Doctoral Thesis

Sidebranch development of xenon dendrites

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Sidebranch Development of Xenon Dendrites

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Abstract

In our experiments, we investigate in situ three-dimensional dendrites during their growth into a pure melt of supercooled xenon. The supercooling, $\Delta T$, is in the range $20 \text{ mK} \leq \Delta T \leq 220 \text{ mK}$. In the studies presented here, we focus on the region where the sidebranches grow. We scale all lengths of a dendrite by the experimentally determined tip radius, $R_{\text{exp}}$. If scaled by $R_{\text{exp}}$, dendrites grown at different $\Delta T$ look similar in shape, i.e., there does not exist any criterion to deduce $\Delta T$ from a normalized shape of a dendrite. It is found that far behind the tip, the width $d$ of the trunk of a dendrite increases linearly with distance $z$ to the tip. At $z = 30 R_{\text{exp}}$, the width of the trunk is $d_{30} = (18 \pm 2) R_{\text{exp}}$. Replacing $R_{\text{exp}}$ by an effective tip radius, $R_{\text{eff}}$, yields a scaling relation between the width of the trunk, $d$, and $R_{\text{eff}}$ at any distance $z$ from the tip, $d = (18\pm2) R_{\text{eff}}$. It is found that the linear increase of $R_{\text{eff}}$ with $z$ corresponds to a temperature increase along the dendrite. Along the dendrite, the temperature rises due to the latent heat released during growth. This thermometric property of $d$ is verified experimentally over the whole range of the supercooling, $\Delta T$.

With increasing distance from the tip, some of the sidebranches disappear by remelting or sintering together, i.e., dynamical coarsening takes place. The sidebranches which survive coarsening determine the envelope of the dendrite. It is found that the shape of the envelope is constant in time. The opening angle of the envelope is determined to $\Phi = 25^\circ \pm 3^\circ$ and is independent of $\Delta T$. The growth velocity of the sidebranches contributing to the envelope is determined to $v_{\text{SB}} = (0.5 \pm 0.05) v_{\text{tip}}$, where $v_{\text{tip}}$ is the growth velocity of the dendrite tip. It is found that $v_{\text{SB}}$ is constant in time, i.e., a sidebranch does not accelerate when one of its neighbors melts back.

Close to the tip, the sidebranches are approximately periodic with spacing $\lambda \approx 3.2 R_{\text{exp}}$. Due to coarsening, the sidebranch spacing increases with increasing distance to the tip. It is found that this increase occurs in discrete
steps and that with increasing distance to the tip, the smaller spacings are taken over by bigger ones. The results of our investigations of the coarsening process are compatible with the scenario of period doubling. Together with the observed sensitivity of the temporal development of the sidebranch spacing to initial conditions, this indicates that dendritic crystallization might be an example for a natural, chaotic system.
Kurzfassung

In unseren Experimenten untersuchen wir in situ dreidimensionale Dendriten während sie in eine reine Schmelze aus Xenon hineinwachsen. Die Unterkühlung $\Delta T$ der Schmelze liegt im Bereich $20 \text{ mK} \leq \Delta T \leq 220 \text{ mK}$. In den hier vorgestellten Untersuchungen konzentrieren wir uns auf den Bereich des Dendriten, wo die Seitenäste wachsen. Wir skalieren alle Längen eines Dendriten mit dem experimentell bestimmten Spitzenradius $R_{\text{exp}}$. Dendriten, die bei verschiedenen $\Delta T$ gewachsen sind, sind einander ähnlich, d.h. es existiert kein Kriterium, um von der Form eines skalierten Dendriten auf $\Delta T$ zu schließen. Unsere Messungen zeigen, dass die Breite $d$ des Stammes weit hinter der Dendritenspitze mit der Entfernung $z$ zur Spitze linear zunimmt. Für $z = 30 R_{\text{exp}}$ beträgt die Breite des Stammes $d_{30} = (18 \pm 2) R_{\text{exp}}$. Ersetzt man $R_{\text{exp}}$ durch einen effektiven Spitzenradius $R_{\text{eff}}$, so erhält man eine Skalierungsbeziehung zwischen $d$ und $R_{\text{eff}}$ für jede Distanz $z$ zur Dendritenspitze, $d = (18 \pm 2) R_{\text{eff}}$. Es hat sich herausgestellt, dass die lineare Zunahme von $R_{\text{eff}}$ mit $z$ einer Temperaturzunahme entlang des Dendriten entspricht. Entlang des Dendriten nimmt die Temperatur zu aufgrund der latenten Wärme, die während des Wachstums freigesetzt wird. Diese thermometrische Eigenschaft von $d$ konnten wir über den ganzen Bereich der Unterkühlung $\Delta T$ verifizieren.

Mit zunehmender Distanz zur Dendritenspitze verschwinden einige Seitenäste indem sie zurückschmelzen oder zusammenwachsen, d.h. es findet eine dynamische Vergrößerung statt. Die Seitenäste, die die Vergrößerung überstehen, bestimmen die Einhüllende des Dendriten. Unsere Messungen zeigen, dass die Form der Einhüllenden zeitlich konstant ist. Der Öffnungswinkel $\Phi$ der Einhüllenden wurde bestimmt zu $\Phi = 25^\circ \pm 3^\circ$ und ist unabhängig von $\Delta T$. Die Wachstumsgeschwindigkeit $v_{SB}$ der Seitenäste, die zur Einhüllenden beitragen, wurde bestimmt zu $v_{SB} = (0.5 \pm 0.05) v_{\text{tip}}$, wobei $v_{\text{tip}}$ die Wachstumsgeschwindigkeit der Dendritenspitze ist. $v_{SB}$ ist zeitlich
konstant, d.h. ein Seitenast zeigt keine Wachstumsbeschleunigung, wenn einer seiner Nachbarn zurück schmilzt.

1. Introduction

Dendritic solidification is an example for the formation of crystals with complex spatial patterns at conditions far from equilibrium. A well-known example for dendrites are snow crystals. Although there are no two snow crystals with the same shape, snow crystals grown at the same conditions of temperature and humidity show the same characteristics, i.e., depending on the solidification conditions they are hexagonal columns or plates or dendrites, etc. [1]. In the case of snow crystals, there are two control parameters which influence the growth process, the concentration of water in the air and the temperature [2]. The simplest case of dendritic solidification is given by the solidification of a pure liquid held at a temperature slightly below its equilibrium freezing point temperature, $T_m$. As it is difficult to control the temperature in the region close to a growing dendrite, usually a special geometry is chosen which can be treated in calculations and can be verified in good approximation in experiments also. In this geometry [3], the dendrite is assumed to grow freely into an infinite volume of melt which has been set to the temperature $T_\infty < T_m$ before the dendrite starts to grow. In this case, the supercooling, $\Delta T = T_m - T_\infty$, is the only control parameter of the system. The latent heat is transported off the growing dendrite into the supercooled melt by diffusion. It is assumed that the growth is limited by this diffusion process only and that kinetics effects are fast. In this case, the growth of a crystal can be described by a solution of the diffusion equation in three dimensions [4, 5]

$$\frac{\partial u}{\partial t} = D\nabla^2 u$$

(1.1)

with boundary conditions given below. $D$ is the thermal diffusivity. Instead of the local temperature, $T$, the dimensionless diffusion field,

$$u \equiv \frac{T - T_\infty}{L/c_p},$$

(1.2)
is used for the mathematical description. This temperature \( u \) is normalized by the unit supercooling, defined as the ratio of the latent heat, \( L \), and the specific heat, \( c_p \). In general, \( u \) satisfies Eq. (1.1) separately in the liquid and in the solid with the respective values of \( D \). In the symmetric model [4], where the material properties in the liquid and in the solid are assumed to be the same, only one diffusion equation has to be solved. This model greatly simplifies some of the mathematics without losing too many important features. Therefore, in the following we restrict our discussion to the symmetric model.

The condition of energy conservation at the moving interface leads to the continuity equation [4]

\[
\mathbf{v}_n = D \left( \nabla u_{\text{solid}} - \nabla u_{\text{liquid}} \right) \cdot \mathbf{n},
\] (1.3)

where \( \mathbf{n} \) is the unit vector normal to the interface, directed into the liquid, and \( \mathbf{v}_n = \mathbf{v} \cdot \mathbf{n} \) is the velocity component normal to the interface. The subscripts indicate on which side of the interface the values of the diffusion field are to be taken. The left-hand side of Eq. (1.3) accounts for the rate at which heat is generated at the boundary, and the right-hand side for the heat flow into the bulk phases on either side of the interface.

Finally, at the interface the boundary condition is [4]

\[
u_n \big|_{\text{interface}} = \Delta - d_0 \kappa - \beta v_n.
\] (1.4)

On the right-hand side, the first term is the dimensionless measure of the supercooling,

\[
\Delta = \frac{\Delta T}{L/c_p}.
\] (1.5)

The second term on the right-hand side of Eq. (1.4) accounts for the deviation of the melting temperature of a curved interface from \( T_m \), known as Gibbs-Thomson effect [6]. The local curvature of the interface, \( \kappa \), is positive for a convex surface, i.e., if the solid bulges into the liquid. The capillary length is defined as

\[
d_0 = \frac{\gamma T_m c_p}{L^2},
\] (1.6)
where $\gamma$ is the interface free energy. The capillary length, $d_0$, is typically of the order of Angstroms and represents a length scale of the dendrite. The third term on the right-hand side of Eq. (1.4) is proportional to the growth rate at the interface.

Eqs. (1.1), (1.3) and (1.4) specify completely the symmetric model of dendritic solidification of a pure substance. The equation of motion for the surface of a dendrite is derived from these equations by eliminating the diffusion field, $u$, in favor of coordinates which describe the position of the moving boundary. The position of the interface is part of the boundary condition and of the solution of the diffusion equation. Therefore, the determination of the shape of a growing dendrite is a moving-boundary problem.

Ivantsov [7] has found that a rotational-symmetric, parabolic needle-crystal is a solution of the moving-boundary problem given by Eqs. (1.1), (1.3) and (1.4). This paraboloid of revolution can be characterized by the radius $R_\text{iv}$ of curvature and the growth velocity $v_\text{tip}$ of the tip. There are two main reasons, why this solution is not satisfactory:

i) For a given $\Delta T$, the Ivantsov solution predicts only a value for the product $R_\text{iv} v_\text{tip}$. It is an experimental fact that for a given $\Delta T$, unique values for the tip radius and $v_\text{tip}$ are selected [8, 9]. The prediction of these two parameters is the so-called selection problem.

ii) In the tip region where no sidebranches are formed, dendrites do not have the shape of a rotational paraboloid [10].

Two steps in the development of the theory of the selection problem may be mentioned, the marginal stability theory and the solvability theory. In the marginal stability theory [11, 12, 13], it is assumed that the operating point of the dendrite occurs where the tip is just marginally stable. This additional condition allows to select the tip radius and the growth velocity of the tip at a given supercooling. The solvability theory for axisymmetric dendrites [4, 14, 15] leads to the result that the anisotropy in the surface tension acts as a singular perturbation, selecting uniquely the shape and the growth velocity of the tip. The selection of the shape and the growth velocity for three-dimensional dendritic crystals of materials with cubic symmetry has
been studied by Ben Amar and Breuer [16]. In the harmonic approximation, capillary effects lead to an nonaxisymmetric shape correction of a rotational paraboloid.

The tip represents only a small part of the whole dendrite and the liquid surrounding the tip is metastable with respect to freezing. In the case of diffusional limited growth, this may lead potentially to a Mullins-Sekerka instability. Mullins and Sekerka [17] have shown that a plane interface, moving at a constant velocity into a metastable phase is unstable against disturbances larger than some characteristic stability length. The interfacial pattern is characterized by the wavelength corresponding to the disturbance growing fastest. Starting with an axisymmetric dendrite, such an instability can initiate in a first step the growth of fins along the dendrite [18, 19]. In a second step, a second instability of the Mullins-Sekerka type destabilizes these fins and sidebranches start to grow at the ridges of these fins.

In Ref. [20], Breuer has studied the contour in the region of the dendrite where four fins are observed in experiments. The calculations have been performed in dimensionless coordinates, where all of the lengths are scaled by the tip radius. In difference to lengths measured in SI-units, all of the dimensionless, relative lengths are denoted with \( \hat{\cdot} \), e.g. \( \hat{x}, \hat{z} \), etc. For materials with cubic symmetry, Breuer has found an analytical solution for the contours of these fins of the form

\[
\hat{z} \propto |\hat{x}|^{\frac{3}{\lambda}},
\]

where the \( z \)-axis is along the growth direction of the dendrite tip and the \( xz \)-plane contains the ridges of two of the four fins.

It has to be mentioned that there exist various tip radii used for scaling in different situations, e.g. the tip radius \( R_{iv} \) of an Ivantsov paraboloid of revolution, the experimentally determined tip radius, \( R_{exp} \), the tip radius \( R_{LM-K} \) in the theory of marginal stability by Langer and Müller-Krumhhaar [11, 12, 13], and so on. Therefore, in each situation one has to consider which tip radius has been used for scaling.

Experimental studies with xenon show that in the tip region, the shape
of three-dimensional dendrites can be described by the tip function [10]

\[ \hat{z} = (0.58 \pm 0.04)|\hat{x}|^{1.67 \pm 0.05}. \] (1.8)

Here, the experimental data have been scaled with the experimentally determined tip radius, \( R_{\text{exp}} \neq R_{\text{iv}} \). Eq. (1.8) is in agreement with the analytical result given in Eq. (1.7). It has been found that in the region of the tip, the experimentally determined shape of a dendrite cannot be fitted by azimuthal harmonics [21].

