Resonating viscosity sensors
micro versus macro approach

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Resonating Viscosity Sensors
Micro versus Macro Approach

A thesis submitted to the
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1999
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ABSTRACT

This thesis reports on the development, fabrication, and characterization of integrated viscosity sensors. In the process of this work, four different miniaturized sensor systems have been investigated and compared with each other. The investigated sensors are a Lamb wave device, a membrane resonator, a thickness shear mode resonator, and a cantilever beam resonator. The four sensor types span an operating frequency range from 200 Hz to 20 MHz. Suitable packages allowing for liquid immersion and future on-line application have been developed.

The sensing principle is based on changes of acoustic wave modes due to a contacting liquid. In all four sensors, piezoelectric transducer elements are used to excite device specific acoustic wave modes. Due to liquid loading the characteristics of the oscillation, such as the resonance frequency and the amplitude of the vibration, are modified. The inverse piezoelectric effect is used to detect and monitor these changes. The shift of the resonance frequency is primarily related to the specific density of the liquid, while changes of the vibration amplitude and the quality factor are related to the liquid viscosity. Besides the micromachined transducer elements based on piezoelectric ZnO films, two commercially available piezoelectric resonators were investigated for comparison.

The Lamb wave sensor and the membrane resonator were fabricated with the bipolar DIMES-01 process at the TU Delft, The Netherlands. This process features optional process modules for the deposition of piezoelectric ZnO films and bulk micromachining. The piezoelectric transducer film consists of a 2.4 μm polycrystalline ZnO layer. A modified fabrication process for the deposition of ZnO films with low tensile stress and high piezoelectric coupling coefficient was developed. The intrinsic stress and the plane strain modulus of the sputtered ZnO films were investigated as a function of the deposition parameters.

The Lamb wave sensors are based on a transmission delay line and consist of a thin silicon membrane of 3 x 8 mm² covered with a piezoelectric transducer layer. An interdigital electrode structure is used to piezoelectrically generate a flexural plate wave in the membrane. A second identical interdigital electrode structure is used to detect the transmitted flexural plate wave. The center frequency and insertion loss of the acoustic wave due to liquid contact were investi-
gated. The design of the interdigital electrodes and the thickness of the ZnO layer were optimized for high piezoelectric conversion coefficients and good transmission characteristics. The thickness of the membrane sandwich was reduced to 8 μm to achieve a lower center frequency of 13.6 MHz.

The investigated piezoelectric membrane resonators are based on the same process as the Lamb wave sensors. Square membranes of $295 \times 295 \, \mu m^2$ with resonance frequencies ranging from 200 - 400 kHz and quality factors of about 90 in air were fabricated. An AC burst signal applied to the center electrode of the membrane excites a transverse membrane vibration. Resonance frequency and vibration amplitude were detected by an electrode located along the membrane edge using the inverse piezoelectric effect and related to the liquid properties.

The thickness shear mode resonator (TSMR) is based on a commercially available, 5 MHz AT-cut quartz of 2.54 cm diameter. The fundamental quartz oscillation mode, a pure transversal shear mode, was characterized with impedance measurements. An electric equivalent circuit model was used to model the impedance characteristic and the calculated equivalent circuit parameters were related to the properties of the liquids.

The commercially available piezoelectric beam resonators are either 2.4 or 4.8 cm long, 1 cm wide, and 1 mm thick. The cantilever is composed of a sandwich of two piezoelectric PZT layers and three metal electrodes. Application of an AC burst signal to one of the PZT layers excites the cantilever into transverse vibrations. The other layer is used to detect the oscillation amplitude. The resonance frequency which is in the order of several hundred Hz and the quality factor were measured and related to the liquid properties.

All sensor systems were characterized with different Newtonian and non-Newtonian liquids and tested for their liquid property sensing capability. The density and shear viscosity of the test liquids were in the range of 0.7 - 1.3 g·cm$^{-3}$ and 0.3 - 1500 mPa·s, respectively. The measurable viscosity range of all sensors ranges up to 1.5 Pa·s. Poor sensor resolution due to heavy damping prevents sensor operation in high viscous liquids. Furthermore, the operation frequencies restrict the application range. High frequency sensors, such as the Lamb wave sensor and the TSMR, are well suited for sensing Newtonian liquids with low viscosity $\eta < 100$ mPa·s. Non-Newtonian liquids tend to respond viscoelastic with increasing probing frequency. Best results for non-Newtonian liquids were achieved with the piezoelectric cantilever, because of its low operation frequency.
ZUSAMMENFASSUNG


Der Lamb-Wave Sensor basiert auf dem Prinzip einer Verzögerungs-Transmissionsleitung und besteht aus einer dünnen Siliziummembran von 3 x 8 mm² mit einer piezoelektrischen Beschichtung. Mit Hilfe einer metallischen Interdigitalstruktur werden piezoelektrisch akustische Wellen in der Membran erzeugt und
Zusammenfassung

mittels einer zweiten, identischen Struktur detektiert. Die Geometrie der Elektroden und die Dicke der ZnO-Schicht wurde in Bezug auf grösstmögliche piezoelektrische Kopplung und gute Transmissionscharakteristik optimiert. Zum Erreichen tieferer Schallwellengeschwindigkeiten wurde die Dicke der Membran auf 8 µm reduziert, wodurch die Resonanzfrequenz auf 13.6 MHz reduziert werden konnte.


Der Scherschwingquartz ist ein 5 MHz AT-cut Quarz mit 2.54 cm Durchmesser. Zur Messung des Schwingungszustandes wurden Impedanzmessungen durchgeführt. Daraus wurden die Parameter für ein elektrisches Ersatzschaltbild extrahiert und mit den Flüssigkeitseigenschaften korreliert.

Der kommerziell erhältliche, piezoelektrische Biegebalken ist 2.4 bzw. 4.8 cm lang, 1 cm breit, 1 mm dick und hat eine Resonanzfrequenz von einigen hundert Hz. Er besteht aus einem Sandwich von zwei PZT Schichten und drei Metallelektroden. Durch Anlegen einer Wechselspannung an eine der Schichten wird eine transversale Schwingung angeregt. Mit Hilfe der zweiten piezoelektrischen Schicht wird das freie Ausschwingen des Balkens detektiert, und die Resonanzfrequenz sowie die Dämpfung errechnet.

Alle Sensoren wurden mit verschiedenen Newtonschen und nicht-Newtonsschen Flüssigkeiten charakterisiert und auf Ihre Anwendbarkeit als Viskositätssensoren überprüft. Der Dichte- und Viskositätsbereich der Flüssigkeiten liegt zwischen 0.7 - 1.3 g·cm⁻³, respektive 0.3 - 1500 mPas. Der messbare Viskositätsbereich aller Sensoren geht bis ca. 1.5 Pas, bevor sie mangels Sensitivität oder zu starker Dämpfung nicht mehr betrieben werden können. Weitere Einschränkungen bezüglich der messbaren Flüssigkeiten ergeben sich durch die verwendeten Frequenzbereiche. Hochfrequente Sensoren eignen sich hauptsächlich für Newtonssche Flüssigkeiten da bei nicht-Newtonsschen viskoelastische Eigenschaften eine zusätzliche Rolle spielen. Aufgrund seiner tiefen Resonanzfrequenz eignet sich der Biegeschwinger am besten zum Messen nicht-Newtonsscher Flüssigkeiten.
1 INTRODUCTION

Only 50 years ago, semiconductor components have started to revolutionize the modern world. What begun with a simple, bulky, bipolar transistor in a lab at Bell in 1947 [1] has become a world market beyond imagination and changed our daily life dramatically. Modern fabrication techniques and continuously improving technology make the price/performance ratio of integrated circuitry (IC) unbeatable and available for a large range of consumers. At the beginning, it was only for the integration of electronic circuitry but soon silicon became a platform for integrated sensors and actuators (ISA) or, in combination with electronic circuitry, the basis for integrated micro electro mechanical systems (iMEMS). The increasing demand of industry and consumers for continuous monitoring, e.g., for product quality control, environmental control, security and safety, and health is pushing the technology further and further. Modern intelligent sensor systems take advantage of the cost effective mass production developed for IC manufacturing and more and more become available in consumer products. Market studies predict at least a doubling of the microsystem technology market by the year 2002 [2]. With the smart sensor systems silicon is up for a new revolution, until they will finally penetrate our every day live and change our way of living [3, 4].

