm\(^{-3}\) ont été mesurées, donnant des moyennes de δ de 0.124 et 0.112. Pour terminer, le détecteur IODE a été employé lors de la campagne de mesure ICIS 2007 où des expériences de nucléation ont été réalisées avec plusieurs types d’aérosols. Les valeurs moyennes de δ ont été comprises entre 0.24 et 0.37. L’intensité des pics enregistrés ont varié entre 5 pW jusqu’à 1000 pW pour quelques cas isolés dans le canal parallèle. Dans les cas où de nombreuses gouttelettes d’eau ont été détectées, les valeurs moyennes du coefficient de dépolarisation sont restées comprises dans une fourchette allant de 0.09 à 0.2. Les cristaux de glace peuvent néanmoins y être différenciés de l’eau liquide.
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<tr>
<th>Acronym</th>
<th>Description</th>
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<tbody>
<tr>
<td>AIDA</td>
<td>Aerosol Interactions and Dynamics in the Atmosphere</td>
</tr>
<tr>
<td>APS</td>
<td>Aerodynamic particle sizer</td>
</tr>
<tr>
<td>CCN</td>
<td>Cloud condensation nuclei</td>
</tr>
<tr>
<td>CID</td>
<td>Canary Islands dust</td>
</tr>
<tr>
<td>CSU</td>
<td>Colorado state university</td>
</tr>
<tr>
<td>DAQ</td>
<td>Data acquisition unit</td>
</tr>
<tr>
<td>DDA</td>
<td>Discrete dipole approximation</td>
</tr>
<tr>
<td>FDTDM</td>
<td>Finite difference time domain method</td>
</tr>
<tr>
<td>FEM</td>
<td>Finite element method</td>
</tr>
<tr>
<td>FSSP</td>
<td>Particle measurement system forward scattering probe</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full width at half maximum</td>
</tr>
<tr>
<td>GO</td>
<td>Geometric optics</td>
</tr>
<tr>
<td>GPMM</td>
<td>Generalized point matching method</td>
</tr>
<tr>
<td>ID</td>
<td>Israel dust</td>
</tr>
<tr>
<td>IN</td>
<td>Ice nuclei</td>
</tr>
<tr>
<td>IODE</td>
<td>Ice optical detector</td>
</tr>
<tr>
<td>IPCC</td>
<td>Intergovernmental Panel on Climate Change</td>
</tr>
<tr>
<td>MOM</td>
<td>Method of moments</td>
</tr>
<tr>
<td>OPC</td>
<td>Optical particle counter</td>
</tr>
<tr>
<td>PINC</td>
<td>Portable ice nucleation chamber</td>
</tr>
<tr>
<td>PMM</td>
<td>Point matching method</td>
</tr>
<tr>
<td>PMT</td>
<td>Photomultiplier tube</td>
</tr>
<tr>
<td>PSL</td>
<td>Polystyrene latex spheres</td>
</tr>
<tr>
<td>SB</td>
<td>Snomax® bacteria</td>
</tr>
<tr>
<td>SD</td>
<td>Saharan dust</td>
</tr>
<tr>
<td>SID</td>
<td>Small ice detector</td>
</tr>
<tr>
<td>SMPS</td>
<td>Scanning mobility particle sizer</td>
</tr>
<tr>
<td>SVM</td>
<td>Separation of variables method</td>
</tr>
<tr>
<td>TMM</td>
<td>T-matrix method</td>
</tr>
<tr>
<td>VIEM</td>
<td>Volume integral equation method</td>
</tr>
<tr>
<td>Acronym</td>
<td>Definition</td>
</tr>
<tr>
<td>---------</td>
<td>------------------------------------------------</td>
</tr>
<tr>
<td>VIPS</td>
<td>Video ice particle sampler</td>
</tr>
<tr>
<td>VSFWF</td>
<td>Vector spherical wave function</td>
</tr>
<tr>
<td>WA</td>
<td>Water breakthrough events</td>
</tr>
<tr>
<td>ZINC</td>
<td>Zurich ice nucleation chamber</td>
</tr>
</tbody>
</table>
### List of Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_{\text{det}}$</td>
<td>$\text{m}^2$</td>
<td>active area of the photomultiplier window</td>
</tr>
<tr>
<td>$a_n$</td>
<td>[-]</td>
<td>Lorenz-Mie coefficients</td>
</tr>
<tr>
<td>$a_{mn}$</td>
<td>$\text{V/m}$</td>
<td>expansion coefficients of the incident electric field</td>
</tr>
<tr>
<td>$b_n$</td>
<td>[-]</td>
<td>Lorenz-Mie coefficients</td>
</tr>
<tr>
<td>$b_{mn}$</td>
<td>$\text{V/m}$</td>
<td>expansion coefficients of the incident electric field</td>
</tr>
<tr>
<td>$C_{\text{sca}}$</td>
<td>$\text{m}^2$</td>
<td>scattering cross section</td>
</tr>
<tr>
<td>$c$</td>
<td>$\text{m/s}$</td>
<td>speed of light in a vacuum</td>
</tr>
<tr>
<td>$c_{mn}$</td>
<td>$\text{V/m}$</td>
<td>expansion coefficients of the internal electric field</td>
</tr>
<tr>
<td>$d$</td>
<td>$\text{m}$</td>
<td>particle diameter</td>
</tr>
<tr>
<td>$d_{\text{eq}}$</td>
<td>$\text{m}$</td>
<td>volume-equivalent sphere particle diameter</td>
</tr>
<tr>
<td>$d_{mn}$</td>
<td>$\text{V/m}$</td>
<td>expansion coefficients of the internal electric field</td>
</tr>
<tr>
<td>$d_{lm}$</td>
<td>[-]</td>
<td>Wigner $d$-functions</td>
</tr>
<tr>
<td>$E$</td>
<td>$\text{V/m}$</td>
<td>electric field</td>
</tr>
<tr>
<td>$E_0$</td>
<td>$\text{V/m}$</td>
<td>constant complex vector of the electric field</td>
</tr>
<tr>
<td>$E_{\theta},E_{\varphi}$</td>
<td>$\text{V/m}$</td>
<td>components of the electric field</td>
</tr>
<tr>
<td>$E_{\theta},E_{\varphi}$</td>
<td>$\text{V/m}$</td>
<td>spherical coordinate components of the electric field vector</td>
</tr>
<tr>
<td>$F$</td>
<td>$\text{m}^2$</td>
<td>Stokes scattering matrix</td>
</tr>
<tr>
<td>$F_{ij}$</td>
<td>$\text{m}^2$</td>
<td>elements of the Stokes scattering matrix</td>
</tr>
<tr>
<td>$g$</td>
<td>[-]</td>
<td>gain function of the photomultiplier tubes</td>
</tr>
<tr>
<td>$H$</td>
<td>$\text{A/m}$</td>
<td>magnetic field</td>
</tr>
<tr>
<td>$H_0$</td>
<td>$\text{A/m}$</td>
<td>constant complex vector of the magnetic field</td>
</tr>
<tr>
<td>$h$</td>
<td>$\text{m}$</td>
<td>particle height</td>
</tr>
<tr>
<td>$h_{n}^{(1)}$</td>
<td>[-]</td>
<td>Hankel functions of the first kind</td>
</tr>
<tr>
<td>$I$</td>
<td>$\text{W/m}^2$</td>
<td>Stokes vector</td>
</tr>
<tr>
<td>$I$</td>
<td>$\text{W/m}^2$</td>
<td>first Stokes vector parameter</td>
</tr>
<tr>
<td>$I_\parallel$</td>
<td>$\text{pW}$</td>
<td>Scattering light intensity from the parallel channel</td>
</tr>
<tr>
<td>$I_\perp$</td>
<td>$\text{pW}$</td>
<td>scattering light intensity from the perpendicular channel</td>
</tr>
<tr>
<td>$j_n$</td>
<td>[-]</td>
<td>spherical Bessel functions of the first kind</td>
</tr>
<tr>
<td>$k$</td>
<td>$\text{m}^{-1}$</td>
<td>(complex) wave vector</td>
</tr>
</tbody>
</table>
\( \mathbf{k}_{\text{Re}} \) [m\(^{-1}\)] real part of the wave vector

\( \mathbf{k}_{\text{Im}} \) [m\(^{-1}\)] imaginary part of the wave vector

\( k \) [m\(^{-1}\)] (complex) wave number

\( k_{\text{Re}} \) [m\(^{-1}\)] real part of the wave number

\( k_{\text{Im}} \) [m\(^{-1}\)] imaginary part of the wave number

\( L \) [-] laboratory frame

\( \mathbf{M}_{mn} \) [-] vector spherical wave functions

\( M_\beta \) [-] weighting factor

\( m \) [-] (complex) relative refractive index of the medium

\( \mathbf{N}_{mn} \) [-] vector spherical wave functions

\( n \) [-] (complex) refractive index relative to vacuum/surrounding medium

\( n_{\text{sph}} \) [-] (complex) refractive index of the medium

\( n_{\text{med}} \) [-] (complex) refractive index of the surrounding medium

\( \hat{n} \) [-] unit vector

\( P \) [-] particle frame

\( P_n \) [-] Legendre polynomial

\( p \) [-] degree of polarization

\( p_C \) [-] degree of circular polarization

\( p_L \) [-] degree of linear polarization

\( p_Q \) [-] degree of linear polarization

\( p_i \) [Pa] ice vapor pressure

\( p_w \) [Pa] water vapor pressure

\( p_{mn} \) [V/m] expansion coefficients of the scattered electric field

\( \mathbf{Q} \) [-] Q-matrix

\( Q \) [W/m\(^2\)] second Stokes parameter

\( q_{mn} \) [V/m] expansion coefficients of the scattered electric field

\( R_{\text{conv}} \) [V/A] current-to-voltage conversion ratio of the preamplifier unit

\( r \) [m] position (radius) vector

\( r \) [m] distance from the origin of a coordinate system

\( r_> \) [m] radius of the smallest circumscribing sphere

\( r_< \) [m] radius of the largest concentric inscribed sphere

\( S \) [m] amplitude matrix

\( S_{ij} \) [m] elements of the amplitude matrix
\( S \ [m^2] \) closed surface area
\( T \ [-] \) T-matrix
\( T_{ij} \ [-] \) elements of the T-matrix
\( T_{ri} \ [-] \) transmittance of an optical element \( i \)
\( t \ [s] \) time
\( U \ [W/m^2] \) third Stokes parameter
\( U_{ctrl} \ [V] \) control voltage of the photomultiplier tube
\( V \ [W/m^2] \) fourth Stokes parameter
\( V \ [m^3] \) volume
\( v \ [m/s] \) phase velocity
\( x \ [-] \) size parameter
\( w \ [-] \) width parameter (peak detection algorithm)
\( y_n \ [-] \) spherical Bessel functions of the second kind
\( Z \ [m^2] \) Stokes phase matrix
\( Z_{ij} \ [m^2] \) elements of the Stokes phase matrix

\( \alpha \ [rad] \) first Euler angle
\( \beta \ [rad] \) second Euler angle
\( \delta \ [-] \) depolarization ratio
\( \delta_L \ [-] \) linear backscattering depolarization ratio
\( \delta_C \ [-] \) circular backscattering depolarization ratio
\( \delta_{Lid} \ [-] \) Lidar depolarization ratio
\( \delta_{\parallel} \ [-] \) parallel depolarization ratio
\( \delta_{\perp} \ [-] \) perpendicular depolarization ratio
\( \delta_{lim} \ [-] \) detectable depolarization threshold
\( \delta_{avg} \ [-] \) depolarization ratio averaged over all orientations
\( \delta_{\parallel avg} \ [-] \) parallel depolarization ratio averaged over all orientations
\( \delta_{\perp avg} \ [-] \) perpendicular depolarization ratio averaged over all orientations
\( \varepsilon \ [F/m] \) electric permittivity
\( \varepsilon_0 \ [F/m] \) electric permittivity of free space
\( \varepsilon \ [-] \) detection efficiency
\( \phi \ [rad] \) azimuth angle
\( \hat{\phi} \ [-] \) unit vector in the \( \phi \) direction
$\Gamma$ [-] aspect ratio

$\lambda$ [m] wavelength

$\mu$ [H/m] magnetic permeability

$\mu_0$ [H/m] magnetic permeability of free space

$\mu_{BG}$ [pW] average background signal

$\omega$ [rad/s] angular frequency

$\pi_n$ [-] angular functions

$\sigma_{BG}$ [pW] standard deviation of the background signal

$\tau$ [pW] Threshold detection limit (peak detection algorithm)

$\tau_n$ [-] angular functions

$\Theta$ [$^\circ$] scattering angle

$\theta$ [rad] polar (zenith) angle

$\hat{\theta}$ [rad] unit vector in the $\theta$ direction

$\psi_n$ [-] Ricatti-Bessel functions

$\zeta_n$ [-] Ricatti-Bessel functions

$\xi$ [mA/W] cathode radiant sensitivity
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INTRODUCTION

1.1 THE ATMOSPHERE

Derived from the Greek ‘ατμος’ (vapor) and ‘σφαιρα’ (sphere), the atmosphere is a layer of gases surrounding the Earth and retained by its gravity. There is no definite boundary between the atmosphere and the outer space. It slowly becomes thinner and fades into space and three quarters of the atmosphere’s mass is within 11 km of the planetary surface. The atmosphere is a mixture of gases containing some liquid and solid particles in suspension. The gaseous part can be divided into two parts: first, the major constituents which follow a vertical distribution, and second, the minor constituents which can have a very different behavior depending on the production, destruction and transport mechanisms. ‘dry air’ is formed by ideal gases that keep a quite constant proportion. For the purpose of meteorology, the composition and the molar mass of the atmosphere’s constituents has been internationally fixed (only CO₂, O₃ and Rn can vary) and are given in Table 1-1 (Lazzarotto et al. 2001).
<table>
<thead>
<tr>
<th>Gas</th>
<th>Volume [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>N₂</td>
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</tr>
<tr>
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<tr>
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<tr>
<td>Ne</td>
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<tr>
<td>O₃</td>
<td>0.4·10⁻⁶</td>
</tr>
<tr>
<td>Rn</td>
<td>6.0·10⁻¹⁸</td>
</tr>
</tbody>
</table>

Table 1-1: International values for the dry air volume percentage.

The average atmospheric pressure at sea level is about 1013 hPa and the pressure decreases exponentially with height, dropping by 50% at an altitude of about 5.6 km. A single equation can model atmospheric pressure through all altitudes even if the temperature changes throughout the atmospheric column, as well as the force of gravity begins to decrease at great altitudes. The temperature of the Earth’s atmosphere varies with altitude among the six different layers shown in Fig. 1-1:

Fig. 1-1: Temperature vertical profile of the earth atmosphere, expressed versus altitude (left scale) and pressure (right scale).
The troposphere is the lowest layer of the atmosphere. It begins at the surface and extends from 7 km (at the poles) to 17 km (at the equator) with some variations due to weather factors. The troposphere is characterized by a negative temperature gradient versus altitude from approximately 0.65°C / 100 m (for moist air) up to 1°C / 100 m (for dry air). This is due to the absorption, mainly by CO₂ and H₂O, of the infrared radiation reflected from the Earth’s surface. There is also a rapid vertical mixing in the troposphere due to solar heating at the surface. The stratosphere starts from the tropopause and is characterized by an increase of temperature with altitude. It ends with the stratopause at an altitude of approximately 50 km where the temperature reaches a maximum of about 0°C. This rise is the result of the solar UV absorption, mainly due to the ozone, which reaches a maximal concentration at a height of about 23 km. This creates a temperature inversion with warmer stratospheric air above the colder air of the top of the troposphere. As a consequence, this restricts considerably the vertical mixing both in the stratosphere and between the troposphere and the stratosphere. The mesosphere, located above the stratopause, extends from 50 km until the mesopause at about 85 km. Although CO₂ is a minor constituent, the infrared emission of this gas is sufficient to induce a temperature decrease where the minimum (~ –90°C) is reached at an altitude of about 90 km. The last region is the thermosphere which extends after the mesopause. The atmosphere is warmed by the UV solar radiation with wavelengths lower than 175 nm. The temperature constantly increases (~ 580°C at 200 km, ~ 720°C at 1000 km) until the thermopause where the temperature gradient starts to become negligible. The altitude of the thermopause is strongly linked to the solar activity.

Considering the Earth as a black body in the infrared and a grey body in the visible (due to a global mean albedo of ~0.3), the mean temperature at the surface should be –18.3 °C. Actually, the global mean temperature on the Earth varies between 11.9°C and 15.3°C due to the presence of compounds in the atmosphere that absorb the infrared radiation emitted by the planet (Larchevêque 2002). These gases called greenhouse gases then re-emit this absorbed radiation in all directions. However, the atmosphere is not completely absorbing and some other transfers take place as well such as heat. More infrared absorption by the atmosphere causes an increase in the temperature since the radiation is re-radiated to the ground (greenhouse effect).
main greenhouse gases are water vapor (H₂O), carbon dioxide (CO₂), ozone (O₃), methane (CH₄), nitrous oxide (N₂O), and halocarbons (CFC). Those gases (except CFCs) have both natural and anthropogenic sources and an increase induces an enhanced greenhouse effect. Although water vapor is the most important greenhouse gas, its strong variability in time and space makes it difficult to determine a clear radiative forcing. The water vapor distribution is mainly driven by air motions and the change of the water physical states, but is only slightly affected by human activities. For the other greenhouse gases, their radiative forcings due to their increases from 1750 to 2005 is estimated to be +2.63 W/m² overall: +1.66 W/m² by CO₂, +0.48 W/m² by CH₄, +0.16 W/m² by N₂O, +0.34 W/m² by halocarbons. The depletion of the stratospheric ozone is estimated to have caused a negative forcing of −0.05 W/m², whereas the radiative forcing of the tropospheric ozone is +0.35 W/m². Those radiative forcings have a high or medium level of scientific understanding (Forster et al. 2007). Further discussions will therefore focus on the aerosol particles and their influence on the atmosphere since their scientific understanding is still not well known.

1.2 THE AEROSOLS

By definition, an aerosol is a stable suspension of a solid or liquid in a gas (Gras 2003). In practical terms, for the ambient atmosphere this includes particles from nanometers to tens, and sometimes even hundreds of micrometers in diameter (from around 10⁻⁹ to 10⁻⁴ m). Aerosol particles may be classified into primary and secondary particles. Primary aerosols are directly emitted into the atmosphere from various sources. Secondary aerosols can be produced from gaseous precursors in the atmosphere (e.g. Finlayson–Pitts and Pitts 2000, Kulmala et al. 2004). They may include inorganic, organic and biological matter such as pollen, viruses, bacteria and plant debris. Most particles contain some water, although cloud elements are normally excluded from this description.

Aerosol particles have both natural sources (e.g.: wind-blown dust and sea spray such as salt) and anthropogenic sources (smokes, fumes and exhausts). In many regions, especially in the northern hemisphere mid-latitudes, anthropogenic components frequently dominate over the natural sources in terms of particle number and mass.
Generally, aerosol particles can either be internally mixed (mixed composition within a particle) or externally mixed (between particles). As a result, aerosols cannot be considered as a gas that is the same wherever it is measured. Particle lifetimes range from days near the surface to several months in the upper troposphere and their spatial and temporal distribution is significantly inhomogeneous. During their lifetimes, the composition of aerosol particles and their size distribution can undergo strong changes by coagulation with other particles, by chemical reactions, by condensation or by evaporation (Seinfeld and Pandis 1998). Their sources are various, ranging from volcanoes, fires, industrial plants and burning fossil fuels. Atmospheric aerosols are very dynamic: They evolve and change their properties from the production point until their last removal by sedimentation (dry deposition) or by wet processes (rain, snow).

1.3 AEROSOLS IN THE ATMOSPHERE AND THEIR INFLUENCES

Tropospheric aerosols have an impact on global climate and human health. Aerosol particles have a significant influence on the global radiation balance by scattering and absorbing the solar radiation, as well as scattering, absorbing and emitting thermal radiation (Gras 2003, Lohmann and Feichter 2005). These mechanisms are referred to as the aerosol direct effect. The radiative forcing of this effect has been newly evaluated by the last Intergovernmental Panel on Climate Change (IPCC) report to ~0.50 W/m² (± 0.40 W/m²) (Forster et al. 2007). The role of aerosol particles in cloud formation has been recognized almost 120 years ago as an essential component of cloud formation (Spurny 2000). Aerosols can affect the climate and change the cloud properties and characteristics by acting as cloud condensation nuclei (CCN) and ice nuclei (IN). This is defined as the aerosol indirect effect and will be discussed in more detail in the next section.

1.3.1 Aerosol indirect effect

According to the IPCC Fourth Assessment Report, the cloud albedo effect of anthropogenic aerosol particles amounts to a global mean forcing of ~0.70 W/m², ranging from ~1.8 to ~0.3 W/m² (see Fig. 1-2) (Forster et al. 2007). The cloud albedo
enhancement is also called *Twomey effect*: it refers to the reflection increase of solar radiation due to the more numerous, but smaller cloud droplets in a cloud whose liquid water content remains constant (Twomey 1959). Another aerosol effect on water clouds is the cloud lifetime effect: The more but smaller cloud droplets reduce the precipitation efficiency, extending therefore the cloud lifetime. Hence, the cloud reflectivity is augmented (Albrecht 1989). This effect is approximately as large as the Twomey effect (Lohmann and Feichter 2005). Finally, the semi-direct effect is the evaporation of cloud droplets caused by air heating due to the absorption of solar radiation by aerosols (Graßl 1979, Hansen et al. 1997). As a result, it produces a warming that can partly offset the cooling due to the indirect aerosol effect. However, a cooling effect can also occur, depending on the location of black carbon particles with respect to the cloud (Penner et al. 2003, Johnson et al. 2004). It has been shown that the cloud droplets were smaller in polluted clouds than in clean clouds over the Atlantic Ocean, but polluted clouds were revealed to be thinner when formed over the continent (Brenguier et al. 2000, Schwartz et al. 2002). Consequently, they are drier than the marine clean clouds (Lohmann and Lesins 2003). As the cloud albedo depends on both, the cloud droplet size, and the cloud thickness, it is difficult to detect an aerosol effect as they partially counteract each other.
In a supercooled water cloud, ice crystals will grow at the expense of cloud droplets if some of these freeze. This process (called Bergeron-Findeisen) takes place because of the lower saturation vapor pressure over ice than over water, leading to a glaciation of the supercooled water cloud. These clouds have a shorter lifetime than pure liquid clouds because precipitation formation is more efficient via the ice phase (Rogers and Yau 1989). Moreover, most precipitation worldwide originates from the ice phase (Lau and Wu 2003). If a fraction of hydrophilic soot aerosol particles is considered to act as contact IN in addition to mineral dust at temperature between 0°C and −35°C, an increase in aerosol concentration from pre-industrial times to present days will lead to a new indirect effect called glaciation indirect effect (Lohmann 2002a). Both warm and glaciation aerosol indirect effects are depicted in Fig. 1-3:
The increase in the contact ice nuclei in the present climate leads to a greater glaciation rate of supercooled clouds in the global climate model ECHAM 4 (Lohmann 2002b). Therefore, the amount of precipitation via the ice phase is enhanced, reducing the cloud cover. The cloud optical depth of mid-level clouds in mid- and high latitudes also drops. As a result, more solar radiation is absorbed within the Earth-atmosphere system. This effect can partially offset the cloud lifetime effect of warm clouds. Considering a supercooled cloud with a given liquid water content: if it contains more cloud droplets due to anthropogenic pollution for example, both the riming and the snowfall rates are smaller (Borys et al. 2003).

1.3.2 Ice nucleation

In mixed-phase clouds, temperatures are too warm for homogeneous freezing of supercooled aerosols or cloud droplets to occur \((T > -35^\circ C)\). To generate precipitation from supercooled liquid water, a solid surface is required to provide a substrate to initiate the formation of ice. The ice formation efficiency as a function of aerosol
particles properties is poorly understood because of the variety of heterogeneous ice nucleation modes. These are summarized in the following picture (Fig. 1-4).

![Diagram of ice nucleation modes](image)

Fig. 1-4: Illustration of the ice nucleation modes described by Vali (1985).

E.g. some materials can initiate ice formation at just a few degrees below the melting point. In addition, various substances can very effectively nucleate ice, going from substances like silver iodide (AgI) to organic macromolecules in the cell membranes of some bacteria. In mixed-phase clouds, ice is nucleated in some fraction of the droplets over a broad temperature range from about −5°C to −40°C. Four nucleation modes have been registered so far (see Fig. 1-4) (Vali 1985). Deposition freezing occurs when water vapor deposits on the aerosol particle, forming ice without any transition through the liquid phase in a supersaturated environment with respect to ice. Condensation freezing occurs when supersaturation with respect to water is reached. Liquid water first forms on the ice nuclei before it freezes. Aerosols can also act as ice nuclei by coming into contact with supercooled water droplets. This mode is called contact freezing and its mechanism is not fully understood, but it is clear that contact freezing is a function of the specific property of the nucleating substance, and not just of a mechanical action of the contact (vibration or shock) (Gőtz et al. 1991). Ice nuclei can also initiate freezing from within a cloud droplet by immersion freezing where the particle is suspended in the interior of the water droplet. Contact nucleation is usually the most efficient process at high temperatures (slightly below 0°C). At lower temperatures, immersion freezing can be more predominant (Lohmann and Feichter 2005). Deposition nucleation is
generally the less efficient mode due to its higher energy barrier than the required one for the freezing nucleation modes. However, this is only true for mixed phase clouds: Deposition freezing can be very efficient for cirrus cloud formation. In addition, ice nuclei may lose their nucleability if foreign gases such as sulfur dioxide (SO$_2$) or ammonia (NH$_3$) occupy their active sites (Pruppacher and Klett 1997).

Aerosols can affect ice clouds in a similar way as the Twomey effect for water clouds. An enhancement in the number of ice nuclei causes this effect. A modification of the ice water content of cirrus clouds can then exert a radiative effect in the infrared (Lohmann and Feichter 2005). However, these effects cannot be quantified yet. The condensation contrails created by exhausts of aircraft engines can be considered as cirrus clouds once they lost their line-shape. The climate forcing of aircraft on the upper tropospheric cirrus clouds can be potentially high (Penner et al. 1999). An increase in ice nuclei due to particles from aircraft emissions in the upper troposphere causes an indirect effect (Lohmann and Feichter 2005, Kärcher et al. 2006). It was found that an increase in fuel consumption in air traffic was correlated with the increase in cirrus cloudiness (Boucher 1999). This cloudiness enhancement was also found over the northern oceans and the United States by analyzing a longer surface dataset (Minnis et al. 2004). Climate model simulations showed that aviation has a significant impact on the number concentrations of black carbon and, therefore, of potential heterogeneous IN (coupled with mineral dust) (Hendricks et al. 2004).