In Ref. [22], Brener and Temkin have investigated the time-dependent behavior of sidebranching deformations by taking into account the nonaxisymmetric shape of a needle crystal with the result that in a frame of reference moving with the dendrite tip, noise-induced wave packets generated in the tip region move along the sides of the dendrite away from the tip while growing in amplitude and spreading. Brener and Temkin have found that for the anisotropic needle crystal the amplitudes of the sidebranches grow faster than in the rotational symmetric case discussed by Langer [23]. In experiments with three-dimensional xenon dendrites, Bisang and Bilgram [10] have found that at a distance \( z \approx 18R_{\text{exp}} \) from the dendrite tip, the amplitude of the sidebranches is about \( 1R_{\text{exp}} \). This experimental result agrees quantitatively and without any fitting parameter with the theoretical predictions by Brener and Temkin. From this experiment it has been concluded that the formation of sidebranches is initiated by thermal fluctuations [10].

Close to the tip, the sidebranches are approximately periodic with a mean spacing [9]

\[ \lambda = (3.2 \pm 0.4)R_{\text{exp}}. \] (1.9)

With increasing distance to the tip, the sidebranches and the trunk of the dendrite are thermally decoupled from the environment and behave like a closed system [5]. Therefore, due to the latent heat released during growth, between the sidebranches the temperature of the liquid is bigger than \( T_{\infty} \) far away from the dendrite. Furthermore, in the region of the sidebranches, the curvature of the surface depends strongly on the position. Due to the Gibbs-Thomson effect, for a convex surface with a large curvature the melting temperature is lower than for such a surface with small curvature and the
Figure 1.1: Contours of a dendrite grown at a supercooling of $\Delta T = 95.8 \text{ mK}$, plotted in a Cartesian frame of reference moving with the dendrite tip. a) One contour of the dendrite. b) A second contour, taken 20 s later is superimposed as a dashed line on the first one. c) Superposition of 300 contours of the same dendrite. The contours are taken during 20 min and the last one is plotted by a white dashed line. In the superposition, the movement of the sidebranches away from the tip leads to two black triangles at either side of the dendrite.

Contrary is the case for a concave surface. This leads to a heat flow between differently curved sidebranches such as the smaller sidebranches disappear by remelting or sintering together, i.e., dynamical coarsening takes place. The study of this coarsening process is part of this work. We will show that the temporal development of the sidebranch spacing is sensitive to initial conditions and that this spacing increases with increasing distance to the tip in discrete steps in compatibility to the scenario of period doubling. However, the physics of the processes taking place during coarsening is only partially understood. For a review on coarsening see Ref. [24].

In our experiments, we investigate three-dimensional xenon dendrites during free growth into a pure, supercooled melt. In the studies presented here, we investigate with high temporal and spatial resolution the region far behind the dendrite tip, where the sidebranches are significant. Fig. 1.1 illustrates the temporal development of a dendrite. In Fig. 1.1a), a contour of a dendrite grown in one of our experiments is shown. We place all of the contours in a frame of reference moving with the tip. In Fig. 1.1b), a second contour of the same dendrite, taken 20 seconds later is superimposed on the first contour.
as a dashed line. Whereas in the tip region the two contours match exactly, far from the tip the contours do not coincide because during the 20 seconds the sidebranches have moved away from the tip. In Fig. 1.1c) finally, 300 superimposed contours of the same dendrite are shown. The last contour is plotted by a white dashed line. This figure reveals three features of the shape of a dendrite.

i) In the tip region, the superimposed contours describe a sharp line. This means, that the shape of the tip stays very stable in time.

ii) The movement of the sidebranches away from the tip leads to two black, well defined triangles on either side of the dendrite. The inner sides of the triangles determine the trunk. The width of the trunk corresponds to the width of the fins but not to the diameter of a rotational cylinder.

iii) The dendrite envelope is determined by the outer sides of the black triangles.

 Whereas the tip and the tip region of a dendrite have been studied in some detail [15, 25, 20], only a few experiments and theoretical studies exist for the region far from the tip. New insight is achieved not only into the coarsening process, but also into the temperature dependence of the width of the trunk and into the shape of the envelope. We will show that the width of the trunk has a thermometric property which can be used to characterize the temporal development of a dendrite. The dendrite trunk is not only an important feature of a freely growing dendrite, it is also a fundamental part of microstructures found in cast alloys [26, 27, 28, 29]. Therefore, apart from its significance in basic research on dendritic solidification, the dendrite trunk is of practical importance too. The shape of the envelope will be shown to be constant in time and independent of temperature, leading to a constant growth velocity of the sidebranches contributing to the envelope. It will be shown that the properties deduced from xenon dendrites can be used to characterize metallic dendrites in cast alloys.

In Sec. 2, we show how to grow dendrites and how we obtain their contours. In Sec. 3, we deduce quantitative results about the shape parameters
of a dendrite in the region far from the tip. In Sec. 4, we compare our results with features found for different materials and solidification techniques.
2. Experiments

In our experiments, we observe in situ the growth of three-dimensional dendrites into a pure melt of supercooled xenon. The experimental setup, depicted in Fig. 2.1, is similar to the one used in previous experiments and a detailed description is given in Ref. [25]. In order to investigate the development of the sidebranches, we have optimized the optical system (illumination tube, periscope) to picture a large region behind the dendrite tip and implemented a linear motion drive to lift the capillary.

For our studies of dendritic solidification we use xenon. Xenon is used as a transparent model substance for metals. In Sec. 4, we will come back to this aspect. In App. A, selected properties of xenon are listed (Tab.A.1). In a liquid nitrogen bath cryostat, the growth vessel is immersed completely into a heat bath (isopentane). Its volume is large enough (≈ 100 cm³) to ensure a free growth of the dendrite. We purify the xenon to 99.9999% before each experiment. The pure melt is supercooled in the range 20 mK ≤ ΔT ≤ 220 mK and during the experiment, the temperature is stabilized to better than ±10⁻⁴ K.

The growth of the dendrite is initiated by means of the capillary injection technique [3]. The growing dendrite is illuminated homogeneously by monochromatic light (λ = 546 nm) and imaged to the chip of a digital camera (768 pixel × 576 pixel) outside the cryostat by means of a periscope. We record the pictures of the growing dendrite on a video tape.

In our experiments, the growth directions of the dendrites are tilted from the direction of the earth gravitational field by an angle in the range 0° ≤ φ ≤ 60°. At the beginning of the free growth of a dendrite, we orient the dendrite by rotating the capillary such as to turn two of the four fins into the object plane of the periscope. A mechanism for a vertical motion of the capillary allows us to move the tip or any other region of the dendrite into the center of the object plane of the periscope. The occasional lifting of the
Figure 2.1: Experimental setup. 1, growth vessel; 2, periscope; 3, illumination system; 4, heat bath (isopentane); 5, tube to provide a laminar flow in the heat bath; 6, stirrer; 7, big mass of stainless steel to reduce the vibrations of the stirrer; 8, liquid nitrogen; 9, adjustable helium gas atmosphere to control the cooling power; 10, heater; 11, temperature sensor; 12, combined rotary and linear motion drive to lift and rotate the capillary; 13, zoom (1x7); 14, CCD-camera; 15, power supply for the camera and output to the video recorder.
capillary does not disturb significantly our measurements.

From a video tape with pictures of a growing dendrite, we digitize up to $N = 300$ frames on a computer in constant time steps $2 \text{s} \leq \Delta t \leq 120 \text{s}$, depending on the growth velocity of the dendrite. From such a digitized picture, as shown in Fig. 2.2a), we extract the contour of the dendrite automatically, i.e., without any influence of the operator. In Fig. 2.2a), the extracted contour of the dendrite is shown by the white line. The optical magnification is calibrated with the outer diameter of the capillary (290 $\mu$m $\pm$ 5 $\mu$m). The precisions $\delta x$ and $\delta z$ of the contour coordinates are $\pm 1$ pixel, which is typically of the order of 10 $\mu$m in the laboratory frame. The optical resolution of the periscope is 1 $\mu$m. Finally, we bring the contour into a coordinate frame of reference, where the dendrite tip stays in the origin and the growth axis is oriented along the $z$–axis as shown in Fig. 2.2b). Contours prepared in that way are the raw data for our investigations. In Tab. B.1 (see App. B), quantitative experimental properties and results of the investigated dendrites are listed.
3. Analysis and Results

In Fig. 3.1, selected shape and growth parameters are depicted characterizing a dendrite. The solid line shows a contour of a dendrite. The tip with the radius $R$ of curvature grows along the main axis of the dendrite with velocity $v_{\text{tip}}$. The growth direction corresponds to the crystallographic [001] direction. The sidebranches grow at a velocity $v_{\text{SB}}$ along the direction of the maximum thermal gradient. This direction is not orthogonal to the main axis of the dendrite but tilted towards the tip by the sidebranch orientation angle $\alpha$. From direct measurement in various contours, we estimate that in our experiments for all of the dendrites the orientation angle of the sidebranches is $\alpha \approx 10^\circ$. Thermal gradients of Ivantsov paraboloids have been calculated by Horvay and Cahn [30]. By considering such a paraboloid as an approximation to the dendrites grown in our experiments, the directions of the calculated, maximum thermal gradients can be compared with the orientations of the sidebranches observed on our dendrites with the result that both have approximately the same orientation with respect to the main axis of the crystals. The parameter $d$ corresponds to the width of the trunk and $\Phi$ is the opening angle of the envelope. Close to the tip, the mean spacing between the sidebranches is $\lambda \approx 3.2R_{\text{exp}}$, Eq. (1.9).

In the studies presented here, we scale the lengths obtained from our experiments by the experimentally determined tip radius, $R_{\text{exp}}$ [9]. The tip function given in Eq. (1.8) does not define a smooth function at $\hat{z} = 0$ and a parabola cannot be fitted in a unique way to a contour. Fitting a parabola to a contour leads to a tip radius decreasing with decreasing fitting height $H$. $R_{\text{exp}}$ is obtained by extrapolating the fitted tip radius to $H \rightarrow 0$ [9]. A correlation between $R_{\text{exp}}$ and the tip function given in Eq. (1.8) has been deduced by Bisang and Bilgram [25]. $R_{\text{exp}}$ decreases with increasing supercooling $\Delta T$ as [9]

$$R_{\text{exp}} = (5.2 \pm 0.4) \Delta T^{-0.83\pm0.03}$$

(3.1)
Figure 3.1: Selected shape and growth parameters characterizing a dendrite. The dendrite tip can be characterized by the radius of curvature, $R$, and the growth velocity $v_{\text{tip}}$ of the tip and the sidebranches by their orientation angle, $\alpha$, their growth velocity, $v_{\text{SB}}$, and close to the tip, where the sidebranches are formed approximately periodically, by the sidebranch spacing, $\lambda$. The opening angle of the envelope, $\Phi$, and the width of the trunk, $d$, describe the integral shape of a dendrite far from the tip.

where $R_{\text{exp}}$ is given in $\mu$m and $\Delta T$ in K.

The scaling property of $R_{\text{exp}}$ is illustrated in Fig. 3.2, where four contours of dendrites, grown at different supercoolings, $\Delta T$, are depicted side by side and superimposed on top of each other. The angle $\phi$ is the angle between the direction of the earth gravitational field and the main axis of the corresponding dendrite during the experiment. In this figure, the lengths are scaled by the respective tip radii, $R_{\text{exp}}$, and the left-hand side of each contour corresponds to the lower side of the dendrite. The supercoolings of the four dendrites vary by a factor of about 10. The sidebranches evolve at $\tilde{\varepsilon} \approx 18$ with a spacing $\tilde{\lambda} \approx 3.2$, Eq. (1.9). Furthermore, in the superposition the contours of all 4 dendrites match in the tip region and the trunks and
Figure 3.2: Scaling property of the tip radius. Contours of four dendrites grown at different supercoolings, $\Delta T$, are depicted side by side and superimposed on top of each other. The lengths are scaled with the experimentally determined tip radii, $R_{\text{exp}}$ [9]. The angle between the direction of the earth gravitational field and the growth direction of the dendrite tip during the experiment is given by $\phi$. The left-hand side of each contour corresponds to the lower side of the dendrite.

$\Delta T$ [mK] 217.5 119.1 50.7 23.6  
$R_{\text{exp}}$ [\mu m] 18.5 30.4 61.8 116.7  
$\phi$ [deg] 23.6 9.7 30.4 50.4

A closer look at this figure reveals that the sidebranches at the lower side of the dendrites are slightly bigger than at the upper side. For all of these dendrites, the angle $\phi$ between their main axis and the direction of the earth gravitational field has been finite. In our experiments, convection takes place in the melt. Therefore, for each dendrite the environment of the lower side is slightly colder than the one of the upper side. Thus, the sidebranches at the upper side are smaller and start to grow a little bit farther away from the tip than the ones on the lower side. In the following, we investigate the upper and lower side of various dendrites separately. In order to simplify the discussion, in these investigations we mirror the contour sides at $x < 0$ to $x > 0$.

In previous studies with xenon, it has been found that the growth velocity
of the dendrite tip is constant in time [25] and depends on the supercooling \( \Delta T \) as [9]

\[
v_{\text{tip}} = (188.8 \pm 8.6) \Delta T^{(1.745\pm0.017)}
\]  

(3.2)

where \( v_{\text{tip}} \) is given in \( \mu m/s \) and \( \Delta T \) in K.