1.1 RHEOLOGY

Rheology is the science of deformation and flow, particularly of liquids. Typical rheological modification processes in our daily life are cooking, baking, mixing, dissolving, and gelatinizing where we unconsciously apply thermal and mechanical processes to selectively change and modify the rheological properties of materials, especially that of foodstuff [5, 6]. And as we all know from our personal experience, continuous monitoring and testing is indispensable for a good result. Therefore, it is not surprising that rheology is an indispensable instrument for continuous foodstuff quality control in industrial manufacturing facilities. However, the application of commercial process rheometers to on-line foodstuff quality control is limited due to their size, cost, and working principles. Up to now only parameters such as pressure, temperature, flow volume, or optical transparency can be reliably monitored on-line. Other parameters such as viscos-
1 Introduction

...taste, particle size distribution, and concentration have to be measured off-line, if it is possible at all. Off-line controls are time consuming and the slow response time can severely increase the manufacturing cost due to:

- Material loss due to insufficient quality of the manufactured product.
- Loss of effective production time due to down time.

Consequently, new methods suitable for continuous on-line foodstuff rheology monitoring have to be developed.

1.2 State of the Art

A large number of test equipment has been developed to examine and characterize liquids. Although the principles for the most common techniques are more than hundred years old, they are still in use [7]. Three categories of rheometers, including the most recent technologies, are briefly discussed in the following:

- Classical rheometers.
- Velocity profile imaging.
- Microsensors applied to fluid property sensing.

1.2.1 Conventional Rheometry

The classical field of shear rheometry is usually subdivided in drag flow and pressure flow methods. Drag flow methods, such as cone-plate, parallel disk, or concentric cylinders, belong to the Couette flow type methods and are well-established off-line methods [7, 8]. Eventually, a side stream setup with on-line capability is possible [7].

The drawbacks of off-line methods are obvious. The time delay between sample collection and data evaluation may be too long, and the sample may be no longer representative for the process conditions. The laboratory condition may not be representative for the process conditions. Moreover, tons of products may already be processed, before the results of an off-line measurement become available.

In pressure driven flows, the natural condition for on-line application methods, pressure difference measurements are made. From the pressure loss over a known distance liquid material properties can be calculated. The calculated results are based on a series of assumptions about the fluid flow. The main shortcomings are...
the difficult technical realization and the assumptions that have to be made about the fluid flow. For these reasons pressure difference methods have only limited on-line application capability.

1.2.2 VELOCITY PROFILE IMAGING

Until recently, no major breakthroughs have been made in on-line rheology and most methods were based on old principles, established almost hundred years ago. Since about a decade new technologies, such as nuclear magnetic resonance imaging (NMRI) [9, 10, 11] or ultrasound velocity profilometry (UVP) [12, 13], have been developed as new methods for continuous and reliable on-line control in the near future.

Both methods are contactless and allow a relatively fast (NMR) or almost instantaneous (UVP) visualization of the flow velocity profile in a tube. In combination with a pressure difference measurement, the complete flow curve over the entire shear rate range in the fluid is obtained instantaneously. The advantages of these methods are manifold [14]:

- Contactless measurements, implying no flow disturbance, on-line cleaning ability, and little influence on the liquid properties.
- Direct flow curve measurements, requiring no assumptions on the flow profile.

The limitations of these methods are only the relatively high cost of these systems for routine on-line application. However, low cost versions are currently under development [15]. Several new approaches, including iMEMS, are currently evaluated as alternatives to the expensive high-end methods.

1.2.3 MICROSENSORS

Most microsensors for liquid property sensing are based on acoustic wave principles. Vibrating rheometers based on rods or tubes operating in torsional oscillation at resonance frequencies in the range of 10 - 500 kHz are reported in [16, 17, 18, 19]. Fibers, oscillating in the 5 kHz range, were also investigated as viscosity sensors [20]. More common are piezoelectric resonators, such as 2 - 10 MHz quartz resonators [21, 22] or delay line configurations operating in the 10 - 100 MHz range [23, 24, 25].
1 Introduction

Among the various reported microsensors only few are silicon based: cantilever structures [26], resonant membranes [27], and Lamb wave devices [28, 29, 30] have been reported. The common sensing principles of these sensors also lead to similar characteristics. The major shortcomings of acoustic wave sensors are:

- High frequency operation range compared to conventional rheometers, viscoelastic effects become more pronounced with increasing frequency and the measurement results can hardly be compared with results of classical rheometers.

- The frequency range is very limited, in most devices the measurements can only be made at a single frequency.

A non-resonant device to measure liquid properties is a silicon based shear stress sensor [31]. The sensor measures the shear stress generated by a flowing liquid at the wall of a tube.

1.3 Micro Versus Macro

Miniaturization of rheometers in order to achieve inexpensive sensors for on-line applications is not straightforward because the measurement conditions change dramatically. Especially in rheology, where the measurement result is a function of the applied liquid deformation the impact of miniaturization is high. Two conceptual changes have to be considered when changing from macro to micro methods.

In conventional rheometers the probed sample volume is usually very large compared to the characteristic structure size in the liquid. Consequently even inhomogeneous liquids, such as suspensions, can be regarded as a quasi continuum. In contrast, microsensors probe only a very small sample volume. Depending on the sensor type the measured liquid layer can be less than 250 nm thick [22]. Typical particle or structure dimensions are often much larger than the sensed volume. Hence, the question rises whether the sensed liquid properties still correspond to those of the bulk material.

The measurement frequencies of conventional rheometers range from 0 - 200 Hz and can be changed continuously. Usually, the liquid behavior is measured for the maximum frequency range possible to get a complete characterization of the liquid under investigation. In contrast, microsensor systems operate over a very lim-
ited frequency range in the high frequency domain, i.e., up to several MHz for acoustic wave devices.

1.4 OUTLINE OF THE THESIS

The goal of this thesis is the development of microsensors for on-line rheometry. A brief outline of the contents and structure of the thesis is given in the following. Chapter 2 introduces the fundamentals of fluid rheology. Simple models for viscous and viscoelastic behavior are discussed and the choice of the sample liquids is motivated. In Chapter 3, the four investigated sensors, a thickness shear mode resonator, a Lamb wave device, a piezoelectric cantilever, and a membrane resonator are presented and their working principle is explained. The basic theory of the different sensors types is outlined and the key points for an optimized sensor design are discussed. Each sensor operates in a different acoustic mode and frequency range. Chapter 4 addresses the fabrication and packaging of the sensors. Especially, the fabrication sequence of the piezoelectric microsensors is described. The piezoelectric ZnO films are characterized by measuring stress, Young's modulus, ZnO composition and orientation. Moreover, the individual packaging solutions for the different sensor types are shown. In Chapter 5, the characterization and performance of the unloaded devices (operation in air) is presented and the various measurement techniques are established. Chapter 6 reports on the test liquid measurements and the performance of the liquid loaded sensors. Various Newtonian and non-Newtonian liquids are characterized and the results are summarized. Advantages and disadvantages of the different sensor types for future on-line applications are discussed. Chapter 7 summarizes the thesis and gives an outlook on the future work.
2 Viscosity

The basic idea for a viscous fluid was first introduced by Sir Isaac S. Newton in 1697 in his famous *Principia Mathematica* [32]: "The resistance which arises from the lack of slipperiness originating in a fluid, other things being equal, is proportional to the velocity by which the parts of the fluid are being separated from each other." But it was not until 1845 that Stokes finally was able to write out this concept in a three-dimensional mathematical form [7].