To conclude, cloud radiative forcing in the present climate cools the climate by approximately 20 W/m$^2$ (Kiehl and Trenberth 1997). Changes of 10% in cloud radiative forcing could thus either cancel or double the anthropogenic greenhouse gas radiative forcing. Both, cloud lifetime, and the radiative transfer changes are the key points contributing to the indirect aerosol effect in climate forcing. This topic inserts itself directly on the main objectives of this work. They will be discussed in details in the next section.
1.4 Objectives

The importance of understanding ice nucleation in cloud microphysics has been recognized since landmark cloud seeding experiments in the 1940s (Schaefer 1947). Despite the fact that much has been learned about ice nucleation in the last half century, there are still large gaps in the understanding of these nucleation processes (Vali 1985, 1991). Even if the aerosol properties affecting the cloud condensation nuclei activation are well understood, there is a lack of knowledge in the ice nucleation capabilities of aerosol particles (Szyrmer and Zawadski 1997, Cantrell and Heymsfield 2005). The theory for heterogeneous nucleation including the molecular nature of its initiation and growth stages is not totally understood. Another important unresolved problem is to understand the link between ice nucleation and cloud properties that affect the climate. Some modeling studies have shown the importance of ice nucleation, but good observations and ice nucleation parametrizations are still missing (Levkov et al. 1995, Rotstayan et al. 2000, Lohmann et al. 2001, Cotton and Field 2002). All these complexities make theoretical approaches quite impractical. Thus, we have to rely on laboratory and field measurements to gain a greater understanding of the ice nucleation mechanisms and to improve their representation in climate models.

A good approach is to perform measurements of ice nucleation processes in the laboratory under controlled conditions. This can be achieved by studying the effectiveness of aerosols at producing ice crystals under different conditions of temperature and humidity. An accepted method is to create a controlled ice supersaturated environment in a diffusion chamber where ice crystals grow by vapor deposition to detectable ice crystals sizes. This chamber can be a static type chamber where the aerosols are placed on an inert substrate. Otherwise, there are also dynamic or continuous flow type chambers, where the aerosols are carried by an air stream between two ice covered plates held at different temperatures. This type of chamber also allows continuous measurements in the laboratory or in the field. The particles are suspended in the flow and therefore do not require the addition of an electric charge. In a diffusion chamber, a cold and a warm wall provide fixed boundary conditions on the temperature and humidity (Schaller and Fukuta 1979). A linear temperature and saturation profile is developed in the steady state between the walls. Consequently, this
results in a steady ice supersaturation whose value depends on the location between the walls. A slow laminar flow parallel to the walls permits growth of ice crystals to a sufficient size for detection at the exit of the chamber. For our experiments, a new instrument to study ice nucleation, the Zurich Ice Nucleation Chamber (ZINC), has been built (Stetzer et al. 2007). It is a continuous flow diffusion chamber and its design is based on the successful Colorado State University (CSU) instrument (Rogers 1988, 1993). One major difference is that the ZINC chamber uses two parallel walls instead of two concentric cylinders. The ice crystals formed inside the chamber and grown to detectable sizes (from 1 µm in diameter) are then counted at its exit with an optical particle counter (OPC).

One of the limitations of the existing continuous flow diffusion chambers is the indirect detection of ice crystals by particle size (Rogers et al. 2001). In the case of the CSU and ZINC chambers, an impactor removes aerosol particles larger than 2 µm from the airstream to identify ice crystals solely based on size. To overcome this limitation, a new detection system was built to distinguish between droplets and ice crystals in order to allow both phases to coexist. Another major element is the condition under which the ZINC chamber, as well as the CSU chamber, is working: ice nucleation experiments can be performed at temperatures between roughly −4°C down to −40°C for the ZINC chamber. At cold temperatures (below −20°C), deposition freezing occurs inside the chamber, meaning that nucleation of ice does not take place via the liquid phase, but occurs directly from the vapor to the solid phase. If higher supersaturation ratio and warmer temperatures are investigated, immersion freezing can take place as well. In this case, we may have both phases present during experiments so that we have to be able to differentiate between them. If water droplets and ice crystals are present simultaneously, it is not possible to distinguish between them only by size. The property of light depolarization can be used in that way, considering that ice particles are nonspherical and thus induce depolarization of the light whereas water droplets do not (Liou and Lahore 1974). If this method works, the evaporation section of the ZINC chamber can be removed. Then the chamber would be shorter which is of advantage for field campaigns.

The depolarization device developed in this thesis follows a similar approach as the one that is used in the AIDA (Aerosol Interactions and Dynamics in the Atmosphere)
chamber where depolarization measurements are performed during ice nucleation experiments. The optical system is comparable as they use 2 convergent lenses separated by a pinhole. The laser used is a 25-mW Ar+ diode working at 488 nm. The splitting of the two polarization component is done with a Glan-Thompson prism (Büttner 2004). Whereas Büttner (2004) focuses on multi-particle events, we may encounter single particle events as IN concentrations might be very low in the field (in the order 10 ℓ⁻¹ at −25°C) (Götz et al. 1991). As the overlap region calculated for our device is 3.02·10⁻⁴ ℓ, we can expect single particle detection. Several experimental applications have been built for single particle detection such as the Small Ice detector (SID) (Hirst et al. 2001), the Particle Measurement System forward scattering probe (FSSP) (Lawson and Cormack 1995) and the Video Ice Particle Sampler (VIPS) (McFarquhar et al. 2002). Other optical systems dealt with ice nucleation of single levitated (Krämer et al. 1996) or free-falling droplets (Wood et al. 2002) where detection of single events using depolarization has been performed. It will be shown that single detection aspects play a major role for light depolarization as well as the particle orientation (chapter 3). Its influence on the depolarization ratio is large (Nicolet et al. 2007). In this thesis, simulation results of single particle depolarization ratios, the design and the software of the ice detector device called IODE (Ice Optical DEtector) will be presented. The IODE device was connected to the bottom of the ZINC chamber where the scattering and polarization of water droplets, aerosol particles and ice crystals were investigated. The results obtained with this detector will be discussed along with perspectives, challenges and possible future work.
2

THEORY OF LIGHT SCATTERING

2.1 LIGHT SCATTERING BY PARTICLES

Various science disciplines such as atmospheric sciences, astronomy, and oceanography have a significant interest in the scattering of light as well as other electromagnetic radiation by small particles (Mishchenko et al. 2000). Engineering sciences are also a field of active research in electromagnetic scattering with specific applications in meteorology, remote sensing and climate research (Kahnert 2003). Gas molecules and suspended particles scatter incident sunlight within the Earth’s atmosphere, giving rise to blue skies, white clouds and many other optical phenomena (Rainbows, coronae and halos). In climatology, the Earth’s radiation budget is strongly influenced by cloud and aerosol scattering and absorption of solar radiation for instance. This radiation is divided into two parts: The short-wave radiation coming from the sun and the long-wave radiation that is emitted from the Earth’s surface. The radiation budget needs to be evaluated and it requires the knowledge of the optical properties of cloud and aerosol particles (Lacis and Mishchenko 1995, Rossow and Schiffer 1999). Another example is the remote sensing of the Earth atmosphere system as it relies on the fraction of radiation scattered and/or emitted by aerosols, clouds and precipitation.
Historical works by Mie (e.g. Mie 1908) and Debye (e.g. Debye 1909) discuss scattering by spherical particles. In particular, the Lorenz-Mie theory can be readily used to determine the light scattering of homogeneous and layered spherical particles composed if isotropic materials are assumed. However, many natural and artificial particles present in the atmosphere have non-spherical shapes. For example, mineral dust and soot aerosols cannot be considered as being spherical, as well as other particles composed of anisotropic materials. Snow and frost crystals can also be considered as non-spherical elements (Mishchenko et al. 2000). Consequently, cirrus clouds and contrail particles also belong into this category. It is now well recognized that the scattering properties of non-spherical particles can drastically differ from those of “equivalent” Mie spheres which are often used as an approximation. Thus, it is very important to clearly understand the role and effect of particle non-sphericity on scattering patterns by measuring and computing adequately the light scattering of these kinds of particles.

A general overview of the basic theory of electromagnetic scattering, absorption, and emission is provided in the next section. Then, the general relationships for matrices describing electromagnetic scattering by small particles will be discussed. A particular accent will be finally put on single particle scattering.

2.1.1 Introduction to electromagnetic scattering

A parallel monochromatic beam of light is by definition an oscillating plane electromagnetic wave. Similarly, a particle is defined as an aggregation of a large number of discrete elementary electric charges. The parallel monochromatic beam propagates in the vacuum without any change in its intensity or polarization state. However, placing a small particle into the beam can cause several distinct effects. Firstly, one of these effects, absorption, is the transfer of some of the energy in the beam by the particle into other forms of energy such as heat. Energy is dissipated from the electromagnetic wave into the medium. For complex systems composed of molecules with a large number of degrees of freedom, many different transitions produce spectral emission lines that are very closely spaced. Thus, the resulting radiation spectrum becomes continuous and includes emitted energy at all frequencies. Secondly, another
phenomenon called *elastic scattering* is defined by the scattering of radiation in all directions at the incident beam frequency. The oscillating electromagnetic field of the incident wave excites the charges to oscillate with the same frequency and therefore radiate secondary electromagnetic waves. The superposition of the overall secondary waves gives the total elastically scattered field. As a consequence, the reduction of the incident beam energy is equal to the sum of the absorbed and scattered energy. This reduction is called *extinction*. In addition to that, the polarization state of the beam after it interacts with the particle may be changed. This phenomenon is called *dichroism* where the extinction rates for different polarization components of the incident beam can be different. Fig. 2-1 summarizes the different effects caused by an electromagnetic wave irradiating a particle.

![Fig. 2-1: Summary of the different elastic and inelastic (Raman, fluorescence) interactions between a particle and an incident light beam.](image)

The classical electromagnetic theory is used as the analytical and numerical basis to describe the scattering properties of media composed of small discrete particles. For our purpose, Maxwell’s equations and relations for time-harmonic macroscopic electromagnetic fields are formulated in order to derive the simplest case: The plane-wave solution. It is related to the basic optical idea of the monochromatic parallel beam of light. This solution naturally leads to the introduction of fundamental quantities such as the refractive index and the Stokes parameters that will be discussed in section 2.1.5. The concept of a quasi-monochromatic beam of light is also defined in detail as well as its implications.
2.1.2 Maxwell’s equations

A fundamental feature of the Maxwell equations concerns the plane-wave solution, allowing a simple traveling-wave solution and given in SI units (Jackson 1998). It represents the transport of electromagnetic energy from one point to another and embodies the concept of a perfectly monochromatic parallel beam of light. Given a homogeneous medium without sources, the solution of a plane electromagnetic wave propagating in such a medium can be written as follows:

\[ E_c(r,t) = E_0 \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t) \]  \hspace{1cm} (2.1)

\[ H_c(r,t) = H_0 \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t) \]  \hspace{1cm} (2.2)

Where \( E_0 \) and \( H_0 \) are constant complex vectors, \( \mathbf{r} \) is the position (radius) vector, \( t \) is the time, \( \omega \) is the angular frequency and \( \mathbf{k} \) is the wave vector. This latter one is also constant and may, in general, be complex:

\[ \mathbf{k} = \mathbf{k}_{\text{Re}} + i\mathbf{k}_{\text{Im}} \]  \hspace{1cm} (2.3)

\( \mathbf{k}_{\text{Re}} \) and \( \mathbf{k}_{\text{Im}} \) are real vectors. Hence we have:

\[ E_c(r,t) = E_0 \exp(-\mathbf{k}_{\text{Im}} \cdot \mathbf{r}) \exp(i\mathbf{k}_{\text{Re}} \cdot \mathbf{r} - i\omega t) \]  \hspace{1cm} (2.4)

\[ H_c(r,t) = H_0 \exp(-\mathbf{k}_{\text{Im}} \cdot \mathbf{r}) \exp(i\mathbf{k}_{\text{Re}} \cdot \mathbf{r} - i\omega t) \]  \hspace{1cm} (2.5)

\( E_0 \exp(-\mathbf{k}_{\text{Im}} \cdot \mathbf{r}) \) and \( H_0 \exp(-\mathbf{k}_{\text{Im}} \cdot \mathbf{r}) \) are the amplitudes of the electric and magnetic waves, respectively and \( \mathbf{k}_{\text{Re}} \cdot \mathbf{r} - \omega t \) is their phase. \( \mathbf{k}_{\text{Re}} \) is normal to the surfaces of constant phase whereas \( \mathbf{k}_{\text{Im}} \) is normal to the surfaces of constant amplitude. The electromagnetic wave is called homogeneous when \( \mathbf{k}_{\text{Re}} \) is parallel to \( \mathbf{k}_{\text{Im}} \) including the case where \( \mathbf{k}_{\text{Im}} = 0 \).
Otherwise, it is called *inhomogeneous*. If the two vectors are parallel, the complex wave vector can be written as:

\[ \mathbf{k} = \hat{n}(k_{Re} + ik_{Im}) \]  

(2.6)

\( \hat{n} \) is a real unit vector in the direction of propagation. Both \( k_{Re} \) and \( k_{Im} \) are real and non-negative. The Maxwell equations for the plane wave can now be given as:

\[ \mathbf{k} \cdot \mathbf{E}_0 = 0 \]  

(2.7)

\[ \mathbf{k} \cdot \mathbf{H}_0 = 0 \]  

(2.8)

\[ \mathbf{k} \times \mathbf{E}_0 = \omega \mu \mathbf{H}_0 \]  

(2.9)

\[ \mathbf{k} \times \mathbf{H}_0 = -\omega \varepsilon \mathbf{E}_0 \]  

(2.10)

Where \( \varepsilon \) and \( \mu \) are the electric permittivity and the magnetic permeability, respectively. Eqs. (2.7) and (2.8) indicate that the plane electromagnetic wave is transverse. Both \( \mathbf{E}_0 \) and \( \mathbf{H}_0 \) are perpendicular to \( \mathbf{k} \). Furthermore, \( \mathbf{E}_0 \) and \( \mathbf{H}_0 \) are mutually perpendicular from Eqs. (2.9) and (2.10) \((\mathbf{E}_0 \cdot \mathbf{H}_0 = 0)\). Eqs. (2.1) to (2.9) give the following relation:

\[ \mathbf{H}_c(\mathbf{r},t) = \mathbf{k}(\omega \mu)^{-1} \times \mathbf{E}_c(\mathbf{r},t) \]  

(2.11)

Therefore, only the electric field (or the magnetic field) can be used to represent a plane electromagnetic wave. If we take the vector product of both sides of Eq. (2.9) with \( \mathbf{k} \) and use Eq. (2.10) together with Eq. (2.7), the following expression can be derived:

\[ \mathbf{k} \cdot \mathbf{k} = \omega^2 \varepsilon \mu \]  

(2.12)

In the case of a homogeneous plane wave, a relation for \( k \) is obtained from Eq. (2.12):
\[ k = k_{\text{Re}} + ik_{\text{Im}} = \omega \sqrt{\varepsilon \mu} = \frac{\omega n}{c} \] (2.13)

Where \( k = 2\pi/\lambda \) is the free-space wave number. The speed of light in a vacuum (c) is given by:

\[ c = \frac{1}{\sqrt{\varepsilon_0 \mu_0}} \] (2.14)

Where \( \varepsilon_0 \) and \( \mu_0 \) are the electric permittivity and the magnetic permeability in free space, respectively. The complex refractive index \( n \) is expressed as following:

\[ n = n_{\text{Re}} + in_{\text{Im}} = c\sqrt{\varepsilon \mu} \] (2.15)

\( n_{\text{Re}} \) is the non-negative real part of \( n \), \( n_{\text{Im}} \) being the non-negative imaginary part. Hence, the plane homogeneous wave has the form:

\[ E_c(r,t) = E_0 \exp\left( -\frac{\omega}{c} n_{\text{Im}} \hat{n} \cdot r \right) \exp\left( i\frac{\omega}{c} n_{\text{Re}} \hat{n} \cdot r - i\omega t \right) \] (2.16)

The real part of the refractive index determines the phase velocity of the wave \( v \):

\[ v = \frac{c}{n_{\text{Re}}} \] (2.17)

For a vacuum, \( n = n_{\text{Re}} = 1 \) and \( v = c \). If the imaginary part of the refractive index is not equal to zero, it determines the decay of the wave amplitude as it propagates through the medium, which is consequently absorbing.
2.1.3 Independent scattering

The electric and magnetic field variations of an electromagnetic wave are linked by the Maxwell’s equations. From the moment, several well-defined restrictions are imposed in order to focus and simplify the discussion of electromagnetic scattering on the most relevant points for this purpose. We limit the treatment of that topic to the scattering by individual particles within a framework of Maxwell’s equations and linear optics. First, only time-harmonic and quasi-monochromatic light is taken into account. It is assumed that the (complex) amplitude of the electric field fluctuates with time much more slowly than the time factor $\exp(-i\omega t)$. Second, electromagnetic scattering occurs without frequency redistribution: the scattered light has the same frequency as the incident light. This assumption excludes inelastic phenomena such as Raman scattering or fluorescence. Third, only scattering in the far-field zone is considered. The propagation of the scattered wave is away from the particle and the electric field vector is oriented perpendicularly to the propagation direction. The scattered field amplitude decreases inversely with distance from the particle. Fourth, we assume that the unbounded host medium surrounding the scatterer is homogeneous, isotropic, non-absorbing and linear. Fifth and most important, only independent scattering and randomly positioned particles are considered. This means that particles are separated widely enough in a way that each of them scatters light exactly in the same way as if no other particles existed. Moreover, no systematic phase relation is assumed between all partial electromagnetic waves scattered by different particles. Thus, the partial wave intensities can be added without regard to phase. To resume, each particle is in the far-field zones of all other particles and scattering by different particles is incoherent.

2.1.4 Reference frame and particle orientation

The directions of incident and scattered waves must be described with respect to a reference frame, as well as the orientation of the particle. In order to express the scattering of a plane electromagnetic wave by a non-spherical particle, this frame is linked to the laboratory and can be defined with a right-handed Cartesian coordinate system $L$ with its origin centered in the middle of the particle (Figure 2-2). This system is referred to as the laboratory reference frame (Watermann 1971). In accordance with
our experimental setup, the $L$ coordinate system is shown in this configuration, where the $x$ axis is pointing out of the sheet, the $y$ axis down and the $z$ axis is directed to the left:

![Fig. 2-2: Laboratory coordinate system](image)

According to Fig. 2-2, the direction of propagation of a transverse electromagnetic wave is defined by the unit vector $\hat{n}$ or with the angles $\theta$ and $\varphi$ where $\theta \in [0, \pi]$ is the polar angle measured from the positive $z$ axis and $\varphi \in [0, 2\pi]$ is the azimuth angle measured from the positive $x$ axis in the clockwise sense when looking in the direction of the positive $z$ axis. $E_\theta$ and $E_\varphi$ are the components of the electric field denoted by $\hat{\theta}$ and $\hat{\varphi}$. Therefore, we have $E_\theta = E_{\theta} \hat{\theta}$ and $E_\varphi = E_{\varphi} \hat{\varphi}$. $\hat{\theta}$ and $\hat{\varphi}$ are the corresponding unit vectors which satisfy $\hat{n} = \hat{\theta} \cdot \hat{\varphi}$.

![Fig. 2-3: Euler angles of rotation $\alpha$ and $\beta$ transforming the laboratory frame $L[x,y,z]$ into the particle frame of the scattering particle $P[x',y',z']$.](image)
A right handed coordinate system $P$, referred to as particle frame, with the same origin as $L$, is then introduced to specify the orientation of the particle with respect to the laboratory reference frame (Figure 2.3). In our case, the particle will be assumed to have an axis of symmetry ($z'$). Therefore, only two angles are needed to describe the orientation of the particle. This system is fixed to the particle and the orientation is determined by two Euler angles of rotation $\alpha$ and $\beta$. These angles transform the coordinate system $L\{x,y,z\}$ into the coordinate system $P\{x',y',z'\}$ and are performed as follows: $\alpha \in [0,2\pi]$ is the anticlockwise rotation about the $z$ axis. $\beta \in [0,\pi]$ is the anticlockwise rotation about the new $y$ axis ($y'$).

2.1.5 Stokes parameters

Usually, it is convenient to use the Stokes parameters of a monochromatic transverse electromagnetic wave to describe its scattering. These parameters can be written as a $4 \times 1$ column vector called the Stokes vector:

$$I = \begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix}$$

The first parameter is equal to the net monochromatic energy flux up to a factor common to all four parameters. $Q$ and $U$ describe the state of linear polarization and $V$ expresses the state of the circular polarization of the wave. These parameters are defined separately as follows:

\[
\begin{align*}
I &= E_\theta E_\theta^* + E_\phi E_\phi^* \\
Q &= E_\theta E_\theta^* - E_\phi E_\phi^* \\
U &= -E_\phi E_\phi^* - E_\theta E_\theta^* \\
V &= i(E_\phi E_\phi^* - E_\theta E_\theta^*)
\end{align*}
\]
Where the asterisk denotes the complex conjugate value (Asano and Sato 1980). The Stokes parameters of a plane monochromatic wave are related by the quadratic identity:

\[ I^2 \equiv Q^2 + U^2 + V^2 \tag{2.20} \]

However, this identity may not be valid for a quasi-monochromatic beam of light. The degree of polarization \( p \) is given by:

\[ p = \frac{\sqrt{Q^2 + U^2 + V^2}}{I} \leq 1, \tag{2.21} \]

and is equal to unity for fully polarized light. In particular, the degrees of linear polarization \( p_L \) and circular polarization \( p_C \) can be defined as:

\[ p_L = \frac{\sqrt{Q^2 + U^2}}{I} \tag{2.22} \]

\[ p_C = \frac{V}{I} \tag{2.23} \]

If \( U \) vanishes, the ratio \( p_Q = -Q/I \) is also often used as a measure of the degree of linear polarization. For unpolarized (natural) light, \( Q = U = V = 0 \). Fig. 2.4 illustrates the main state of light polarization (linear and circular) and the related Stokes parameters:
2.1.6 Amplitude and Phase matrices

Let us consider the same wave as defined before and incident upon a non-spherical particle in a direction \( \hat{n}^{\text{inc}} \):

\[
E^{\text{inc}}(\mathbf{r}) = E^{\text{inc}}_{\theta}(\mathbf{r}) \hat{\theta}^{\text{inc}} + E^{\text{inc}}_{\phi}(\mathbf{r}) \hat{\phi}^{\text{inc}}
\]  

(2.24)

Where the time dependence factor \( \exp(-i\omega t) \) is omitted. Due to the linearity of the Maxwell’s equations and boundary conditions, the scattered electric field can be expressed linearly in terms of the incident electric field. In the far-field region \( kr >> 1, r = |\mathbf{r}| \) where \( \mathbf{r} \) is the radius vector with its origin in the origin of the laboratory coordinate system, the scattered wave becomes spherical and is given by (Tsang et al. 1985):

\[
E^{\text{sca}}(\mathbf{r}) = E^{\text{sca}}_{\theta}(r, \hat{n}^{\text{sca}}) \hat{\theta}^{\text{sca}} + E^{\text{sca}}_{\phi}(r, \hat{n}^{\text{sca}}) \hat{\phi}^{\text{sca}}
\]  

(2.25)

\[
\hat{n}^{\text{sca}} = \frac{\mathbf{r}}{r}
\]  

(2.26)
Now it is possible to express the electric field vector components of the scattered wave as a function of the electric field vector components of the incident wave using the so-called amplitude matrix ($S$).

\[
\begin{bmatrix}
E_{\phi}^{\text{sca}} \\
E_{\varphi}^{\text{sca}}
\end{bmatrix}
= \frac{\exp(ikr)}{r} S(\hat{n}^{\text{sca}}; \hat{n}^{\text{inc}}; \alpha, \beta)
\begin{bmatrix}
E_{\phi}^{\text{inc}} \\
E_{\varphi}^{\text{inc}}
\end{bmatrix}
\]  

(2.27)

This 2 × 2 matrix depends on several factors such as the directions of incidence and scattering light, the size, morphology and composition of the scattering particle and its orientation given by the Euler angles $\alpha$ and $\beta$. The amplitude matrix is the primary quantity that defines the single-scattering law. From the definitions of the $S$ matrix and the Stokes parameters, the transformation of the latter of the incident plane wave into those of the scattered wave considering light scattering by a single particle is given by:

\[
I^{\text{sca}} = \frac{1}{r^2} Z(\hat{n}^{\text{sca}}; \hat{n}^{\text{inc}}; \alpha, \beta) I^{\text{inc}}
\]  

(2.28)

\[
I^{\text{sca}} = \frac{1}{r^2} Z(\theta^{\text{sca}}, \phi^{\text{sca}}; \theta^{\text{inc}}, \phi^{\text{inc}}; \alpha, \beta) I^{\text{inc}}
\]  

(2.29)

Where $Z$ is a 4 × 4 phase matrix where generally all 16 elements are non-zero. However, these elements can be expressed in terms of only 7 independent real constants resulting from the four complex elements of the amplitude matrix minus an irrelevant phase (van de Hulst 1957). There are a number of linear and quadratic inequalities that can be used for testing the elements of the obtained phase matrix considering a collection of particles (Hovenier and van der Mee 2000). The most important of them are:

\[
Z_{11} \geq 0
\]  

(2.30)

\[
|Z_{ij}| \leq Z_{11}, \; i,j = 1,\ldots,4.
\]  

(2.31)
2.1.7 Scattering matrix

The scattering matrix $F$ relates the Stokes parameters of the incident and scattered beams defined with respect to the scattering plane. This plane is determined by the unit vectors $\hat{n}^{\text{inc}}$ and $\hat{n}^{\text{sca}}$ (van de Hulst 1957, Bohren and Huffmann 1983). The easiest case of the scattering matrix is to direct the $z$ axis of the laboratory reference frame $L$ along the incident beam ($\theta^{\text{inc}} = 0$). The principal azimuthal plane is superposed with $\varphi^{\text{inc}} = \varphi^{\text{sca}} = 0$. Then, the scattering matrix is proportional to the phase matrix $Z(\theta^{\text{sca}} = \varphi^{\text{sca}} = \theta^{\text{inc}} = \varphi^{\text{inc}} = 0)$. All 16 elements of $F$ are generally non-zero and depend on the particle orientation with respect to the incident and scattered beams.