### 3.1 Nomenclature

In the following investigations, we characterize a sidebranch by its maximum in the \( x \)–direction and the two minima bordering it, as it is shown in Fig. 3.3. The solid line shows a contour of a dendrite and the dots and diamonds display the minima and the maxima of the sidebranches, respectively. The fat line is the length \( l_{i,20} \) of the sidebranch with the 6th maximum at the right-hand side of the contour.
maxima in a dendrite contour are denoted by

\[ \begin{align*}
\text{minima: } & \quad x_{i,n}^{\text{min}} = \left( x_{i,n}^{\text{min},1}, x_{i,n}^{\text{min},2} \right), \\
\text{maxima: } & \quad x_{i,n}^{\text{max}} = \left( x_{i,n}^{\text{max},1}, x_{i,n}^{\text{max},2} \right),
\end{align*} \tag{3.3, 3.4} \]

where the index \( n = 1, 2, ..., N \) refers to the contour number (\( N \) is the total number of extracted contours) and \( \tilde{i}, i = 1, 2, ... \) are the numbers of the minima and the maxima counted from the tip, respectively. With the time step \( \Delta t \) between two consecutive contours, we can ascribe to each contour its relative time of extraction, \( t_n = n \Delta t \).

In order to decide whether a sidebranch with maximum \( x_{i,n}^{\text{max}} \) is growing or melting, we calculate its length defined by

\[ l_{i,n} = \sqrt{\left( x_{i,n}^{\text{min}} - x_{i,n}^{\text{max}} \right)^2 + \left( z_{i,n}^{\text{min}} - z_{i,n}^{\text{max}} \right)^2}, \tag{3.5} \]

where \( \tilde{i} \) is the number of the lower bordering minimum of the sidebranch (see Fig. 3.3). A sidebranch keeps growing during \( \Delta t \), if its length satisfies the criterion \( \Delta l_{i,n}^{\text{g}} = l_{i,n+1} - l_{i,n} > 0 \). \( i \) and \( i' \) are not necessarily the same, because during the time \( \Delta t \), new sidebranches might have evolved, leading to \( i' = i + (\text{number of new sidebranches}) \). The analogue criterion for a remelting sidebranch is \( \Delta l_{i,n}^{\text{r}} < 0 \).

In order to determine the growth velocity \( v_{SB} \) of the sidebranches with high resolution, we choose \( \Delta t \) as small as possible. The lower limit for the choice of \( \Delta t \) is given by the ratio of the spatial inaccuracy \( \delta z \) of the contour coordinates and the growth velocity of the dendrite tip, \( v_{\text{tip}} \), as explained below. To investigate the temporal development of \( v_{SB} \) of a sidebranch, it is necessary to identify the sidebranch in various contours \( n \) at times \( t_n \). We characterize a sidebranch by its maximum and its two minima bordering it. Therefore, on the computer we identify a sidebranch during its temporal development by determining its lower minimum and its maximum in subsequent contours. Fig. 3.4 shows the temporal development of a dendrite, where 8 contours are plotted in the laboratory frame of reference, where the dendrite tip moves at the constant velocity, \( v_{\text{tip}} \). The time step between two
consecutive contours is $\Delta t = 15$ s and the distance between the contour tips is about $110 \, \mu m$. This figure shows that in the laboratory frame of reference, the minima of the sidebranches stay approximately in place while the maxima move laterally according to the growth of the corresponding sidebranch. Therefore, in a frame of reference moving at the velocity $v_{\text{tip}}$, with the dendrite tip, with increasing time the sidebranches move away from the tip and the distance of a minimum or a maximum from the tip increases during $\Delta t$ by $\Delta z = \Delta t v_{\text{tip}}$. Therefore, to determine a minimum or a maximum in two subsequent contours on the computer, it is necessary that $\Delta z > \delta z$. This yields the condition $\Delta t > \delta z / v_{\text{tip}}$. 

Figure 3.4: Temporal development of the dendrite #4 in the laboratory frame of reference. The distance between the tips of two subsequent contours is about $110 \, \mu m$. The arrows point at remelting sidebranches.
Additionally to our definitions of melting and growing sidebranches, we define an *active sidebranch* as being bigger than the next active sidebranch closer to the tip [31]. This term is used for the investigation of the envelope. This definition corresponds to an additional condition for a growing sidebranch, because a sidebranch stopping to be active still satisfies the condition of a growing sidebranch for some time.

### 3.2 Trunk

According to Fig. 3.1, we determine the width $d$ of the trunk by means of the minima of the sidebranches, $x_{\text{min}}^i = (x_{\text{min}}^{i,n}, z_{\text{min}}^{i,n})$. Although the trunk of a dendrite can be identified in a single digitized picture (see Fig. 2.2), one single contour does not provide enough minima for a precise determination of $d$. Therefore, for each dendrite we take into account all $N$ contours which have been extracted.

In Fig. 3.4, it can be recognized that the minima of growing sidebranches stay approximately in place whereas the ones of remelting sidebranches move laterally away from the main axis of the dendrite. We have found that this lateral movement may be induced in two ways. Either a remelting sidebranch sinters together with one of its neighbors or the contour between such a sidebranch and one of its neighbors cannot be resolved anymore. Consequently, the minima of remelting sidebranches do not correspond to the dendrite trunk as shown in Fig. 1.1c). Therefore, for the determination of the width $d$ of the trunk we consider the minima of the growing sidebranches only.

In Fig. 3.5, we consider a dendrite in a frame of reference moving with the tip. The minima of all of the growing sidebranches are displayed by dots. The solid line shows one contour of the dendrite and the dashed line is a plot of the tip function given by Eq. (1.8). This figure reveals 5 features of a dendrite.

i) In the *tip region*, i.e., below $\hat{z} \approx 18$, no sidebranches are formed. The contour can be described by the tip function given in Eq. (1.8).

ii) For $\hat{z} \gtrsim 25$, the $x$-coordinates of the minima develop approximately linearly with $z$, defining the *trunk region*. 
Figure 3.5: Minima of all of the growing sidebranches. The solid line displays one of $N = 195$ contours of the dendrite #4. The black dots are the minima $x_{\text{min}}^n$ of all of the growing sidebranches, the dashed line depicts the tip function, $z = 0.58 |x|^{5/3}$, describing the contour at the dendrite tip and in the inset, the white dots are the mean minima, $\bar{x}_{\text{min}}$, averaged over consecutive $z$-intervals of size $\delta_z = 0.9 \exp$ (see Fig. 3.6a)).
iii) The range $18 < z < 25$ defines a transition region.

iv) Due to coarsening, the number of growing sidebranches per length decreases with increasing distance from the tip. With increasing $z$, this leads to a decreasing number of minima per length which can be used to determine the width of the trunk.

v) On the left-hand side of the contour ($x < 0$), the sidebranches are slightly bigger than on the right-hand side. For this dendrite, the angle $\phi$ between its main axis and the direction of the earth gravitational field has been $\phi \approx 27.2^\circ$ and the side of the contour at $x < 0$ corresponds to the lower side of the dendrite during the experiment. As discussed on p. 15, due to convection the sidebranches on the upper side of a dendrite are smaller and start to grow a little bit further away from the tip than the ones on the lower side.

In the following, we determine the value $z_{\text{trunk}} \approx 25$ for the onset of the trunk region. First, in order to account for the decreasing number of data points per length with increasing $z$, we determine a set of points $\bar{x}_{\text{min}} = (\bar{x}_{\text{min}}, \bar{z}_{\text{min}})$ which are equidistant along $z$ by averaging the $x$-coordinate of the minima in consecutive, discrete $z$-intervals of constant size $\delta z$. In the range $0.7 R_{\exp} \leq \delta z \leq 1.5 R_{\exp}$, a homogeneous distribution of the $\bar{x}_{\text{min}}$ is achieved. Then, we fit the dimensionless, mean minima in various $z$-intervals $[z_l, z_u]$ by

$$ \hat{\bar{z}}_{\text{min}} = A + B \hat{\bar{x}}_{\text{min}}. $$

(3.6)

We vary systematically the fitting intervals $[z_l, z_u]$ by increasing successively the lower boundary $z_l$ and keeping $z_u$ fixed at $z_{\text{min}}$ farthest away from the tip. In the trunk region, $\bar{x}_{\text{min}}$ develops approximately linearly with $\bar{z}_{\text{min}}$, whereas in the transition region this is not the case (see inset of Fig. 3.5). Therefore, by varying systematically the fitting interval, for $z_l$ in the transition region, we expect the fitted slope $B$ to depend on $z_l$ and for $z_l$ in the trunk region, we expect $B$ to be constant. Consequently, the onset of the trunk region, $z_{\text{trunk}}$, can be determined by setting

$$ z_{\text{trunk}} = \hat{z}_{\text{sat}}, $$

(3.7)
Figure 3.6: Determination of the distance from the tip where the trunk region begins. a) The solid line displays the right-hand side of the contour shown in Fig. 3.5. The dots are the mean minima $\hat{x}_{\text{min}} = (\hat{x}_{\text{min}}, \hat{z}_{\text{min}})$, obtained by averaging the $x-$coordinates of the minima in Fig. 3.5 over consecutive $z-$intervals of size $\delta_z = 0.9 R_{\text{exp}}$ and plotted as white dots in the inset in Fig. 3.5. The value $z_t$ is the $z-$coordinate of the mean minimum farthest away from the tip. b) Slope $B$ of the fitted mean minima ($\hat{z}_{\text{min}} = A + B \hat{x}_{\text{min}}$) as a function of the lower boundary $\hat{z}_t$ of the fitting interval $[\hat{z}_t, \hat{z}_{\text{sat}}]$. The value $\hat{z}_t = \hat{z}_{\text{sat}}$ where $B$ saturates at its maximum value $B_{\text{sat}}$ corresponds to the onset of the trunk region.
with $z_{\text{sat}}$ as the value of $z_l$ where the slope $B$ saturates. This procedure is illustrated in Fig. 3.6 for the dendrite shown in Fig. 3.5. In Fig. 3.6a), the mean minima $\tilde{X}_{\text{min}}$ are shown and in b), the fitted slope $B$ is plotted vs $\hat{z}_l$. Fig. 3.6b) is representative for all of the dendrites investigated. With increasing $\hat{z}_l$, the slope $B$ increases and saturates at $B_{\text{sat}} \approx 22$ for $\hat{z}_l = \hat{z}_{\text{sat}} \approx 26$. For all of the dendrites, the linear correlation of the mean minima in intervals $[z_l, z_u]$ with $z_l \geq z_{\text{sat}}$ is close to 1. Therefore, we conclude that in the trunk region, the linear fitting function given in Eq. (3.6) describes the trunk well.

Finally, we use $A_{\text{sat}}$ and $B_{\text{sat}}$ to describe the trunk in the trunk region, namely

$$ \hat{z} = A_{\text{sat}} + B_{\text{sat}} \hat{x} $$ (3.8)

The fitted values $A_{\text{sat}}$, $B_{\text{sat}}$ and $\hat{z}_{\text{sat}}$ of all of the dendrites investigated are listed in Tab. B.1 (see App. B).

For all of the dendrites investigated, $z = 30 R_{\text{exp}}$ lies in the trunk region. Therefore, using Eq. (3.8) we determine the half width $d_{30}/2$ of the trunk for dendrites grown at various supercoolings, $\Delta T$, at a distance $z = 30 R_{\text{exp}}$ from the tip. In Fig. 3.7, $d_{30}/2$ is plotted vs the tip radius, $R_{\text{exp}}$. The upwards and downwards directed triangles refer to the upper and lower sides of the dendrites, and the dashed line is the fitted function

$$ \frac{d_{30}}{2} = (9.0 \pm 1.0) R_{\text{exp}}. $$ (3.9)

where the data, $d_{30}/2$, have been weighted according to the accuracies of the fitted slope, $B_{\text{sat}}$. In agreement with our qualitative observation in Fig. 3.2, where for dendrites grown at various supercoolings the trunks of the superimposed, scaled contours coincide, we find that at a distance $z = 30 R_{\text{exp}}$ from the tip the width $d_{30}$ of the trunk scales with the tip radius, $R_{\text{exp}}$.

A closer look at Fig. 3.7 reveals that the value of $d_{30}/2$ for the upper side (▲) of a dendrite is usually slightly bigger than for the lower side (▼). This difference is correlated with the orientation $\phi$ of the dendrite during the experiment and is a further indication that the growth of sidebranches is influenced by convection (see p. 15). In Fig. 3.7, this is confirmed by the equal
results for $\frac{d_{30}}{2}$ of both sides of the only dendrite with $\phi = 0^\circ$ ($R_{\text{exp}} = 36.5\mu m$, dendrite #6). By fitting the values $\frac{d_{30}}{2}$ for the upper and lower sides separately with $\frac{d_{30}}{2} = 9 (R_{\text{exp}} + \delta R_\uparrow)$ and $\frac{d_{30}}{2} = 9 (R_{\text{exp}} + \delta R_\downarrow)$, respectively, we obtain a mean shift $\delta R = \delta R_\downarrow - \delta R_\uparrow \approx 0.1 R_{\text{exp}}$. Using Eq. (3.1), we estimate the difference in the supercooling between the upper and lower side of a dendrite by means of $\delta R$ to be of the order of $\delta(\Delta T)/\Delta T \approx 10\%$. As $\delta(\Delta T)/\Delta T$ is only a small effect we cannot determine a clear dependence of $d$ on $\phi$.