As discovered by Newton, viscosity is a measure of the flow ability of materials. Any material, be it gas, liquid, or solid, is subject to deformation and thus flows. The science of studying liquid flow and solid deformation behavior is called rheology, from the Greek verb *ρεύνω*, to flow. However, rheology is usually restricted to the study of the fundamental relations, called the constitutive relations, between force and deformation in materials, primarily liquids.

2.1 Introduction

To discuss the concept of viscosity we consider only the most elementary example of shear flow in which a relative displacement of parallel infinitely thin planes of fluid occurs [8]. Such a flow is called simple shear flow and is shown schematically in Fig. 2.1.

Given the coordinate system in Fig. 2.1, the fluid velocity components are

\[ v_x = v_x(z), \quad v_y = 0, \quad v_z = 0. \quad (2.1) \]

The measure of the local dynamic state of deformation is the shear stress \( \sigma = \sigma_{xy} \) and the measure of the local kinematic state of deformation is the shear rate \( \dot{\gamma} \) which is defined as

\[ \dot{\gamma} = \frac{d}{dz} v_x(z). \quad (2.2) \]
The simplest constitutive equation for the shear flow of a fluid is

\[ \sigma = \eta \dot{\gamma}, \]  

(2.3)

where \( \eta \) is the Newtonian shear viscosity. The Newtonian shear viscosity \( \eta \) is constant for a given temperature and pressure and independent of the shear rate. Such fluids are called Newtonian and are represented with a dash pot in mechanical equivalent models, as shown in Tab. 2.1.

Otherwise, a generalized Newtonian fluid, i.e., a purely viscous fluid with shear rate dependent shear viscosity \( \eta(\dot{\gamma}) \), is defined. Even the generalized Newtonian fluid model cannot describe the so-called memory effects which may occur during processing of a fluid of interest. These effects are of elastic nature or due to processes involving structural changes of the fluid. The influence of the fluid memory on the flow process is characterized with the Deboiah number

\[ De = \frac{\text{characteristic fluid time}}{\text{characteristic process time}}, \]  

(2.4)

which is the ratio of the characteristic time of the fluid to the characteristic time of the process. For fluid deformation processes that are slow compared to the characteristic fluid time, e.g., a relaxation time, the influence of the fluid memory...
on the flow process can be neglected and the rheological properties of the fluid can be approximated by the generalized Newtonian fluid.

2.2 Viscosity and Viscoelasticity

Newtonian fluids, defined through their linear relationship between stress and shear rate, represent a very special class of fluids. However, most fluids behave non-Newtonian. Fig. 2.2 shows some examples of flow curves and their classification. The simplest mathematical model used to describe the flow curves of types 1.-3. in Fig. 2.2 is the power law-model of Ostwald-de Waele

\[ \sigma = k\dot{\gamma}^n, \]  

(2.5)

where \( k \) and \( n \) are the rheological parameters of the model. For \( n = 1 \) a Newtonian flow curve is obtained. In contrast, flow curves with \( n < 1 \) and \( n > 1 \) describe so called shear thinning and shear thickening liquids, respectively.

The corresponding rheological model for viscoplastic flow curves is the Herschel-Bulkley model

\[ \sigma = \sigma_y + k\dot{\gamma}^n, \]  

(2.6)

with the additional rheological parameter \( \sigma_y \) describing the yield stress of the fluid. A set of flow curves for fluids with yield stress, e.g., thixotropic fluids, are the curves 4.-6. in Fig. 2.2.

Often liquids do not obey these simple laws and exhibit very complicated rheological behavior. As mentioned before, memory effects such as creep or relaxation may occur. These effects can be described as a mixture between viscous and solid material behavior. A simple model for a purely solid material is Hook's law

\[ \sigma = G\gamma, \]  

(2.7)

where \( G \) is the elastic modulus and \( \gamma \) the strain. The equivalent mechanical model for this behavior is a spring. Whether the fluid exhibits more viscous or more elastic behavior depends on the kind of experiment performed. For example simple shear flow experiments are not suited to measure viscoelastic properties. For
this purpose the material is exposed to a periodically oscillating shear stress according to

$$\sigma(\omega) = \sigma_0 e^{i\omega t}$$  \hspace{1cm} (2.8)

where $\sigma_0$ is the amplitude of the shear stress and $\omega$ the angular frequency. The strain

$$\gamma(\omega) = \gamma_0 e^{i(\omega t - \delta)}$$  \hspace{1cm} (2.9)

caused by the varying stress $\sigma(\omega)$ is monitored. Similar to Eqn. (2.7), the complex modulus of elasticity
2.2 Viscosity and Viscoelasticity

\[ G' = G' + iG'' = \frac{\sigma(\omega)}{\gamma(\omega)} \]  

(2.10)

can be defined as the ratio of the complex shear stress and the complex strain of the material, where \( G' \) is the storage modulus and \( G'' \) the loss modulus. In a similar way, the concept of a complex viscosity \( \eta^* \) can be introduced as

\[ \eta^* = \eta' - i\eta'' = \frac{\sigma(\omega)}{\gamma(\omega)} \]  

(2.11)

where \( \eta' \) is the so-called dynamic viscosity and \( \eta'' \) is the elastic part of the complex viscosity. For small angular frequency \( \omega \), the dynamic viscosity \( \eta' \) converges to the shear viscosity

\[ \eta = \lim_{\omega \to 0} \eta'(\omega). \]  

(2.12)

An empirical relation between the shear viscosity \( \eta \) and the dynamic viscosity \( \eta' \) is given by the Cox-Merz rule [8]

\[ \eta(\dot{\gamma}) = \left| \eta^\alpha(\omega) \right| = \left( \eta^2 + \left( \frac{G''}{\omega} \right)^2 \right)^{1/2} \]  

(2.13)

where \( \omega = \dot{\gamma} \). The simplest models to describe linear viscoelasticity are linear combinations of Hookean elastic and Newtonian viscous behavior. A series combination of the corresponding mechanical equivalent models, as shown in Tab. 2.1, results in the Maxwell model of a viscoelastic fluid [8]

\[ \tau \sigma + \sigma = \eta \dot{\gamma}. \]  

(2.14)

where

\[ \tau = \frac{\eta}{G} \]  

(2.15)
Viscosity

has the dimension of time, \( \tau \) is called relaxation time and characterizes the rate of stress decay within the solid. As long as the relaxation time \( \tau \ll \omega^{-1} \), the liquid behaves almost purely viscous, as can be seen in Tab. 2.1 for \( \eta^* \) of a Maxwell liquid. For inverse measurement frequencies \( \omega^{-1} \) in the range of the relaxation time \( \tau \), viscoelastic effects become more and more dominant. Finally, in the case of \( \omega^{-1} \gg \tau \) the fluid behaves purely elastic, i.e., like a solid.

Likewise, a linear viscoelastic solid is described by the Kelvin model

\[
\sigma = G\gamma + \eta \dot{\gamma},
\]

which corresponds to a parallel combination of a spring and a dashpot. Such a material behaves like a solid for low angular frequencies and exhibits more viscous behavior with increasing angular frequency. A summary of the presented models is given in Tab. 2.1.

The models presented above are the most elementary models for viscoelasticity and reveal the close relation between viscosity and elasticity in fluids, without discussing the origin of these effects. Most fluids show a much more complex rheological behavior [7].