The concept of the scattering matrix is particularly useful when all orientations of the particles are of equal probability, e.g. particles are randomly oriented. If each particle has a plane of symmetry and/or particles and their mirror particles are present in equal number, such a scattering medium is called *macroscopically isotropic* and *symmetric*. Due to the symmetry, the scattering matrix $F$ for such a media is invariant with respect to the choice of the scattering plane. It depends only on the angle between the incident and the scattered beam defined by $\Theta = \theta^{\text{sca}}$:

$$F(\Theta) = \frac{4\pi}{C^{\text{sca}}} \langle Z(\Theta,0;0,0) \rangle$$

$$C^{\text{sca}} = 2\pi \int_0^\pi \sin(\Theta) \langle Z_{11}(\Theta,0;0,0) \rangle \, d\Theta$$

Where $C^{\text{sca}}$ is the average scattering cross section per particle. The proportionality constant $4\pi / C^{\text{sca}}$ comes from the usual normalization condition on the phase function. For macroscopically isotropic and symmetric media, the average scattering cross section is independent of the direction and polarization of the incident beam. The scattering matrix for this media has a block-diagonal structure (van de Hulst 1957):
\[
F(\Theta) = \begin{bmatrix}
F_{11}(\Theta) & F_{12}(\Theta) & 0 & 0 \\
F_{12}(\Theta) & F_{22}(\Theta) & 0 & 0 \\
0 & 0 & F_{33}(\Theta) & F_{34}(\Theta) \\
0 & 0 & -F_{34}(\Theta) & F_{44}(\Theta)
\end{bmatrix}
\]

(2.34)

As a direct consequence of Eqs. (2.30) and (2.31), we have the following inequalities:

\[
F_{11} \geq 0
\]

(2.35)

\[
|F_{ij}| \leq F_{11}, \ i,j = 1,\ldots,4.
\]

(2.36)

For spherically symmetric particles, the structure of the scattering matrix is simplified further. The amplitude matrix is always diagonal and we have:

\[
F(\Theta) = \begin{bmatrix}
F_{11}(\Theta) & F_{12}(\Theta) & 0 & 0 \\
F_{12}(\Theta) & F_{11}(\Theta) & 0 & 0 \\
0 & 0 & F_{33}(\Theta) & F_{34}(\Theta) \\
0 & 0 & -F_{34}(\Theta) & F_{33}(\Theta)
\end{bmatrix}
\]

(2.37)

This type of scattering matrix appears in the standard Lorenz-Mie theory of light scattering by homogeneous isotropic spheres. Therefore, the matrix shown in Eq. (2.37) is referred to as the Lorenz-Mie scattering matrix. Particularly, we have the following expressions for exact backward (\(\Theta = \pi\)) and forward (\(\Theta = 0\)) scattering:

\[
F_{33}(0) = F_{11}(0) \quad \text{and} \quad F_{33}(\pi) = -F_{11}(\pi)
\]

(2.38)

If the particles are not spherically symmetric and do not form a macroscopically isotropic and mirror-symmetric medium, the scattering matrix \(F\) does not have the simple block-diagonal structure of Eq. (2.34). All 16 elements of \(F\) can be non-zero and depend on the incidence direction and the orientation of the scattering plane rather than
only on the scattering angle. The phase matrix depends on the specific values of the azimuthal angles of the incidence and scattering directions rather than on their differences. These effects can directly indicate the presence of oriented particles lacking spherical symmetry. For example, measurements of interstellar polarization are used in astrophysics to detect preferentially oriented dust grains. Different values of extinction for different polarization components of the transmitted starlight are obtained (Martin 1978). In the same way, the depolarization of radiowave signals propagating through the Earth’s atmosphere may indicate the presence of partially aligned non-spherical hydrometeors (Oguchi 1983).

If non-spherical particles are randomly oriented and form a macroscopically isotropic and mirror-symmetric medium, then the phase matrix depends only on the difference between the azimuthal angles of the incidence and scattering directions. The scattering matrix becomes block-diagonal (Eq. (2.34)) and depends only on the scattering angle. Moreover, \( F \) possesses almost the same structure as the Lorenz-Mie scattering matrix (Eq. (2.47)). However, the key point is that the Lorenz-Mie identities \( F_{22}(\Theta) = F_{11}(\Theta) \) and \( F_{44}(\Theta) = F_{33}(\Theta) \) generally do not hold for non-spherical particles. Therefore, the most reliable indicator of particle non-sphericity is the linear depolarization ratio \( \delta_L \):

\[
\delta_L = \frac{F_{11}(\pi) - F_{22}(\pi)}{F_{11}(\pi) + F_{22}(\pi)}
\]  

(2.39)

Although it is not yet well exploited (Woodward et al. 1998), the circular depolarization ratio \( \delta_C \) given in terms of \( \delta_L \) can also be used to indicate particle non-sphericity (Mishchenko and Hovenier 1995):

\[
\delta_C = \frac{2\delta_L}{1-\delta_L}
\]  

(2.40)

When assuming randomly oriented particles (scattering matrix), we have \( F_{12} = F_{21} \). If we consider a single particle in a fixed orientation (phase matrix), then we have \( Z_{12} \neq Z_{21} \), leading to a different definition of the depolarization ratio that will be discussed in the
next section. One exception is for perfect backscattering ($\Theta = 180^\circ$) as shown in the appendix A where $Z_{12} = Z_{21}$.

### 2.2 The Depolarization Technique

The interaction of light with aerosols or cloud and precipitation particles (hydrometeors) has often been studied since they reveal a large variety of optical displays. The appearance of halo arcs, rainbows and coronas actually represent concentrations of reflected, refracted or diffracted natural light in the scattering interactions of atmospheric particles of various sizes, shapes and orientations. For example, the scattering geometry is peculiar to each display, the reflections from randomly polarized sunlight can become partially, or even totally polarized (Sassen 1987a, Können and Tinbergen 1998). Similarly, considering a completely polarized light source, a change in the polarization state of the scattered light might be expected. As previously discussed, the main goal of this project is focused on the differentiation between ice and water particles in cloud physics applications. The effect of particle non-sphericity can be described by the structure modification of the scattering matrix $F$ as mentioned in section 2.1.7 using the concept of light depolarization.

The technique of light depolarization is commonly used (e.g. in lidar applications) to differentiate between water droplets and ice crystals by determining the degree of the depolarization of the scattered light using an incident linearly polarized light source. Liquid water particles are assumed to be spherical and therefore cause no depolarization. Ice crystals can be considered as being non-spherical and cause partial depolarization of the scattered light (Liou and Lahore 1974). In other words, let us consider a polarized laser beam where its electromagnetic field is parallel to the scattering plane (parallel receiver channel). The scattered light from water droplets will induce a scattered intensity only in the parallel receiver channel. However, scattered light from ice crystals will generate intensity in both the parallel and the perpendicular channel. Back- and side scattering from spherical particles results from external and internal reflections, and surface waves, which produce no depolarization. Similarly, scattering into the forward direction is governed by diffraction and grazing reflections which do not involve any depolarization. On the other hand, scattering from non-
spherical particles may result in part from non-depolarizing specular (external) reflections, but many strong contributions may also be made by multiple internal reflections at the particle faces. These phenomena are more pronounced in the backward and side directions since forward scattering is still dominated by diffraction (Sassen and Liou 1979).

Since the 1970s, the depolarization technique has been used in lidars and in remote sensing applications to detect ice crystals in clouds (Sassen 1974, Sassen 1977, Sassen and Liou 1979, Gobbi 1998). Radar backscattering measurements are also widely used to characterize non-spherical particles in atmospheric remote sensing applications (Aydın 2000, Bringi and Chandrasekar 2001) and biomedicine (Schmitt and Xiang 1998, de Boer et al. 1999). For example, the foundation of fuzzy logic systems was reviewed for the classification of hydrometeor types based on polarimetric radar observations (Liu and Chandrasekar 2000, Straka et al. 2000). Focusing on atmospheric research, lidar depolarization observations are used to distinguish between various types of polar stratospheric clouds (Browell et al. 1990). More recently, a depolarization device was built for the AIDA chamber (Büttner 2004) to measure the depolarization in situ from ice crystals and aerosols in the chamber. Due to technical considerations, measurements are not performed at a perfect backscattering angle ($\Theta = 176^\circ$). The next section will point out at the theoretical aspects of the depolarization ratio for single particles. Fixed orientations have also to be taken into account because a single particle has a specific orientation when it is detected. This is not the case in the AIDA chamber experiments as collections of particles that are randomly oriented inside the vessel are investigated.

### 2.2.1 Depolarization ratio for single particles in a fixed orientation

The depolarization ratio has been defined in several ways. Here it is defined as the intensity of the scattered light in the perpendicular channel over the total amount of scattered light assuming an incident light polarized parallel to the scattering plane (Isawaka et al. 2003, Sakai et al. 2003):

$$\delta = \frac{I_{\text{sca}}^{\perp}}{I_{\text{sca}}^{\parallel} + I_{\text{sca}}^{\perp}}$$  \hspace{1cm} (2.41)
This definition has the advantage of being normalized \( \delta \in [0,1] \). \( \delta \) allows us to describe the depolarization for non-spherical particles such as certain aerosols or ice crystals. This experimental definition can be linked to the theoretical one that can be derived from the elements of the phase matrix. Let us consider a single particle with its orientation fixed and a completely linearly polarized laser source. The light is polarized parallel to the scattering plane. The Stokes vector of the incident light is then given by \([1,1,0,0]^\top\). The Stokes vector of the scattered light is then given by (Mishchenko et al. 2000):

\[
\begin{bmatrix}
I^\text{sca} \\
Q^\text{sca}
\end{bmatrix} = \frac{1}{r^2} \begin{bmatrix}
Z_{11} & Z_{12} \\
Z_{21} & Z_{22}
\end{bmatrix} \begin{bmatrix}
I^\text{inc} \\
Q^\text{inc}
\end{bmatrix}
\]

(2.42)

and

\[
\begin{aligned}
I^\text{sca} &= I^\text{sca}_\parallel + I^\text{sca}_\perp \\
Q^\text{sca} &= I^\text{sca}_\parallel - I^\text{sca}_\perp
\end{aligned}
\]

(2.43)

Considering that \( Q^\text{inc} = I^\text{inc} \), \( I^\text{sca} = (Z_{11}+Z_{12})I^\text{inc} \) and \( Q^\text{sca} = (Z_{21}+Z_{22})Q^\text{inc} \), the following definition of the depolarization of the light results from Eqs. (2.42) and (2.43):

\[
\delta_\parallel = \frac{I^\text{sca}_\perp}{I^\text{sca}} = \frac{Z_{11}+Z_{12}-Z_{21}-Z_{22}}{2(Z_{11}+Z_{12})}
\]

(2.44)

Similarly, when using a light source polarized perpendicular to the scattering plane, the Stokes vector becomes \([1,-1,0,0]^\top\) so \( Q^\text{inc} = -I^\text{inc} \). Consequently, the depolarization can be defined as:

\[
\delta_\perp = \frac{I^\text{sca}_\perp}{I^\text{sca}} = \frac{Z_{11}-Z_{12}+Z_{21}-Z_{22}}{2(Z_{11}-Z_{12})}
\]

(2.45)

If we consider this definition for a randomly oriented collection of particles, we have \( Z_{12} = Z_{21} \) and the expression given in Eq. (2.53) is the same as the one given in previous
studies where the (parallel) depolarization ratio was defined as the perpendicular intensity over the total (parallel and perpendicular) intensity (Iwasaka et al. 2003, Sakai et al. 2003, Büttner 2004).
3

MODELING STUDIES

3.1 INTRODUCTION TO NUMERICAL SOLUTIONS OF LIGHT SCATTERING

In order to compute the scattered electromagnetic field, all theoretical and numerical techniques are based on solving Maxwell’s equations either analytically or numerically. Exact solutions have traditionally been reduced to trying to solve the vector Helmholtz equation for the time-harmonic electric field. The separation of variables technique has been used in one of the few coordinate systems in which this equation is separable. Unfortunately, this technique results in an analytical solution only for a few simple cases. Independently, Lorenz (1890), Love (1899), Mie (1908) and Debye (1909) derived the solution for an isotropic homogeneous sphere. This solution has been extended to concentric multilayered spheres (Wait 1963, Mikulski and Murphy 1963, Bhandari 1985), radially inhomogeneous spheres (Wyatt 1962) and optically active (chiral) spheres (Bohren 1974). A solution for electromagnetic scattering by a homogeneous isotropic infinite circular cylinder was found by Wait (1955). An extension to optically active cylinders was made by Bohren (1978). Finally, Oguchi (1973) and Asano and Yamamoto (1975) derived a general solution for homogeneous isotropic spheroids. However, the solution for the simplest finite non-spherical particles, spheroids is already so complex that it behaves like a numerical solution. Some exact numerical solutions often behave
like an analytical solution by means of expanding the incident and scattered fields in a set of orthogonal eigenfunctions with convenient properties.

The scattering properties of homogeneous or layered spheres can be easily computed through the Lorenz-Mie theory, using one of the efficient computer algorithms (Wiscombe 1980, Bohren and Huffmann 1983). The main advantage of this theory is to treat non-spherical particles as if they were spheres to which Lorenz-Mie results are applicable. However, the assumption of sphericity is not reliable in atmospheric applications (see chap. 2): spherically symmetric particles are not capable of producing such optical phenomena as halos, pillars, and zenith-enhanced lidar backscattering as observed for ice crystals (Greenler 1990, Platt 1977). Concerning modeling studies of non-spherical particles, previous works and studies about their light scattering properties were pioneered in the 1970s (Oguchi 1973, Asano and Yamamoto 1975, Purcell and Pennypacker 1973). Light scattering by spheroidal particles were performed since the end of the 1970s (Asano 1979, Asano and Sato 1980). These studies were only performed for a collection of randomly oriented particles (Asano 1979, Muinonen et al. 1989). Later, investigations of scattering properties of hexagonal ice crystals using the ray tracing technique were also made where the particle size relative to the wavelength is large (Takano and Jayaweera 1985), Cai and Liou 1982, Rockwitz 1989).

3.2 COMPUTATIONAL TECHNIQUES

3.2.1 Spherical particles

Since the advent of computers, Mie calculations have been implemented in various computer languages. Mie calculations require difficult computations of many special functions that are not readily available in most computer languages. Secondly, the calculations have to calculate slowly converging series before a satisfactory result can be reached. Many researchers in the past few years made improvements in computational algorithms (Infeld 1947, van de Hulst 1957, Dave 1969, Lentz 1976, Wiscombe 1980).

The quantities required for the actual formulas of Mie scattering are given by the following relations (Infeld 1947, Kerker 1969):
\[ S_{11}(\eta) = \sum_{n=1}^{N} \frac{2n+1}{n(n+1)} \left[ a_n \pi_n(\eta) + b_n \tau_n(\eta) \right] \]  

(3.1)

\[ S_{22}(\eta) = \sum_{n=1}^{N} \frac{2n+1}{n(n+1)} \left[ a_n \tau_n(\eta) + b_n \pi_n(\eta) \right] \]  

(3.2)

\[ S_{11} \text{ and } S_{22} \text{ are the scattering amplitude functions. } a_n \text{ and } b_n \text{ are the Mie coefficients, and } \pi_n \text{ and } \tau_n \text{ are the angular eigenfunctions. } a_n, \ b_n, \ \pi_n \text{ and } \tau_n \text{ are expressed as (Infeld 1947):} \]

\[ a_n = \frac{\psi_n'(mx)\psi_n(x) - m\psi_n(mx)\psi_n'(x)}{\psi_n'(mx)\zeta_n(x) - m\psi_n(mx)\zeta_n'(x)} \]  

(3.3)

\[ b_n = \frac{m\psi_n'(mx)\psi_n(x) - \psi_n(mx)\psi_n'(x)}{m\psi_n'(mx)\zeta_n(x) - \psi_n(mx)\zeta_n'(x)} \]  

(3.4)

\[ \pi_n(\eta) = P_n^\prime(\eta) \]  

(3.5)

\[ \tau_n(\eta) = \eta \pi_n(\eta) - (1 - \eta^2) \pi_n'(\eta) \]  

(3.6)

Where \( P_n \) is a Legendre polynomial and \( \eta = \cos(\Theta) \). \( x = \pi d/\lambda \) is the size parameter where \( d \) is the particle diameter and \( \lambda \) the wavelength. \( \psi_n(x) \) and \( \zeta_n(x) \) are the Ricatti-Bessel functions given by:

\[ \psi_n(x) = x j_n(x) \]  

(3.7)

\[ \zeta_n(x) = x(j_n(x) - iy_n(x)) \]  

(3.8)

Where \( m \) is the relative refractive index of the sphere and is equal to \( n_{\text{sph}} / n_{\text{med}} \) (with \( n_{\text{sph}} \) the complex refractive index of the sphere and \( n_{\text{med}} \) that of the surrounding medium. \( j_n(x) \) and \( y_n(x) \) are the real-valued spherical Bessel functions of the first kind and the second kind, respectively (Mishchenko et al. 2002):
The challenge is to determine the Bessel functions for Mie scattering and weaknesses can appear in the complex Bessel function algorithms that can lead to large errors. Two potential pitfalls can occur at that level. Due to non-convergence, calculation of $\psi_n(mx)$ can first overflow if $mx$ has an imaginary part bigger than 100 (single precision) or 720 (double precision) (Du 2004). Second, if the order of $\psi_n(x)$ is bigger than $|x|$. Nowadays, ultra high precision Mie codes can overcome these difficulties.

### 3.2.2 Non-spherical particles

The numerical techniques for computing electromagnetic scattering by non-spherical particles are very numerous. Many of them have been rederived several times under different names and these methods can basically be classified into two categories. First, the differential equation methods compute the scattering field by solving the vector wave equation either in the frequency, or in the time domain. Second, integral equation techniques are based on the surface or volume integral counterparts of Maxwell’s equations. There are some exceptions known as hybrid methods that can be derived using different approaches. Most numerical techniques require knowledge of ensemble-averaged quantities such as the optical cross sections and scattering matrix elements, whereas others calculate the scattered electric field for a single particle in a fixed orientation. Although some traditional versions of many techniques are applicable only to homogeneous, isotropic, optically inactive particles, several extensions to anisotropic and inhomogeneous scatterers exist. In the rest of this section, several methods that have extensive practical applications will be described. Particles will be characterized with the following parameters: a) The size parameter $x = 2d_{eq} / \lambda$, where $d_{eq}$ is the diameter of the surface- or volume-equivalent sphere (volume-equivalent is always used in this study) and b) The aspect ratio $\Gamma$, which expresses the degree of the particle non-
sphericity. The definition of $\Gamma$ is given in Fig. 3-1 for several types of non-spherical particles:

![Fig. 3-1: Illustration of a prolate spheroid (a), oblate spheroid (b), circular cylinder (c) and hexagonal cylinder (d). $\Gamma$ is the aspect ratio, $a$ is the equatorial axis length, $b$ the polar axis length, $d$ the cylinder diameter and $h$ its height.]

A) Separation of variables method for spheroids

Oguchi (1973) and Asano and Yamamoto (1975) pioneered the separation of variables method (SVM) for single homogeneous, isotropic spheroids. This method was then significantly improved by Voshchinnikov and Farafonov (1993). The principle is to solve the electromagnetic scattering problem for an oblate or a prolate spheroid in the respective spheroidal coordinate system. It is based on expanding the incident, internal, and scattered fields in vector spheroidal wave functions (VSWFs). As these functions are not orthogonal to the spheroidal surface, this procedure results in an infinite set of linear algebraic equations for the unknown expansion coefficients (internal and scattered fields). Hence, the resolution has to be done numerically.

The main advantage of SVM is that it can produce very accurate results. However, the system of linear equations becomes large and ill conditioned for spheroids significantly larger than the wavelength and/or for large refractive indices. Furthermore, the computation of vector spheroidal wave functions is a tricky mathematical and numerical problem, especially for absorbing particles. The main limitation of this method is that it is applicable only to spheroidal particles.
### B) Finite element method

The finite element method (FEM) is a differential equation method that computes the scattered time-harmonic electric field. It solves numerically the Helmholtz equation subject to boundary conditions at the particle surface (Morgan and Mei 1979, Silvester and Ferrari 1996). The particle is embedded in a finite computational domain that is discretized into many small-volume cells called *elements* (about 10 to 20 elements per wavelength). The unknown field values are specified at the nodes of these elements. The differential equation is converted into a matrix equation through the boundary conditions for these unknown node field values. Electric fields at the nodes interact only with their neighbors and the resulting matrix equation is sparse and banded. Thus, it significantly reduces the computational effort. Although electromagnetic scattering in the far-field zone is an open-space problem, the FEM is always implemented in a finite computational domain to limit the number of unknowns to a size that can be accommodated by the computer memory. Therefore, approximate absorbing boundary conditions have to be imposed at the outer boundary of the computational domain.

Important advantages of the FEM are that it permits the simulation of arbitrarily shaped and inhomogeneous particles. This method is also simple in concept and execution. However, FEM computations are spread over the entire computational domain rather than confined to the scatterer itself. As a result, these computations can be time consuming and limited to size parameters less than about 10. The finite spatial discretization and the approximate absorbing boundary conditions make FEM poorly suitable for achieving high numerical accuracy.

### C) Finite difference time domain method

The finite difference time domain method (FDTDM), as well as the FEM, calculates the electromagnetic scattering in the time domain by directly solving Maxwell's time-dependent curl equations (Yee 1966, Kunz and Luebbers 1993, Taflove 1998). The space and time derivatives of the electric and magnetic fields are approximated using a finite difference scheme with space and time discretizations. These discretizations are selected in a way that they limit computational errors and ensure numerical stability of the
algorithm. In the same way as the FEM, the scattering particle is embedded in a finite computational domain and absorbing boundary conditions are used to model scattering in the open space. The fields are specified at spatial grid points with discretization density similar to that needed for the FEM. The system of equation updates the fields explicitly as the grid values at the previous and current time steps are used to calculate the values at the next time step. Thus, no large system of linear equations needs to be solved. The memory storage requirement is proportional to the total number of grid points. The FDTDM has become recently popular because of its conceptual simplicity and ease of implementation.

D) Point matching method

The point matching method (PMM) is a differential equation technique. In this method, the incident and internal fields are expanded in vector spherical wave functions regular at the origin. The scattered field outside the scatterer is expanded in outgoing VSWFs. The unknown expansions coefficients of the internal and scattered fields are found by truncating the expansions to a finite size and matching the fields at the surface of the scatterer with the use of boundary conditions. In the simple version of the method, the fields are matched at as many points on the surface as there exist unknown expansion coefficients (Oguchi 1973). However, the validity of the PMM depends on the applicability of the Rayleigh hypothesis (e.g. Lewin 1970, Millar 1973, Bates 1975). The generalized point matching method (GPMM) ameliorates this problem by forming an overdetermined system of equations for the unknown expansion coefficients. This is done by matching the fields in the least-squares sense at a significantly greater number of surface points than the number of unknowns (Morrison and Cross 1974, Oguchi and Hosoya 1974, Al–Rizzo and Tranquilla 1995a).

E) Volume integral equation method

Here, the scattering of a plane electromagnetic wave by an object of volume $V$ can be described by an integral equation where the internal electric field is unknown (Shifrin 1951, van Bladel 1961). The internal electric field can be physically represented at each
point of the volume $V$ as a sum of the incident field and the field induced by sources at all interior points. The interior region is discretized into $N$ small cubic cells with about 10 to 20 cells per wavelength. The field within each cell is assumed to be constant. This procedure is repeated for each of the $N$ unknown internal field values and results in a matrix equation that must be solved numerically. Once the internal field is obtained, the external field can be found. Finally, the scattered field is computed by subtracting the incident field from the external one. This version of the volume integral equation method (VIEM) is known as the method of moments (MOM) (Harrington 1968).

The major advantages of volume integral equation methods are that they satisfy the radiation condition at infinity and are confined to the scatterer itself. Thus, there have fewer unknowns than the differential equation methods. VIEMs can be applied to inhomogeneous, anisotropic and optically active scatterers (e.g. Su 1989). The main inconveniences are a low computational accuracy and slow improvement with increasing $N$, as well as the fast central processing unit time growth with increasing size parameter. Several modifications of the method of moments have been developed under different names, such as the discrete dipole approximation that will be discussed later. The main difference between these techniques is the way in which they treat the self-interaction term.

\( F) \quad \textit{Discrete Dipole Approximation} \)

The former concept of the discrete dipole approximation (DDA) is based on partitioning a particle into a number $N$ of elementary polarizable units called dipoles (Purcell and Pennypacker 1973). The electromagnetic response of the dipoles to the local electric field is assumed to be known. The field that excites a dipole is a superposition of the external field and the fields scattered by all other dipoles. Consequently, it is possible to write a system of $N$ linear equations for $N$ fields exciting the $N$ dipoles. The numerical solution of this system is used to compute $N$ partial fields scattered by the dipoles and the total scattered field.

The biggest advantage of the DDA method is to be applicable to arbitrarily shaped, inhomogeneous, and anisotropic particles. On the other hand, limited numerical
Modeling studies

accuracy, especially for scattering matrix elements, is one of the major disadvantages. There is also slow convergence of results with increasing $N$ and the need to repeat the entire calculation for each new direction of incidence (Singham 1989, Draine and Flatau 1994, Okamoto et al. 1995, Liu and Illingworth 1997). These factors have made DDA computational time consuming, especially for particle size and/or orientation distributions, and have limited the particle size parameter to relatively small values. Finally, the last method (T-matrix) will be discussed more in details in the next section (3.2.3).

3.2.3 The T-matrix method

The T-matrix method (TMM) was initially introduced by Waterman (1965, 1971) as a technique for computing electromagnetic scattering by single, homogeneous, non-spherical particles based on the Huygens principle. This principle was formerly known as the extended boundary condition, the null-field method, and the Schelkunoff equivalent current method. The T-matrix approach has proven to be extremely powerful by relating expansions of incident and scattered waves in vector spherical wave functions. It has drastically expanded its realm and it now includes electromagnetic, acoustic, and elastodynamic wave scattering by single and compound scatterers, multiple scattering in discrete random media, and scattering by gratings and periodically rough surfaces (Varadan and Varadan 1980, Tsang et al. 1985, Varadan et al. 1988).