If we replace $R_{\text{exp}}$ by an effective tip radius, $R_{\text{eff}}$, the scaling relation given
in Eq. (3.9) is valid not only at a distance \( z = 30 R_{\exp} \) but at any distance from the tip, namely

\[
d = (18 \pm 2) R_{\text{eff}}.
\]  

(3.10)

In the following, we deduce the physical meaning of \( R_{\text{eff}} \). According to Eq. (3.8), \( d \) increases linearly with \( z \). Using Eq. (3.10), this increase corresponds to an increase of the effective tip radius, \( R_{\text{eff}} \). From Eq. (3.1) follows that an increase of the tip radius is correlated to a decrease of the supercooling. Therefore, we conclude that along the dendrite the temperature rises. Such a temperature rise is expected as latent heat is released during the growth of the dendrite. It can be determined by calculating the effective supercooling, \( \Delta T_{\text{eff}} \), from the effective tip radius, \( R_{\text{eff}} \), at a given distance \( z \) from the tip.

Now, we show the correctness of this interpretation of \( R_{\text{eff}} \) by means of Fig. 3.8 which is obtained as follows. First, we determine the mean trunk, \( \bar{z} = \bar{A}_{\text{sat}} + \bar{B}_{\text{sat}} \bar{x} \), by averaging the experimentally determined values \( A_{\text{sat}} \) and \( B_{\text{sat}} \) over both sides of all of the dendrites. We plot this mean trunk \( (\bar{z} = -145 + 20 \bar{x}) \) in absolute units of \( \mu m \) using the tip radius \( R_{8,8} \) of the dendrite \#8 for rescaling:

\[
z = -4400 + 20 x.
\]  

(3.11)

Second, using Eqs. (3.10) and (3.11), we assign to each \( z \)-value in this plot of the mean trunk an effective tip radius, \( R_{\text{eff}} \), and using Eq. (3.1), an effective supercooling, \( \Delta T_{\text{eff}} \). Finally, we plot the contours of various dendrites onto the mean trunk according to their supercoolings, \( \Delta T \), by assuming that \( \Delta T_{\text{eff,30}} = \Delta T \), where \( \Delta T_{\text{eff,30}} \) is the effective supercooling at a distance \( z = 30 R_{\exp} \) from the tip. This plot is shown in Fig. 3.8, where the fat dashed lines display the mean trunk given in Eq. (3.11) and the solid lines the contours of the dendrites. The quantitative agreement of the trunks in the contours with the mean trunk confirms our interpretation, where the increase of the effective tip radius, \( R_{\text{eff}} \), with increasing distance from the tip corresponds to an increase of the temperature along the dendrite and that \( \Delta T_{\text{eff,30}} \approx \Delta T \). This thermometric property of \( d \) is verified over the whole range of the supercooling, \( \Delta T \). By means of the mean trunk, one can
estimate the temperature increase per length along a dendrite to be of the order of about 5%. One might imagine that the combined contours in Fig. 3.8 represent the contour of one hypothetical, extraordinary long dendrite.
Figure 3.8: Thermometric property of the dendrite trunk. The solid lines show contours of dendrites grown at different supercoolings, $\Delta T$, and the dashed lines display the mean trunk, $z = -4400 \pm 20x$. The contours have been shifted vertically with respect to each other by assuming that at $z = 30R_{exp}$ behind the tip the effective supercooling, $\Delta T_{eff,30}$, equals $\Delta T$. 
3.3 Envelope

In this section, we present two studies about the shape of the envelope. In Sec. 3.3.1, we characterize the envelope by its opening angle $\Phi$ and in Sec. 3.3.2, we investigate the growth velocity $v_{SB}$ of the sidebranches contributing to the envelope. For each dendrite, we take into account all of the extracted contours.

3.3.1 Opening Angle $\Phi$ of the Envelope

We determine the opening angle $\Phi$ of the envelope (see Fig. 3.1) from the maxima $x_{\text{max}}^{i,n} = (x_{\text{max}}^{i,n}, z_{\text{max}}^{i,n})$ of the active sidebranches. If all of the sidebranches would grow with the same, constant velocity during the whole development of the dendrite and if it would be possible to determine $x_{\text{max}}^{i,n}$ at any desired accuracy, then the opening angle $\Phi$ could be determined by measuring two of the maxima only. However, during the development of a dendrite not all of the sidebranches survive and thus not all of the sidebranches grow at a constant velocity. Furthermore, the precision of the determination of the $x_{\text{max}}^{i,n}$ and $z_{\text{max}}^{i,n}$ is limited. The precision of the data can be improved by an averaging process. We average the maxima of the active sidebranches in two ways, illustrated in Fig. 3.9. In a), the full dots show the maxima $x_{\text{max}}^{i,n}$ of all of the growing sidebranches. The $x-$coordinates of these maxima, $x_{\text{max}}^{i,n}$, are averaged over $z-$intervals of constant size $\delta_z$, leading to equidistant mean maxima, $\bar{x}_{\text{max}} = (\bar{x}_{\text{max}}, \bar{z}_{\text{max}})$, indicated by the open dots. Similar considerations as in the case of the determination of the width of the trunk (Sec. 3.2) lead to the choice $\delta_z = 0.9 R_{\text{Exp}}$. The dashed line is a linear fit of the mean maxima, $\bar{x}_{\text{max}}$, leading to the value $\Phi^a$ for the opening angle of the envelope. Fig. 3.9b) illustrates the second averaging method where the temporal developments of the maxima of the active sidebranches are considered. At both sides of the dendrite, the full dots show the temporal development of the maximum of an active sidebranch $j$, $1 \leq j \leq N_{\text{active}}$ ($N_{\text{active}}$ is the total number of active sidebranches). The dashed line is the fitted linear regression through the data plotted at the right-hand side of the contour and is used to determine the opening angle $\Phi_j$ of the corresponding sidebranch. Averaging of the $\Phi_j$ over all $N_{\text{active}}$ sidebranches leads to the value $\Phi^b$, where the $\Phi_j$ are
Figure 3.9: Two methods for the determination of the opening angle of the envelope. The contour in both graphs is the same (dendrite #4). In a), the full dots display the maxima, $\hat{\eta}_{i\text{max}}^l$, of all of the active sidebranches in all of the contours. The open dots are the mean maxima, $\hat{\eta}_{\text{max}}$, over consecutive $z-$intervals of size $\delta_z = 0.9 R_{\text{exp}}$. The dashed line displays the fitted linear regression through the mean maxima at $x > 0$ leading to the opening angle $\Phi^a$. In b), at either side of the contour the dots display the temporal development of the maximum of an individual active sidebranch $j$, $1 \leq j \leq N_{\text{active}}$. The dashed line is the fitted linear regression through the data plotted at the right-hand side, leading to the opening angle $\Phi_j$. Averaging of all of the $\Phi_j$ over all $N_{\text{active}}$ active sidebranches leads to $\Phi^b$. 
Figure 3.10: Experimentally determined opening angle of the envelope as a function of the supercooling $\Delta T$ of the dendrites. The upwards and downwards directed triangles display the values obtained at the upper and lower sides of the dendrites, respectively. The full and open triangles show the results $\Phi^a$ and $\Phi^b$, respectively, obtained with the two averaging methods illustrated in Fig. 3.9a) and b). The dashed line represents the mean value $\Phi = 25^\circ$ over all of the displayed data.

weighted according to the lifetimes of the sidebranches, i.e., it is given more weight to a sidebranch being active during the whole temporal development of the dendrite than to a sidebranch which disappears close to the tip.

In Fig. 3.10, $\Phi^a$ and $\Phi^b$ for the opening angle of the envelope obtained for various dendrites are plotted vs supercooling, $\Delta T$, by open and full triangles, respectively. The upwards and downwards directed triangles refer to the opening angle obtained at the upper and lower sides of the dendrites. Furthermore, the results are listed in Tab. B.1 in the appendix. With both methods, we find that the opening angle $\Phi$ of the envelope is independent of $\Delta T$. The mean values $\overline{\Phi^a} = 26^\circ \pm 2^\circ$ and $\overline{\Phi^b} = 24^\circ \pm 2^\circ$ agree with each other. The dashed line in Fig. 3.10 corresponds to the mean value, $\overline{\Phi}$, over all of the dendrites and both methods:

$$\overline{\Phi} = 25^\circ \pm 3^\circ.$$  \hspace{1cm} (3.12)

A closer look at Fig. 3.10 reveals, that the opening angle at the upper side
(Δ and ▲) of a dendrite is usually a little bit smaller than at the lower side (▽ and ▼). This effect corresponds with the convectionally induced smaller sidebranches at the upper part than at the lower part (see p. 15). This is confirmed by the only dendrite with orientation \( φ = 0° \) (\( ΔT = 95.7\text{ mK} \), dendrite #7), where the results of the opening angle for both sides of the dendrite are the same.

### 3.3.2 Growth Velocity \( v_{SB} \) of Active Sidebranches

We determine the growth velocity \( v_{SB}^{jn} \) of an active sidebranch \( j \) \( (1 \leq j \leq N_{active}) \) by comparing the coordinates of the sidebranch maximum in two consecutive contours taken with an interval size \( Δt \) between each other. In the laboratory frame of reference, the growth velocity is

\[
\begin{align*}
v_{SB}^{jn} &= \left( v_{SBx}^{jn}, v_{SBz}^{jn} \right), \\
v_{SBx}^{jn} &= \frac{x_{\text{max}}^{n+1} - x_{\text{max}}^{jn}}{Δt} \\
v_{SBz}^{jn} &= \frac{z_{\text{max}}^{n+1} - z_{\text{max}}^{jn}}{Δt} - v_{\text{tip}}
\end{align*}
\]

In these equations, the index \( j = 1, 2, ..., N_{active} \) refers to the number of the active sidebranch, which is usually different from the number \( i \) of the maximum in the \( n \)th contour (see Sec. 3.1). The growth velocity of the dendrite tip, \( v_{\text{tip}} \), is given in Eq. (3.2).

We find that the growth velocity \( v_{SB}^{jn} = |v_{SB}^{jn}| \) of an active sidebranch \( j \) is constant in time, i.e., \( v_{SB}^{jn} = v_{SB}^{jn} \). Furthermore, we find that for all of the active sidebranches of a dendrite \( v_{SB}^{jn} \) is the same. By averaging the velocities \( v_{SB}^{jn} \) over all \( N_{active} \) active sidebranches, we obtain the mean value, \( v_{SB} \).

In Fig. 3.11, we compare \( v_{SB} \) for dendrites grown at various supercoolings, \( ΔT \), with the growth velocity of the tip, \( v_{\text{tip}} \). The upwards and downwards directed triangles refer to the upper and lower sides of the dendrites. The
Figure 3.11: Experimentally determined growth velocity $v_{SB}$ of the active sidebranches as a function of the growth velocity $v_{tip}$ of the dendrite tip. The upwards and downwards directed triangles refer to the upper and lower sides of the dendrites, respectively. The dashed line shows the fitted result $v_{SB} = 0.5 v_{tip}$.

The dashed line is the linear fit

$$v_{SB} = (0.50 \pm 0.05) \ v_{tip}.$$  (3.16)

This result can be compared with the result given in Eq. (3.12) for the opening angle $\Phi$ of the envelope by means of the trigonometric relation

$$v_{tip} \cdot \sin \Phi = v_{SB} \cdot \cos(\Phi - \alpha),$$  (3.17)

where $\alpha \approx 10^\circ$ is the sidebranch orientation angle (see p. 13). The geometric disposition of the parameters in Eq. (3.17) is illustrated in Fig. 3.1. The determination of $v_{SB}$ from the trigonometric relation given in Eq. (3.17) by means of $\Phi$, $v_{tip}$ and $\alpha$ leads to the same result as the direct measurement described above.
A closer look at Fig. 3.11 reveals again, that the growth velocities at the upper sides of the dendrites are generally smaller than at the lower sides. This observation is consistent with the usually larger opening angles at the lower sides of the dendrites than at the upper sides, which has been discussed in Sec. 3.3.1 in the context of convection.

Furthermore, for a few dendrites we observe one or two sidebranches which produce sidebranches by themselves. This observation of so-called secondary sidebranches is very rare in our experiments because we focus on a limited number of sidebranches in order to achieve a high optical resolution. However, we find that as soon as a sidebranch \( j \) starts to produce sidebranches by itself, its growth velocity, \( v_{SB}^{j} \), increases towards \( v_{\text{tip}} \), whereas the growth velocity \( v_{SB}^{(2nd)} \) of the secondary sidebranches amounts again to about 0.5 \( v_{\text{tip}} \). Furthermore, the orientation angle \( \alpha \) of the primary sidebranch tends from 10° towards 0°.
3.4 Coarsening

In the last step of our studies of the region far behind the dendrite tip, we investigate the coarsening process using three different methods. In Secs. 3.4.1 and 3.4.2, we approach coarsening by numerical investigations and in Sec. 3.4.3, we combine the results obtained in the previous sections with qualitative studies of the temporal development of contours.