Tab. 2.1: Summary of the four basic models for linear viscoelastic material behavior.

<table>
<thead>
<tr>
<th>Model</th>
<th>Type</th>
<th>Mechanical model</th>
<th>( G^* )</th>
<th>( \eta^* )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hook</td>
<td>elastic solid</td>
<td>( G )</td>
<td>( \frac{-iG}{\omega} )</td>
<td></td>
</tr>
<tr>
<td>Newton</td>
<td>viscous liquid</td>
<td>( i\omega \eta )</td>
<td>( \eta )</td>
<td></td>
</tr>
<tr>
<td>Maxwell</td>
<td>viscoelastic liquid</td>
<td>( \frac{\eta \omega^2 (\tau + i\omega^{-1})}{1 + \omega^2 \tau^2} )</td>
<td>( \frac{\eta (1-i\omega \tau)}{1 + \omega^2 \tau^2} )</td>
<td></td>
</tr>
<tr>
<td>Kelvin</td>
<td>viscoelastic solid</td>
<td>( G + i\omega \eta )</td>
<td>( \eta - \frac{iG}{\omega} )</td>
<td></td>
</tr>
</tbody>
</table>
2.3 MODEL SYSTEMS

The choice of a good model system is a key point for this work, since part of the devices investigated operate in the high frequency range where viscoelastic effects, as explained in Section 2.2, may occur. In the following, the choice of sample liquids is motivated and the liquid properties are characterized. The steady shear viscosity $\eta$ of the sample liquids was either measured with a Couette type rotational shear rheometer CS 50 from Bohlin Instruments, Cirencester, England, or tabulated data was used [33, 34].

Standard Newtonian liquids frequently used to characterize high frequency devices are H$_2$O/glycerol solutions. Arbitrary shear viscosities in the range of 1 - 1500 mPa·s at room temperature can be mixed by changing the weight concentration $\Psi$ of glycerol in H$_2$O, while the corresponding density $\rho_f$ changes from 1 - 1.29 g·cm$^{-3}$. The solutions behave Newtonian even up to frequencies of 84 MHz [35]. The temperature dependence of the shear viscosity of glycerol is tabulated in [33] and behaves approximately according to the Andrade-Eyring model [7]

$$\eta = \alpha e^{E_\eta/(RT)},$$

where $\alpha$ is a pre-exponential function, $E_\eta$ the activation energy for viscous flow, $R$ the gas constant, and $T$ the absolute temperature. Data and fit for glycerol are shown in Fig. 2.3.

A common Newtonian test liquid in rheological science and industry is poly(dimethylsiloxane) (PDMS). The shear viscosity of PDMS increases with increasing length of the polymer chains, whereas the density of about 0.97 g·cm$^{-3}$ hardly changes. The chosen PDMS samples with steady shear viscosity $\eta$ ranging from 10 - 5000 mPa·s were supplied by Wacker Chemie GmbH, Liestal, Switzerland. In contrast to H$_2$O/glycerol mixtures, PDMS shows increasing viscoelastic behavior in the high frequency range with increasing length of the polymer chains.

Mixtures of acetone and ak10, a PDMS with a shear viscosity of 9.3 mPa·s at 25 °C, were used to cover the low viscosity range from 0.3 - 9.3 mPa·s. The shear viscosity $\eta$ of acetone/ak10 mixtures strongly varies with the weight concentra-
Fig. 2.3: Temperature characteristics of the shear viscosity $\eta$ of glycerol. The data [33] is described by the Arrhenius-Eyring function with a flow activation energy of $E_\eta = 67 \text{kJ}\cdot\text{mol}^{-1}$.

As model system for suspensions, H$_2$O/glycerol mixtures exhibit a similar behavior, as can be seen in Fig. 2.4.

As model system for suspensions, H$_2$O with different volume concentrations $\Phi$ of 20 nm glass beads was used. A starting solution with 40% weight concentration of particles, supplied by ABCR, Karlsruhe, Germany, was used to prepare diluted solutions. The shear viscosity depends on the particle volume concentration $\Phi$ of the glass beads in H$_2$O and can be described with the Maron-Pierce model [8]

$$\eta_r = \left(1 - \frac{\Phi}{\Phi_{\text{max}}}ight)^{-2}, \quad (2.18)$$

where

$$\eta_r = \frac{\eta}{\eta_{\Phi = 0}} \quad (2.19)$$
Fig. 2.4: Shear viscosity $\eta$ at 20 °C of acetone/ak10 and $H_2O$/glycerol mixtures as a function of the weight concentration $\Psi$ of glycerol in $H_2O$ and ak10 in acetone, respectively. ak10 is a PDMS with shear viscosity of $\eta = 9.3$ mPa·s at 25 °C.

is the relative viscosity, and $\Phi_{max}$ the maximum volume concentration of glass beads in $H_2O$. The density and shear viscosity of the suspensions ranges from 1 - 1.3 g·cm$^{-3}$ and 1 - 15 mPa·s, respectively. The fit to the measured data, shown in Fig. 2.5, reveals a maximum particle concentration of $\Phi_{max} = 0.29$.

Because of the small size of the glass beads, the Brownian motion impedes sedimentation and the suspension appears homogenous even for small probing volumes. A summary of the test liquids and their properties is given in Tab. 2.2.

To illustrate the large range of shear viscosities, a number of familiar materials are listed in Tab. 2.3 with their shear viscosities.
2 Viscosity

Fig. 2.5: Shear viscosity $\eta$ of suspensions of $H_2O$ with 20 nm glass beads as a function of volume concentration $\Phi$ of glass beads. From the Maron-Pierce fit, a maximum volume concentration $\Phi_{\text{max}} = 0.29$ is calculated.

Tab. 2.2: Summary of test liquid properties at room temperature. Density $\rho$, shear viscosity $\eta$, and sound velocity $c$ are listed.

<table>
<thead>
<tr>
<th></th>
<th>$\rho_\ell$ [g cm$^{-3}$]</th>
<th>$\eta$ [mPa s]</th>
<th>$c_\ell$ [m s$^{-1}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H_2O$/glycerol</td>
<td>1 - 1.29</td>
<td>1 - 1500</td>
<td>1485 - 1915</td>
</tr>
<tr>
<td>PDMS</td>
<td>0.96 - 0.97</td>
<td>10 - 5500</td>
<td>$\approx$ 980</td>
</tr>
<tr>
<td>Acetone/ak10</td>
<td>0.79 - 0.93</td>
<td>0.3 - 11.3</td>
<td>980 - 1196</td>
</tr>
<tr>
<td>Suspensions</td>
<td>1 - 1.3</td>
<td>1 - 15</td>
<td>1485 - 1500</td>
</tr>
</tbody>
</table>
Tab. 2.3: Shear viscosity of some familiar materials at room temperature.

<table>
<thead>
<tr>
<th>Liquid</th>
<th>Viscosity [Pa·s]</th>
<th>Liquid</th>
<th>Viscosity [Pa·s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass</td>
<td>$10^{40}$</td>
<td>Glycerol</td>
<td>$10^9$</td>
</tr>
<tr>
<td>Asphalt</td>
<td>$10^8$</td>
<td>Water</td>
<td>$10^{-3}$</td>
</tr>
<tr>
<td>Honey</td>
<td>$10^1$</td>
<td>Air</td>
<td>$10^{-5}$</td>
</tr>
</tbody>
</table>
2 Viscosity
3 VIBRATING SENSOR THEORY

In this chapter the fundamentals of liquid property sensing with acoustic wave (AW) sensors, resonant membranes, and cantilevers are introduced. First, the physics of the unloaded devices is explained with simplified mechanical models. Then the theory is expanded to devices operated in Newtonian liquids. Different aspects to optimize the performance of the Lamb wave sensor and the membrane resonator are discussed. The advantage of the microsensors investigated in this work is that only the flat unstructured surface of the back of a membrane has to be exposed to the liquid and, in contrast to passive sensors like shear stress sensors [31], no additional equipment is required to generate a fluid flow.