Nowadays, the T-matrix method is known to be a powerful tool for computing electromagnetic scattering by single, homogeneous non-spherical particles (Mishchenko et al. 2000) as well as for spheroids, cylinders and Chebyshev particles of different shape parameters (Mishchenko and Sassen 1998). Benchmark numbers for particles in fixed and random orientations were reported by Mishchenko (1991a), Kuik et al. (1992), Mishchenko et al. (1996a), Mishchenko and Mackovski (1996), Hovenier et al. (1996), and Wielandt et al. (1997). They cover a range of equivalent-sphere size parameters from a few units up to 60. Computations for homogeneous and layered spheroids, finite circular cylinders, Chebyshev particles and two-sphere clusters in random orientation were reported and analyzed by Mugnai and Wiscombe (1980, 1986, 1989), Wiscombe and
Mugnai (1986, 1988), Kuik et al. (1994), Mishchenko and Travis (1994b, 1994c), Mishchenko and Hovenier (1995), Mishchenko et al. (1995, 1996a, 1996b) and Quirantes (1999). This method was also extended to non-axisymmetric particles such as hexagonal columns (Havemann and Baran 2001, Baran and Havemann 2001, Havemann et al. 2003, Kahnert 2003), superellipsoids, rounded cubes and realistically shaped particles (Wriedt 2002). It was shown to be more efficient and accurate than other frequently used techniques, and a greater range of size parameters can be computed. Moreover, it compares favorably with other frequently used techniques in terms of efficiency and accuracy in many applications. It is the only method that has been used in systematic surveys of non-spherical scattering based on calculations for thousands of randomly oriented particles. First, the general concept of the T-matrix will be introduced. Second, a focus will be made on computation for single particles.

A) The T-matrix approach

Considering the scattering of a plane electromagnetic wave by a single particle (Eq. (2.26)), the incident and scattered fields are expended in vector spherical wave functions as follows:

\[
E^{\text{inc}}(r) = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} [a_{mn} R_g M_{mn}(k r) + b_{mn} R_g N_{mn}(k r)]
\]

(3.11)

\[
E^{\text{sca}}(r) = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} [p_{mn} M_{mn}(k r) + q_{mn} N_{mn}(k r)], \quad r > r_>
\]

(3.12)

Where \(k\) is the wavenumber and \(\lambda\) the wavelength in the surrounding medium. \(r_>\) is the radius of the smallest circumscribing sphere of the scattering particle centered at the origin of the coordinate system (Fig. 3-2):
The functions $R_g M_{mn}$ and $R_g N_{mn}$ are regular (finite) at the origin, while the use of the outgoing functions $M_{mn}$ and $N_{mn}$ in Eq. (3.12) ensures that the scattered field satisfies the so-called radiation condition at infinity (i.e., the transverse component of the scattered electric field decays as $1/r$, whereas the radial component decays faster than $1/r$ with $r \to \infty$). The requirement $r > r_\ast$ in (3.12) means that the scattered field is considered only outside the smallest circumscribing sphere of the scatterer. The Rayleigh hypothesis (e.g. Bates 1975, Paulick 1990) assumes that the scattered field can be expanded in outgoing wave functions not only in the outside region but also in the region between the particle surface and the circumscribing sphere (Fig. 3-2). Because the validity range of this hypothesis is poorly known and is in fact questionable, the requirement $r > r_\ast$ in Eq. (3.12) is important in order to make sure that the Rayleigh hypothesis is not implicitly invoked (Lewin 1970). The vector spherical wave functions $M_{mn}$ and $N_{mn}$ are expressed as:

\[
M_{mn}(kr) = \frac{1}{i^m} d^m_n h_n^{(1)}(kr) C_{mn}(\theta) \exp(i m \phi) \tag{3.13}
\]

\[
N_{mn}(kr) = \frac{1}{i^m} d^m_n \left\{ \frac{n(n+1)}{kr} h_n^{(1)}(kr) P_{mn}(\theta) + \frac{1}{kr} \left[ kr h_n^{(1)}(kr) \right] B_{mn}(\theta) \right\} \exp(i m \phi) \tag{3.14}
\]

\[
B_{mn}(\theta) = \frac{1}{\sin \theta} \frac{d}{d \theta} d^n_{m0}(\theta) + \frac{i m}{\sin \theta} d^n_{0m}(\theta) \tag{3.15}
\]
\[
C_{mn}(\theta) = \frac{im}{\sin \theta} d^n_{0m}(\theta) - \frac{d}{d\theta} d^n_{0m}(\theta)
\]  
(3.16)

\[
P_{mn}(\theta) = r \frac{d^n_{0m}(\theta)}{r}
\]  
(3.17)

\[
d_n = \sqrt{\frac{2n+1}{4\pi(n+1)}}
\]  
(3.18)

\[d^n_{im}(\theta)\] are Wigner \(d\) functions defined by Varshalovitch et al. (1988). The expansion coefficients of the plane incident wave are given by the following analytical formulas (Tsang et al. 1985):

\[
a_{mn} = 4\pi(-1)^m i^n d_n C^*_{mn}(\theta^{\text{inc}}) E_0^{\text{inc}} \exp(-im\varphi^{\text{inc}})
\]  
(3.19)

\[
b_{mn} = 4\pi(-1)^m i^{n-1} d_n B^*_{mn}(\theta^{\text{inc}}) E_0^{\text{inc}} \exp(-im\varphi^{\text{inc}})
\]  
(3.20)

Where the asterisk indicates the complex conjugation. Owing to the linearity of Maxwell’s equations and the boundary conditions, the relation between the scattered field coefficients \(p_{mn}\) and \(q_{mn}\) on one hand, and the incident field coefficients \(a_{mn}\) and \(b_{mn}\) on the other hand must be linear. This relation is expressed by a transition matrix (or T-matrix) \(T\) as follows (Watermann 1971, Tsang et al. 1985):

\[
p_{mn} = \sum_{n'=-\infty}^{\infty} \sum_{m'=-\infty}^{\infty} \left[ T^{11}_{nn'm'n'} a_{m'n'} + T^{12}_{nn'm'n'} b_{m'n'} \right]
\]  
(3.21)

\[
q_{mn} = \sum_{n'=-\infty}^{\infty} \sum_{m'=-\infty}^{\infty} \left[ T^{21}_{nn'm'n'} a_{m'n'} + T^{22}_{nn'm'n'} b_{m'n'} \right]
\]  
(3.22)

In matrix notation, Eqs. (3.21) and (3.22) can be rewritten as:
\[
\begin{bmatrix}
p \\
q
\end{bmatrix} = T \begin{bmatrix}
a \\
b
\end{bmatrix} = \begin{bmatrix}
T^{11} & T^{12} \\
T^{21} & T^{22}
\end{bmatrix} \begin{bmatrix}
a \\
b
\end{bmatrix}
\]

(3.23)

and it forms the basis of the T-matrix approach. If \( T \) is known for a given scatterer, Eqs. (3.19), (3.20, (3.21), (3.22) and (3.12) give the scattered field and, as a consequence, the amplitude matrix appearing in Eq. (2.37). Using the large argument asymptotic for spherical Hankel functions:

\[
h_n^{(1)}(kr) \approx \frac{(-1)^{n+\frac{1}{2}} \exp(ikr)}{kr}, \quad kr \gg n^2
\]

(3.24)

We obtain the following expression for the amplitude matrix in dyadic notation:

\[
S(\hat{n}^{\text{sca}}, \hat{n}^{\text{inc}}) = \frac{4\pi}{k} \sum_{n,m} \int \cos^{n-1}(\theta) d\theta \left\{ i^n \exp[i(m\phi^{\text{sca}} - m'\phi^{\text{inc}})] \right\} x
\]

(3.25)

\[
\begin{bmatrix}
T^{11} & T^{12} \\
T^{21} & T^{22}
\end{bmatrix} \begin{bmatrix}
C_{mn}(\theta^{\text{sca}}) + iB_{mn}(\theta^{\text{sca}}) \\
C^{*}_{m'n'}(\theta^{\text{inc}})
\end{bmatrix}
\]

\[
\begin{bmatrix}
T^{12} & T^{22}
\end{bmatrix} \begin{bmatrix}
C_{mn}(\theta^{\text{sca}}) + iB_{mn}(\theta^{\text{sca}}) \\
B^{*}_{m'n'}(\theta^{\text{inc}})
\end{bmatrix}
\]

At that point, it is possible to compute any scattering characteristic with knowledge of the amplitude matrix.

A fundamental feature of the T-matrix approach is that it depends only on the physical and geometrical characteristics of the scattering particle (refractive index, size, shape, and orientation). For example, it reduces exactly to the Lorenz-Mie theory when the scattering particle is a homogeneous or layered sphere composed of isotropic materials. This method is completely independent of the incident and scattered fields. Therefore, the T-matrix needs to be computed only once and then it can be used for any direction of incidence and scattering and for any polarization state of the incident field.
B) Computation for single particles

The standard scheme for computing the T-matrix for single homogeneous scatterers in the particle reference frame is called the extended boundary condition method (EBCM). It is based on the vector Huygens principle (Watermann 1971) and the general problem is to find the field scattered by an object bounded by a closed surface $S$ (Fig. 3-1). The Huygens principle establishes the following relationship between the incident field $E^{\text{inc}}(\mathbf{r})$, the total external field $E(\mathbf{r})$, and the surface field on the exterior of $S$:

\begin{align}
E^{\text{inc}}(\mathbf{r}) &= 0 + \int_S E(\mathbf{r} \text{, } \mathbf{r} \text{ outside } S) \tag{3.26} \\
E^{\text{int}}(\mathbf{r}) &= 0 + \int_S E(\mathbf{r} \text{, } \mathbf{r} \text{ inside } S) \tag{3.27}
\end{align}

The integral term involves the unknown surface field on the exterior of $S$. The incident and the scattered waves are expanded in regular and outgoing VSWFs, respectively, according to Eqs. (3.11) and (3.12). The internal field can also be expanded in VSWFs regular at the origin:

\[ E^{\text{int}}(\mathbf{r}) = \sum_{m=1}^{\infty} \sum_{n=-\infty}^{\infty} \left[ c_{mn} R_m^g M_{mn}(mk\mathbf{r}) + d_{mn} R_m^g N_{mn}(mk\mathbf{r}) \right], \quad \mathbf{r} \text{ inside } S \tag{3.28} \]

$m$ is the refractive index of the particle relative to that of the surrounding medium. The surface field on the exterior of $S$ can be expressed in the surface field on the interior of $S$ considering the boundary conditions. The application of Eqs. (3.11), (3.26), (3.27) and (3.28) to points with $r < r_c$ gives the following matrix equation:

\[
\begin{bmatrix}
a \\
b
\end{bmatrix} =
\begin{bmatrix}
Q^{11} & Q^{12} \\
Q^{21} & Q^{22}
\end{bmatrix}
\begin{bmatrix}
c \\
d
\end{bmatrix} \tag{3.29}
\]
The elements of the $Q$-matrix are simple surface integrals of VSWFs products that depend only on the particle size, shape and refractive index. The unknown expansion coefficients of the internal field $c$ and $d$ can be found by inverting this matrix equation, given the known expansion coefficients of the incident field $a$ and $b$. Similarly, a matrix expression for points with $r > r_\ast$ using Eqs. (3.12), (3.26), (3.27) and (3.28) can be found and is given by:

$$\begin{bmatrix} p \\ q \end{bmatrix} = \begin{bmatrix} R_gQ^{11} & R_gQ^{12} \\ R_gQ^{21} & R_gQ^{22} \end{bmatrix} \begin{bmatrix} c \\ d \end{bmatrix}$$

(3.30)

Where the elements of the $R_gQ$ matrix are also given by simple integrals over the particle surface and depend only on the particle characteristics. The comparison between Eqs. (3.23), (3.29) and (3.30) leads to:

$$T = -R_gQ^{-1}$$

(3.31)

Finally, Eq. (3.23) gives the expansion coefficients of the scattered field and, thus, the scattered field itself.

### 3.3 RESULTS

The public domain T-matrix code from *M. I. Mishchenko* (Mishchenko 2000) was used to calculate the depolarization ratio of non-spherical particles in a fixed orientation. The results are published in Nicolet et al. (2007). The major differences as compared to previous studies is that we have not dealt with a collection of randomly oriented particles with a given concentration, but only with single particles that have to be detected as such. These single particles are positioned in a specific orientation so that the scattered light for a given orientation has to be considered. The source code gives the phase and the amplitude matrices depending on the particle type (shape, size, orientation and refractive index), the wavelength of the light source and the direction of the incident and scattered beam.
The scattering angle is 175° ($\theta_{\text{inc}} = 5^\circ, \theta_{\text{sca}} = 180^\circ, \varphi_{\text{inc}} = \varphi_{\text{sca}} = 90^\circ$). Due to technical considerations, it is not possible to align the laser on the optical axis of the detector. The laser beam has to be directed alongside the optical elements in the backward direction (lenses, pinhole and mirror). Ice crystals are considered in this work with $n = 1.319 + 2.61 \cdot 10^{-9}i$ (Warren 1984). They are assumed to be circular cylinders, which is an approximation for hexagonal columns (Baran and Havemann 2001). Using circular columns allows determination of the particle orientation by only two Euler angles instead of three, as they are axisymmetric. This leads to a simplification of the particle orientation definition and to a shorter computation time. However, complex interference effects that manifest themselves for hexagonal particles at fixed orientations may not be resolved. The particle diameters $d$ (equivalent-volume sphere) used in our simulations vary between 0.13 and 4 µm and the aspect ratios $\Gamma = d/h$ (where $d$ is the diameter and $h$ the height of the particle) vary between 0.3 and 3. As the model becomes unstable for particle size bigger than 4 µm, no results can be obtained beyond this limit. Moreover, the computation time for larger size parameters increases dramatically. Due to the axial symmetry, the intervals of the Euler angles necessary to represent all orientations are $\alpha \in [0, \pi/2]$ and $\beta \in [0, \pi]$. The calculations are made in 5° steps for each $\alpha$ and $\beta$.

In order to evaluate if there is a clear dependency of the depolarization ratio on the size of the particles and for better comparison with other literature, all values for each orientation are averaged to have a global mean depolarization ratio. We are taken into account the cylindrical projection of the plots for the averaging process. With $\alpha$ being independent of the area covered by each pair of $(\alpha, \beta)$, each ratio with a given value of $\alpha$ is weighted, considering the area that it covers in reality (see section 3.3.1). As we see, the depolarization ratio for $\beta$ close to 0° and 180° (poles) are weighted less than values for $\beta = 90^\circ$ (equator), which are used for normalization. In that way, the weighting factor $M_\beta$ simply equals the absolute value of the sine of $\beta$. Thus, the averaged value of the depolarization ratio ($\delta_{\text{avg}}$) for all orientations can be determined, using:

$$M_\beta = |\sin(\beta)|, \text{ and}$$

(3.32)
\[ \delta_{\text{avg}} = \frac{1}{\sum_{\beta} \delta(\alpha, \beta) M_{\beta}} \]  

(3.33)

This gives an indication of an average value of \( \delta \) that can be expected as a function of the size of the particle. This definition corresponds to other studies that have been made for randomly oriented particles (Mishchenko and Sassen 1998, Büttner 2004, Zakharova and Mishchenko 2000).

All simulations made with the T-matrix method are summarized in Tables 3-1 and 3-2. Results of the averaged depolarization ratio (parallel and perpendicular) and the averaged relative intensity given by the phase matrix elements \( Z_{11} \) and \( Z_{12} \) are listed here for several sizes and aspect ratio:

<table>
<thead>
<tr>
<th>( \Theta )</th>
<th>( d ) [( \mu \text{m} )]</th>
<th>( \Gamma )</th>
<th>( \delta_{\parallel}^{\text{ave}} ) (( \sigma ))</th>
<th>( \delta_{\perp}^{\text{ave}} ) (( \sigma ))</th>
<th>( Z_{11}+Z_{12} ) (( \sigma ))</th>
<th>S or A</th>
</tr>
</thead>
<tbody>
<tr>
<td>175°</td>
<td>0.13</td>
<td>1.0</td>
<td>0.000 (0.000)</td>
<td>0.000 (0.000)</td>
<td>0.000 (0.0000)</td>
<td>A</td>
</tr>
<tr>
<td>175°</td>
<td>0.52</td>
<td>1.0</td>
<td>0.017 (0.000)</td>
<td>0.018 (0.000)</td>
<td>0.004 (0.0000)</td>
<td>A</td>
</tr>
<tr>
<td>175°</td>
<td>0.8</td>
<td>1.0</td>
<td>0.091 (0.014)</td>
<td>0.093 (0.014)</td>
<td>0.015 (0.0000)</td>
<td>S</td>
</tr>
<tr>
<td>175°</td>
<td>1.0</td>
<td>1.0</td>
<td>0.138 (0.028)</td>
<td>0.135 (0.019)</td>
<td>0.029 (0.0002)</td>
<td>S</td>
</tr>
<tr>
<td>175°</td>
<td>1.2</td>
<td>1.0</td>
<td>0.178 (0.033)</td>
<td>0.184 (0.028)</td>
<td>0.046 (0.0008)</td>
<td>A</td>
</tr>
<tr>
<td>175°</td>
<td>1.6</td>
<td>1.0</td>
<td>0.218 (0.037)</td>
<td>0.226 (0.034)</td>
<td>0.093 (0.0035)</td>
<td>S</td>
</tr>
<tr>
<td>175°</td>
<td>2.0</td>
<td>1.0</td>
<td>0.231 (0.035)</td>
<td>0.254 (0.043)</td>
<td>0.141 (0.0087)</td>
<td>S</td>
</tr>
<tr>
<td>175°</td>
<td>2.5</td>
<td>1.0</td>
<td>0.231 (0.035)</td>
<td>0.243 (0.039)</td>
<td>0.236 (0.0319)</td>
<td>A</td>
</tr>
<tr>
<td>175°</td>
<td>3.0</td>
<td>1.0</td>
<td>0.179 (0.027)</td>
<td>0.195 (0.027)</td>
<td>0.392 (0.0702)</td>
<td>S</td>
</tr>
<tr>
<td>175°</td>
<td>3.5</td>
<td>1.0</td>
<td>0.191 (0.033)</td>
<td>0.180 (0.026)</td>
<td>0.639 (0.2182)</td>
<td>A</td>
</tr>
<tr>
<td>175°</td>
<td>4.0</td>
<td>1.0</td>
<td>0.160 (0.027)</td>
<td>0.182 (0.025)</td>
<td>0.980 (0.7226)</td>
<td>S</td>
</tr>
</tbody>
</table>

Table 3-1: Values of the parallel and perpendicular depolarization ratio \( \delta_{\parallel}^{\text{ave}} \) and \( \delta_{\perp}^{\text{ave}} \) of isometric ice cylinders (\( \Gamma = 1 \)) for different particle diameters. \( Z_{11}+Z_{12} \) shows the relative scattered intensity averaged over all orientations. Depolarization values are either mentioned in the following sections of this chapter (S) or in the appendix A (A). \( \Theta \) is the scattering angle.
Table 3-2: Values of the parallel and perpendicular depolarization ratio $\delta_{\text{avg}}^\| \text{ and } \delta_{\text{avg}}^\perp$ of 2-µm diameter ice cylinders for different particle aspect ratios. $Z_{11}+Z_{12}$ shows the relative scattered intensity averaged over all orientations. Depolarization values are either mentioned in the following sections of this chapter (S) or in the appendix A (A). $\Theta$ is the scattering angle.

These results on the depolarization ratio of single particles in a fixed orientation are discussed in this section where the influence of size and aspect ratio is considered. The simulations marked with “S” are shown more in detail later in this section whereas those marked with “A” are illustrated in Appendix A. Other calculations were done at a perfect backscattering angle ($\Theta = 180^\circ$) to see the difference in the depolarization ratio. These values are also listed in appendix A, as well as results obtained for prolate and oblate spheroids.

### 3.3.1 Depolarization ratio with varying size

Before discussing the results in detail, it is important to note that the contour plots presented in this chapter are not area conservative. Due to the symmetry of the assumed particle model, all orientations are obtained with $\alpha \in [0,\pi/2]$ and $\beta \in [0,\pi]$, this corresponds to a quarter of a sphere. Fig. 3-3 shows a regular cylindrical projection as will be used in the following where $\alpha$ is the longitude and $\beta$ the latitude. Note that the areas near the poles are strongly overemphasized.
Fig. 3-3: Visualization of the regular cylindrical projection. $\alpha$ and $\beta$ are the Euler angles. The areas for small and large values of $\beta$ are actually smaller than the ones for $\beta$ around 90°.

The following result (Fig. 3-4) shows the depolarization ratio $\delta_1$ as a function of size for an aspect ratio of 1. This value was taken because it is the aspect ratio expected in our case, considering that small crystals ($d < 10 \; \mu m$) are thought to grow nearly isometrically ($\Gamma = 1$) (Young 1993). This assumption is based on measurements with a limited resolution but when new instruments that can detect small crystals become available, this aspect may be redefined.
Different aspects can be noticed in Fig. 3-4. First, all values of the depolarization ratio i.e. from 0 (no depolarization) to 1 (totally reversed depolarization) may occur for the same particle but in different orientations. That means that even a non-spherical particle can behave like a spherical particle in terms of light scattering from a linearly polarized light source, but only for specific orientations. On the other hand, a
depolarization value close to 1 can be observed in other orientations, so that the particle can almost totally reverse the polarization. This is a unique feature of single particle detection as a collection of randomly oriented particle never produces depolarization ratios higher than 0.41 (upper value for a prolate spheroid with $\Gamma = 1.2$) (Mishchenko and Sassen 1998). Furthermore, a more irregular structure in the distribution of $\delta_\parallel$ can be observed with increasing particle size. The regions with high depolarization ratios become more numerous but are smaller. Many alternations between areas of high and low $\delta_\parallel$ occur especially as a function of $\beta$. This can be explained by the fact that varying $\beta$ leads to a much stronger change in the angle of incidence between the incoming light and the particle while varying $\alpha$ introduces a rotation of the incident plane of polarization rather than the angle of incidence. Measurements made at $\theta^{\text{inc}} = 0^\circ$ (perfect backscattered light: $\Theta = 180^\circ$) showed an axisymmetrical distribution at $\beta = 90^\circ$. In this case the $\hat{n}^{\text{inc}}$ vector pointing in the direction of the incident beam has the same direction as the $z$ axis. For $\beta = 90^\circ$ the $z'$ axis is perpendicular to $\hat{n}^{\text{inc}}$ and the $\hat{n}^{\text{aca}}$ vector (direction of the scattered light), hence the symmetry at this angle. An example is given in Fig. 3-5. Like in the 175$^\circ$-scattering angle, the scattering characteristics are more strongly modified by varying $\beta$, causing large depolarization differences. Therefore, a small tilt or rotation of a large particle can induce a large difference in the depolarization ratio.

![Fig. 3-5: Perpendicular depolarization ratio ($\delta_\perp$) as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$ for cylinders of diameter $d = 2 \ \mu m$ and aspect ratio $\Gamma' = 1$. The areas of the figures are non-conservative (see text for details).](image)
On the other hand, basically all simulations exhibit the same large areas with low depolarization for values of $\alpha$ close to 90° and $\beta$ close to 0° and 180°. Fig. 3-6 shows the same representation, but for a light source with polarization perpendicular to the scattering plane ($\delta_\perp$):

Fig. 3-6: Perpendicular depolarization ratio ($\delta_\perp$) for cylinders with an aspect ratio $\Gamma = 1$. The panels represent $\delta_\perp$ for different particle diameters as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$. The areas of the figures are non-conservative (see text for details).
Concerning the depolarization ratio $\delta_\perp$, the same considerations as for $\delta_\parallel$ are also valid in this case, given the similar patterns.

### 3.3.2 Depolarization ratio with varying aspect ratio

In this section, we discuss the evolution of the depolarization ratios for different aspect ratios with a fixed particle diameter of 2 µm. This corresponds to the lower size limit that we expect for the ice crystals grown in the ZINC chamber (Stetzer et al. 2007). The simulations made for aspect ratios from 0.3 to 3 are shown in Fig. 3-7:
Fig. 3-7: Perpendicular depolarization ratio for particles with \(d = 2\ \mu m\) for different aspects ratios as a function of the orientation of the particle given by the Euler angles \(\alpha\) and \(\beta\). The areas of the figures are non-conservative (see text for details).

For the same particle size, the structure is similar for the different depolarization ratios considered (\(\delta_{||}\) and \(\delta_{\perp}\)). This is logical as this structure is size dependent (cf. Figs. 3-4 and 3-6). On the other hand, the variation of the aspect ratio does not seem to induce a major modification in this structure where low values alternate with high ones quite rapidly. This shows that \(\delta\) is mostly influenced by the orientation but not by the aspect ratio. Moreover, the same typical areas of non-depolarizing orientations appear as in Fig. 3-4.
and 3-6. Globally, the appearance of the distribution is more or less the same, except for columnlike particles ($\Gamma = 0.3$) where a more extended region of high depolarization occurs at $\beta$ between $60^\circ$ and $120^\circ$ for $\alpha$ between $30^\circ$ and $80^\circ$.

### 3.3.3 Mean depolarization ratio versus particle size and aspect ratio

All values obtained for the averaged depolarization ratio listed in tables 3-1 and 3-2 are discussed in the following section. The next figure (Fig. 3-8) illustrates the occurrence frequency of $\delta^\perp_{\text{avg}}$ values obtained for four different types of particles. This representation is made with respect of the weighting function expressed above that describes the non-conservative area patterns. This shows the fraction of ice particles that can be detected considering a given depolarization limit $\delta_{\text{lim}}$. This defines the values above which a particle can be detected as a depolarizing one (i.e. an ice crystal). $\delta_{\text{lim}}$ depends on the background depolarization of molecules and other particles in the sampled volume as well as the detectable intensity of the perpendicular channel.