3.4.1 Gaussian Superposition

In a first approach to the coarsening process, we investigate the vertical spacing $\lambda_{i,n}$ between consecutive, growing sidebranches $i,n$ and $i+1,n$ in all of the extracted contours $n = 1, 2, \ldots, N$ of a dendrite. We determine $\lambda_{i,n}$ from the $z-$coordinate of the maxima $x_{i,n}$ of the growing sidebranches:

$$\lambda_{i,n} = z_{i+1,n}^{\max} - z_{i,n}^{\max}. \tag{3.18}$$

The development of the sidebranch spacing along a dendrite can be described, if $\lambda_{i,n}$ is determined as a function of the distance $z_{i,n}$ from the tip, defined by

$$z_{i,n} = \frac{z_{i+1,n}^{\max} + z_{i,n}^{\max}}{2}. \tag{3.19}$$

In Fig. 3.12a), a digitized video picture of a dendrite is shown and in b), one extracted contour ($n = 130$) is plotted in units of the tip radius, $R_{\text{tip}}$. The dots display the maxima of the growing sidebranches at the right contour side. Finally, c) shows the points $(\lambda_{i,n}, z_{i,n})$ obtained at the right-hand side of the dendrite from all of the contours. Fig. 3.12c) reveals three features which are representative for all of the dendrites investigated:

i) The sidebranch spacings $\lambda_{i,n}$ take discrete values at about 3.5, 6, 9.5, ...

ii) The distance $z_{i,n}$ closest to the tip, indicated by arrows, increases with increasing $\lambda_{i,n}$.

iii) The $z-$range, where the points for the sidebranch spacings $\lambda_{i,n}$ of about 9.5 are found is smaller than the ranges for the sidebranch spacings of about 3.5 and 6.
Figure 3.12: Investigation of the sidebranch spacing along a dendrite. In a), one digitized video frame of the dendrite #1 is shown. In b), the extracted contour \((n = 130)\) of the dendrite is plotted by a solid line and the dots display the maxima of the growing sidebranches at the right-hand side, \(x > 0\). At a distance \(\hat{z}_{4,130}\) from the tip, the sidebranch spacing \(\hat{\lambda}_{4,130}\) between the 4th and 5th growing sidebranch is indicated schematically. In c), all of the sidebranch spacings \(\hat{\lambda}_{i,n}\) of this side of the dendrite are mapped at their corresponding positions \(\hat{z}_{i,n}\) behind the dendrite tip.
The third feature is an artifact. With increasing distance from the tip and especially with increasing distance between the sidebranches, it becomes difficult to trace the temporal development of the individual sidebranches on the computer. Together with the increasing sidebranch spacing with increasing distance from the tip, this leads to a reduced number of data points per length. Therefore, we believe that the sidebranch spacing \( \lambda_{i,n} \approx 9.5 \) continues to higher values of \( \tilde{\lambda}_{i,n} \) than it does in Fig. 3.12c).

A closer look at Fig. 3.12 reveals furthermore, that in the region of the first sidebranch spacing (\( \tilde{\lambda}_{i,n} \approx 3.5 \)) at a distance \( \tilde{z}_{i,n} \gtrsim 20 \), many points are found within a relatively large range \( 2 \lesssim \tilde{\lambda}_{i,n} \lesssim 4 \). This cluster of points splits up at \( \tilde{z}_{i,n} \gtrsim 28 \) into two at \( \tilde{\lambda}_{i,n} \approx 3 \) and \( \tilde{\lambda}_{i,n} \approx 4 \), i.e., on the one hand, the sidebranch spacing \( \tilde{\lambda}_{i,n} \) is constant at a value of about 3 and on the other hand, with increasing \( \tilde{z}_{i,n} \) the sidebranch spacing drifts continuously towards a bigger value at about 4. A similar behavior can be observed for the second sidebranch spacing at \( \tilde{\lambda}_{i,n} \approx 6 \), although it cannot be recognized so easily because there are less points than in the range \( 2 \lesssim \tilde{\lambda}_{i,n} \lesssim 4 \). This continuous drift of the sidebranch spacing with increasing distance from the tip has been observed for all of the dendrites investigated and correlates with the linear increase of the width of the trunk along the dendrite, as will be discussed later.

Now, we determine the discrete values of the sidebranch spacings, identified by eye in Fig. 3.12c), as follows. In a first step, in order to include the measuring errors in the investigation of the temporal development of the sidebranch spacings we assign to each data point \((\lambda_{i,n}, \tilde{z}_{i,n})\) a Gaussian function

\[
\tilde{f}^{i,n}(\lambda) = \frac{1}{\sigma \sqrt{2 \pi}} e^{-\frac{1}{2} \frac{(\lambda - \lambda_{i,n})^2}{\sigma^2}},
\]

where the standard deviation \( \sigma \) is given by the lateral resolution of our image analysis (see Sec. 2). For all of the dendrites, it is of the order of 10 \( \mu \text{m} \).

In a second step, we sum up the Gaussian functions, \(\tilde{f}^{i,n}(\lambda)\), in \(z\)-intervals of size \( \delta_z = 5 R_{\text{exp}} \). For the sidebranch spacings shown in Fig. 3.12c), the sums of the Gaussian functions, \(f_z(\tilde{\lambda})\), scaled to a standard amplitude, are plotted by fat lines in Fig. 3.13. Finally, we fit the sums of the Gaussian functions, \(f_z(\lambda)\), with a small number \(N_z(\text{Gauss})\) of Gaussian functions, \(g_z^j(\lambda)\),
Figure 3.13: Method of the superimposed Gaussian functions. The fat solid lines display the sums of the Gaussian functions, $f_{z}(\hat{\lambda})$, obtained by summing up the Gaussian functions, $f^{i,n}(\hat{\lambda})$, assigned to the experimentally determined points $(\hat{\lambda}^{i,n}, \hat{z}^{i,n})$ plotted in Fig. 3.12c). The fat dashed lines display the fits with $N_{z(Gauss)}$ Gaussian functions, $g_{z}^{j}(\hat{\lambda})$. $N_{z(data)}$ refers to the number of points $(\hat{\lambda}^{i,n}, \hat{z}^{i,n})$ having contributed to $f_{z}(\hat{\lambda})$.

In Fig. 3.13, the fitted Gaussian functions, $g_{z}^{j}(\lambda)$, are plotted by fat dashed lines and at the right-hand side, there are given the number $N_{z(data)}$ of points $(\hat{\lambda}^{i,n}, \hat{z}^{i,n})$ having contributed to $f_{z}(\hat{\lambda})$ and the number $N_{z(Gauss)}$ of Gaussian functions used for fitting. We find that only a small number of Gaussian functions, $g_{z}^{j}(\lambda)$, is necessary to describe the experimental data, $f_{z}(\hat{\lambda})$. In a given $z-$interval, the $g_{z}^{j}(\hat{\lambda})$ describe quantitatively the distributions of the values of the sidebranch spacing. In Tab. C.1 in the appendix, there are listed the values of the centers, $\hat{\lambda}_{z}^{j}$, standard deviations, $\sigma_{z}^{j}$, and intensities, $I_{z}^{j}$, of the $g_{z}^{j}(\hat{\lambda})$ fitted to the $f_{z}(\lambda)$ in Fig. 3.13.
In Fig. 3.13, for \( z = 10 \) three peaks at 3.0 ± 0.5, 3.6 ± 0.5 and 5.9 ± 0.7, corresponding to the spatial period of the perturbations at the ridges of the fins close to the tip. With increasing \( z \), the first peak stays at \( \lambda \approx 3 \), the second one drifts from 3.6 to 4.2 and a longer period appears at about 9.6. In Fig. 3.13, the main branch drifts from 3.6 to 4.2. This drift takes place during the temporal development of the dendrite where in the region of the trunk, the effective supercooling, \( \Delta T_{\text{eff}} \), decreases. It can be attributed to this decrease in \( \Delta T_{\text{eff}} \) along the dendrite within ±8% (see App. C).

In order to increase the number of experimental data, \( f_{i,n}(\lambda) \), for the determination of the distributions of the sidebranch spacings, \( f_x(\lambda) \), we evaluate the points (\( \lambda^{i,n}, z^{i,n} \)) of all of the dendrites investigated together. In Fig. 3.14, the solid lines show the sums of the Gaussian functions, \( f_x(\lambda) \), obtained from the \( f_{i,n}(\lambda) \) at the upper sides of all of the dendrites. At the right-hand side of the graph, there are given the numbers \( N_{i,n} \) of points (\( \lambda^{i,n}, z^{i,n} \)) contributing to the corresponding \( f_x(\lambda) \). Similar as in the Figs 3.12c) and 3.13, this figure, Fig. 3.14, reveals three tendencies:

i) The distance \( \tilde{z} \) closest to the tip where a peak appears for the first time (indicated by arrows), tends to increase with \( \lambda \).

ii) With increasing \( \tilde{z} \), at smaller values of \( \tilde{\lambda} \) the distribution peaks tend to disappear whereas new peaks are generated at larger values of \( \tilde{\lambda} \).

iii) The distribution peaks tend to occur at discrete values of \( \tilde{\lambda} \).

Whereas the first and the third tendency has been observed already in Fig. 3.12c), the second one indicates that with increasing distance from the tip, the smaller sidebranch spacings are taken over by bigger ones. However, in some regions of this figure, the discrete peaks spread and fuse together such as it is not possible to deduce any clear scenario for the coarsening process. This spreading and fusion of peaks is due to the different orientations of the individual dendrites with respect to the direction of the earth gravitational field during the experiments. In the paragraph above, it has been shown that a continuous drift in the sidebranch spacing with increasing distance from the tip is due to the decrease of the effective supercooling,
Figure 3.14: Distribution of the sidebranch spacings at the upper side of all of the investigated dendrites. The solid lines display the distributions $f_z$ of the sidebranch spacing $\lambda$ in dimensionless units. The numbers on the right-hand side of the graph refer to the number $N_z(\text{data})$ of data points $(\hat{\lambda}^{i,n}, \hat{z}^{i,n})$ contributing to the corresponding distribution $f_z(\lambda)$. 
Analysis and Results

$\Delta T_{\text{eff}}$, along the dendrite. In Sec. 3.2, it has been shown that the decrease of $\Delta T_{\text{eff}}$ along the dendrite is influenced by convection (see Fig. 3.7 and p. 23). Because the dendrites have been oriented differently during the experiments, the continuous drifts in the sidebranch spacing with increasing distance to the tip of the dendrites are different too. Therefore, the superposition of the Gaussian functions, $f_{i}^{(n)}(\lambda)$, of all of the dendrites leads to a spreading or fusion of peaks in the sums of the Gaussian functions, $f_{s}(\lambda)$.

With the method of the superimposed Gaussian functions we have found that the sidebranch spacings tend to occur at $3.5 R_{\exp}, 6 R_{\exp}, 9.5 R_{\exp}, \ldots$ and that the continuous drifts in the sidebranch spacings are due to the temperature increase along the dendrite. Furthermore, we have found that with increasing distance from the tip, the smaller sidebranch spacings are taken over by bigger ones.

3.4.2 Fourier Transform

A second approach to the coarsening process is based on the wave like shape of a contour line along the sidebranches. According to the results of the previous section, a Fourier transform of such a contour line is supposed to show the spatial periodicity corresponding to the sidebranch spacings at about $3.5 R_{\exp}, 6 R_{\exp}, 9.5 R_{\exp}, \ldots$. However, there are two properties of dendrites which complicate a Fourier transform of a contour line:

i) The sidebranches have thick bellies and narrow bases.

ii) As mentioned already in Sec. 3.1, the sidebranches do not grow orthogonally to the main axis of the dendrite.

We construct a single valued function by replacing the natural contour by its outermost or innermost contour line. This construction is illustrated in Fig. 3.15, where the fat solid and dashed lines show the outermost and the innermost contour lines of the natural contour, respectively. Using the outermost contour line, low frequencies obtained by fast Fourier transform (FFT) contain informations about the sidebranch spacing. High frequencies are due to sharp corners generated by the construction of a single valued function. The FFT spectrum using the innermost contour line, containing
even more sharp corners than the outermost contour line, appears to be very noisy.

In Sec. 3.4.1, we have found that with increasing distance from the tip the smaller sidebranch spacings are taken over by the larger ones. To detect this effect by means of FFT, we analyze a time sequence of a given region of a dendrite, i.e., a region which is fixed in the laboratory frame of reference. Such a time sequence is shown in Fig. 3.16a), where four contours of the dendrite #8 are plotted in a frame of reference moving with the tip. At the top of each contour, its number, n, is given and the region of the dendrite, selected for the investigation by FFT, is surrounded by a box. The time step between two consecutive contours n and n+1 is Δt = 30 s. The 12 FFT spectra obtained from the selected region in the contours n = 22, 23, ..., 34 are displayed in Fig. 3.16b). The solid lines represent the Fourier amplitudes, A, as a function of the wavelengths, $\lambda$, corresponding to the sidebranch spacing. The data of subsequent contours are shifted vertically each time by 2 units relative to each other. The dashed lines indicate the temporal development of the wavelengths at about 3 and 6. At the right-hand side, the extraction
Figure 3.16: Investigation of the sidebranch spacing by means of fast Fourier transform (FFT). a) Temporal sequence at the right-hand side ($x > 0$) of the dendrite #8. At the top of each contour, its number, $n$, is given. The selected region surrounded by a box is investigated by means of FFT during its temporal development in the contours $n = 22, 23, \ldots, 34$. b) 12 Fourier amplitudes obtained from the outermost envelope of the selected region. The amplitudes, $A$, are shifted each time by 2 units and plotted vs the wavelength, $\lambda$. At the right-hand side, the extraction time $t_n = n \Delta t$ of the corresponding contour is given ($\Delta t = 30\,\text{s}$).
time $t_n = n \Delta t$ of each contour is given. This figure is representative for all of the FFT spectra obtained for different dendrites and shows, that with increasing time, the wavelength at about $\delta$ increases in amplitude, i.e., it becomes more and more important.

In App. C, a calculation of the Gaussian peaks, drifting with increasing distance from the tip, shows that this drift corresponds to the linear increase of the width of the trunk with increasing distance from the tip. We obtain the same result within $\pm 5\%$ from an analogous calculation of the drift in Fig. 3.16 for the wavelength $\hat{\lambda}$ at about $\delta$.