Additionally, two alternative, non-silicon based, resonant “microsensors” were investigated to overcome some of the limitations of the Lamb wave sensor and the membrane resonator. However, all the resonant sensor systems have one drawback in common: they are restricted to a limited frequency range and operate at high frequencies compared to standard rheometers with typical frequency ranges < 200 Hz [7].

3.1 ACOUSTIC WAVE SENSORS

The variety of AW devices [36, 37] can principally be divided into three classes, namely bulk, surface, and plate AW devices. Each of these classes incorporates a number of subclasses, depending on propagation direction, polarization, and many other details influencing the AW propagation, such as gratings or guiding layers. A good survey on AWs in solids is given in [38]. In the following, some of the most common AW types are introduced and shown schematically with propagation direction (big arrow) and polarization or particle displacement at the device surface (small arrow).

A very familiar example for bulk AWs with longitudinal polarization are acoustic sound waves in solids. A bulk AW device with transversal polarization, schemati-
3 Vibrating Sensor Theory

Fig. 3.1: Schematic deformation of a bulk AW device, such as the AT-cut quartz TSMR used in quartz crystal microbalances (QCM).

...cally shown in Fig. 3.1, is the thickness shear mode resonator (TSMR). Usually, TSMRs are made of thin AT-cut quartz slides and feature wave propagation in the direction perpendicular to the surface and in-plane shear deformation. Quartz based TSMRs are widely used as frequency determining elements due to the outstanding mechanical material properties and the piezoelectricity of quartz [39].

In contrast to the bulk AWs, which are characterized by an oscillation of the entire device volume, surface AWs are confined to the surface of a solid, i.e., the oscillation amplitude decays exponentially within a few wavelength into the substrate. An example for a surface AW with wave polarization parallel to the surface is the surface transverse wave (STW). The corresponding surface AW with polarization perpendicular to the surface is the Rayleigh wave (SAW). A schematic of both types is shown in Fig. 3.2. Surface AWs can be experienced during earthquakes, where the fast travelling STW causes almost no harm and the slower Rayleigh wave causes most of the damage. Rayleigh wave devices are widely used for high frequency filter applications and can be found in TVs or mobile phones.

Acoustic plate modes (APM) occur if the wave guiding solid is less than a few acoustic wavelengths thick. Two examples, a shear horizontal acoustic plate mode (SH-APM) and a flexural plate mode (FPM), also called Lamb wave, are shown in Fig. 3.3.

Tab. 3.1 provides a compact survey of the different AW types, including their operating frequency range, application areas and geometric dimensions relative to the acoustic wavelength $\lambda$. 

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3.1 Acoustic Wave Sensors

Fig. 3.2: Schematic of a surface transverse wave (STW) or Love wave (LW) and a Rayleigh wave (SAW). STW devices use gratings and LW devices a thin film to guide and confine the AW along the surface. The oscillation decays within a few wavelengths into the substrate.

Fig. 3.3: Schematic deformation of a SH-APM and a FPW or Lamb wave. For SH-APM the plate thickness is about 3 - 10 wavelengths. For FPW the plate thickness is much smaller than the wavelength.

AW propagation in solids is affected by adjacent materials and, therefore, well suited to sense these material properties. Of course, the different AW types respond distinctively to a given load. To compare the sensing capabilities of the different AW types the relative sensitivity $S^r_g$ defined as

$$S^r_g = \frac{12g}{g\phi_x}$$

(3.1)
Tab. 3.1: Comparison of the most common AW types and the applicable media.

<table>
<thead>
<tr>
<th>Device</th>
<th>Wave type</th>
<th>Plate thickness</th>
<th>Frequency [MHz]</th>
<th>Media</th>
</tr>
</thead>
<tbody>
<tr>
<td>TSM</td>
<td>bulk</td>
<td>λ/2</td>
<td>5 - 10</td>
<td>gas, liquid</td>
</tr>
<tr>
<td>SAW</td>
<td>surface</td>
<td>&gt; λ</td>
<td>30 - 300</td>
<td>gas</td>
</tr>
<tr>
<td>STW</td>
<td>surface</td>
<td>&gt; λ</td>
<td>30 - 300</td>
<td>gas, liquid</td>
</tr>
<tr>
<td>FPW</td>
<td>plate</td>
<td>&lt; λ</td>
<td>5 - 20</td>
<td>gas, liquid</td>
</tr>
<tr>
<td>SH-APM</td>
<td>plate</td>
<td>3 - 10 λ</td>
<td>25 - 200</td>
<td>gas, liquid</td>
</tr>
</tbody>
</table>

is used. \( x \) is the quantity of interest, and \( g \) the measured parameter. As an example, \( S_{\Delta m}^f \) provides the relative change of the resonance frequency to a mass load \( \Delta m \) per unit area.

The mass sensitivity \( S_{\Delta m}^f \) of common AW types is compared in Tab. 3.2. Generally, bulk AWs are less sensitive to surface mass changes than surface AW modes because only a small part of the AW energy is close to the interfacing surface. The response of AWs to viscous liquids is additionally accompanied by AW attenuation due to viscous energy dissipation. In Tab. 3.3, the resulting sensitivity is the same for all AW types. This is because the viscous energy dissipation is calculated based on the same dissipation mechanism which is independent of the type of device and, therefore, depends only on the liquid properties.

A very convenient way to generate AW is based on the piezoelectric effect, where electric fields generate stress and strain fields in a piezoelectric substrate material and vice versa. The linear relation between mechanical and electric fields is described with the constitutive equations of piezoelectricity [40]

\[
S = dE + s^E T, \quad (3.2)
\]

\[
D = e^T E + dT \quad (3.3)
\]
3.1 Acoustic Wave Sensors

**Tab. 3.2:** Resonance frequency shift $\Delta f$ as a function of the added mass per unit area $\Delta m$ and the mass sensitivity $S_f^{\Delta m}$ for several AW sensors with substrate density $\rho$ and thickness $d$, respectively. The parameters $c_{TSM}$, $c_{SAW}$, and $c_{STW}$ depend on material properties of the AW devices.

<table>
<thead>
<tr>
<th>Mode</th>
<th>Response</th>
<th>Sensitivity $S_f^{\Delta m}$</th>
<th>Typical value for $S_f^{\Delta m}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>TSM</td>
<td>$\Delta f = -c_{TSM} f^2 \Delta m$</td>
<td>$-c_{TSM} f$</td>
<td>14 for 6 MHz quartz</td>
</tr>
<tr>
<td>SAW</td>
<td>$\Delta f = -c_{SAW} f^2 \Delta m$</td>
<td>$-c_{SAW} f$</td>
<td>200 for 158 MHz quartz</td>
</tr>
<tr>
<td>STW</td>
<td>$\Delta f = -c_{STW} f^2 \Delta m$</td>
<td>$-c_{STW} f$</td>
<td>180 for 250 MHz quartz</td>
</tr>
<tr>
<td>FPW</td>
<td>$\Delta f = -\frac{1}{2\rho d} f \Delta m$</td>
<td>$-\frac{1}{2\rho d}$</td>
<td>380 for 5.5 MHz composite plate</td>
</tr>
<tr>
<td>SH-APM</td>
<td>$\Delta f = -\frac{1}{2\rho d} f \Delta m$</td>
<td>$-\frac{1}{2\rho d}$</td>
<td>19 for 101 MHz quartz, mode $n = 1$</td>
</tr>
</tbody>
</table>

*The factors $c_{STW}$ also depend on the grating of the STW device.*

**Tab. 3.3:** Sensitivity of the AW attenuation factor $\alpha$ to changes in liquid density $\rho_l$ and viscosity $\eta$ calculated for Newtonian liquids.