Fig. 3-8: Occurrence of the depolarization ratio $\delta^\perp_{\text{avg}}$ considering an isometric ($\Gamma = 1$) ice crystal with diameter of 2 µm (upper panel, left) and 1 µm (upper panel, right), a column ice crystal ($\Gamma = 0.3$, lower panel, left), and a plate ice crystal ($\Gamma = 2$, lower panel, right), both with a diameter of 2 µm.
Between 35% and 60% of all cases produce a depolarization ratio ≤ 0.05 corresponding to the limit of detectable depolarization (\(\delta_{lim}\)). The occurrence shows a decreasing trend with increasing \(\delta_{\perp}\). There is also a large difference in the non-detectable fraction (\(\delta < \delta_{lim}\)) for different sizes (see the top right and top left panels). \(\delta_{lim}\) determines the number of occurrences below the detection limit. On the other hand, the aspect ratio does not change \(\delta\) significantly. A decrease in the detection limit induces an increase in the relative occurrence of detectable ice particles (\(\delta > \delta_{lim}\)). Table 3-3 illustrates the fraction of particles that generate a depolarization ratio between 0 and \(\delta_{lim}\) for the 4 types previously exposed in Fig. 3-8.

<table>
<thead>
<tr>
<th>(\delta_{lim})</th>
<th>(\Gamma = 1, d = 2 \mu m)</th>
<th>(\Gamma = 1, d = 1 \mu m)</th>
<th>(\Gamma = 0.3, d = 2 \mu m)</th>
<th>(\Gamma = 2, d = 2 \mu m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.02</td>
<td>0.204</td>
<td>0.417</td>
<td>0.261</td>
<td>0.317</td>
</tr>
<tr>
<td>0.03</td>
<td>0.246</td>
<td>0.486</td>
<td>0.299</td>
<td>0.363</td>
</tr>
<tr>
<td>0.04</td>
<td>0.290</td>
<td>0.547</td>
<td>0.332</td>
<td>0.392</td>
</tr>
<tr>
<td>0.05</td>
<td>0.349</td>
<td>0.600</td>
<td>0.363</td>
<td>0.417</td>
</tr>
</tbody>
</table>

Table 3-3: Relative occurrence of particles with a depolarization ratio ≤ \(\delta_{lim}\) as a function of size, aspect ratio, and \(\delta_{lim}\).

With a detection limit of 0.02, the frequency of non-detectable ice crystals can be reduced to almost 20% for a 2-\(\mu m\) diameter isometric cylinder. However, considering a diameter of 1 \(\mu m\) with the same aspect ratio leads to a pretty poor result, even with \(\delta_{lim} = 0.02\) where the relative occurrence of non-detectable ice crystals remains above 40%. This case represents a lower size limit as we expect particle sizes larger than 1 \(\mu m\) in the ZINC chamber. Fig. 3-9 shows the evolution of \(\delta_{avg}\) with size for isometric cylinders (\(\Gamma = 1\)) as compared to the application of depolarization in contrails (Mishchenko and Sassen 1998). The vertical bars refer to the ± variance.
A positive correlation between the size and the depolarization ratio can be noticed up to a diameter of 2 µm, which corresponds to a size parameter $x = \pi d/\lambda$ of 15.4 for our laser wavelength of 407 nm. For larger sizes, there is only a slight decrease of the depolarization ratio with increasing size. As the particle diameter becomes much larger than the wavelength, the Mie regime influence is decreasing with increasing particle size. This regime where the particle diameter is close to the wavelength defines a transition domain: There is no clear dependence of particle size on the scattered light intensity and it results to oscillations in intensity with increasing particle size (Penndorf 1962). The depolarization ratio is then no longer dependent on the size. In addition, geometric optics (GO) predicts size-parameter independent depolarization ratios for non-absorbing particles, as considered here (Mishchenko and Hovenier 1995). Also, multiple internal reflections in very large particles are not the only mechanism of producing depolarization and are not necessarily the ones producing maximal $\delta$ values (Liou and Lahore 1974). Mishchenko and Sassen (1998) present similar trends when the depolarization is plotted against the size parameter for perfect backscattering angle ($\Theta = 180^\circ$) at $\lambda = 532$ nm (Mishchenko and Sassen 1998). For the same scattering angle $\Theta$, ice crystals produce more depolarization using a shorter wavelength but the difference tends to decrease for high size parameters (from $x > 15$). In all cases, it is no longer
possible to obtain a detectable depolarization value for particle size parameters smaller than 1. Considering a wavelength of 407 nm, higher values are produced at $\Theta = 180^\circ$ than $175^\circ$ where differences up to 0.05 occurred for size parameters higher than 15. In that case, the variation between $\bar{\delta}_\parallel$ and $\bar{\delta}_\perp$ are negligible, so only the former is shown in Fig. 3-9. In addition, the differences regarding data of Mishchenko and Sassen (1998) can also be explained by the discrete integration over all orientations that was done in our study. Due to the model stability, it was not possible to show results for sizes larger than 4 $\mu$m ($x > 30$).

Concerning the influence of the aspect ratio, Fig. 3-10 shows that this factor does not play a large role in the mean depolarization ratio $\bar{\delta}$ for a given size (here $d = 2 \mu$m). Only needles and oblate particles with $\Gamma = 2$ have slightly higher depolarization ratios than more compact particles. In both cases, $\delta_\parallel$ and $\delta_\perp$ are very close which confirms that the polarization orientation of the light beam is not playing a large role either. This was also observed by Mishchenko and Sassen (1998) despite their values being generally higher and a weak increase occurring at higher aspect ratios.

![Fig. 3-10: Evolution of the mean depolarization ratio with increasing aspect ratio. The black triangles show the results of Mishchenko and Sassen (1998) for randomly oriented cylinders with $\Gamma = 1$ and $\lambda = 532$ nm. As they used the typical lidar definition of the depolarization $\delta_{Lid} = I_\perp/I_\parallel$, the values are converted into $\bar{\delta}$ using $\bar{\delta} = \delta_{Lid}/(1+\delta_{Lid})$. The vertical bars refer to the $\pm$ variance.](image-url)
4

THE ICE OPTICAL DETECTOR DEVICE

4.1 DESCRIPTION OF THE ICE OPTICAL DETECTOR DEVICE (IODE)

The optical system built to detect ice crystals and water droplets at the bottom of the ZINC chamber is composed of several elements that will be described separately in the next sections. After being illuminated by a polarized laser light source, the light scattered from the particles is collected with an optical system composed of two lenses and a pinhole. The two polarization components are then split with a polarization prism and the light intensity is measured by a photomultiplier for each channel. The signal is finally analyzed after amplification and a software retrieves the peaks as well as the depolarization ratios.

In the ZINC chamber, the particles fall down in a laminar flow. Turbulence does not have to be taken into account. As the particles may be randomly distributed over the width of the chamber, they have to be detected at any location in the detection region. The overlap region (area resulting from the intersection between the laser beam and the detector field of view i.e. the area where the particles can be detected) must cover the whole width. Coated windows are mounted on both apertures to keep the chamber tight
during measurements in order to avoid contamination. Therefore, all optical components are located outside the chamber. The scattering plane is vertical in this configuration and the polarization can be easily switched in parallel or perpendicular position to the cited plane in order to get $\delta_\parallel$ or $\delta_\perp$, respectively. The whole detection system is shown in Fig. 4-1. All elements except the preamplifier units are housed inside a black box and are thus shielded from the parasitic natural light.

![Fig. 4-1: Schematic of the IODE instrument setup, coupled with the ZINC chamber.](image)

4.1.1 The laser

In our experimental setup, the laser light source is a linearly polarized (100:1) 50 mW GaN diode laser operating at 407 nm (Oxxius OXV-405). It offers high power stability and its low-noise characteristics make it ideal for sensing applications. In addition, its compactness allows for easy integration. The output power can be adjusted manually or modulated through analog and TTL controls. Collimation is already included in the
laser so that the beam divergence remains at 0.6 mrad. The laser ray is tilted downwards with an angle of 5° with respect to the horizontal plane to have a scattering angle $\Theta$ of 175°. The beam (Gaussian type) diameter has an elliptical shape. The specifications of the laser are listed in Table 4-1.

<table>
<thead>
<tr>
<th>Wavelength *1</th>
<th>407.0 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>CW Power *1</td>
<td>51 mW</td>
</tr>
<tr>
<td>Power stability *1</td>
<td>± 0.25%</td>
</tr>
<tr>
<td>Optical Noise *1</td>
<td>± 0.3%</td>
</tr>
<tr>
<td>Beam diameter (elliptical)</td>
<td>1 x 2.5 mm</td>
</tr>
<tr>
<td>Beam divergence *1</td>
<td>3.6 mm</td>
</tr>
<tr>
<td>Polarization ratio</td>
<td>100:1</td>
</tr>
<tr>
<td>Dimensions:</td>
<td></td>
</tr>
<tr>
<td>Length</td>
<td>156 mm</td>
</tr>
<tr>
<td>Diameter</td>
<td>34 mm</td>
</tr>
</tbody>
</table>

*1 from manufacturing test report

Table 4-1: Specifications of the laser Oxxius OXV-405 used for the IODE detector device.

4.1.2 Optical elements

The two side-apertures of the ZINC chamber are closed with specially designed windows to keep it tight. A special antireflection and waterproof coating (AR) is applied on every window to increase their transmittance up to 99.6%. Each window has a diameter of 10 mm and a thickness of 6 mm. On each side, two windows are mounted on KF flanges: one window for the laser beam entrance (backscatter side) and exit (forward side) and the other two for the detectors of the scattered light. We chose to have two separate windows for each flange to avoid parasite light reflected from the laser entrance window to enter the detection system. The KF flanges are made of black anodized aluminum to reduce reflections coming from them. They are also heated during experiments to avoid condensation of moisture on the windows due to the cold operation temperatures of the chamber. Pictures of the windows mounted on KF flanges are shown in appendix B.

The main optical system is composed of two quartz plano-convex convergent lenses separated by a pinhole. The first lens (the closest to the window: LOT-Oriel model 3-41220) has a focal length of 25 mm whereas the second lens focal length (LOT-Oriel model 3-41210) is 19 mm. Both have a diameter of 12.7 mm and a transmittance of
about 93%. The pinhole (Melles Griot GmbH model 04 IDC 000) is placed at the focal point of both lenses. It defines the detector field-of-view and therefore prevents reflected parasite light to enter the detector system. The scattered light is then reflected at 90° with a coated mirror (Melles Griot 02 MPQ 007/038). It is made of fused silica with a protected silver coating (/038) to reach a reflectance higher than 95%. The light is then split into its two polarization components (parallel and perpendicular) with a α-BBO Wollaston polarizing prism (United Crystals Company). It is AR-405 nm coated, the aperture is 10 × 10 × 10 mm and its transmittance is >90%. It has the advantage over the other polarizing prism types of splitting the scattered ray almost symmetrically. Another advantage is to avoid multiple reflections in the prism if the angles are not precisely adjusted. This would lead to cross-talk from the parallel into the perpendicular channel. The angle between the two resulting beams is ~16° and a scheme of the Wollaston prism is illustrated in Fig. 4-2.

![Fig. 4-2: Wollaston prism: the parallel (I_v) and perpendicular (I_h) polarization components are split in the interface of the two orthogonal prisms with perpendicular optic axis.](image)

Finally, before detection by the photomultipliers (PMTs), both rays pass through bandpass filters (Andover Corp. 405FS10-12.5) that stop wavelengths different from 407 nm to eliminate other light sources from reaching the PMTs. As the filters were designed for a wavelength of 405 nm, the transmission is 47% at 407 nm (49.6% at the center wavelength of 405.2 nm). The FWHM (full width at half maximum) is 10 nm.

### 4.1.3 Photomultiplier tubes

Photomultiplier tubes are extremely sensitive detectors of light that multiply the signal produced by incident light by a “gain” factor. They are constructed from a vacuum tube
that houses a photocathode, several dynodes, and an anode. Incident photons strike the photocathode located at the entry window of a PMT where electrons are produced. These electrons are then directed towards several dynodes in series and the process of secondary emission multiplies them. The geometry of the dynode chain is such that a cascade occurs with an ever-increasing number of electrons being produced at each stage. Finally, the anode is reached where the accumulation of charge results in a sharp current pulse indicating the arrival of a photon at the photocathode. A schematic of a photomultiplier tube is shown in Fig. 4-3:

![Fig. 4-3: Schematic of a photomultiplier tube (PMT).](image)

The model used for our detector (Hamamatsu H5783P) has a metallic package and delivers high gain and a high-speed response. This PMT model provides an internal compact high-voltage power supply circuit, making the module easy to use. It is ideal for photon counting under extremely low light conditions. The photomultiplier specifications are listed in Table 4-2. The gain can be set from $2.81 \cdot 10^2$ to $2.30 \cdot 10^6$ by applying different control voltages $U_{\text{ctrl}}$ from 0.25 V to 0.9 V.
### Table 4-2: Specifications of the Hamamatsu H5783P photomultiplier tubes used in the parallel and the perpendicular channels of the IODE detector.

| Parameters                                      | PMT (||) | PMT (⊥) | comments                                           |
|-------------------------------------------------|----------|---------|----------------------------------------------------|
| Input voltage                                   | +11.5 V  | +15.5 V |                                                    |
| Max. input voltage                              | +18 V    |         |                                                    |
| Max. input current                              | 9 mA     |         |                                                    |
| Max output signal current                        | 100 µA   |         |                                                    |
| Max. control voltage                            | +1.0 V   |         | (input impedance: 100 kΩ)                         |
| $U_{ctrl}$ adjustment range                     | +0.25 V  | +0.9 V  | (recommended)                                     |
| Effective area                                   | Ø 8 mm   |         |                                                    |
| Peak sensitivity wavelength                     | 420 nm   |         |                                                    |
| Cathode radiant sensitivity ($\xi$)              | 62 mA/W  |         | (measured at 420 nm)                              |
| Cathode luminous sensitivity *1                  | 66.0 µA/lm | 69.1 µA/lm | (Light source: tungsten filament operated at 2856 K) |
| Anode luminous sensitivity *1                   | 66.6 A/lm | 101.0 A/lm | ($U_{ctrl} = +0.8$ V)                            |
| Anode dark current *1                            | 0.06 nA  | 0.09 nA | ($U_{ctrl} = +0.8$ V)                            |
| Anode Ripple Interference *1                    | 1.08 mV  | 1.06 mV | (peak-to-peak, no signal)                        |
| Circuit current *1                               | 5.4 mA   | 5.4 mA  | (no signal)                                       |
| Gain ($g$) *1                                    | 10.1 x 10^5 | 14.6 x 10^5 | ($U_{ctrl} = +0.8$ V)                            |
| Dark counts *1                                   | 4 s⁻¹    | 11 s⁻¹  |                                                    |
| Rise time                                       | 0.78 ns  |         | ($U_{ctrl} = +0.8$ V)                            |
| Transit time                                     | 5.4 ns   |         |                                                    |

**note:**

*1 from manufacturer final test sheet

The gain function $g$ can be calculated from a logarithmic extrapolation curve that is illustrated in Fig. 4-4:

![Fig. 4-4: Curve of the Amplification gain calibration.](image-url)
4.1.4 Preamplifier units and data acquisition device

The output signal of each photomultiplier tube is connected to a preamplifier unit (Hamamatsu C7319). As the output signal is current-type, the preamplifier furnishes current-to-voltage conversion coupled with signal amplification. It has three switchable conversion ratios $R_{\text{conv}}$: $10^5$, $10^6$ and $10^7$ volts per ampere and also two bandwidths of 20 kHz and 200 kHz with switch selecting. The path shown in Fig. 4-5 expresses the conversion steps of the signal processing from the PMT photocathode to the preamplifier unit in order to obtain a voltage signal:

![Signal-processing stages from the scattered light entering the photocathode to the voltage type signal.](image)

The complete transformation from the voltage signal to physical units can be obtained by taking the inverse path shown in Fig. 4-5, leading to:

$$I[\text{pW}] = I[V] \cdot 10^{12} \cdot (\xi \cdot g \cdot R_{\text{conv}})^{-1} \quad (4.1)$$

To obtain and analyze the signal of the preamplifier outputs, a 4-channel analog input data acquisition unit (DAQ) is used to provide simultaneous sampling with integrated signal conditioning with 16-bit resolution. It provides an USB interface and consists of two components: a National Instrument 9215 module and an USB-9162 USB carrier. The maximum sampling rate is 100 kHz per channel and the typical operating voltage range is 10.4 V. With the USB connection, real-time signals can directly be available on a computer.

The optical elements, the photomultiplier tubes, and the laser are all fixed on a custom-made breadboard with a cutout for the ZINC chamber. The laser is attached on a special holder where the position, the height, the azimuthal and the horizontal angle of the beam can be finely tuned. Fig. 4-6 illustrates a picture of the whole device.
4.2 **The IODE Software**

4.2.1 Peak detection algorithm

A LabView program was made for real-time data analysis of the detection signals. It includes a peak detection algorithm for particle detection and direct measurements of depolarization ratios for individual particles. The LabView software coupled with the DAQ module allows us to visualize, record, and treat real-time data obtained for the parallel and perpendicular channels simultaneously. The sampling rate (number of points per unit of time) can be directly defined on the LabView interface. According to simulations made by Stetzer (2007) with the Fluent computational fluid dynamics software, the average velocity of particles inside the ZINC chamber is 0.1 m/s. It rises up to 0.4 m/s at the height of the detector windows due to the width shrinking. Considering a laser beam diameter of 2.5 mm (parallel setup), the interaction time between the beam and a particle is ~6 ms. With a given sampling rate of 10 kHz, one particle will then be sampled with approximately 60 points. For the perpendicular setup, the beam diameter
is 1 mm, leading to an interaction time of 2.4 ms. The signal peak is theoretically defined with 24 points which is still sufficient for a good peak detection.

The peak detection algorithm scans the signal from the photomultipliers and searches for peaks in the following way: A peak is considered valid if it has the following characteristics: First, the elements of a sequence of points begins below a threshold limit \( \tau \), then exceeds it for some time, and then returns to a value below \( \tau \). Second, the interval that exceeds \( \tau \) is greater than or equal to a given width \( w \). The peak-finding algorithm then fits a parabolic function to the data points above \( \tau \). Using a least-squares algorithm, the peak detection algorithm finally returns amplitudes (intensities) and locations (time) of the peaks.

The determination of the best value for the threshold limit \( \tau \) is done by the analysis of the background signal. If we assume that the background is white noise, its probability density function then follows a normal distribution. Some measurements of the background are shown in Fig. 4-7 where the experimental density function of a background signal of average \( \mu_{BG} \) and standard deviation \( \sigma_{BG} \) is plotted. The theoretical normal distributions \( \mathcal{N}(\mu_{BG}; \sigma_{BG}) \) are also plotted.
The threshold value $\tau$ for each channel ($\tau_\parallel$ and $\tau_\perp$) can then be determined with the application of the statistical law relative to a normal distribution given by:

$$\tau = \mu_{BG} + n\sigma_{BG}$$  \hspace{1cm} (4.2)
Where \( n \) is an integer. If the peak amplitude is equal to \( \tau \), the probability that it is effectively a peak and therefore does not belong to the background signal is illustrated in Table 4-3 as a function of \( n \).

<table>
<thead>
<tr>
<th>( n )</th>
<th>probability</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>69.2%</td>
</tr>
<tr>
<td>2</td>
<td>95.4%</td>
</tr>
<tr>
<td>3</td>
<td>99.7%</td>
</tr>
<tr>
<td>4</td>
<td>99.99%</td>
</tr>
</tbody>
</table>

Table 4-3: Probability that a peak signal does not belong to the background, as a function of the integer \( n \).

The definitions introduced in the peak detection algorithm discussions and its related parameters are shown in Fig. 4-8.

In this example, the dots represent the real-time data obtained from the photomultiplier. The background intensity was 260 pW (standard deviation: 11 pW), which results in a threshold value \( \tau \) of 293 pW for \( n = 3 \). The width \( w \) was set to 15. Once the signal crosses the red line (\( \tau \)) and if the number of points above \( \tau \) is equal or greater than \( w \), the peak is considered as valid. The peak terminates when the signal falls again below \( \tau \). All points above the threshold value (blue dots) are then fitted with the
parabolic function. The peak intensity (after subtraction of the background) and its location are determined at the maxima of the function. We can see that a secondary peak also crosses the threshold line at approximately 0.7 s, but there are less than 15 points that are above τ. Therefore, this peak is not detected.

To determine the depolarization ratio, both peaks corresponding to the parallel and perpendicular channel have to appear at the same time, considering a tolerance time interval Δt that should be set within an optimum value. Peak detection is made first in the parallel channel. If a peak in the perpendicular channel is then located at the same time ± Δt, δ can be obtained from both peak intensities using Eqs. (4.2) and (4.3). If no corresponding peak in the perpendicular channel is found, δ is set to zero.

4.2.2 Data filtering

The background signal is a result of the scattering of molecules, residual particles inside the overlap detection area and scattering from the chamber walls and other elements of the system. As low intensity signal levels due to small particle sizes are expected (see chapter 1), it is important to detect single particles that are partly merged into the noisy background. T-matrix calculations made for water droplets and cylindrical ice crystals show intensities ranging from less than 0.05 pW up to around 10 pW, considering particles sizes from 1 µm to 6 µm in diameter. These measurements show a strong variability as a function of the particle orientation (ice cylinders) and the distance to the detector (see Eq. (2.38)). Other results of the phase matrix elements $Z_{11}$ and $Z_{12}$ for water droplets and polystyrene latex spheres (PSL) are listed in appendix C.

Lowpass filtering can be used in that way by suppressing the noise and keeping peak characteristics in terms of intensity, location and width. Moreover, the standard deviation of the background is lowered and therefore the possible threshold limit τ. Hence, the detection efficiency for small particles increases. It reduces the probability of possible occurrence of parasite peaks (peaks due to the noise and not showing the presence of a particle). Figure 4-9 shows an example of the background signal with and without filtering using different cutoff frequencies:
Fig. 4-9: Background signal of the parallel channel at \( g = 1.01 \times 10^3 \) and filtered data using cutoff frequencies of 1500 Hz, 500 Hz and 100 Hz of the lowpass filter.

The values of the average, the standard deviation and the threshold limit as a function of the cutoff frequency are illustrated in Table 4-4.

<table>
<thead>
<tr>
<th></th>
<th>no filter</th>
<th>200 Hz</th>
<th>500 Hz</th>
<th>1000 Hz</th>
<th>1500 Hz</th>
<th>2000 Hz</th>
<th>3000 Hz</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \mu ) [V]</td>
<td>1.0716</td>
<td>1.0716</td>
<td>1.0716</td>
<td>1.0716</td>
<td>1.0716</td>
<td>1.0716</td>
<td>1.0716</td>
</tr>
<tr>
<td>( \sigma ) [V]</td>
<td>0.0914</td>
<td>0.0260</td>
<td>0.0360</td>
<td>0.0414</td>
<td>0.0505</td>
<td>0.0576</td>
<td>0.0710</td>
</tr>
<tr>
<td>( \tau ) [V]</td>
<td>1.3458</td>
<td>1.1496</td>
<td>1.1796</td>
<td>1.1958</td>
<td>1.2229</td>
<td>1.2445</td>
<td>1.2844</td>
</tr>
</tbody>
</table>

Table 4-4: Values of the average, the standard deviation and the threshold detection limit \( \tau \) for 6 different cutoff frequencies considering a parallel background signal at \( g = 3.68 \times 10^4 \).

Finally, the background in both channels is included in the experimental definition of the depolarization ratios that can be used in our device:

\[
\delta_\parallel = \frac{I_{\parallel}^{\text{sca}} - I_{\parallel}^{\text{BG}}}{(I_{\parallel}^{\text{sca}} - I_{\parallel}^{\text{BG}}) + (I_{\parallel}^{\text{sca}} - I_{\parallel}^{\text{BG}})}
\]  
(4.3)

\[
\delta_\perp = \frac{I_{\perp}^{\text{sca}} - I_{\perp}^{\text{BG}}}{(I_{\perp}^{\text{sca}} - I_{\perp}^{\text{BG}}) + (I_{\perp}^{\text{sca}} - I_{\perp}^{\text{BG}})}
\]  
(4.4)

Where \( I_{\parallel}^{\text{BG}} \) and \( I_{\perp}^{\text{BG}} \) stand for the average background intensities in the parallel and perpendicular channels (with respect to the scattering place). An illustration of the software interface is given in Appendix D.
5

FIRST EXPERIMENTS AND RESULTS

5.1 CALIBRATION AND FIRST TESTS

The objectives of these first experiments were to see if we can differentiate between spherical and non-spherical particles. Then, the effects of changing the tuning parameters, e.g. the width $w$, the lowpass filter frequency $f$ and the PMT gain $g$ will be investigated. We will also see if experiments are reproducible, i.e. if the same results are found throughout two experiments at the same conditions in terms of aerosol type, size, concentration, and with the same physical conditions. The main goal is to determine the optimal values of the parameters mentioned above for further ice nucleation experiments with the ZINC chamber.

5.1.1 PSL particles

Tests were done with spherical latex PSL particles with known diameters of 0.67 µm, 1 µm, 2 µm and 3 µm. The main objective for using PSL spheres was first to see if they are detectable with the IODE detector and second, to check the sphericity of these latex particles. As no depolarization is expected for these particles, we need to ensure that no peaks in the perpendicular channel will be found. Another point is to determine if
particles of this range of sizes are detectable as we expect to have ice crystal sizes greater than about 1 µm in diameter during ice nucleation experiments.

PSL particles were generated by a wet dispersion technique utilizing an atomizer. A suspension of latex spheres is first diluted with demineralized water. The particles were then produced by the atomizer and injected into a simulation chamber. This chamber was specially built for these tests. It has the same dimension as the ZINC chamber at the level of the detection: the distance between the two walls is 10 mm for a width of 300 mm. The chamber height is 56 mm. Consequently, the overlap area for detection is the same. This permits to test different arrangements and tunings of the optical elements of the detector. An aerosol particle sizer (APS, model: TSI 3321) was connected at the bottom of the chamber to obtain the concentration and the size distribution of the particles detected. Figure 5-1 illustrates the setup of the PSL experiments and Fig. 5-2 shows the size distribution of 1-µm diameter PSL particles obtained from the APS at two different concentrations:
First experiments and results

As stated above, the experiment was performed with particles of 1 µm diameter. The parameters of the detector were set as following: the current-to-voltage conversion ratio $R_{\text{conv}}$ at $10^7$ V/A and for the parameters related to the peak algorithm parameters, $w = 5$ and $n = 3$. The sheath air pressure of the atomizer was 1 bar and the sampling flow of the APS was always fixed at 5 lpm (4 lpm of sheath air and 1 lpm for the air containing the aerosols). Given a particle concentration of 27 cm$^{-3}$, this leads to a rate of 450 particles per second. Fig. 5-3 depicts the peaks registered in both parallel and perpendicular channels, using a PMT gain of $1.01 \cdot 10^3$ (left panel) and $7.65 \cdot 10^3$ (right panel). For this experiment, no lowpass filter is applied.
Fig. 5-3: Parallel and perpendicular peak intensities obtained for PSL 1-µm diameter particles for a control voltage \( U_{\text{ctrl}} \) of 0.3 V (left panel) and 0.4 V (right panel).