### 3.4.3 Contours

Because the dynamical process of coarsening leads to a very complex temporal development of a dendrite contour far behind the tip, we have not been able to determine the scenario for coarsening with the methods of superimposed Gaussian functions (Sec. 3.4.1) or Fourier transforms (Sec. 3.4.2). Therefore, we investigate now the temporal development of a region of a dendrite directly in the contours. In Fig. 3.17, 7 halves of contours of the dendrite #4 are depicted, plotted in a frame of reference moving with the dendrite tip. At the top of each contour, the corresponding contour number, $n = 140, 142, \ldots, 152$, is given. To reduce the effects of perturbations, for our studies of coarsening we have selected a region close to the tip where the sidebranches are formed regularly. In Fig. 3.17, this region is marked by a fat line. It is stationary in the laboratory frame of reference. In each contour, the dots display the maxima and the dashed lines indicate the spacings of the growing sidebranches. In the next paragraph, we study the development of the selected region.

In the contour 140, all of the 7 sidebranches are growing and increase in size approximately linearly with distance from the tip. In this contour part, the distance $\hat{\lambda}^{\delta n}$ between all of the sidebranches is $\hat{\lambda} \approx 3.2$. The sidebranches keep growing at a constant velocity up to the contour 142, where a new sidebranch evolves. In the contour 144, the 3$^{rd}$ and the 4$^{th}$ as well as the 5$^{th}$ and the 6$^{th}$ sidebranches have the same length. In the contour 148 finally, the 4$^{th}$ and the 6$^{th}$ sidebranch remelt. According to our definition of $\lambda^{\delta n}$
Figure 3.17: Temporal development of a selected region of a dendrite. The solid lines display 7 half's of contours at the lower side of the dendrite #4 in a frame of reference moving with the dendrite tip. At the top of each contour, its number, \( n = 140, 142, \ldots, 152 \), is given. A selected contour part is marked by a fat line. The dots display the maxima of the growing sidebranches and the dashed lines indicate the corresponding spacings \( \lambda^{|n} \) between the growing sidebranches.

As the vertical distance between the growing sidebranches (see p. 34), the spacing which has initially been the same between all 8 sidebranches is no longer homogeneous, i.e., between the 3\(^{\text{rd}}\) and the 5\(^{\text{th}}\) and between the 5\(^{\text{th}}\) and the 7\(^{\text{th}}\) sidebranch, the distance has doubled and is now about 2 \( \lambda \). In the contour 152 finally, the 7\(^{\text{th}}\) sidebranch remodels as next, leading to a further doubling of the distance between the 5\(^{\text{th}}\) and the 8\(^{\text{th}}\) sidebranch, i.e., the sidebranch spacing has increased to about 4 \( \lambda \). This example illustrates that coarsening sets in at a distance of about 40 \( R_{\text{exp}} \) from the tip and that in this limited frame of observation, during the dynamical process of coarsening the distance between the growing sidebranches doubles as

\[
\lambda \longrightarrow 2 \lambda \longrightarrow 4 \lambda.
\]  

(3.21)

A further doubling step (8 \( \lambda \)) has been observed on other dendrites. In the next paragraph, we discuss this experimental observation given in Eq. (3.21)
on a hypothetical, ideal dendrite.

For an ideal dendrite, the sidebranches increase linearly in length with distance from the tip. This follows from the triangular shape of the envelope or the constant growth velocity of the active sidebranches (see Secs. 3.3.1 and 3.3.2). If a sidebranch of an ideal dendrite remelts, the growth conditions of its neighbors are improved, as the melting sidebranch is no longer a source of latent heat. Due to the constant growth velocity of the active sidebranches, the improved growth conditions lead to the formation of bellies on the surviving sidebranches. Because the lower neighbor of the remelting sidebranch is bigger than its lower neighbor, the latter one remelts as next. For an ideal dendrite, this period doubling happens \textit{ad infinitum}, i.e.,

\[
\lambda \rightarrow 2\lambda \rightarrow 4\lambda \rightarrow ... \rightarrow 2^n\lambda \rightarrow ...
\]  

(3.22)

This scenario of period doubling for an ideal, hypothetical dendrite is compatible with our observation on regularly formed dendrites, Eq. (3.21). However, in Fig. 3.12c) we have observed a sidebranch spacing at about \(9.5 R_{\text{exp}}\) and in Fig. 3.14, distribution peaks occur at about \(15 R_{\text{exp}}, 18 R_{\text{exp}}, \ldots\). In units of the initial spacing \(\lambda \approx 3.2 R_{\text{exp}}\), such sidebranch spacings correspond to about \(3\lambda, 5\lambda, 6\lambda, \ldots\). A similar case is found in the contour 148 in Fig. 3.17, where in the selected region the 7th sidebranch remelts instead of the 8th one. This deviation from the ideal case of period doubling can be understood by considering the history of the dendrite, displayed in Fig. 3.18.

The solid lines are plots of contours halves \((n = 108, 111, \ldots, 139)\) of the same side of the dendrite #4 which is plotted in Fig. 3.17. Again, the distances between the growing sidebranches are indicated by dashed lines and the maxima of the growing sidebranches are marked by dots. The region selected in Fig. 3.17 by a fat line, is marked again by a fat line in the last contour with number 139. In the next paragraph, we discuss the history of the selected region in the contours 108 to 139.

Due to a small disturbance close to the tip, in the contour 108 the second sidebranch, marked by a gray color, has about the same length as the first sidebranch, marked by a striped pattern. Both sidebranches have appeared just a few contours before. In the contour 111, the gray sidebranch remelts and in the contour 114, its upper and lower neighbors have thickened already distinctively. Now, we trace the development of the lower, thickened
Figure 3.18: History of the selected region of the dendrite #4 plotted in Fig. 3.17. The solid lines depict 12 contours halves of the same side of the dendrite as in Fig. 3.17. At the top of each contour, its number, \( n = 108, 111, \ldots, 139 \), is given. The region, selected in Fig. 3.17 by a fat line, is marked in the contour 139 by a fat line too. The dots display the maxima of the growing sidebranches and the horizontal dashed lines indicate the sidebranch spacings. To facilitate the tracing of different sidebranches over all of the contours, three of them are marked by a grey color and a dashed or pointed pattern.
neighbor marked by the striped pattern. Because this sidebranch has become relatively big, its next neighbor towards the tip starts to remelt right after its formation close to the tip, whereas in the unperturbed case, a first remelting of sidebranches sets in at a distance of about $40R_{\text{exp}}$ from the tip (see Fig. 3.18). On the other hand, the remelting of this sidebranch amplifies the thickening of the already big, striped sidebranch above it. Finally, the striped sidebranch thickens such as even its neighbor after next towards the tip remelts, leading to a distance $\lambda_{\text{in}} \approx 3\lambda$ between the growing sidebranches. The melting of the second next sidebranch in this tripling mode occurs very slowly, i.e., on the computer, this sidebranch is detected as melting only in the contour 137. This sidebranch melts so slowly, because it is still slightly bigger as its next neighbor towards the tip, marked by a dotted pattern. However, in the meantime, the dotted sidebranch thickens such as its next two neighbors towards the tip remelt in the contour 135, similar as it happened with the striped sidebranch one cycle earlier. Therefore, a spacing of about $3\lambda$ occurs again. Finally, the third next neighbor of the dotted sidebranch is just the $8^{\text{th}}$ sidebranch in the selected contour part plotted in Fig. 3.17. Due to the two tripling modes in the two cycles before, this $8^{\text{th}}$ sidebranch is still a little bit thicker as it would be in the ideal case. This is just enough for it to win the competition with its lower neighbor, so that in the selected contour part, the $7^{\text{th}}$ sidebranch remelts rather than the $8^{\text{th}}$ one.

To summarize the discussion in the paragraph above, a slight disturbance in the growth of a sidebranch at the very first stage of its evolution may influence the growth of several sidebranches. The lower neighbor of such a sidebranch might thicken such as two of its lower neighbors remelt instead of only the next one, leading to a tripling of the initial sidebranch spacing, i.e., $\lambda \rightarrow 3\lambda$. Such a remelting may persist over several cycles until the sidebranches are no longer influenced by the initial perturbation. From this observation, we conclude that the temporal development of the sidebranch spacing is sensitive to initial conditions. Without perturbations at the early stages of sidebranching, coarsening sets in at a distance of about $z \approx 40R_{\text{exp}}$ from the tip (see Fig. 3.17).

Now, we combine the insight into the coarsening process, obtained from the direct investigation of contours, with the information contained in Fig. 3.14.
For a regularly spaced dendrite, coarsening sets in at a distance of about 
\( z \approx 40R_{\exp} \) from the tip and the observed development of the sidebranch 
spacing is compatible with the scenario of period doubling given in Eq. (3.22) 
for an ideal, hypothetical dendrite. A small disturbance close to the tip may 
lead to a tripling of the initial spacing. If we assume period doubling as the 
predominant scenario of coarsening, with increasing distance to the tip such 
a tripled spacing may double likewise, i.e., 
\[ 3\lambda \rightarrow 6\lambda \rightarrow 12\lambda \rightarrow \ldots \]
In fact, sidebranch spacings of about 6\( \lambda \) and 12\( \lambda \) correspond to distribution 
peaks at about 18\( R_{\exp} \) and 36\( R_{\exp} \) in Fig. 3.14. In this figure, further dis-
tribution peaks at about 15\( R_{\exp} \), 16\( R_{\exp} \), 22\( R_{\exp} \) and 32\( R_{\exp} \) may be due 
to further disturbances or due the continuous shifts of sidebranch spacings 
with increasing distance to the tip, correlated to the increase of temperature 
along a dendrite.
4. Discussion

In Sec. 4.1, we discuss the experimental results on the width $d$ of the trunk of a dendrite. In Sec. 4.2, we consider the results on the opening angle $\Phi$ of the envelope and on the growth velocity $v_{SB}$ of the active sidebranches. In Sec. 4.3 finally, we discuss the results on the coarsening scenario.

4.1 Trunk

From experimental studies we have deduced a scaling relation between the width of the trunk, $d_{30}$, at a distance $z = 30 R_{\exp}$ from the tip and the tip radius $R_{\exp}$ of the dendrite, Eq. (3.9). Replacing $R_{\exp}$ by an effective tip radius, $R_{\text{eff}}$, yields the scaling relation between the width of the trunk, $d$, and $R_{\text{eff}}$ at any distance $z$ from the tip, Eq. (3.10). From Eq. (3.10) and the observed linear increase of $d$ with distance from the dendrite tip, Eq. (3.8), we have concluded that along a dendrite the temperature rises, i.e., the effective supercooling $\Delta T_{\text{eff}}$ decreases. This temperature increase is due to the latent heat released during the growth of the dendrite and amounts to about 5% per length. Using the thermometric property of $d$, we have combined dendrites grown at various supercoolings to one hypothetical dendrite (see Fig. 3.7). Such a long xenon dendrite has never been grown in an experiment, it would break down by its own weight.

In Sec. 1, we have mentioned that dendritic solidification influences the properties of cast alloys. Now, we will show that the relation $d = (18 \pm 2) R_{\text{eff}}$, Eq. (3.10), is applicable to metallic alloys.

In Refs. [28, 29], Grugel has studied directional solidification of Al-Si alloys. In these experiments, the growth rates are similar to the ones in our experiments with xenon. Therefore, as in our experiments, in the experiments presented in Refs. [28, 29], the trunk is screened by the sidebranches and thus close to the liquid where the space between the dendrites is relatively big, no
interaction takes place between the trunks. Grugel has measured the width $d$ of the trunk from micrographs and calculated the tip radius $R_K$ from the supercooling according to Ref. [5]. From Figs. 2. and 3. in Ref. [29], for the region close to the liquid one can deduce the proportionality $d \approx (20 \pm 5) R_K$ which is in good agreement with our observation on freely growing xenon dendrites, Eq. (3.10). But in the mushy zone where the crystals are close to each other, interaction of the dendrites cannot be excluded. Studies of Corbett et al. [32] on Pb-Sn alloys during directional solidification lead to a similar result. Therefore, we conclude that the width $d$ of the trunk is an integral parameter [9] which is independent of material and can be used to describe quantitatively the temporal development of freely grown dendrites as well as the microstructure in directionally solidified metallic alloys if the results are scaled by the tip radius.

Detailed studies of grain refinement during the rapid solidification of levitated supercooled melts are presented in Refs. [26, 27, 33, 34] and a model has been developed, where the width $d$ of the trunk is assumed to be proportional to the tip radius $R_{LM-K}$:

$$d = c R_{LM-K}$$

(4.1)

For the binary alloys Ni-Cu [26] and Ni-C [34, 27], $d$ has been measured from micrographs of solidified samples and $R_{LM-K}$ has been calculated from the supercooling, $\Delta T$, using the marginal stability criterion. For these systems, the proportionality factor has been estimated to $c = 40 \pm 20$ [33]. This value has been used to model the evolution of microstructures.

During rapid solidification, where many dendrites grow in a small droplet of supercooled melt, two features prevent an interaction of the dendrite trunks. First, due to the high growth rate of the order of m/s, in metallic alloys the thermal diffusion length, of the order of $10^{-6}$ m, is small compared to the distance between the dendrites. Second, the trunk of a dendrite is screened by the sidebranches. Therefore, the experiments performed with levitated droplets of alloys can be compared with our experiments on xenon and the model of grain refinement can be refined with our experimentally determined proportionality factor, $c = 18 \pm 2$, given in Eq. (3.10).
4.2 Envelope

The experimental results obtained by the investigation of the active side-branches show that the opening angle $\Phi$ of the envelope is $25^\circ \pm 3^\circ$, Eq. (3.12). It is independent of the supercooling and of the age of the dendrite within the frame of observation. This result is consistent with the observation, that the growth velocity $v_{SB}$ of the active sidebranches is constant in time and $v_{SB} = (0.5 \pm 0.05) v_{tip}$, Eq. (3.16), where $v_{tip}$ is the growth velocity of the dendrite tip. A constant value of $v_{SB}$ means, that an active sidebranch does not accelerate when one of its neighbors starts to remelt. Taking into account $v_{SB}$, $v_{tip}$ and the orientation angle $\alpha \approx 10^\circ$ of the sidebranches, $\Phi$ can be calculated to be about $25^\circ$ as found by direct measurement. As soon as a sidebranch starts to produce sidebranches by itself, its velocity tends towards $v_{tip}$ and $\alpha$ towards $0^\circ$. In this case, an opening angle $\Phi \approx 45^\circ$ is expected in a region far behind the dendrite tip.