<table>
<thead>
<tr>
<th>Mode</th>
<th>Sensitivity $S_{\alpha}^{\rho_l}$</th>
<th>Sensitivity $S_{\alpha}^{\eta}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>TSM</td>
<td>$\frac{1}{2\rho_l}$</td>
<td>$\frac{1}{2\eta}$</td>
</tr>
<tr>
<td>SH-APM</td>
<td>$\frac{1}{2\rho_l}$</td>
<td>$\frac{1}{2\eta}$</td>
</tr>
<tr>
<td>FPM</td>
<td>$\frac{1}{2\rho_l}$</td>
<td>$\frac{1}{2\eta}$</td>
</tr>
</tbody>
</table>
where $T$ and $S$ are the stress and strain tensors, $D$ and $E$ the dielectric displacement and electric field vectors and $d, s^E, e^T$ the piezoelectric strain, the elastic compliance, and dielectric tensor, respectively. Superscripts $E$ and $T$ indicate material parameters for constant electric field and constant stress, respectively. The material property tensors $d, s$ and $e$ determine the AW type for a given substrate and electric field distribution. A detailed discussion of piezoelectrically driven AW and SAW devices can be found in [38, 41]. Typical monocrystalline piezoelectric substrate materials are LiNbO$_3$, LiTaO$_3$, and quartz. For thin film applications, PZT and ZnO are commonly used. Other thin film materials, such as AlN and CdS, have lower piezoelectric coupling coefficients and, therefore, are less common. An approach to circumvent the drawbacks of these thin film materials by using monocrystalline quartz films has been presented recently [42].

### 3.1.1 Thickness Shear Mode Resonator

Due to their numberless application areas and their unsurpassed material properties, AT-cut quartz TSMRs have been extensively investigated after 1921, when the first quartz crystal controlled oscillator was described [39]. Only five years afterwards, the first broadcast transmitter used a quartz crystal control [43].

A TSMR typically consists of a thin AT-cut quartz disk with circular electrodes on both sides, as shown in Fig. 4.13. In the following, the theory for a TSMR having one surface in contact with a viscous liquid of infinite extension is discussed [44].

**TSM Resonator Theory**

Due to the piezoelectric properties and the crystalline orientation of the AT-cut quartz, an applied voltage between the electrodes results in a pure shear deformation [44]. For convenience, a coordinate system is chosen such that the shear deformation is along the $x$-direction in the $x$-$y$ plane, as shown in Fig. 3.4.

The resulting shear stress $\sigma_{xz}$ also lies along the $x$-direction and the stress-strain relation in the quartz slab is then given by [44]

$$
\sigma_{xz} = \frac{F_x(z, t)}{A} = \frac{\partial}{\partial z} u_x(z, t),
$$

(3.4)
3.1 Acoustic Wave Sensors

Fig. 3.4: Schematic of the deformation of an AT-cut quartz slab of thickness \(dz\) and surface area \(A\) due to a force \(F_x\) acting in the \(x\)-direction.

where \(\mu\), \(u_x(z,t)\), and \(F_x(z,t)\) are the piezoelectrically stiffened shear modulus, the displacement along \(x\), and the net force action on a slab of thickness \(dz\) and area \(A\), respectively. Combining Eqn. (3.4) with Newton's second law

\[
\frac{\partial}{\partial z} F_x(z,t)dz = \rho q A dz \frac{\partial^2}{\partial t^2} u_x(z,t),
\]

(3.5)

where \(\rho q\) is the quartz density, leads to the Helmholtz wave equation

\[
\frac{\partial^2}{\partial z^2} u_x(z,t) = \frac{\rho q}{\mu} \frac{\partial^2}{\partial t^2} u_x(z,t)
\]

(3.6)

with the general steady state solution

\[
u_x(z,t) = u_+ e^{i(\omega t - kz)} + u_- e^{i(\omega t + kz)}.
\]

(3.7)

\(u_+\) and \(u_-\) denote the amplitudes of shear waves traveling in positive and negative \(z\)-direction, respectively, and

\[
k = \omega \left(\frac{\rho q}{\mu}\right)^{1/2}
\]

(3.8)
3 Vibrating Sensor Theory

is the AW propagation constant. Applying the boundary condition for an unconstrained lower crystal surface at \( z = 0 \), i.e., \( F_y(0,t) = 0 \), implies that \( u_+ = u_- = u_0 \). Therefore, the strain in the x-direction becomes

\[
u_x(z, t) = 2u_0 \cos(kz)e^{i\omega t}.
\]

(3.9)

In the simplest case, the free resonator, the upper surface at \( z = h \) is also unconstrained, i.e., \( F_y(h,t) = 0 \). Applying this boundary condition to Eqn. (3.9) provides the resonance condition \( k \cdot h = n \cdot \pi \). Inserting the resonance condition into Eqn. (3.8) leads to the angular resonance frequencies

\[
\omega_n = \frac{n\pi(\frac{1}{h})^{1/2}}{\sqrt{\rho \mu}}.
\]

(3.10)

where \( n \) is the mode number. Actual disk shaped quartz crystal resonators with circular electrodes have finite dimensions and exhibit maximal shear vibrating amplitudes in the centre and diminishing amplitudes towards the edge [45].

RESPONSE OF A TSMR TO NEWTONIAN LIQUIDS

Since the discovery of the sensing capability of TSMRs [46], they are extensively used as mass monitoring devices and liquid property monitors [44, 47]. In the case of a liquid contacting the upper surface of the quartz, the unconstrained boundary condition is no longer valid. Instead, a shear stress is acting on the surface due to the liquid viscosity.

The differential equation for the shear velocity \( v_x(z,t) \) of an incompressible, Newtonian liquid with shear viscosity \( \eta \) and density \( \rho_l \) can be derived from the Navier-Stockes equation and is given by [44]

\[
\frac{\partial^2}{\partial z^2} v_x(z, t) = \frac{\rho_l}{\eta} \frac{\partial}{\partial t} v_x(z, t).
\]

(3.11)

For the liquid contacting the quartz surface at \( z = h \), a non-slip boundary condition is assumed, i.e., the liquid is firmly bound to the surface such that the velocity at the interface \( v_x(h,t) \) is the same for quartz and liquid. Furthermore, \( v_x(z,t) \) vanishes for \( z \to \infty \). Thus, the solution of Eqn. (3.11) becomes
3.1 Acoustic Wave Sensors

\[ v_x(z, t) = v_0 e^{-k_l(z - h)} e^{j\omega t}, \quad (3.12) \]

where \( v_0 \) is the maximum velocity at the quartz/liquid interface and

\[ k_l = (1 + i) \left( \frac{\omega \rho_l}{2 \eta} \right)^{1/2} \quad (3.13) \]

the complex wave number describing a damped shear wave radiated into the liquid by the oscillating surface. The effective thickness of the sheared liquid layer \( \delta_v \), i.e., the distance from the quartz surface, at which the shear wave amplitude is reduced to \( e^{-1} \) of its value at the interface, is

\[ \delta_v = \left( \frac{2\eta}{\omega \rho_l} \right)^{1/2}. \quad (3.14) \]

Furthermore, the shear stress on the liquid side of the interface must be equal with opposite sign to the shear stress on the quartz side. Combining Eqn. (2.3) and Eqn. (3.4) leads to the resonance condition of a TSMR in contact with a Newtonian liquid

\[ \tan \omega h \left( \frac{\rho_q}{\mu} \right)^{1/2} = -\left( \frac{\omega \rho_l \eta}{2 \rho_q \mu} \right)^{1/2}. \quad (3.15) \]

Assuming only small changes \( \Delta \omega \) of the free angular resonance frequency \( \omega_0 \), this equation can be solved. The resulting frequency shift \( \Delta f \) of the TSMR resonance frequency due to a viscous liquid can then be approximated by

\[ \Delta f = -f_0^{3/2} \left( \frac{\rho_l \eta}{\pi \rho_q \mu} \right)^{1/2}. \quad (3.16) \]

where \( f_0 = \omega_0 (2\pi)^{-1} \) is the free resonance frequency.
Fig. 3.5: Schematic displacement of an AT-cut quartz and an adjacent liquid. $\delta_v$ indicates the effective thickness of the sheared liquid boundary layer.