The peak intensities found in the parallel channel are almost the same, using a PMT gain of \( 1.01 \times 10^3 \) and \( 7.65 \times 10^3 \) which translates into a control voltage \( U_{\text{ctrl}} \) of 0.3 V and 0.4 V, respectively. The average intensities are 25 pW for the lower gain and 21 pW for the higher one. In both cases, no significant peak in the perpendicular channel was registered, proving the latex particles are indeed spherical particles and that these spherical particles do not generate any depolarization signal. However, three peaks were found in the second case \( (U_{\text{ctrl}} = 0.4 \text{ V}) \) but this might stem from foreign particles present in the simulation chamber as it is not completely air tight. The particle number concentration was assumed to be constant \((\pm 2 \text{ cm}^{-3})\). The main difference between these two experiments is the number of parallel peaks detected. More particles are counted with a higher control voltage. Increasing the PMT gain leads to a better detection efficiency for particle detection. Therefore, more parallel peaks are counted, using \( g = 7.65 \times 10^3 \). However, only 230 peaks in about 145 seconds were found in this latter experiment, leading to a rate of \( 0.095 \text{ cm}^{-3} \). The detection efficiency \( \varepsilon \) can be calculated, using:

\[
\varepsilon = \frac{\text{conc. (IODA)}}{\text{conc. (APS)}}
\]  

\( (5.1) \)
Where conc.(IODE) is the particle concentration obtained with the IODE device and conc.(APS) is the one from the aerodynamic particle sizer. Considering a particle concentration of 27 cm\(^{-3}\) given by the APS, the detection efficiency \(\varepsilon\) obtained is 3.5\(\times\)10\(^{-3}\). This rather low result can be explained by the turbulences occurring at the outlet of the atomizer in the simulation chamber. As the APS inlet and the atomizer outlet are not linked and worked with different flowrates, it is difficult to ensure a laminar flow of particles when they pass through the laser beam. Moreover, the simulation chamber is not air tight and its main function is to align the detector optical elements. Many particles crossed the edge of the laser beam or even did not cross it at all. Nevertheless, the main goal of this experiment was not to have quantitative results, but was firstly to see if particles are detectable and secondly if only parallel peaks were found.

In experiments with PSL particles, two different sizes of particles with the same settings of the detector were used. The gain was set to 7.65\(\times\)10\(^3\) and the current-to-signal conversion ratio \(R_{\text{conv}}\) to 10\(^7\) V/A. The peak detection algorithm parameters were the same as in the previous experiments (\(w = 5\) and \(n = 3\)) and no lowpass filter was applied in the measurements. Fig. 5-4 shows the results obtained with a particle diameter of 0.67 \(\mu\)m (top panel) and 2 \(\mu\)m (bottom panel). The particle concentrations were 30 cm\(^{-3}\) and 14 cm\(^{-3}\), respectively.

![Fig. 5-4: Parallel peak intensities for PSL particles with diameter of 0.67 \(\mu\)m (top panel) and 2 \(\mu\)m (bottom panel). The gain was set to 7.65\(\times\)10\(^3\).](image-url)
Similarly to the previous experiment, the main difference is the number of peaks detected. We found 182 peaks with 0.67 µm diameter PSL latex spheres on one side, and 1206 peaks for 2 µm diameter particles. Although ten times more peaks were found for the 2-µm particles case, the average intensities of both types were almost the same. An average intensity of 13 pW for the 0.67-µm particles and 12 pW for the 2-µm ones were recorded. There is no clear size dependence because we are in the domain of the Mie scattering regime, where particle sizes are comparable to the wavelength (407 nm) in our case. For much larger particles \((d \gg \lambda)\), the Rayleigh approximation can be used and the scattered light intensity is proportional to the square of the particle diameter. However, this is not the case in our situation. Some isolated peaks exhibit a much higher intensity than the average. This might result from simultaneous scattering events where two or more particles are detected at the same time. Another possibility is that some particles could have been closer to the detector but were still in the overlap region where they can be detected. Being closer to the detector leads to a larger peak intensity as the intensity is proportional to the square of the distance (see Eq. (2.38)).

Experiments made with PSL latex spheres were also performed in order to understand the differences after applying two lowpass filters with frequencies of 1000 Hz and 2000 Hz. The detector parameters were the same than for the previous experiment, except that the width \(w\) was set to 20 to avoid detection of parasite peaks as much as possible. The particle concentration was 14 cm\(^{-3}\) and the PSL particles used had a diameter of 3 µm. Fig. 5-5 illustrates the peak intensities (left side) and their respective occurrence histograms (right side) with a 1000-Hz lowpass filter (upper panel) and a 2000-Hz lowpass filter (bottom panel).
First experiments and results

Fig. 5-5: Parallel peak intensities (left panels) and their respective occurrence histograms (right panels) using a 1000-Hz lowpass filter (upper panels) and a 2000-Hz lowpass filter (lower panels). The experiments were performed with 3-µm diameter PSL particles with a concentration of 14 cm⁻³.

In the 1000-Hz lowpass filter experiment, the particles were injected into the simulation chamber during about 100 seconds (between 110 and 210 s on the relative time scale). 470 peaks were recorded, giving a detection rate of 0.28 cm⁻³ with an average intensity of 7.8 pW ($\tau = 5.3$ pW). On the other hand, using the 2000-Hz lowpass filter reduced the number of peaks to 333 recorded peaks during a 170-s duration, lowering the detection rate to 0.12 cm⁻³. The average intensity was 9.3 pW for a detection threshold of $\tau = 7.2$ pW. The detection efficiencies were 0.02 and $8.5 \cdot 10^{-3}$ for $f = 1000$ Hz and 2000 Hz respectively, which is higher than for the previous experiment but still very low. As discussed above, simultaneous detection events and turbulences cause particle losses. Particles can also pass besides the laser beam without crossing it. As it was explained in more detail in chapter 4, decreasing the lowpass frequency leads to decrease the detection threshold value. Consequently, more peaks are detected at 1000 Hz than 2000 Hz. However, the average intensities are lower with the 1000-Hz filter. This is due to the flattening of short duration peaks where their intensity are not
perfectly translated when using the 1000-Hz filter. The effect of applying several lowpass filters on different simulated peak types is shown in appendix E.

After these three tests with PSL particles, we proved that spherical particles can be detected with our device, which almost entirely only produced signals in the parallel channel. Therefore, almost no depolarization signals were found during these experiments. We also varied the gain settings and the lowpass filters in the measurements. On the other hand, the width parameter $w$ is not so relevant as values between 5 and 20 did not seem to have a large influence on the number of peaks detected. A low value of $w$ leads to a detection of short peaks that not necessarily belong to real particle signals as high values may cut off the detection of short duration peaks related to particles crossing the edge of the beam. Considering the peak duration of ice crystals crossing in the middle of the laser beam in the ZINC chamber (~6 ms), a peak can be defined with approximately 60 peaks (sampling frequency of $10^4$ Hz). As we have to take into account particles passing through the edges of the beam, resulting in shorter peak durations with lower intensities, a value of $w$ between 10 and 20 should be convenient to detect peaks in that case. To strengthen this affirmation, further experiments were made with the ZINC chamber and results will be discussed in section 5.2. The next section will compare the data obtained during the PSL spheres experiments to the results from T-matrix simulations.

5.1.2 Comparisons with simulation results

Spherical, homogeneous and isotropic particles ($\Gamma = 1$) were simulated using the T-matrix method as discussed in section 3.2.3. The method reduces to the exact Lorenz-Mie theory for these particles. For latex spheres, we used a refraction ratio $n$ of 1.59 according to the manufacturer specifications. Having a linearly polarized light at 407 nm, the equation to determine the scattered intensity from a PSL latex sphere following Eq. (2.38) becomes:

$$I^{\text{sc}}[W] = \frac{1}{r^2} (Z_{11} + Z_{12}) \cdot A_{\text{det}} \cdot Tr \cdot I^{\text{inc}}$$  \hspace{1cm} (5.2)
Where $r$ is the distance between the particles and the photomultipliers ($50 \pm 1$ cm). $Z_{11}$ and $Z_{12}$ are the amplitude matrix elements determined by the T-matrix method, $A_{\text{det}}$ is the area of the detector (i.e., the active area of the PMT window) ($4.42 \cdot 10^{-5}$ m). The incident light energy follows a Gaussian distribution and the value taken is the power density integrated over the whole area of the laser cross-section ($1.85 \cdot 10^4$ W/m$^2$) according to the manufacturer specifications. $T_r$ is the overall attenuation of the scattered signal which is:

$$T_r = \prod_i T_{r_i} \quad (5.3)$$

$T_r$ is the product of all transmittances of the optical elements $T_{r_i}$ and was calculated to be 0.38. The calculated values for $I_{\text{scattering}}$ are plotted as a function of the particle diameter with the experimental values obtained from the PSL experiments in Fig. 5-6.

![Graph showing parallel peak intensities of PSL particles obtained experimentally (black circles) and according to T-matrix calculations (white circles). The vertical bars show the standard deviation of the experimental values.](image)

There is no clear size dependence on the intensities calculated with the T-matrix method. Particles with diameters between 1 µm and 3 µm backscatter approximately the same amount of light and 4-µm diameter spheres even scatter less. Particles larger than 4 µm lead to an increase in intensity and we might expect that it continues to rise further for larger sizes. Hence, we can consider that we are in the Mie regime for these sizes. However, the experimental results obtained with PSL latex spheres show much
larger intensities for particles with diameters of 0.67 \( \mu \text{m} \), 1 \( \mu \text{m} \), 2 \( \mu \text{m} \) and 3 \( \mu \text{m} \) than the theoretical results. These differences probably result from forward scattering of particles that is reflected by the walls and enters the optical system. As forward scattering can be higher than backward scattering by about 3 or 4 orders of magnitude, this effect cannot be neglected. Finally, we may have simultaneous scattering of spheres that are detected as one particle (coincidence events) and also scattering from parasite particles present inside the chamber.

5.1.3 Water droplets

Water droplet measurements were performed with a contact freezing chamber called Collision Chamber. This chamber follows a similar design as the ZINC chamber where the two walls are cooled to the same temperature. This chamber is also 10 mm thick, but it is narrower than the ZINC chamber (200 mm). Its length is variable as it is composed of several single elements that can be assembled together. It was designed to study the formation of ice crystals through contact freezing. For this experiment, only water droplets were injected into the chamber. Fluent simulations have shown that the flow in this chamber is laminar and droplets therefore follow a given streamline. It is possible to align the laser beam in such a way that it crosses the streamline of these droplets. The water drops are created by a piezoelectric droplet generator. The droplet size is \( \sim 75 \mu \text{m} \) and the droplet generator was operated at a rate of 100 droplets per second. The IODE detector is placed horizontally at the bottom of the chamber.

Before running the droplet generator, the intensity and the standard deviation of the background were measured in order to determine the threshold limit \( \tau \) for peak detection. A 1000-Hz lowpass filter was applied and the PMT gain \( g \) was set to \( 1.01 \cdot 10^3 \) (\( U_{\text{cut}} = 0.3 \text{ V} \)) with a conversion rate of the preamplifier of \( 10^7 \text{ V/A} \). The low bandwidth filter was used in order to decrease the noise of the signal. With \( n = 3 \) and measurements without the lowpass filter, the mean background signal was 0.163 V (260 pW) for the parallel channel. The standard deviation was 0.0228 V (36 pW) leading to \( \tau = 0.231 \text{ V} \) (369 pW). Concerning the filtered real-time signal, the mean background stayed unvaried whereas its standard deviation lowered to 0.007 V (11 pW), leading to a threshold value \( \tau \) of 0.184 V (293 pW). The width \( w \) was set to 30 in both channels.
sample of a real-time signal of 1.5 s in both the parallel and perpendicular channel is shown in Fig. 5-7. The left panel illustrates the non-filtered real-time data while the right panel shows the filtered data using the 1000-Hz cutoff frequency.

Peaks only appear in the parallel channel as expected since the water droplets do not depolarize light. Assuming a droplet settling velocity of about 15 cm/s according to fluent simulations, the interaction time between one water drop and the laser beam is approximately 17 ms considering the laser beam configuration with its polarizing plane parallel to the scattering plane (diameter of 2.5 mm). Some of the peaks illustrated in Fig. 5-7 have a width of about 15 to 20 ms, which is indicative of single droplet detection. Other peaks have a shorter width like the highest one. This may be due to a droplet that passed at the edge of the laser beam resulting in a shorter interaction time between the beam and the particle. Thus, the peak width is smaller. On the other side, some coincidence events (simultaneous detection of several droplets at the same time) occurred and can be seen in a much greater peak width and also a higher intensity (Fig. 5-7 at 1.2 s for example). Two experiments were made at the same operation conditions to see if they are reproducible, i.e. if comparable results can be found. All peaks registered for these experiments are shown in Fig. 5-8 with their respective frequency charts.
In the experiment 1, 6760 peaks were detected which translates into 71 water droplets per second. Given that the droplet generator was working at a rate of 100 particles per second, some losses occurred at the level of the detection as not all particles were detected ($\varepsilon = 0.71$). There are two main reasons for that. First, some coincidence events happened during the measurements as seen in Fig. 5-7. Several water droplets are counted as only one single peak. Secondly, some particles might have fallen beside the laser beam and were therefore not recorded.

A statistical analysis of the recorded peaks shows an average value of 105.6 pW with a large distribution ranging from 33.5 pW up to 1243.3 pW. These large differences are due to particles crossing the edges of the laser beam, to the distance between the droplet and the photomultiplier ($r$) at its detection as the scattered light intensity decreases quadratically with increasing $r$, and due to coincidence events. The experiment 2 gave similar results: about 74.3 droplets per second were detected (7099 peaks overall, $\varepsilon = 0.743$) with intensity values ranging from 33.5 pW up to 1369.8 pW. The average intensity was 105.7 pW. The intensity histograms of both experiments are also very similar, showing that these tests with steady conditions are reproducible.
5.2 EXPERIMENTS WITH THE ZINC CHAMBER

Tests with the IODE detector at the bottom of the ZINC chamber were realized in the laboratory where 300-nm montmorillonite particles were used as ice nuclei. The main objective is to see if ice crystals are detectable, i.e. whether depolarization peaks can be resolved. The montmorillonite particles were generated with a fluidized bed generator (TSI model 3400A) and where size is selected with a scanning mobility particle sizer (SMPS). A dilution system located after the fluidized bed generator is employed to have a low concentration of monodispersed aerosol particles. The experimental setup, including the SMPS and the dilution system is illustrated in Fig. 5-9. Varying the dilution valve located just before the fluidized bed modifies the aerosol concentration. A water based condensation particle counter (TSI model 3782) was connected in parallel to the ZINC chamber after the SMPS to measure the particle concentration.

![Schematic diagram of the experimental setup generating a monodisperse flux of montmorillonite particles for ZINC experiments.](image)

The temperature of the two walls was set to –15°C. The montmorillonite aerosol particles are then injected inside the chamber at the flowrate of 1 lpm, adding 2 \( \times \) 4.5 lpm of sheath air. The background for both parallel and perpendicular channels is determined at that moment. After that, the cold wall temperature is decreased to increase the supersaturation ratio until activation of particles occurs. The activation was detected at a sample temperature of 240.5 K and a relative humidity with respect to ice RH\(_i\) of 141.5%. These conditions were then fixed for the rest of the experiments in order to be able to compare results obtained with different values of \( \omega \) and different particle concentrations. The gains \( g \) of the parallel and perpendicular channel were set to 1.01 \( \times \) 10\(^3\) and 2.99 \( \times \) 10\(^3\), respectively. The current-to-voltage conversion ratio \( R_{\text{conv}} \) was
fixed to $10^7$ for all ZINC experiments and a lowpass filter of frequency $f = 500$ Hz was applied to the measurements. For this experiment, we first varied the width parameter $w$ for a given particle concentration of 20 cm$^{-3}$. Then the concentration was varied for a fixed $w$. We finally determined the fraction of activated particles by dividing the number of ice crystals found (i.e. the number of simultaneous parallel and perpendicular peaks, giving a depolarization signal) by the total number of particles injected into the chamber. The results are summarized in Table 5-1:

<table>
<thead>
<tr>
<th>$n$</th>
<th>$w$</th>
<th>concentration</th>
<th>activated frac. (IODE)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>5</td>
<td>20 cm$^{-3}$</td>
<td>6.2 %</td>
</tr>
<tr>
<td>3</td>
<td>10</td>
<td>20 cm$^{-3}$</td>
<td>6.9 %</td>
</tr>
<tr>
<td>3</td>
<td>15</td>
<td>20 cm$^{-3}$</td>
<td>6.7 %</td>
</tr>
<tr>
<td>3</td>
<td>30</td>
<td>20 cm$^{-3}$</td>
<td>3.4 %</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>20 cm$^{-3}$</td>
<td>1.7 %</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>30 cm$^{-3}$</td>
<td>1.9 %</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>34 cm$^{-3}$</td>
<td>2.2 %</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>47 cm$^{-3}$</td>
<td>2.7 %</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>70 cm$^{-3}$</td>
<td>1.1 %</td>
</tr>
</tbody>
</table>

Table 5-1: Activated fraction determined for different parameter values of the integer $n$, the width $w$ and the particle concentration.

As stated in section 5.1.1, changing the width parameter $w$ does not influence the number of peaks detected significantly given here by the activated fraction. From $w = 30$, the activated fraction falls down approximately to half of its values than for lower $w$ because some short-duration ice peaks (related to a small width value) are no more detected by the peak algorithm. As a conclusion, changing the width parameter between 5 and 15 during ZINC experiments does not have any noticeable influence. If $n$ and $w$ remain the same, an increase in the activated fraction with increasing particle concentration is observed. As they still remain low (between 20 cm$^{-3}$ to 47 cm$^{-3}$), the particle counts vary from 5 s$^{-1}$ to 23 s$^{-1}$. The relation between the peak counts and the particle concentration is plotted in Fig. 5-10:
A linear correlation between these two parameters is valid if we are dealing with low aerosol concentrations. However, when concentrations are too high, coincidence events might occur that can limit the increase of the overall number of peaks (saturation). This already occurred at aerosol concentrations of 70 cm$^{-3}$ and 83 cm$^{-3}$. The following experiments (A and B) were performed with the same conditions of temperature and supersaturation ratio, but with different aerosol concentrations. The parallel, perpendicular peaks, and depolarization ratios are depicted in Fig. 5-11 and Table 5-2 lists the average values of the peak intensities and the depolarization ratios.
Table 5-2: Related statistical analysis of the experiment A and B shown in Fig. 5-11.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>experiment A</th>
<th>experiment B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aerosol concentration</td>
<td>83 cm(^{-3})</td>
<td>12 cm(^{-3})</td>
</tr>
<tr>
<td>Concentration of ice nuclei</td>
<td>0.96 cm(^{-3})</td>
<td>0.38 cm(^{-3})</td>
</tr>
<tr>
<td>Fraction activated (IODE)</td>
<td>1.2 %</td>
<td>3.2 %</td>
</tr>
<tr>
<td>Mean parallel intensity</td>
<td>79.9 pW</td>
<td>77.6 pW</td>
</tr>
<tr>
<td>Mean perpendicular intensity</td>
<td>10.7 pW</td>
<td>10.0 pW</td>
</tr>
<tr>
<td>Mean depolarization ratio</td>
<td>0.124</td>
<td>0.112</td>
</tr>
</tbody>
</table>

All parameters in experiment A and B yield comparable values of mean intensities and depolarization ratios. As expected, a larger concentration is found in the first case where the aerosol concentration is almost seven times higher. However, the increase in counts is not as large as the aerosol increase. As a result, the activated fraction is lower during experiment A. This might be a result of slight changes in temperature and humidity condition inside the chamber, as these parameters cannot remain exactly constant because of temperature fluctuations in the cooling system. Coincidence events might have also occurred in experiment A where the aerosol concentration was 83 cm\(^{-3}\).
Experiments made with PSL latex spheres particles, water droplets and ice crystals have proven that differentiation between spherical and non-spherical single particles can be done with the IODE detector. The property of depolarization allows us to distinguish between the liquid and the ice phase without detecting aerosol particles because of their sub-micrometer sizes. It was also possible to determine the optimal parameters to be used for the peak detection algorithm and the optical detectors for further ice nucleation experiments.
THE ICIS 2007 WORKSHOP

Ice nucleation experiments discussed here were carried out during the International Workshop on Comparing Ice Nucleation Measuring systems (ICIS 2007) that was held in Karlsruhe, Germany. The main goal of this workshop was to compare different ice nucleation devices such as the ZINC and the CSU chamber (Rodgers 1988) during ice nucleation experiments. Several aerosol types were used, like Arizona test dust, soot particles, Canary Islands dust (CID), Israel dust (ID), Saharan dust (SD), and Snomax® bacteria (SB) (Morris et al. 2004). This latter product stems from cells of *P. syringae* bacteria that have been freeze-dried and then killed by gamma radiation. The aerosol particles were sampled mostly from the NAUA chamber, which is part of the AIDA facility. The first one was a small-sized (3.7 m³) evacuable aerosol chamber made of stainless steel operating at ambient temperature (stability: ± 2 K) (Schnaiter et al. 2006). The AIDA chamber is a cylindrical vessel made of 2 cm thick aluminum walls and has a height of 7 m, a diameter of 4 m, and a volume of 84 m³ (Möhler et al. 2003). Aerosols were generated and made available in the NAUA chamber. While sampling from the NAUA chamber was always possible, sampling from the AIDA chamber, however, was restricted because it depended on the expansion cycles processed within the AIDA chamber.

Experiments with the ZINC chamber were done in the following way: Activation sequences were made by decreasing the temperature of the cold wall and setting the
warm wall to a constant temperature, leading to an increase of the supersaturation inside the chamber. Each activation cycle was realized with a different warm wall temperature such that every activation occurred at a different temperature. The results of a characteristic activation sequence are shown and discussed in the following sections of this chapter. Ice nucleation experiments with Snomax® bacteria will be discussed first, followed by Israel dust, Saharan dust, and Canary Islands dust. Every activation experiment will be denoted with a code containing the letters of the aerosol type, following by the number of the activation (for example, SD4: fourth activation with Saharan dust). The aerosol flowrate was fixed at 0.2 lpm for all experiments, as first tests with Arizona test dust showed a high aerosol concentration. Meanwhile, the sheath air flowrate was set to $2 \times 4.9$ lpm.

### 6.1 SNOMAX® BACTERIA (SB)

During all experiments, a 500-Hz lowpass filter was applied. This represents an optimal value as the background signal is sufficiently smoothed without modifying the peak retrievals too much. The gain was set to $1.27 \times 10^3$ and $5.33 \times 10^3$ for the parallel channel and the perpendicular one, respectively. The conversion rate was fixed to $10^7$ V/A for both. The Preamplifier low bandwidth was used in order to decrease the noise of the signal. The width $w$ was tuned to 15 and the integer $n$ to 3 for the threshold determination for both channels. A part of a 1.5-s duration real-time signal during the 4th activation experiment for both the parallel and perpendicular is shown in Fig. 6-1.
All peaks in the parallel and perpendicular channel are recorded during the activation cycles. The depolarization ratios are then directly derived from the peak intensities by the software. One typical activation event is described in Fig. 6-2. The temperature of the warm wall was set to 263 K.
Fig. 6-2: Scheme of a typical activation experiment (Snomax® bacteria 6th activation: SB6). Panel A shows the temperature and humidity profiles calculated from the wall temperatures and flows during the experiment. Panel B illustrates the raw data from the optical particle counter as a color-coded intensity spectrum. The activated fraction of aerosol particles calculated from the OPC data is plotted in panel C. The parallel and perpendicular intensities of the IODE detector are given in panel D and E. Panel F illustrates the depolarization ratio obtained from both intensities.

Panel 6-2A shows the temperature (T) and humidity profiles (with respect to ice RH_i and to water RH_w) inside the chamber of the whole dataset containing the activation. The humidity is calculated by using the parametrizations from Murphy and Koop (Murphy and Koop 2005). Panel 6-2B illustrates the OPC raw data as a color-coded size intensity plot. Individual size distributions are obtained every 5 seconds. The activated fraction derived from the OPC data is shown in panel 6-2C. The number of activated particles is
obtained as following: The size spectra integration from channel 10 upward (to eliminate electronic noise from the detector) gives the number of total particles on one hand, and on the other hand the number of activated particles is calculated by integrating the same data set but approximately from channel 90 to 160 upward, depending on the aerosol size distribution. The ratio of these two numbers is then the activated fraction. Panels 6-2D and 6-2E show the individual peak intensities from the parallel and the perpendicular channel obtained with the IODE detector, respectively. Finally, panel 6-2F depicts the depolarization ratio obtained from the intensities as discussed above.

Snomax® bacteria activates around water activation. As this is within the regime of immersion freezing, we may have ice crystals and water droplets simultaneously. Due to the evaporation section in the lower part of the ZINC chamber, these water droplets formed in the upper part should evaporate and we should only have ice particles detected by the OPC and the IODE detector. However, if the relative humidity with respect to water is above a certain critical value, the water droplets do not have sufficient time to evaporate entirely due to their large sizes. Therefore, they are detected with both detectors. This is called a ‘water breakthrough’ event where we expect detection of ice crystals and water droplets at the same time although the evaporation section of the chamber is always sub-saturated with respect to water.