In Ref. [31], Huang and Glicksman have studied the sidebranch structure of succinonitrile (SCN) dendrites. There, the growth rates of sidebranches are measured to be about 40% of the growth rate $v_{tip}$ of the tip, which is in rough agreement with our result for the growth velocity $v_{SB}$ of the active sidebranches. Furthermore, in a first region behind the tip, where the sidebranches do not yet produce sidebranches by themselves, in Fig. 10. of Ref. [31], the opening angle of the envelope, $\Phi$, can be estimated to be about $24^\circ$, which is in agreement with our observations on xenon dendrites. In a region further away from the tip, where all of the active sidebranches produce sidebranches by themselves, in Fig. 10. of Ref. [31], $\Phi$ is about $41^\circ$. This is compatible with our observation that the growth velocity $v_{SB}$ of sidebranches producing sidebranches by themselves tends towards the growth velocity $v_{tip}$ of the dendrite tip. From Fig. 5. in Ref. [31], one can deduce an orientation angle $\alpha \approx 10^\circ$ of the sidebranches which is in agreement with the orientation of the sidebranches in our experiments with xenon.

On SCN dendrites grown in microgravity experiments by Glicksman et al. [35], Li and Beckermann [36] have fitted the lateral positions $x_{max}$ of the maxima of the sidebranches in dependence of the distance $z$ from the tip by a power law with the result, $\hat{x}_{max} = 0.668 \hat{z}^{0.859}$, where $\hat{x}_{max}$ and $\hat{z}$ are
given in units of the experimentally determined tip radius, \( R_{\text{exp}} \). This result yields a power law for the growth velocity \( v_{\text{SB}} \) of the sidebranches, namely, \( v_{\text{SB}}/v_{\text{tip}} = 0.574 \tilde{z}^{-0.141} \). These power laws for \( \dot{x}_{\text{max}} \) and \( v_{\text{SB}}/v_{\text{tip}} \) do not agree with the linear shape of the envelope and the constant growth velocity of the active sidebranches deduced from our experiments with xenon. A comparison of the data leads to two facts which may be the reasons for this difference.

i) From Fig. 4. in Ref. [36], one can deduce that the sidebranches start to grow at a distance \( \tilde{z} \approx 20 \) from the tip, which is in agreement with observations on xenon dendrites [9]. However, Li and Beckermann have fitted data for \( \tilde{z} > 10 \), i.e., not only the maxima of the growing sidebranches have been fitted but also the width of the fins in the tip region where no sidebranches are yet formed. In this region, the data are not linear but can be described by the tip function given in Eq. (1.8). In our studies on the other hand, we have fitted the maxima of all of the active sidebranches only at distances \( \tilde{z} \gtrsim 25 \), where the lengths of the sidebranches grow linearly with distance from the tip.

ii) Fig. 2. in Ref. [37] shows pictures of dendrites grown during the microgravity experiment of Glicksman et al. [35]. In these pictures, several dendrites grow parallel and close to each other. Therefore, instead of having grown freely, the dendrites investigated by Li and Beckermann did compete with each other leading to smaller sidebranches than it would be the case during free growth.

Trivedi and Somboonsuk [38] have investigated pattern formation during directional solidification of transparent, binary systems. A SCN-acetone solution has been chosen as a model substance for binary alloys. Fig. 2 in Ref. [38] shows a dendritic array in a steady state. From this figure, close to the liquid one can estimate the opening angle \( \Phi \) of the envelope to be about 25° and the orientation angle \( \alpha \) of the sidebranches to be of the order of 10°. These values for a dendritic array of a binary solution in a steady state correspond with our observations on freely growing xenon dendrites. Therefore, we conclude that in the region close to the liquid, dendrites grown by directional solidification technique show the same behavior as freely grown dendrites. A similar agreement has been found in Sec. 4.1 for metallic alloys.
From this discussion we conclude that the growth velocity $v_{SB}$ of active sidebranches and the opening angle $\Phi$ of the envelope are integral parameters [9] to characterize a dendrite independently of material and solidification technique.

### 4.3 Coarsening

Our experimental results on the dynamical process of coarsening for three-dimensional xenon dendrites are compatible with the scenario of period doubling for an ideal, hypothetical dendrite, $\lambda \rightarrow 2\lambda \rightarrow 4\lambda \rightarrow \ldots \rightarrow 2^n\lambda \rightarrow \ldots$, Eq. (3.22), where $\lambda \approx 3.2R_{exp}$ is the initial sidebranch spacing. We have found, that the temporal development of the sidebranch spacing is sensitive to initial conditions. In the context of period doubling, due to this sensitivity shifted doubling modes may occur, $3\lambda \rightarrow 6\lambda \rightarrow 12\lambda \rightarrow \ldots \rightarrow 3 \cdot 2^n\lambda \rightarrow \ldots$ which are compatible with our observations (see Fig. 3.14).

Huang and Glicksman [31] have investigated the sidebranch spacing of pure SCN dendrites. In Figs. 10. and 11. of Ref. [31], the sidebranch spacings are plotted as a function of the distance from the tip. No information on the temporal development is given, therefore it is not possible to deduce any coarsening scenario from these figures. However, the measured sidebranch spacings are compatible with the coarsening scenario of period doubling, as it is the case with our results obtained for xenon dendrites.

Trivedi and Somboonsuk [38] have investigated pattern formation during directional solidification of a SCN-acetone solution. The temporal development of the solid-liquid interfacial pattern is shown in Fig. 1.a) through d). One can correlate the temporal development of these interfacial patterns with the development of the sidebranches along a dendrite. In both cases, a Mullins-Sekerka instability first leads to periodic disturbances of the smooth interface. Then, coarsening leads to an increase of the typical wavelengths of the disturbances and finally, independent dendrites develop. In Fig. 1.a) through d) of Ref. [38], one can recognize that in the case of initially regularly spaced bumps, with increasing time every second bump tends to remelt and that in the case, where the bumps are disturbed at their early stages of
evolution, two or even three neighbored bumps remelt with increasing time. This observation is compatible with the scenario of period doubling, as it is the case with our results deduced from on freely growing xenon dendrites and it reflects the same sensitivity of the temporal development of the sidebranch spacing to initial conditions. Such a sensitivity to initial conditions it typical for chaotic systems.

For a nonlinear, dynamical system, period doubling is a scenario leading into chaos [39]. The nonlinearity of a dendritic system, the sensitivity of the temporal development of the sidebranch spacing to initial conditions and the compatibility of the observed coarsening scenario with period doubling, as found in our investigations and compatible with the investigations in Refs. [31, 38], indicate that dendritic crystallization might be an example for a natural, chaotic system.
## A. Xenon

Table A.1: Selected properties of xenon

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<thead>
<tr>
<th>Property</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Molecular weight</td>
<td>$M_m$</td>
<td>131.30</td>
<td>g/mol</td>
</tr>
<tr>
<td>Triple-point temperature</td>
<td>$T_t$</td>
<td>161.3897</td>
<td>K</td>
</tr>
<tr>
<td>Triple-point pressure</td>
<td>$p_t$</td>
<td>0.816901</td>
<td>bar</td>
</tr>
<tr>
<td>Triple-point molar volume (l)</td>
<td>$V_m(l)$</td>
<td>44.31</td>
<td>cm$^3$/mol</td>
</tr>
<tr>
<td>Triple-point molar volume (s)</td>
<td>$V_m(s)$</td>
<td>38.59</td>
<td>cm$^3$/mol</td>
</tr>
<tr>
<td>Heat of fusion</td>
<td>$L$</td>
<td>2299</td>
<td>J/mol</td>
</tr>
<tr>
<td>Melting entropy</td>
<td>$\Delta S_m$</td>
<td>14.24</td>
<td>J/mol K</td>
</tr>
<tr>
<td>Jackson α-factor</td>
<td>$\alpha_{\text{Jackson}}$</td>
<td>1.71</td>
<td>**</td>
</tr>
<tr>
<td>Specific heat (l)</td>
<td>$c_p(l)$</td>
<td>44.6</td>
<td>J/mol K</td>
</tr>
<tr>
<td>Specific heat (s)</td>
<td>$c_p(s)$</td>
<td>36.0</td>
<td>J/mol K</td>
</tr>
<tr>
<td>Thermal conductivity (l)</td>
<td>$\lambda_l$</td>
<td>0.734 $\cdot$ 10$^{-3}$</td>
<td>W/cm K</td>
</tr>
<tr>
<td>Thermal conductivity (s)</td>
<td>$\lambda_s$</td>
<td>4.76 $\cdot$ 10$^{-3}$</td>
<td>W/cm K</td>
</tr>
<tr>
<td>Thermal diffusivity (l)</td>
<td>$D_{th}(l)$</td>
<td>7.29 $\cdot$ 10$^{-4}$</td>
<td>cm$^2$/s</td>
</tr>
<tr>
<td>Thermal diffusivity (s)</td>
<td>$D_{th}(s)$</td>
<td>4.96 $\cdot$ 10$^{-3}$</td>
<td>cm$^2$/s</td>
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<td>Refractive index (l)</td>
<td>$n_l$</td>
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</tr>
<tr>
<td>Refractive index (s)</td>
<td>$n_s$</td>
<td>1.4560</td>
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</tr>
<tr>
<td>Solid-liquid interfacial free energy</td>
<td>$\gamma_{sl}$</td>
<td>1.073 $\cdot$ 10$^{-8}$</td>
<td>J/cm$^2$</td>
</tr>
<tr>
<td>Capillary length</td>
<td>$d_0$</td>
<td>4.9 $\cdot$ 10$^{-8}$</td>
<td>cm</td>
</tr>
<tr>
<td>Unit of supercooling</td>
<td>$\Theta$</td>
<td>59.2</td>
<td>K</td>
</tr>
</tbody>
</table>

1 (l) liquid; (s) solid

Materials with $\alpha < 2$ are usually not faceting in contact with the melt.
### B. Dendrites

Tab. B.1: Quantitative experimental properties of investigated dendrites

<table>
<thead>
<tr>
<th>Dendrite</th>
<th>#1</th>
<th>#2</th>
<th>#3</th>
<th>#4</th>
<th>#5</th>
<th>#6</th>
<th>#7</th>
<th>#8</th>
<th>#9</th>
<th>#10</th>
<th>#11</th>
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<tbody>
<tr>
<td>$\Delta T$ (≤ 1%) [mK]</td>
<td>65.5</td>
<td>16.9</td>
<td>217.5</td>
<td>207.5</td>
<td>61.7</td>
<td>41.3</td>
<td>95.7</td>
<td>119.1</td>
<td>50.7</td>
<td>23.6</td>
<td>139.3</td>
</tr>
<tr>
<td>$R_{\text{exp}}$ (≤ 1%) [µm]</td>
<td>50.0</td>
<td>153.9</td>
<td>18.5</td>
<td>19.2</td>
<td>52.5</td>
<td>73.3</td>
<td>36.5</td>
<td>30.4</td>
<td>61.8</td>
<td>116.7</td>
<td>26.7</td>
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<tr>
<td>$N$</td>
<td>146</td>
<td>76</td>
<td>85</td>
<td>195</td>
<td>190</td>
<td>76</td>
<td>300</td>
<td>37</td>
<td>42</td>
<td>71</td>
<td>15</td>
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<tr>
<td>$\Delta t$ (≤ 1%) [s]</td>
<td>30</td>
<td>60</td>
<td>10</td>
<td>5</td>
<td>15</td>
<td>60</td>
<td>1</td>
<td>30</td>
<td>120</td>
<td>120</td>
<td>30</td>
</tr>
<tr>
<td>$\phi$ (≤ 1%) [grad]</td>
<td>41.8</td>
<td>26.1</td>
<td>23.6</td>
<td>27.2</td>
<td>42.8</td>
<td>39.9</td>
<td>0</td>
<td>9.7</td>
<td>30.4</td>
<td>50.4</td>
<td>60</td>
</tr>
<tr>
<td>$z_{\text{frame}}$</td>
<td>50</td>
<td>20</td>
<td>90</td>
<td>70</td>
<td>80</td>
<td>50</td>
<td>60</td>
<td>110</td>
<td>70</td>
<td>40</td>
<td>150</td>
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</tbody>
</table>

**Trunk**

<table>
<thead>
<tr>
<th>$z_{\text{sat}}$ (±10%)</th>
<th>▲ 24 *</th>
<th>20</th>
<th>28</th>
<th>27</th>
<th>14</th>
<th>30</th>
<th>27</th>
<th>25</th>
<th>18</th>
<th>35</th>
</tr>
</thead>
<tbody>
<tr>
<td>▼ 23</td>
<td>9</td>
<td>20</td>
<td>27</td>
<td>27</td>
<td>13</td>
<td>26</td>
<td>30</td>
<td>22</td>
<td>17</td>
<td>27</td>
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</table>