Fig. 3.5 schematically shows the displacement of a TSMR with an adjacent liquid layer. A typical value for the effective thickness of the sheared liquid boundary layer of $\text{H}_2\text{O}$ at $f_0 = 5 \text{ MHz}$ is $\delta_v = 250 \text{ nm}$.

This simple model has been extended to the first complete continuum electromechanical model for an AT-cut quartz resonator contacted by a single viscoelastic medium of arbitrary thickness [48]. The model was further extended to a TSMR loaded with an additional mass layer and related to the equivalent circuit representation of an AT-cut quartz in the vicinity of its resonance frequency [49].

**Equivalent Circuit of a TSM Resonator**

As frequency determining element in oscillator circuits, the TSMR has an equivalent electric circuit representation. A common electrical circuit representation of a loaded TSMR is the modified Butterworth-van-Dyke equivalent circuit shown in Fig. 3.6 [49].

The total admittance $Y$ of the equivalent circuit, without considering parasitic capacitances, is composed of a static and motional branch yielding
3.1 Acoustic Wave Sensors

Fig. 3.6: Electrical circuit representation of a liquid loaded TSMR based on a modified Butterworth-van-Dyke equivalent circuit. The motional branch represents the elements arising from the mechanical motion, the static branch includes the static electric circuit elements.

\[ Y = i\omega C_0 + \frac{1}{Z_m} \]  

where \( Z_m \) is the impedance of the motional branch given by

\[ Z_m = (R_1 + R_2) + i\omega (L_1 + L_2 + L_3) + \frac{1}{i\omega C_1}. \]

Each of the equivalent circuit elements has a distinct physical interpretation. \( C_0 \) in the static branch is the capacitance of the parallel plate capacitor formed between the two electrodes. The elements in the motional branch correspond to the mechanical properties of the materials. \( C_1 \) is related to the elasticity of the quartz, \( R_1 \) and \( R_2 \) represent the viscous losses, and the inductances \( L_1, L_2, \) and \( L_3 \) describe the mass inertia of the respective materials. Since the piezoelectric tensor couples the electric and mechanical domain each equivalent circuit element can be expressed as a function of the related material properties. The final results for a quartz TSMR with two identical circular electrodes of radius \( r \) are presented below [49].

The static electrode capacitance \( C_0 \) is
3 Vibrating Sensor Theory

\[ C_0 = \frac{\varepsilon_{22} \pi r^2}{h}, \quad (3.19) \]

where \( \varepsilon_{22} \) is the dielectric permittivity of the quartz. The capacitance \( C_1 \) represents the elasticity of the quartz and is given by

\[ C_1 = \frac{8 \kappa^2 C_0}{n \pi}, \quad (3.20) \]

where \( \kappa^2 \) is the electromechanical coupling coefficient for quartz and \( n \) the mode number. The inductance parameters \( L_1, L_2 \) and \( L_3 \) for quartz, liquid and adjacent mass layer are

\[ L_1 = \frac{1}{\omega_0^2 C_1}, \quad (3.21) \]

\[ L_2 = \frac{\omega_0 L_1}{n \pi} \left( \frac{2 \rho_i h}{\omega \mu \rho_q} \right)^{1/2}, \quad (3.22) \]

and

\[ L_3 = \frac{2 \omega_0 L_1 \rho_s}{n \pi (\mu \rho_q)^{1/2}}, \quad (3.23) \]

where \( \omega_0 \) and \( \omega \) are the angular series resonance frequencies of the unloaded and loaded quartz, and \( \rho_s \) the surface mass density of the mass layer, respectively. The resistance \( R_1 \) describes the viscous energy loss within the quartz given by

\[ R_1 = \frac{\eta_q}{\mu C_1} \left( \frac{\omega}{\omega_0} \right)^2, \quad (3.24) \]
where \( \eta_q \) represents the effective quartz viscosity. The viscous energy loss within the liquid is

\[
R_2 = \frac{\omega_0 L_1}{n \pi} \left( \frac{2 \omega_0 \eta_q}{\mu \rho_q} \right)^{1/2}.
\] (3.25)

From the equivalent circuit representation the series resonance frequency \( f_s \) and the quality factor \( Q \) of the loaded quartz can be calculated as

\[
f_s = \frac{1}{2 \pi ((L_1 + L_2 + L_3)C_1)^{1/2}}
\] (3.26)

and

\[
Q = \frac{L_1 + L_2 + L_3}{R_1 + R_2}.
\] (3.27)

Since the entrained liquid layer volume is much smaller than the oscillating quartz volume, \( L_2 \ll L_1 \) holds. Furthermore, the viscous loss in the fluid dominates the acoustic energy loss in the quartz such that \( R_1 \ll R_2 \). Neglecting additional mass layers, the \( Q \)-factor can be approximated with

\[
Q = \frac{\mu \rho_q}{2 \omega_0 \eta_q} \left( \frac{2 \omega_0 \eta_q}{\mu \rho_q} \right)^{1/2}.
\] (3.28)

This expression is independent of the electrode geometry. It also shows that the measurement of the series resonance frequency \( f_s \) and the quality factor \( Q \) for the described sensor geometry is not sufficient to independently determine the density \( \rho_l \) and viscosity \( \eta \). Only the product \( \rho_l \eta \) can be measured, as can be verified from Eqn. (3.16) and Eqn. (3.28). With a structured electrode, where liquid is trapped in small cavities and treated as component of a mass layer, the separation of \( \rho_l \) and \( \eta \) becomes possible, as presented in [50]. The AT-cut quartz material
properties commonly used are $\rho_f = 2651 \text{ kg·m}^{-3}$, $\mu = 2.947 \cdot 10^{10} \text{ N·m}^{-2}$, $\kappa^2 = 7.74 \cdot 10^{-3}$, $\varepsilon_{22} = 40.32 \cdot 10^{-12} \text{ F·m}^{-1}$, and $\eta_f = 3.5 \cdot 10^{-4} \text{ kg·m}^{-1}·\text{s}$.

### 3.1.2 LAMB WAVE SENSOR

The Lamb wave device investigated in this work consists of a thin membrane made of Si, SiO$_2$, Al and ZnO with two Al interdigital transducers (IDT) on top, as shown in Fig. 3.7. An AC voltage is applied to one IDT. Due to the piezoelectric effect, the resulting stress and strain fields excite a FPW within the membrane. The Lamb wave propagates to the other IDT where the inverse piezoelectric effect is used to convert the stress and strain fields of the AW in an electrical output signal.

![Schematic cross-section of a Lamb wave sensor.](image)

Fig. 3.7: Schematic cross-section of a Lamb wave sensor. Applying an AC voltage to the input IDT generates a Lamb wave due to the mechanical stress and strain induced by the piezoelectric effect. The wave propagates to the other IDT where the acoustic signal is converted to an electric output signal via the inverse piezoelectric effect.