The particle activation started at about $t = 7630$ s ($T = 257.5$ K and $\text{RH}_i = 110.2\%$). This is revealed in the OPC raw data (panel B) where several counts appear in channels 210 and higher. Everything before that time is considered as background (air loaded with aerosols). Consequently, this is the background signal taken into account for the IODE peak algorithm tuning (determination of the threshold $\tau$). The start and the increase of the activated particles fraction (panel C) was also remarkable as the curve rose from 0 at about 7625 s to 0.1 between roughly $t = 7800$ to 8000 s. The activation was also indicated with peaks in both the parallel and perpendicular channels found by the IODE detector. However, the first appearance of ice in the IODE detector had a delay of about 5 s in comparison to the optical particle counter data. This might be due to the OPC data that have a 5-s time resolution so that the late counts during one interval could have been indicated at the beginning of that interval, i.e. earlier. During the activation, the activated fraction remained more or less
The concentration of Snomax® bacteria is assumed to be constant all along the experiment. Meanwhile, the temperature and the relative humidity continued to decrease, respectively increase, showing that a higher supersaturation did not cause further activation. Concerning the IODE results, some large isolated peaks occurred, especially in the parallel channel. This may be due to two main reasons: First, coincidence events seemed to happen. Simultaneous detection of several particles leads to a much higher intensity and to a miscount of peak numbers. This is mostly the case when concentrations of particles are pretty high (more than roughly 70 cm⁻³). Secondly, as all the ice particles are distributed over the entire width of the chamber at the level of the detector, closer particles generate a higher scattered intensity due to the dependence of the scattering on the distance \( r \) to the detector (see Eq. (2.38)). A rise in the perpendicular channel intensity also appeared during the activation run because of two reasons: The formation of bigger ice crystals along the experiment and/or a change in the particle shape as a result of varying temperature and relative humidity with respect to ice (Pruppacher and Klett 1997). Thus, an increase in the depolarization ratio was denoted.

The activation ended up shortly before \( t = 8100 \) s, which is well indicated in both the OPC and the IODE detectors. Another way to visualize the activation is to plot the mean values and the standard deviation of the two IODE channels. The detector software allows making this determination by taking the average of all the sampled individual points from the real-time signal every 0.5 s. These profiles are illustrated in Fig. 6-3.
In the upper panel, it is hard to see a strong rise, especially in the perpendicular channel. On the contrary, the activation is nicely seen in terms of standard deviations (lower panel). However, it remains difficult to obtain the exact timing of the start and termination of the activation. This method will be beneficial or superior in case of high concentrations of aerosol particles where peak detection becomes tricky. Table 6-1 indicates the values obtained during the whole experiment from the IODE detector (intensities and depolarization ratios) and includes the OPC raw data.

Fig. 6-3: Real-time signal average values (upper panel) and their standard deviations (lower panel) calculated within a 0.5-s time interval for the Snomax 6th activation experiment.
The average values of all cases (Table 6-1) indicate that the large ice particles formed inside the chamber scatter more light than the non-depolarizing particles. These can be either water droplets or other particles that do not depolarize light. The question remains if we really sample water droplets at the bottom of the chamber, given that all the particles passed through the evaporation section of the chamber where ice saturation prevails. It is possible that un-activated aerosol particles that took up water are falsely detected as non-depolarizing particles. These organic particles may deliquesce and no efflorescence occurs after that as we are well above the efflorescence point in the evaporation section. Therefore, the particles exhibit a spherical shape and are large enough to be detected. Some small ice crystals might also exhibit a spherical shape, but even low asphericities can induce strong depolarization signals (Mishchenko and Sassen 1998, Leisner 2006). The OPC counted a total amount of 14,951 particles during the activation whereas the IODE registered 10,564 non-spherical particles during the same time interval. According to previous calibration tests made in the laboratory, the OPC has a detection efficiency between 50% and 60%. Therefore, the sum of all particles detected by the IODE is close to the real total number of particles. This includes also coincidence events and particles not crossing the laser beam. Adding the number counts with $\delta_\parallel < 0.05$ (74 counts, which is negligible here) allows a comparison between the experimental data and the modeled one (Nicolet et al. 2007) for a 2-μm diameter particle with an aspect ratio of 1 (Fig. 6-4).

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**Table 6-1:** Statistical analysis of the OPC and IODE data during the Snomax 6th activation experiment (SB6).

<table>
<thead>
<tr>
<th></th>
<th>Sum of counts</th>
<th>average [pW]</th>
<th>std. dev. [pW]</th>
</tr>
</thead>
<tbody>
<tr>
<td>OPC</td>
<td>14,951</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>$I_\parallel$ (ice)</td>
<td>10,564</td>
<td>39.48</td>
<td>40.74</td>
</tr>
<tr>
<td>$I_\parallel$ (non-depol.)</td>
<td>16,811</td>
<td>24.18</td>
<td>15.47</td>
</tr>
<tr>
<td>$I_\perp$</td>
<td>10,564</td>
<td>11.83</td>
<td>8.29</td>
</tr>
<tr>
<td>$\delta_\perp$</td>
<td>-</td>
<td>0.250</td>
<td>0.112</td>
</tr>
</tbody>
</table>


The ICIS 2007 workshop

Fig. 6-4: Simulated occurrence of the depolarization ratio $\delta_\perp$ considering a cylindrical ice crystal with an aspect ratio of 1 and a diameter of 2 µm (from Nicolet et al. 2007) (left panel) and depolarization ratio $\delta_\perp$ obtained during the Snomax® 6th activation experiment (SB6) for the whole IODE dataset (right panel).

The fraction between 0 and 0.05 is comparable (30% vs. 35%) but the average value obtained from the experiment is significantly higher. The right panel shows a skewed distribution but it looks closer to a normal distribution centered at about 0.25, ranging from 0.011 to 0.821. The differences may be caused by the differences in shape and size of the real ice crystals in the ZINC chamber as compared to the idealized shapes used in the simulations. Nevertheless, the ice particles should theoretically be randomly oriented, given their settling velocity (~0.4 m/s) and a diameter range between 1 and 10 µm (Hallett 1987). Moreover, the ice particles were modeled as cylinders and they should have a more complex shape in reality. Unfortunately, it is not possible to independently determine the size and shape of these parameters and to have clear information about the size and the aspect ratio of these particles. However, the same decrease as calculated can be observed at $\delta_\perp \approx 0.4$ with very few occurrences of high depolarization (Nicolet et al. 2007).
6.2 **ISRAEL DUST (ID)**

In the case of an activation made with Israel dust particles, the concentrations and the size of the aerosol particles were much larger as shown in Fig. 6-5.

![Figure 6-5: Scheme of the Israel dust 4th activation experiment (ID4).](image)

The starting point of the activation is clearly identifiable on the activated fraction panel. In that case, the activated fraction constantly increased until reaching the minimum temperature (238 K) and the maximum humidity with respect to ice (157%). The same evolution occurred for the parallel and perpendicular peak intensities. This translates into a rise in the particle size in a similar way like in the SB6 activation experiment. On
the contrary, the depolarization ratio stayed at the same level, showing that this parameter is no more dependent on the size, as it is the case for large size parameters (Nicolet et al. 2007).

6.3 SAHARAN DUST (SD)

For this experiment, Saharan dust aerosols were injected into the ZINC chamber. The humidity conditions inside the chamber were tuned to high values, peaking at RH$_i$ = 173% and RH$_w$ = 133% with the lowest temperature at 246.5 K. Fig. 6-6 depicts the same type activation panels as discussed before.

![Figure 6-6: Scheme of the Saharan dust 9th activation experiment (SD9).](image)
Shortly before \( t = 7500 \) s, a ‘water breakthrough’ event as defined in section 6.1 appeared. Water droplets which survived the evaporation section appear in the OPC raw data as every bin had a large amount of counts (cyan-green colored bins from channel 50 to 200 in panel B. The activated fraction rapidly increased to values of about 0.5 and remained constant all along the activation until the temperature was increased. In the IODE data, the peak structure was significantly different than the previous ice activations discussed. In the parallel channel, a large increase can be seen whereas the perpendicular peak signals stayed constant globally. The presence of higher peaks indicated the presence of ice crystals among the water drops. Many low perpendicular signals can be considered as part of the background signal. Therefore, the resulting depolarization ratios leveled off at low values, as discussed later (Fig. 6-16). This trend suggests the following explanation: The mixture of water droplets and ice particles caused this overall slight rise in the average signal intensity. It was high enough to detect a high number of low intensity peaks. The statistical analysis of this activation experiment shown in Fig. 6-7 and Table 6-2 establishes that the ice crystals detected are largely underrepresented within all particles. Saturation occurred with the IODE detector as the total number of OPC counts went largely beyond the non-depolarizing particle counts.
Fig. 6-7: Histograms of parallel intensities (ice and water particles, upper panels), perpendicular intensities (bottom left panel) and depolarization ratios (bottom right panel) during the 9th Saharan dust activation experiment for the whole IODE dataset.

<table>
<thead>
<tr>
<th></th>
<th>Sum of counts</th>
<th>average [pW]</th>
<th>std. dev. [pW]</th>
</tr>
</thead>
<tbody>
<tr>
<td>OPC</td>
<td>356,703</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>$I_i$ (ice)</td>
<td>2680</td>
<td>136.67</td>
<td>48.55</td>
</tr>
<tr>
<td>$I_i$ (non-depol.)</td>
<td>38,964</td>
<td>83.95</td>
<td>50.41</td>
</tr>
<tr>
<td>$I_\perp$</td>
<td>2680</td>
<td>15.66</td>
<td>4.25</td>
</tr>
<tr>
<td>$\delta_\perp$</td>
<td>-</td>
<td>0.114</td>
<td>0.048</td>
</tr>
</tbody>
</table>

Table 6-2: Statistical analysis of the OPC and IODE data during the Saharan dust 9th activation experiment (SD9).

Although ice crystals were in the minority, they scattered more light than water drops and exhibit a quasi-normal distribution in the frequency plot. The water droplets scattering intensity showed nearly a double structure: We suggest that the first peak is related to single particle detection whereas the second maximum distribution (95 pW) corresponds to coincidence events. Still, the perpendicular channel consisted almost entirely of low intensities. This could indicate a particular shape of the ice crystals that does not generate a strong depolarization effect, for example horizontally aligned plates, or columns whose vertical axis ($z$) have the same orientation than the $z$ axis of the
laboratory frame (Nicolet et al. 2007). As it is not possible to monitor the shape of these particles, it is not possible to confirm that. The large gap between the OPC and the IODE counts is caused by a saturation effect that did prevent the IODE detector from counting all particles. In the real-time averaged signal for both channels, the water activation can be clearly identified, too (Fig. 6-8).

![Figure 6-8](image)

*Fig. 6-8: Real-time signal average values (upper panel) and their standard deviations (lower panel) calculated within a 0.5 s-time interval for the Saharan dust 9th experiment.*

The average and standard deviation plots illustrate a large increase in the parallel channel. The perpendicular one is slightly rising – sufficiently to get all the low peaks as previously discussed – but nothing comparable to the parallel channel. In case of high concentrations, the average-standard deviation method is a good tool to show the presence of water droplets and/or ice crystals.
The next figure (Fig. 6-9) shows activation at a lower temperature (234 K), with a humidity with respect to ice close to 180%. In this case, no water breakthrough occurred even though relative humidity with respect to ice up to 120% was reached.

![Graph of Saharan dust - 5\textsuperscript{th} activation](image)

**Fig. 6-9:** Scheme of the 5\textsuperscript{th} Saharan dust activation experiment (SD5).

The peak occurring in the activated fraction plot at approximately 8700 s is the result of frost coming directly from the ice layers on both walls. During activation experiments, ice crystals are growing from the walls. The temperature differences set throughout activation processes cause small displacements of the chamber, including vibrations inside the chamber that make this frost falling. Consequently, these crystals are detected by both the optical particle counter and the IODE detector. In this experiment, the event was dismissed in the IODE data by post-treatment and no peaks are visible in
the related panels of Fig. 6-9. Concerning the depolarization ratio, we see an increase along the activation run until the minimum temperature was reached. As discussed before, this might originate from different particle sizes and shape that are preferred at different temperatures and humidities (Pruppacher and Klett, 1997).

6.4 CANARY ISLANDS DUST (CID)

During experiments using Canary Islands dust, the unactivated dust particles size was even smaller than for Saharan dust as illustrated in the panel b) of Fig. 6-10 that show the OPC and IODE data of the 4th activation experiment (CID4). This allowed a better discrimination of the activated fraction and the probability to have single particle detection with the IODE device was higher. The gain for the parallel channel was set to $1.59 \cdot 10^3$ whereas the one for the perpendicular channel was fixed at $3.65 \cdot 10^3$. 
The IODE data for peak intensities and depolarization ratios were missing between $t = 2593$ and 2638 s which was due to a software bug where no data were recorded during this time interval. However, it was possible to detect the termination of the activation at about 2660 s. The number of activated particles increased during the experiment whereas the depolarization ratio globally declined from $t = 2440$ to 2480 s before starting to rise again. The values of the depolarization ratio are generally lower than those obtained for the Saharan dust (at the end of the activation), the Israel dust and the Snomax® particles. We suppose therefore that the aerosol type has an influence on the ice crystal shape and size and thus the amount of depolarization. Moreover, it is possible that they have different properties in terms of their active sites, but it is not possible to confirm this affirmation at this point.
6.5 **Overall Discussions**

During the ICIS 2007 workshop, several kinds of aerosol particles were investigated and it was possible to determine their critical saturation ratios for activation from the activation experiments that were performed for each aerosol type. A threshold to detect the onset of freezing must be set above a background level. It is obtained from the number of counts with the OPC in channels above approximately 90 to 160, depending of the aerosol size distribution, and we chose a threshold activated fraction of 1% (Stetzer et al. 2007). Since data from the optical particle counter are available only every 5 seconds and particles can theoretically be activated in different parts of the chamber, they need shorter or longer time until they reach the OPC. Thus, the inaccuracy of the exact time of ice nucleation is estimated as 10 seconds at the minimum.

Mean temperatures and supersaturations for the onset of freezing are then collected for all activations and plotted in Fig. 6-11 for the different dust types. The solid line represents water saturation which was calculated using the parametrizations for the partial water vapor pressure \( p_w \) and the ice vapor pressure \( p_i \) from Murphy and Koop (2005).

![Fig. 6.11: Critical saturation ratios determined with the OPC for Arizona test dust (ATD), Canary Islands dust, Saharan dust, and Israel dust from ZINC experiments using a 1%-activated fraction as threshold for the onset of freezing. The water saturation \((p_w/p_i)\) line is calculated using the parametrizations for \( p_w \) and \( p_i \) from Murphy and Koop (2005).](image)
A sharp decrease can be seen for all the three dust types, decreasing from RH$_i$ between 165% and 175% to RH$_i$ between 115% and 120%, (except for Saharan dust at RH$_i$ ≈ 140%) for the decrease of temperature from 245 K to 242 K. This was also the case for the Arizona test dust experiments that were performed in the first three days of the workshop without the IODE detector. Israel dust, Canary Islands dust and Saharan dust have similar activation properties, except Saharan dust which does not activate as good at lower temperatures. This may be due to the different surface roughness and chemical composition of the Saharan dust. This was shown by Möhler et al. (2006) where the critical ice saturation ratio already increased at T ≈ 210 K and RH$_i$ between 100% and 110%, reaching almost 120% at a temperature of 222 K. The cooling power of the warm wall cryostat limited the investigation to temperatures above 233 K. Thus, it was not possible to detect the stabilization of critical RH$_i$ observed by Möhler et al. for temperatures lower than 233 K as well as the decrease of the ice supersaturation ratio for the Saharan dust.

Fig. 6-12: Critical saturation ratios for Snomax® and soot particles from ZINC experiments using a 1%-activated fraction as threshold for the onset of freezing. The plain line shows the water saturation line using the parametrizations from Murphy and Koop (2005) and the dotted line illustrates the breakthrough section where water droplets survive the evaporation section of the ZINC chamber (blue diamonds).
The figure 6-12 illustrates the same plot for soot particles and Snomax® bacteria. The different behavior of these two types of particles is clearly evident. On one hand, soot can act as IN according to laboratory experiments made in a cloud chamber for temperatures ranging from –5°C to –20°C (Gorbunov et al. 2001). They are also involved in heterogeneous freezing processes from aircraft exhaust that lead to an increase in cirrus cloud coverage (Hansen and Toon 1997). As seen in Fig. 6-12, supersaturation ratios of 60% with respect to ice are necessary for temperatures around 240 K. However, we can detect a decrease in RH_1 for lower temperature, reaching values of about RH_1 = 127% at T = 235 K. On the other hand, Snomax® particles can nucleate ice at temperatures as warm as –1°C (Morris et al. 2004). As it was not possible to go to temperatures that high due to technical limitations, we started the activation scan from 264 K to 246 K and we found a linear increase in RH_1 from 109% to 118%. This is due to immersion freezing that prevails for activation at warmer temperatures. As it requires at least saturation with respect to water, RH_1 increases with decreasing temperatures to follow the water saturation line parametrized by Murphy and Koop (2005). We confirmed with this experiment that Snomax® bacteria are good ice nuclei at warm temperature.

Concerning the water activation experiments discussed previously, they were all recorded in the same way than the other activations and plotted also in Fig. 6-12 (blue diamonds). This defines the ‘breakthrough’ threshold above which water droplets which had formed inside the chamber are surviving the passage through the evaporation section. As all points follow a clear trend line given by the dotted line, these water breakthrough events are not dependent on the aerosol type, but are only due to the technical characteristics of the ZINC chamber. It is therefore unique for this chamber. Another important point to investigate is the comparison between the critical saturation ratios obtained with the OPC at a 1%-activated fraction threshold and the first significant ice peak recorded with the IODE detector. This is shown in Fig. 6-13:
Fig. 6-13: Critical saturation ratios for Israel dust, Canary Islands dust and Snomax® particles from ZINC experiments using a 1%-activated fraction as threshold for the onset of freezing. The full symbols indicate the OPC data determination whereas the open symbols show the IODE determination. The plain line shows the water saturation line using the parametrizations from Murphy and Koop (2005).

We see that the data is generally similar, but the values differ a bit, as both detectors are not located at the same place, creating a time shift. Moreover, we have to consider the OPC 5-s sampling rate discussed above as well as the uncertainties concerning the temperature and saturation values. Another point is that the first ice crystal determined with the IODE detector defines the critical saturation ratios whereas a given proportion of activated particles are taken into account for the OPC data. As a result, the values obtained with the IODE detector are generally found at a higher temperature and lower supersaturation than for the optical particle counter. Therefore, the IODE activation points on Fig. 6-13 should be located below the ones from the OPC. Most points obtained with the IODE detector tend to show this, except for 2 points (one for Israel dust and one for Snomax particles): This may come from the background correction that had to be made with a data analysis program used for the OPC raw data. As Israel dust was very large, the background correction was difficult to make and the precision in the activated fraction determination was not optimal. For Snomax®, the first ice crystals seen had a delay in comparison with the OPC data determination as earlier ice particles were not seen by the detector. This is due to their sizes as they are still too small to be detected with the IODE.
Considering the differences between the OPC and the IODE detector, Fig. 6.14 shows the counts obtained by both detectors during the 6\textsuperscript{th} activation of Snomax\textsuperscript{®} (SB6) for an integration time of five seconds.

Fig. 6.14: OPC counts (black solid line) and ice peaks counts obtained from the IODE detector (magenta solid line) for the Snomax\textsuperscript{®} 6\textsuperscript{th} activation experiment (SB6) and considering an integration time of 5 seconds.

From $t = 7710$ and 8000 s, the number of ice peaks obtained with the IODE detector ranges between 50% and 97% of the total counts from the OPC data. If we consider that the total OPC counts represent the exact number of ice crystals formed, we suggest that the fraction of the particles not detected by the IODE device either stems from ice particles not crossing the laser beam, or ice crystals having an orientation that does not sufficiently depolarize light to be detectable (see chapter 3). After that, the ice peak counts went beyond the OPC counts. This might originate from a change in the background level taken into account in the peak detection algorithm. An increase of the background level lead causes more ice particles to be counted instead of ice crystals. A small move of the ZINC chamber can be due to the walls cooling at different temperatures, causing a disalignment of the detector optics. As a result, it can cause a higher signal in both channels as some frost on the inner chamber walls can reflect some light from the laser beam edges directly into the optical system. There were some gaps
in the IODE data, as some peaks were not recorded during several time intervals and these incomplete counts were therefore omitted. Fig. 6-15 depicts the total counts of both detectors for the Snomax® 7th experiment (SB7) where water activation occurred.

![Graph](image-url)

**Fig. 6-15:** Optical particle counter counts (black solid line), water peaks (blue line) and ice peaks counts obtained from the IODE detector (magenta solid line) for the Snomax® 7th activation experiment (SB6) and considering an integration time of 5 seconds.

This activation experiment can be divided in two parts: The first one concerns only ice crystals as no water droplets survived the ZINC evaporation section. The ice peak counts from the IODE range between 35% and 97% of the total OPC counts, with values mainly around 70%. This situation is comparable to the one described in Fig. 6-14 for the 6th activation experiment.

The most interesting part is the ‘water breakthrough’ occurring at \( t = 740 \) s: From that time, the counts from the OPC and the water peaks determined with the IODE device drastically increased. The temperature was 253.6 K and we had a relative humidity with respect to ice of 136% (112.7% with respect to water). From \( t = 790 \) s, the IODE water droplets counts started to stay constant at values of approximately 1100 counts every five seconds, showing the saturation effect of the IODE detector whereas the OPC counts continued to climb until approximately \( t = 820 \) s. The water peak remained more or less constant until the end of the water activation shortly before
900 s. At the same time, the total number of the OPC counts varied a little bit more before plunging at \( t \approx 905 \) s. During the water activation, the number of ice peaks continued to rise, reaching a maximum at \( t = 845 \) s (274 peaks). The number of ice crystals then began to steadily decrease until the end of the activation at about 1000 s.

Concerning experiments with a breakthrough of water droplets, it is not possible to only rely on the OPC data as it does not distinguish between the water and the ice phase. Consequently, this leads to miscounts of the number of total particle and we cannot determine the number of ice particles present during these events. We show that ice crystals are still visible with the IODE detector when water breakthrough events occur. Thus, it is possible to show and to follow the formation of ice crystals along these processes. Hence the evaporation section can be removed from the ZINC chamber for further experiments as it was shown that differentiation between ice crystals and water droplets can be done during activation experiments at every level of saturation ratio. An additional information to identify water breakthrough events is to make a statistical analysis on the depolarization ratios of all ice crystals during a whole activation experiment and this is especially noticeable in case of high aerosol concentration events. A boxplot chart in Fig. 6-16 summarizes some activation experiments.
Fig 6-16: Summary boxplot chart resuming depolarization ratios $\delta_\perp$ obtained for some activation experiments made during the ICIS 2007 workshop (ID: Israel dust, SD: Saharan dust, CID: Canary Islands dust, SB: Snomax® bacteria). The box represents the 25%, 50%, and 75% quartiles and the whiskers show the 5% and 95% percentiles. The number stands for the experiment number and the “WA” mark indicates water breakthrough events.

For Israel dust, Saharan dust and Canary Islands dust, water breakthrough events (WA) are clearly identifiable from the ice activation ones. The difference is smaller for the Snomax® bacteria as concentrations of ice and water particles were much lower than for dust. However, the background perpendicular peak intensities were quite comparable. Finally, other activation schemes are shown in appendix F.
7

DISCUSSION

7.1 MODELING STUDIES

Previous experimental works on depolarization, such as lidar applications, or measurements in the AIDA chamber were done by investigating a collection of particles and not single particles. In our application with single particles we do not need to consider any scattering effects of simultaneous scattering by two or more particles. The depolarization ratio for single scattering events can strongly vary as a function of the particle orientation. For the same particle, either a non-depolarized signal or a very high depolarization can occur, depending on the angles $\alpha$ and $\beta$. As the depolarization technique is mainly used to differentiate between spherical and non-spherical particles, the results from single particles above show that this is not always possible with this technique. The relative occurrence of non-detectable ice particles ($\delta < \delta_{\text{lim}}$) varies with size, aspect ratio and the detection limit considered, ranging from 0.204 up to 0.6 for a 2-\(\mu\)m diameter isometric cylinder for $\delta_{\text{lim}} = 0.05$ (see Fig. 3-8 and table 3-3). The aim is to have a detection system as sensitive as possible with a very low background in order to decrease $\delta_{\text{lim}}$.

The non-depolarizing areas ($\alpha \approx 90^\circ$ and $\beta \approx 0^\circ$ and $180^\circ$) observed in all single particle cases relate to the cross section that is seen from the $\hat{n}_{\text{sca}}$ vector. For $\beta$ close to
0° and 180°, the bottom and the top of the particle are seen so that it appears as a sphere from an observer placed at the origin of the laboratory coordinate system and looking in the direction of the \( \hat{\mathbf{n}}_{\text{ex}} \) vector. Nevertheless, as real particles are expected to exhibit a hexagonal shape, it could still be possible to obtain a significant depolarization even for these values of \( \beta \). For \( \alpha \) close to 90° and \( \beta \approx 90° \), the particle appears as a square as the observer looks in the same direction as previously defined. The particle looks as a rounded rectangle for other values of \( \beta \) until it is seen as a sphere for \( \beta \approx 0° \) and 180° as discussed above. By analogy, the former configuration is the same as for lidar backscattering by quasi-horizontally oriented ice crystal plates where no significant depolarization is obtained (Yang et al. 2003). This spherical shape might also have an influence on the non-depolarizing results in these specific orientations. In all non-depolarizing areas, the angle of incidence between the laser beam and the particle is always close to 90° (\( \alpha \) close to 90° and \( \beta \) close to 0° and 180°). This limits the number of internal reflections and therefore may generate less depolarization (Sassen and Liou 1979). All these conclusions are strengthened by the fact that both the size and the aspect ratio do not have any influence on the scattering of these regions.