<table>
<thead>
<tr>
<th></th>
<th></th>
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<th></th>
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<th></th>
<th></th>
<th></th>
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</thead>
<tbody>
<tr>
<td>▼ -225 *</td>
<td>-104</td>
<td>-111</td>
<td>-32</td>
<td>-43</td>
<td>-14</td>
<td>-98</td>
<td>-98</td>
<td>-73</td>
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</table>

<table>
<thead>
<tr>
<th>$B_{\text{sat}}$ (±10%)</th>
<th>▲ 10</th>
<th>22</th>
<th>21</th>
<th>24</th>
<th>6</th>
<th>29</th>
<th>20</th>
<th>33</th>
<th>10</th>
<th>31</th>
</tr>
</thead>
<tbody>
<tr>
<td>▼ 30</td>
<td>*</td>
<td>16</td>
<td>16</td>
<td>8</td>
<td>9</td>
<td>20</td>
<td>15</td>
<td>14</td>
<td>14</td>
<td>-∞</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$\frac{d_{\text{sat}}}{R_{\text{exp}}}$ (±10%)</th>
<th>▲ 9.4</th>
<th>8.5</th>
<th>8.5</th>
<th>9.1</th>
<th>8.6</th>
<th>9.1</th>
<th>9.4</th>
<th>9.6</th>
<th>9.9</th>
<th>12.9</th>
<th>8.1</th>
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<tbody>
<tr>
<td>▼ 8.2 *</td>
<td>8.6</td>
<td>9.2</td>
<td>8.0</td>
<td>7.8</td>
<td>9.2</td>
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<td>8.5</td>
<td>10.4</td>
<td>7.6</td>
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<td></td>
</tr>
</tbody>
</table>

**Envelope**

<table>
<thead>
<tr>
<th>$N_{\text{active}}$</th>
<th>▲ 24</th>
<th>3</th>
<th>*</th>
<th>87</th>
<th>33</th>
<th>8</th>
<th>63</th>
<th>34</th>
<th>19</th>
<th>4</th>
<th>11</th>
</tr>
</thead>
<tbody>
<tr>
<td>▼ 36</td>
<td>3</td>
<td>*</td>
<td>89</td>
<td>39</td>
<td>9</td>
<td>61</td>
<td>27</td>
<td>18</td>
<td>6</td>
<td>15</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$q^{(a)}$ (±10%) [grad]</th>
<th>▲ 30.5</th>
<th>25.5</th>
<th>22.6</th>
<th>22.0</th>
<th>27.2</th>
<th>21.0</th>
<th>26.4</th>
<th>27.4</th>
<th>*</th>
<th>*</th>
<th>20.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>▼ 27.2</td>
<td>31.0</td>
<td>28.4</td>
<td>28.1</td>
<td>38.0</td>
<td>26.6</td>
<td>26.2</td>
<td>30.5</td>
<td>*</td>
<td>*</td>
<td>36.5</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$q^{(b)}$ (±10%) [grad]</th>
<th>▲ 25.8</th>
<th>21.7</th>
<th>21.8</th>
<th>20.0</th>
<th>18.4</th>
<th>26.6</th>
<th>23.5</th>
<th>23.4</th>
<th>23.5</th>
<th>19.3</th>
<th>15.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>▼ 26.1</td>
<td>31.3</td>
<td>26.4</td>
<td>25.0</td>
<td>32.5</td>
<td>29.4</td>
<td>23.5</td>
<td>26.6</td>
<td>30.5</td>
<td>28.7</td>
<td>24.1</td>
<td></td>
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</tbody>
</table>

**Growth Velocity of Active Sidebranches**

<table>
<thead>
<tr>
<th>$v_{\text{SB}}/v_{\text{tip}}$ (±10%)</th>
<th>▲ 0.51</th>
<th>*</th>
<th>0.49</th>
<th>0.51</th>
<th>0.51</th>
<th>0.57</th>
<th>0.67</th>
<th>0.54</th>
<th>0.55</th>
<th>0.53</th>
<th>*</th>
</tr>
</thead>
<tbody>
<tr>
<td>▼ 0.54</td>
<td>*</td>
<td>0.52</td>
<td>0.54</td>
<td>0.63</td>
<td>0.64</td>
<td>0.66</td>
<td>0.57</td>
<td>0.57</td>
<td>0.54</td>
<td>*</td>
<td></td>
</tr>
</tbody>
</table>
Refer to the upper and lower side of the dendrite, respectively;

**Dendrite**

- **ΔT**  Supercooling of the dendrite;
- **R_{exp}**  Experimentally determined tip radius of the dendrite;
- **N**  Number of video frames digitized during the growth of the dendrite;
- **Δt**  Time step between two consecutive digitized video frames;
- **ϕ**  Angle between the direction of the earth gravitational field and the main axis of the dendrite during the experiment;
- **\hat{z}_{frame}**  Approximate length of the imaged dendrite in units of R_{exp};

**Trunk** (Sec. 3.2)

- **z_{sat}**  Value of the lower boundary z_l of the fitting interval [z_l, z_u], where the fitted slope saturates;
- **A_{sat}**  Fitted value of the parameter A in the interval [z_{sat}, z_u];
- **B_{sat}**  Fitted slope B of the linear regression in the interval [z_{sat}, z_u];
- **\hat{w}_{2}/R_{exp}**  Half width of the trunk at a distance \( z = 30 R_{exp} \) behind the dendrite tip, scaled by the tip radius R_{exp};

**Envelope** (Sec. 3.3.1)

- **N_{active}**  Number of active sidebranches;
- **ϕ^{(a)}**  Opening angle of the envelope determined by investigation of all of the active sidebranches;
- **ϕ^{(b)}**  Opening angle of the envelope determined by investigation of the temporal development of the maxima belonging to the active sidebranches;

**Growth velocity of the active sidebranches** (Sec. 3.3.2)

- **v_{SB}/v_{tip}**  Growth velocity of the active sidebranches, scaled by the growth velocity v_{tip} of the dendrite tip.
C. Gaussian Superposition

<table>
<thead>
<tr>
<th>( z )</th>
<th>10</th>
<th>15</th>
<th>20</th>
<th>25</th>
<th>30</th>
<th>35</th>
</tr>
</thead>
<tbody>
<tr>
<td>( N_z(\text{data}) )</td>
<td>18</td>
<td>65</td>
<td>148</td>
<td>111</td>
<td>53</td>
<td>11</td>
</tr>
<tr>
<td>( N_z(\text{Gauss}) )</td>
<td>3</td>
<td>4</td>
<td>6</td>
<td>5</td>
<td>4</td>
<td>3</td>
</tr>
<tr>
<td>( \lambda_1^2 )</td>
<td>3.0</td>
<td>2.9</td>
<td>2.2</td>
<td>2.0</td>
<td>3.2</td>
<td>3.0</td>
</tr>
<tr>
<td>( \sigma_1^2 )</td>
<td>0.5</td>
<td>0.7</td>
<td>0.5</td>
<td>0.8</td>
<td>0.7</td>
<td>0.5</td>
</tr>
<tr>
<td>( I_1^2 ) [a.u.]</td>
<td>2.5</td>
<td>4.1</td>
<td>1.0</td>
<td>0.5</td>
<td>2.7</td>
<td>1.0</td>
</tr>
<tr>
<td>( \lambda_2^2 )</td>
<td>3.6</td>
<td>3.6</td>
<td>3.0</td>
<td>3.2</td>
<td>4.2</td>
<td>3.0</td>
</tr>
<tr>
<td>( \sigma_2^2 )</td>
<td>0.5</td>
<td>0.7</td>
<td>0.7</td>
<td>0.7</td>
<td>0.5</td>
<td>0.6</td>
</tr>
<tr>
<td>( I_2^2 ) [a.u.]</td>
<td>5.0</td>
<td>4.8</td>
<td>4.8</td>
<td>4.9</td>
<td>5.0</td>
<td>4.9</td>
</tr>
<tr>
<td>( \lambda_3^2 )</td>
<td>5.0</td>
<td>6.0</td>
<td>3.8</td>
<td>4.0</td>
<td>5.1</td>
<td>6.1</td>
</tr>
<tr>
<td>( \sigma_3^2 )</td>
<td>0.7</td>
<td>0.6</td>
<td>0.7</td>
<td>0.6</td>
<td>0.5</td>
<td>0.7</td>
</tr>
<tr>
<td>( I_3^2 ) [a.u.]</td>
<td>1.7</td>
<td>1.9</td>
<td>4.8</td>
<td>4.7</td>
<td>1.8</td>
<td>2.5</td>
</tr>
<tr>
<td>( \lambda_4^2 )</td>
<td>9.5</td>
<td>5.0</td>
<td>5.7</td>
<td>0.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \sigma_4^2 )</td>
<td>0.5</td>
<td>0.6</td>
<td>0.8</td>
<td>0.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( I_4^2 ) [a.u.]</td>
<td>0.5</td>
<td>0.7</td>
<td>1.2</td>
<td>2.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \lambda_5^2 )</td>
<td>6.3</td>
<td>9.7</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \sigma_5^2 )</td>
<td>0.5</td>
<td>0.6</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( I_5^2 ) [a.u.]</td>
<td>0.9</td>
<td>0.4</td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>( \lambda_6^2 )</td>
<td>9.6</td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>( \sigma_6^2 )</td>
<td>0.5</td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( I_6^2 ) [a.u.]</td>
<td>0.9</td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>

Tab. C.1: Fitted Superimposed Gaussian Functions in Fig. 3.12c)
\( \hat{z} \) Lower boundary of the summation interval, scaled by the tip radius \( R_{\text{exp}} \);
\( N_{z(\text{data})} \) Number of experimentally determined points \((\lambda^i, z^i)\) having contributed to the sums of the Gaussian functions, \( f_z(\lambda) \);
\( N_{z(\text{Gauss})} \) Number of Gaussian functions \( g_j(\lambda) \) used to fit \( f_z(\lambda) \)
\( \lambda_j^i \) Location of the Gaussian peak of \( g_j(\lambda) \);
\( \sigma_j^i \) Standard deviation of the Gaussian function \( g_j(\lambda) \);
\( I_j^i \) Intensity of the Gaussian peak of \( g_j(\lambda) \) scaled to a standard height.

The gray colors marking the locations \( \lambda_j^i \) of some of the Gaussian peaks help to trace their development with increasing \( \hat{z} \).

From Tab C.1 it follows that the peak of \( g_{10}^0(\lambda) \) located at \( \lambda_{10}^0 = 3.6 \pm 0.5 \) in the first interval beginning at \( \hat{z} = 10 \), drifts to \( \lambda_{30}^0 = 4.2 \pm 0.5 \) in the interval beginning at \( \hat{z} = 30 \). With the introduction of an effective tip radius, \( R_{\text{eff}} \), similar as in Eq. (3.8) in the case of the trunk, we can rescale the peak \( \lambda_{30}^0 \) into units of its effective tip radius, \( R_{\text{eff},30} \), at a distance \( z = 30 R_{\text{exp}} \) from the tip by means of \( \lambda_{30}^0 \approx 3.6 \tilde{R}_{\text{eff},30} \), leading to

\[
\tilde{R}_{\text{eff},30} \approx \frac{\lambda_{30}^0}{3.6} \approx \frac{4.2 R_{\text{exp}}}{3.6} \approx 1.17 R_{\text{exp}}.
\]

Now, we calculate the effective tip radius, \( R_{\text{eff},30} \), by means of the widths of the trunk, \( d_{10} \) and \( d_{30} \), at \( z = 10 R_{\text{exp}} \) and \( 30 R_{\text{exp}} \) behind the dendrite tip, respectively. The linear development of the trunk of this dendrite, \( \hat{z} = -67 + 10 \hat{z} \) (see Tab. B.1), leads to \( d_{10} \approx \frac{(10+67) R_{\text{exp}}}{10} = 7.7 R_{\text{exp}} \) and \( d_{30} \approx \frac{(30+67) R_{\text{exp}}}{10} = 9.7 R_{\text{exp}} \). Making use of the scaling relation between \( d \) and \( R_{\text{eff}} \), Eq. (3.10), we calculate the effective tip radius, \( R_{\text{eff},30} \), distance \( z = 30 R_{\text{exp}} \) from the tip by means of \( d_{30} \approx 7.7 R_{\text{eff},30} \), leading to

\[
R_{\text{eff},30} \approx \frac{d_{30}}{7.7} \approx \frac{9.7 R_{\text{exp}}}{7.7} \approx 1.26 R_{\text{exp}}.
\]

The calculated effective tip radius in the case of the sidebranches, \( \tilde{R}_{\text{eff},30} \) in Eq. (C.1) agrees with the one calculated in the case of the trunk, \( R_{\text{eff},30} \) in Eq. (C.2) within \( \pm 8\% \). Therefore, we conclude that the continuous drifts in the sidebranch spacings correspond to the increased temperature along the dendrite.
Bibliography


Curriculum Vitae

April 1969
Born in Luzern, Switzerland

1976-1982
Primary school in Luzern

1982-1990
High school (Kantonsschule Luzern and Gymnasium St. Klemens in Ebikon, LU)

June 1990
Maturity type B

1990-1995
Study of physics at the ETH Zürich

October 1995
Diploma work in experimental physics at the ETH Zürich in the group of Prof. H. R. Ott on
“Josephson effect in Bi$_2$Sr$_2$CaCu$_2$O$_8$”

1996-2000
Research and teaching assistant at the laboratory for Solid State Physics at the ETH Zürich in the Freezing and Melting team of Prof. J. H. Bilgram within the group of Prof. H. R. Ott

2000
Ph.D. thesis at the ETH Zürich on “Sidebranch Development of Xenon Dendrites”
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