The presence of very high depolarization ratios, on the other hand, can be explained by multiple internal reflections inside a crystal. For bigger particles, a slight change in the orientation can result in a strong difference in the depolarization, so these internal reflections can be highly modified depending of the angle of incidence of each internal interaction inside the particle and the occurrence of these reflections. For smaller sizes, it is more difficult to derive an explanation for this as we are closer to the Mie regime. This regime might also have an influence on the distribution of the depolarization ratio for small particles \( (d < 1 \, \mu m) \) which is more regular. On the contrary, the larger the particles are, the lower is the influence of the size. This might be due to a weaker influence of the Mie oscillations. By averaging all orientations for a given particle, it was possible to determine the impact of size and aspect ratio on the mean calculated depolarization values and to compare them with former studies. Still, these comparisons have to be regarded with caution, as both the scattering angle and the wavelengths are slightly different. First, the depolarization ratio is positively correlated with size up to a critical particle diameter where it reaches a peak depolarization value of \( \bar{\delta} \approx 0.23 \) to 0.25. For larger particles, the trend tends to decline slowly. This evolution can also be seen in Mishchenko and Sassen (1998) where a
collection of randomly oriented ice particles was simulated. In this study, a weaker
decrease for particles with size parameters larger than 20 was observed. The
wavelength used and the different angle of incidence ($\theta^{inc}$) can explain the differences
between these simulations. Krämer et al. (1996) showed a mean depolarization value of
0.23 (regarding the definition used in our study) at 632.8 nm for single ice crystal
events, resulting from freezing of droplets with an initial diameter of 50 µm in diameter
(Krämer et al. 1996). Regarding the aspect ratio $\Gamma$, no clear effect of this parameter could
be observed. Even particles with $\Gamma$ close to 1 show depolarization ratios higher than 0.2.

7.2 EXPERIMENTAL RESULTS

A depolarization detector device (IODE) was designed and built to distinguish between
water droplets and ice particles which can coexist in a diffusion chamber. Ice crystals
depolarize the incident polarized light whereas spherical water drops do not. The
parallel and the perpendicular component (to the incident light polarization) of the
scattering light intensities are measured and the depolarization ratio is determined.
Peaks in both channels are counted with a peak detection algorithm. The signal noise is
reduced with a lowpass filter. This lowers the standard deviation of the background
signal and therefore increases the detection efficiency.

First tests were performed with spherical latex particles (PSL) with several
known diameters to test if the detector is capable to detect these particles. We also
checked if PSL particles yield a zero signal in the perpendicular channel as they should
and if small size spheres (0.67 µm in diameter) could also be detected in order to define
the lower limit of the detectable size. After that, water droplets were used for other
experiments to confirm results obtained from the PSL sphere tests. We showed that
both latex spheres and water droplets of larger sizes (~75 µm in diameter) can be
detected and do not generate significant peaks in the perpendicular channel, but only a
parallel component as expected from the theory. We succeeded in detecting particles
smaller than 1 µm, which is the theoretical lower limit of ice crystal sizes in the ZINC
chamber. Concerning peak detection, it was possible to determine the influence of every
tunable parameter such as the peak width $w$, the number $n$ of standard deviations
included for determining the threshold value $\tau$, the gain $g$, and the lowpass filter
frequency $f$. It was then possible to obtain the optimal values for these parameters for further experiments to improve the peak detection algorithm. The width $w$ is not the most sensitive parameter as values between 10 and 20 (number of points for the parabolic function approximation) gave comparable results. The integer number $n$ was usually set to 3, which represents a probability that the signal does not belong to the background of 99.7%. With a higher value, it is possible that low signals coming from very small particles in a high background condition may not be detected. It is not possible to know exactly the best frequency to use for lowpass filtering, but we showed that un-filtered data resulted in a higher detection threshold limit $\tau$, while low frequencies flatten the real-time signal too much, decreasing the detection efficiency in that way. For PSL experiments, frequencies of 1000 Hz and 2000 Hz were used, where we conclude that the former one was better regarding the detection efficiency. For the ZINC experiments, we set the lowpass filter frequency to 500 Hz as we expected a higher concentration of larger particles. It was also noted that the detector is not sensitive enough for detecting the aerosol particles of 300 nm in diameter.

The IODE device was then used with the ZINC chamber during the ICIS 2007 workshop in order to see if water droplets and ice crystals can be distinguished during ice nucleation experiments. The idea was to couple the IODE device with the ZINC chamber to have additional information about ice activation onsets and terminations as well as on ice particle concentration. One other task was also to check if ice crystals are still detectable among a large concentration of water droplets (during water breakthrough occurrences). Several activation experiments were discussed for different aerosol types. The first one, where ice activation occurred with a low particle concentration (Snomax® bacteria), the second one was ice activation with a higher particle concentration (Israel dust), the third one with a water breakthrough event (Saharan dust). Activation experiments realized with Canary Islands dust were also discussed. It was possible to identify the start and termination of each activation event with the IODE detector. In the case of ice activation, simultaneous increases in peak intensities in both parallel and perpendicular channels were found during the activation processes. The resulting depolarization ratios showed values ranging from 0.02 up to 0.8, which translates into single ice particle detection in most of the cases according to theory (Nicolet et al. 2007). Considering the Israel dust experiment, it was also possible to determine the activation onset. On the other hand, water droplets were well
determined in the third case (Saharan dust). Even though the particle concentration was very high, causing some saturation problems with the peak detection algorithm, it was possible to identify the presence of liquid particles by using the 0.5-s average real-time values together with their standard deviations. This alternative solution works well for high particle concentrations. A clear increase in the parallel channel was seen, whereas the perpendicular channel signal exhibited no significant rise. It was also shown that the ice particles scatter more light than water droplets and have a broader intensity distribution. Thus, they can be discriminated from water droplets.

It was also possible to determine critical saturation ratios with the IODE detector. The first ice peak event is taken as reference whereas a 1%-activated fraction is taken into account from the OPC raw data. Consequently, the IODE determination for saturation ratios usually preceded the detection by the OPC, occurring at higher temperatures and lower relative humidities than for the OPC. The most visible difference between both detectors is seen during water breakthrough events. It is impossible to distinguish between ice crystals and water droplets with the OPC as detection only is a function of particle size. During these events, we observed a large increase in the total number of counts which is the sum of all detectable particles. The IODE detector can clearly show the presence of ice particle among water droplets (see Fig. 6-15). The sharp increase in the OPC and in the IODE non-depolarizing particle counts occurring when critical ‘breakthrough’ RHw is exceeded is due to activation and subsequent detection of water droplets. Meanwhile, the concentration of ice particles continues to rise up in the same way as in all other activation experiments without water breakthrough occurrences. The water breakthrough conditions do not depend on the aerosol type, but are only a function of the experimental setup of the ZINC chamber and its evaporation section (length, sub-saturation with respect to water). Therefore, it is possible to parametrize it with a polynomial fit.

Some uncertainties remain. First, not every particle can be counted with the device as some particles can pass besides the laser beam and therefore not cross it. The alignment of the laser beam passing through the chamber plays a major role in order to miss as few particles as possible. In the presence of high particle concentrations, several particles may falsely be counted as a single event. In case of ice crystals, some specific particle orientations do not induce a significant depolarization ratio, causing them to be
miscounted as water droplets. A change in the background intensity during experiments can lead to a misinterpretation of the threshold values in the peak detection algorithm. These variations are caused by small displacements of the chamber, owing to the walls cooling at different temperatures: While the warm wall stayed at the same temperature, the cold wall is cooled down, creating a move due to the temperature difference up to 2 mm at the bottom of the chamber. This can cause a miscount of peaks and to apparent background peaks even if there is no activation. For the moment, the readjustments have to be done manually. Finally, particle detection in high concentration conditions is a problem because of a saturation of the detector causing the IODE detector to count significantly less particles than the OPC.
This study allowed us to obtain information about the depolarization ratio of single ice particles in order to investigate if it is theoretically and practically possible to distinguish between ice crystals and water droplets for single particle detection. Different types of ice crystals were modeled with varying aspect ratios and sizes. Moreover, different orientations of the ice particles were studied to infer their influence on depolarization. It was found that the orientation is of major importance, where values of $\delta$ can vary from 0 (no depolarization) to 1 (totally reversed polarization). As the orientation of a particle is directly linked to the angle of incidence between the faces of the crystals and the laser beam, the depolarization ratio depends mainly on this parameter. It is possible to detect an ice crystal in most of the cases with this method. However, detection is not possible in some orientations as they do not cause significant depolarization. Concerning the relevance of assuming cylindrical columns to represent ice crystals in our modeling study, the relative error of the depolarization ratio between cylindrical and hexagonal columns is about 6% for a size parameter of 11.5 and an aspect ratio of unity (Baran et al. 2001). For smaller particles ice cylinders underestimate $\delta_{\text{avg}}$ and for larger ones, $\delta_{\text{avg}}$ is overestimated. This means that we might expect higher values for $\delta$ at $x < 11.5$ and lower values for bigger size parameters. There are still some limitations by comparing the simulated data to experimental data: most remote sensing investigations were done with much larger particles than studied here and retrievals are made at different concentrations of particles where multiple
scattering can play a significant role (Sassen 1974, Sassen and Liou 1979, Gobbi et al. 2000, Gobbi et al. 2003, Sassen et al. 2003, Yang et al. 2003, Iwasaka et al. 2003, Sakai et al. 2003). The differences in scattering angles and wavelengths complicate the comparison even if the same trends of the depolarization ratio with increasing size and shape parameters can be seen.

Concerning the peak detection algorithm, the challenge is to reduce the background signal and its standard deviation as much as possible to have the best possible detection accuracy (i.e. to lower $\tau$). Several improvements can be made to increase this detection accuracy. It is possible to modify the 2-lens optical system to avoid any light scattering from the walls by reducing the optical aperture of the detector. For the first version, the beam divergence was 1.1°, giving a scattered beam diameter close to 10 mm. In this configuration, some signal might have been lost, as the opening aperture of the PMT windows were 8 mm. However, we had to limit the distance between the particles and the detectors ($r$) as much as possible because low light conditions were expected. With the following specifications (focal lengths of 100 mm and 38 mm, pinhole diameter of 0.8 mm), the beam divergence is 0.6° with a scattered beam diameter of 1.2 mm at the level of the PMT windows. Therefore, we can ensure that the scattered rays can totally be centered on the photomultiplier windows without any cross-talk between the two channels. A forward scattering channel is planned to allow counting and sizing the particles with a better efficiency. We have seen that the scattered intensities were low (less than 0.5 pW in the worst cases) in the backward direction. Nevertheless, intensities can be about 2 or 3 orders of magnitude higher in the forward direction ($\Theta = 5^\circ$) (see appendix C for T-matrix modeling results). It is not possible to have depolarization measurements in the forward direction, but making use of data from the forward channel in addition to the backward ones (parallel and perpendicular) allow us to have more reliable information about the particle number concentrations. This will be the scope for further improvements of the detector. However, it is also possible to count aerosol particles as forward detection is more efficient as backward (see appendix C for details). Forward scattering measurements can also be used for particle sizing, but the particles have to be located in the same distance from the detector all the time, which is not the case for the ZINC setup. Another significant improvement is the automation of the threshold limit settings. These are still made and adjusted manually when variations of the background
intensity occur. Finally, having all optical elements in a portable black box can be a good alternative solution. It could be attached directly to the windows of the ZINC chamber. Therefore, the small chamber displacements discussed in chapter 7 would not be important anymore as the optical detector will no longer depend on these movements (as it will not stand on the breadboard).

The main idea is also to get rid of the evaporation section of the ZINC chamber. We have seen that discrimination can be made between ice crystals and water droplets. Moreover, it was possible to detect ice particles among water drops during water breakthrough events and to have quantitative results, too. This is important for further airborne measurements where the ZINC chamber has to be shortened to match the requirements for aircraft use. This PINC chamber (Portable Ice Nucleation Chamber) is built to perform airborne measurements. The improved IODE device will therefore be connected to this chamber for ice particle detection.

Considering the first successful tests and the results obtained during the ICIS 2007 workshop, the IODE device is a promising tool to detect ice crystals and water droplets simultaneously in cloud diffusion chambers. The depolarization method appears to be a powerful tool, as most single ice particles can be detected without directly considering the intensity of the scattered light. Here, this method is performed from the new perspective of detecting single particles where as previous studies only considered a collection of particles of a given concentration. Attaching the IODE detector directly to the ZINC chamber windows (without using a breadboard for the optical elements) can avoid background intensity changes during experiments. Moreover, this technique is promising for laboratory (for instance for cloud diffusion chambers) and airborne instruments when investigating mixed phase clouds where single water droplets and ice crystals can co-exist, which need to be detected and differentiated.
parallel and perpendicular depolarization ratios

Fig. A-1: Perpendicular depolarization ratio for particles with \( d = 2 \, \mu m \) for different aspects ratios as a function of the orientation of the particle given by the Euler angles \( \alpha \) and \( \beta \). The scattering angle \( \Theta \) is 175° and the areas of the figures are non-conservative (see text for details).
Fig. A-2: Parallel and perpendicular depolarization ratios ($\delta_||$ and $\delta_\perp$) for cylinders with an aspect ratio $\Gamma = 1$. The panels represent $\delta_||$ and $\delta_\perp$ for different particle diameters as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$. The scattering angle $\Theta$ is 175° and the areas of the figures are non-conservative (see text for details).
Fig. A-3: Parallel depolarization ratio for particles with $d = 2$ μm for different aspects ratios from 0.3 to 1.6 as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$. The scattering angle $\Theta$ is 175° and the areas of the figures are non-conservative (see text for details).
Fig. A-4: Parallel depolarization ratio for particles with $d = 2\ \mu m$ for different aspects ratios from 1.8 to 3 as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$. The scattering angle $\Theta$ is $175^\circ$ and the areas of the figures are non-conservative (see text for details).
Parallel and perpendicular depolarization ratios

Fig. A-5: Parallel and perpendicular depolarization ratios ($\delta_\parallel$ and $\delta_\perp$) for oblate spheroids (upper panels) and cylinders (lower panels) with an aspect ratio $\Gamma = 2$ and a particle diameter $d = 2 \mu m$. The scattering angle $\Theta$ is $180^\circ$. The panels represent $\delta_\parallel$ and $\delta_\perp$ for different particle diameters as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$. The areas of the figures are non-conservative (see text for details).
Fig. A-6: Parallel and perpendicular depolarization ratios ($\delta_||$ and $\delta_\perp$) for oblate spheroids (upper panels) and cylinders (lower panels) with an aspect ratio $\Gamma = 2$ and a particle diameter $d = 2 \mu m$. The scattering angle $\Theta$ is $175^\circ$. The panels represent $\delta_||$ and $\delta_\perp$ for different particle diameters as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$. The areas of the figures are non-conservative (see text for details).
Fig. A-7: Relative scattered intensity ($Z_{11} + Z_{12}$) for cylinders with an aspect ratio $\Gamma = 1$. The panels represent $Z_{11} + Z_{12}$ for different particle diameters as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$. The scattering angle $\Theta$ is $175^\circ$ and the areas of the figures are non-conservative (see text for details).
Fig. A-8: Relative scattered intensity \((Z_{11}+Z_{12})\) for particles with \(d = 2 \mu m\) for different aspect ratios from 0.3 to 1.6 as a function of the orientation of the particle given by the Euler angles \(\alpha\) and \(\beta\). The scattering angle \(\Theta\) is 175° and the areas of the figures are non-conservative (see text for details).
Fig. A-9: Relative scattered intensity \((Z_{11}+Z_{12})\) for particles with \(d = 2 \mu m\) for different aspect ratios from 1.8 to 3 as a function of the orientation of the particle given by the Euler angles \(\alpha\) and \(\beta\). The scattering angle \(\Theta\) is 175° and the areas of the figures are non-conservative (see text for details).

Fig. A-10: Relative scattered intensity \((Z_{11}+Z_{12})\) as a function of the orientation of the particle given by the Euler angles \(\alpha\) and \(\beta\) for an oblate spheroid of diameter \(d = 2 \mu m\) and aspect ratio \(\Gamma = 2\) \((\Theta = 180°)\). The areas of the figures are non-conservative (see text for details).
Fig. A-11: Relative scattered intensity ($Z_{11} + Z_{12}$) as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$ for an oblate cylinder of diameter $d = 2 \, \mu\text{m}$ and aspect ratio $\Gamma = 2$ ($\Theta = 180^\circ$). The areas of the figures are non-conservative (see text for details).

Fig. A-12: Relative scattered intensity ($Z_{11} + Z_{12}$) as a function of the orientation of the particle given by the Euler angles $\alpha$ and $\beta$ for an oblate spheroid of diameter $d = 2 \, \mu\text{m}$ and aspect ratio $\Gamma = 2$ ($\Theta = 175^\circ$). The areas of the figures are non-conservative (see text for details).
Fig. A-13: Relative scattered intensity \((Z_{11}+Z_{12})\) as a function of the orientation of the particle given by the Euler angles \(\alpha\) and \(\beta\) for an oblate spheroid of diameter \(d = 2\ \mu\text{m}\) and aspect ratio \(\Gamma = 0.5\ (\Theta = 175^\circ)\). The areas of the figures are non-conservative (see text for details).
appendix B

The IODE Detector Windows

Fig. B-1: Pictures of the windows mounted on the KF flange for the backscattering direction.
Fig. B-2: Pictures of the windows mounted on the KF flange for the forward scattering direction.
## T-MATRIX RESULTS FOR LATEX SPHERES AND WATER DROPLETS

### Table C-1

| $\Theta$ | $d_{eq}$ [µm] | $\Gamma$ | $\delta_{||}^{\text{avg}}$ (σ) | $\delta_{\perp}^{\text{avg}}$ (σ) | $Z_{11}+Z_{12}$ (σ) |
|----------|----------------|--------|-------------------------------|-------------------------------|-------------------|
| 180°     | 0.5            | 1      | 0.015 (0.000)                 | 0.015 (0.000)                 | 0.005 (0.0000)    |
| 180°     | 0.8            | 1      | 0.097 (0.018)                 | 0.097 (0.018)                 | 0.016 (0.0000)    |
| 180°     | 1              | 1      | 0.151 (0.030)                 | 0.151 (0.030)                 | 0.031 (0.0002)    |
| 180°     | 1.2            | 1      | 0.192 (0.037)                 | 0.192 (0.037)                 | 0.049 (0.0010)    |
| 180°     | 1.6            | 1      | 0.250 (0.041)                 | 0.250 (0.041)                 | 0.112 (0.0057)    |
| 180°     | 2              | 1      | 0.287 (0.050)                 | 0.287 (0.050)                 | 0.191 (0.0183)    |
| 180°     | 2.5            | 1      | 0.265 (0.043)                 | 0.265 (0.043)                 | 0.344 (0.0686)    |
| 180°     | 3              | 1      | 0.239 (0.042)                 | 0.239 (0.042)                 | 0.568 (0.2401)    |
| 180°     | 3.5            | 1      | 0.224 (0.036)                 | 0.224 (0.036)                 | 0.809 (0.5549)    |
| 180°     | 4              | 1      | 0.219 (0.030)                 | 0.219 (0.030)                 | 1.154 (0.9127)    |

Table C-1: Values of the parallel and perpendicular depolarization ratios $\delta_{||}^{\text{avg}}$ and $\delta_{\perp}^{\text{avg}}$ of isometric ice cylinders ($\Gamma = 1$) for different particle diameters at a perfect backscattering angle ($\Theta = 180°$). $Z_{11}+Z_{12}$ shows the relative scattered intensity averaged over all orientations.

### Table C-2

| $\Theta$ | $d_{eq}$ [µm] | $\Gamma$ | $\delta_{||}^{\text{avg}}$ (σ) | $\delta_{\perp}^{\text{avg}}$ (σ) | $Z_{11}+Z_{12}$ (σ) |
|----------|----------------|--------|-------------------------------|-------------------------------|-------------------|
| 180°     | 2              | 2      | 0.336 (0.065)                 | 0.336 (0.065)                 | 0.118 (0.0053)    |
| 180°     | 2              | 2      | 0.372 (0.074)                 | 0.372 (0.074)                 | 0.143 (0.0130)    |
| 175°     | 2              | 2      | 0.248 (0.043)                 | 0.325 (0.062)                 | 0.059 (0.0010)    |
| 175°     | 2              | 0.5    | 0.269 (0.041)                 | 0.279 (0.042)                 | 0.050 (0.0016)    |

Table C-2: Values of the parallel and perpendicular depolarization ratios $\delta_{||}^{\text{avg}}$ and $\delta_{\perp}^{\text{avg}}$ of 2 µm-diameter ice cylinders for different particle aspect ratios. $Z_{11}+Z_{12}$ shows the relative scattered intensity averaged over all orientations. Calculations were made for spheroids (1) and cylinders (2). $\Theta$ is the scattering angle.
Table C-3: Relative scattered intensity \((Z_{11} + Z_{12})\) of PSL latex spheres \((n = 1.59)\) as a function of the particle diameter. Calculations were made considering a wavelength of 407 nm and a scattering angle of 175°.

<table>
<thead>
<tr>
<th>Diameter (µm)</th>
<th>(Z_{11})</th>
<th>(Z_{12})</th>
<th>(Z_{11} + Z_{12})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.67</td>
<td>0.0885</td>
<td>0.0038</td>
<td>0.0923</td>
</tr>
<tr>
<td>1</td>
<td>0.8258</td>
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<td>0.8941</td>
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<td>0.0672</td>
<td>0.9653</td>
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<td>0.3637</td>
<td>0.8395</td>
</tr>
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<td>0.6062</td>
<td>0.5144</td>
<td>1.1206</td>
</tr>
<tr>
<td>3</td>
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<td>-0.2246</td>
<td>1.0356</td>
</tr>
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<td>3.5</td>
<td>3.4130</td>
<td>-1.3814</td>
<td>2.0316</td>
</tr>
<tr>
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<td>-4.2575</td>
<td>0.2436</td>
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<tr>
<td>4.5</td>
<td>9.1775</td>
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<td>2.3658</td>
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<td>-4.5138</td>
<td>7.8103</td>
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<td>5.5</td>
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<tr>
<td>7</td>
<td>3.8888</td>
<td>0.2215</td>
<td>4.1103</td>
</tr>
</tbody>
</table>

Table C-4: Relative scattered intensity \((Z_{11} + Z_{12})\) of PSL latex spheres \((n = 1.59)\) as a function of the particle diameter. Calculations were made considering a wavelength of 407 nm and a scattering angle of 5°.

<table>
<thead>
<tr>
<th>Diameter (µm)</th>
<th>(Z_{11})</th>
<th>(Z_{12})</th>
<th>(Z_{11} + Z_{12})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.67</td>
<td>1.5349</td>
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<tr>
<td>1</td>
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<td>1.5</td>
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<td>137.6092</td>
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<tr>
<td>7</td>
<td>286.0461</td>
<td>-3.0057</td>
<td>283.0404</td>
</tr>
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</table>
### Table C-5: Relative scattered intensity ($Z_{11}+Z_{12}$) of water droplets ($n = 1.343$) as a function of the particle diameter. Calculations were made considering a wavelength of 407 nm and a scattering angle of 175°.

<table>
<thead>
<tr>
<th>diameter (µm)</th>
<th>$Z_{11}$</th>
<th>$Z_{12}$</th>
<th>$Z_{11} + Z_{12}$</th>
</tr>
</thead>
<tbody>
<tr>
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<tr>
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<td>0.1038</td>
</tr>
<tr>
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<td>0.2209</td>
<td>0.7272</td>
</tr>
<tr>
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<td>0.2270</td>
<td>0.0618</td>
<td>0.2888</td>
</tr>
<tr>
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<td>-0.2200</td>
<td>0.4521</td>
</tr>
<tr>
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<td>-0.4375</td>
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</tr>
<tr>
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<tr>
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<td>2.3845</td>
</tr>
<tr>
<td>7</td>
<td>0.6035</td>
<td>-0.4528</td>
<td>0.1507</td>
</tr>
</tbody>
</table>

### Table C-6: Relative scattered intensity ($Z_{11}+Z_{12}$) of water droplets ($n = 1.343$) as a function of the particle diameter. Calculations were made considering a wavelength of 407 nm and a scattering angle of 5°.

<table>
<thead>
<tr>
<th>diameter (µm)</th>
<th>$Z_{11}$</th>
<th>$Z_{12}$</th>
<th>$Z_{11} + Z_{12}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.67</td>
<td>2.7338</td>
<td>-0.0017</td>
<td>2.7321</td>
</tr>
<tr>
<td>1</td>
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<td>9.1443</td>
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<tr>
<td>1.5</td>
<td>10.5745</td>
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<tr>
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<td>80.9930</td>
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<td>50.6750</td>
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<tr>
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<td>131.2693</td>
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<tr>
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<td>134.8836</td>
</tr>
<tr>
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<td>-2.7505</td>
<td>71.8120</td>
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<tr>
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<td>121.0611</td>
<td>-5.6547</td>
<td>115.4064</td>
</tr>
<tr>
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<td>-3.9366</td>
<td>0.6946</td>
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<td>4.0418</td>
<td>36.1015</td>
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<td>0.7854</td>
<td>76.8409</td>
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<td>126.6022</td>
<td>-8.8607</td>
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<tr>
<td>7</td>
<td>154.2684</td>
<td>-4.4500</td>
<td>149.8184</td>
</tr>
</tbody>
</table>
Fig. D-1: Software interface of the peak detection algorithm designed with LabView. From up to down, the 1st panel shows the real-time parallel and perpendicular signals, the 2nd panel depicts the peaks found in both channel, the 3rd panel indicates the depolarization peaks obtained from the parallel and perpendicular peaks and the 4th panel shows the average real-time signal calculated every 0.5 second.
Fig. E-1: Real-time data of the parallel channel filtered with a lowpass filter of different frequencies from 75 Hz to 1500 Hz.
Fig. F-1: Scheme of the Snomax® 4th activation experiment (SB4).
Fig. F-2: Scheme of the Snomax® 7th activation experiment (SB7).
Fig. F-3: Scheme of the Snomax® 11th activation experiment (SB11).
Fig. F-4: Scheme of the Saharan dust 6th activation experiment (SD6).
Fig. F-5: Scheme of the Israel dust 8th activation experiment (ID8).
Fig. F-6: Scheme of the Canary Islands dust 2nd activation experiment (CID2).
Other activation schemes

Fig. F-7: Scheme of the Canary Islands dust 7th activation experiment (CID7).
REFERENCES


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CURRICULUM VITAE

Last name        NICOLET
First name       Mathieu

Date of birth    20th of July 1979
Birthplace       Schiers, Switzerland
Citizen          Cugy (FR), Switzerland

Country          Switzerland and Czech Republic
Profession       Engineer in environmental sciences
Languages        French
                  English (spoken and written)
                  German (good notions)

Formation

1995 – 1998     Certificate of Maturity in Science, College of
                Chamblandes, Lausanne.

1998 – 2004     Studies at the Swiss Federal Institute of Technology,
                Lausanne (science of environment)

2004            Diploma in environment engineering
                (Implementation of depolarisation measurements at
                355 nm on the Jungfraujoch LIDAR)

                Institute of Technology, Zurich (Institute for Atmospheric
                and Climate Science)