### LAMB WAVE PROPAGATION THEORY

A simplified analytical model for AWs propagating in a uniform thin plate is presented subsequently. The one dimensional differential equation describing the surface normal displacement $u_z(x,t)$ for a thin plate can be derived by balancing
the forces of motion acting on a plate element [51, 52]. Assuming a thin plate with \( h \ll \lambda \) and small plate displacement \( u_z(x, t) \ll h \), where \( h \) is the plate thickness and \( \lambda \) the acoustic wavelength, one obtains

\[ D \frac{\partial^4}{\partial x^4} u_z(x, t) - T h \frac{\partial^2}{\partial x^2} u_z(x, t) + \rho_p h \frac{\partial^2}{\partial t^2} u_z(x, t) = 0, \]  

(3.29)

where \( D, T, \rho_p \) are the membrane bending moment, the in-plane stress, and the mass density of the plate, respectively. The bending moment \( D \) is

\[ D = \frac{E h^3}{12(1 - \nu^2)}, \]  

(3.30)

where \( E, \nu \) are the Young’s modulus and the Poisson’s ratio of the plate, respectively. Other contributions such as rotary inertia effects and shear corrections were found to be negligible for small deflections \( u_z(x, t) \ll h \) and thin plates \( h \ll \lambda \) [28, 29]. Eqn. (3.29) can be solved with a harmonic traveling FPW with normal displacement

\[ u_z(x, t) = u_0 e^{i(\omega t - kx)}, \]  

(3.31)

where \( u_0, \omega, \) and \( k = 2\pi \lambda^{-1}, \) are the AW amplitude, its the angular frequency, and the wave number, respectively. Substitution in Eqn. (3.29) yields

\[ c = k \left( \frac{D + Thk^{-2}}{\rho_p h} \right)^{1/2}, \]  

(3.32)

for the phase velocity \( c \) of a FPW. The corresponding group velocity \( c_{gr} \) is

\[ c_{gr} = \frac{\partial}{\partial k} \omega(k) = c + \frac{Dk^2}{c\rho_p h}. \]  

(3.33)
3 Vibrating Sensor Theory

and about twice the phase velocity $c$ for $T = 0$. A more detailed discussion of AW propagation in thin plates, including rotary inertia and shear correction terms is given in [29].

Due to the anisotropic material properties of single crystal silicon, the phase velocity of the Lamb wave depends on the propagation direction. With a software package dedicated to calculate the propagation of AWs in anisotropic, multilayered substrates [53, 54], the angle dependent Lamb wave velocity was calculated for composite, stress free membranes with 2.4 $\mu$m ZnO, 0.6 $\mu$m Al, 0.1 $\mu$m SiO$_2$ and a (100) silicon epitaxial layer of 4.6 $\mu$m and 8.6 $\mu$m thickness, respectively. The layer thicknesses were normalized with a wavelength $\lambda = 80$ $\mu$m, as given in Tab. 3.5. The phase velocity $c$ reaches a maximum in the $<110>$ direction and a minimum in the $<100>$ direction, as shown in the polar plot of Fig. 3.8.

Fig. 3.8: Orientation dependent phase velocity $c$ of Lamb waves in (100) Si membranes with 4.6 $\mu$m and 8.6 $\mu$m Si thickness within the (100) plane. The minimum and maximum propagation velocity are in the $<100>$ and $<110>$ direction, respectively.
3.1 Acoustic Wave Sensors

Assuming an AW length of 80 μm, the minimal and maximal phase velocities \( c_{\text{min}} \) and \( c_{\text{max}} \), with resulting center frequencies are listed in Tab. 3.4. Lowest center frequencies are obtained for a 4.6 μm thick membrane with a FPW orientation along the <100> direction.

**Tab. 3.4:** Minimal and maximal phase velocity \( c_{\text{min}} \) and \( c_{\text{max}} \), as well as minimal and maximal resonance frequency \( f_{0\text{min}} \) and \( f_{0\text{max}} \), of Lamb waves with acoustic wave length \( \lambda = 80 \mu \text{m} \) for (100) Si membranes.

<table>
<thead>
<tr>
<th>Thickness</th>
<th>( c_{\text{min}} ) [m·s(^{-1})]</th>
<th>( c_{\text{max}} ) [m·s(^{-1})]</th>
<th>( f_{0\text{min}} ) [MHz]</th>
<th>( f_{0\text{max}} ) [MHz]</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.6 μm Si</td>
<td>1081</td>
<td>1123</td>
<td>13.5</td>
<td>14.0</td>
</tr>
<tr>
<td>8.6 μm Si</td>
<td>1650</td>
<td>1711</td>
<td>20.6</td>
<td>21.4</td>
</tr>
</tbody>
</table>

**INVISID LIQUID**

Lamb waves operated in liquids change the phase velocity and the wave amplitude due to liquid density \( \rho_l \), sound velocity \( c_p \), and viscosity \( \eta \). The response of a Lamb wave to an inviscid liquid load is derived by considering the additional forces that impede the normal plate motion \( u_z(x,t) \) in Eqn. (3.31) as described in detail in [55]. The fluid motion \( v(x,z,t) \) can be calculated from the wave equation of the fluid velocity potential \( \psi(x,z,t) \)

\[
\Delta \psi = \frac{1}{c_p^2} \frac{\partial^2 \psi}{\partial t^2}
\]  

(3.34)

by taking the gradient

\[
v(x,z,t) = \nabla \psi.
\]

(3.35)

The boundary condition is given by the plate surface motion \( v_z \). The final solution for the phase velocity \( c \) of a Lamb wave in contact with an inviscid liquid is
3 Vibrating Sensor Theory

\[ c = k \left( \frac{D}{\rho_p h + \rho_i \delta_p} \right)^{1/2}, \]  
(3.36)

where \( \delta_p \) is the skin depth of the evanescent pressure wave in the fluid, given by

\[ \delta_p = \frac{\lambda}{2\pi} \left( 1 - \frac{c^2}{c_l^2} \right)^{-1/2}. \]  
(3.37)

The phase velocity \( c \) of the Lamb wave is therefore determined by the mass of the membrane and the mass of the evanescent-moving liquid layer. As long as \( c < c_l \), \( \delta_p \) is real and describes the skin depth of an evanescent pressure wave in the liquid. Since no energy is carried away by the evanescent wave, the Lamb wave propagates loss free in an inviscid liquid. For \( c > c_l \), wave propagation is no longer loss-free. The fluid velocity field \( v(x,z,t) \) of an inviscid liquid for one period of a FPW is plotted in Fig. 3.9.

\[ \lambda/2 \]
\[ \lambda \]
\[ \text{Fluid} \]
\[ \text{Plate} \]

Fig. 3.9: Velocity field \( v(x,z,t) \) of an inviscid fluid near the surface of a Lamb wave device.

**VISCOSOUS LIQUIDS**

For a viscous liquid the in-plane component of the Lamb wave couples viscously to the fluid, generating motion in a viscous boundary layer of thickness \( \delta_p \) [56],

40
3.1 Acoustic Wave Sensors

similarly to Eqn. (3.14). Consequently, the velocity profile parallel to the surface of the Lamb wave device is given by

\[ v_x(z, t) = v_0 e^{-z/\delta_v} e^{i(\omega t - z/\delta_v)}, \]

(3.38)

analogous to Eqn. (3.12). The motion of the entrained liquid layer can be split up in a part that moves in-phase and a part that moves out-of-phase with respect to the plate motion. The in-phase component of the entrained layer can be interpreted as an additional contribution to the mass loading of the plate, whereas the out-of-phase component gives rise to viscous dissipation. The effective mass per area that oscillates in phase with the plate wave can be calculated from the in-phase contribution of \( v_x(x, z, t) \) times \( \rho_l \) and integration over the liquid

\[ \rho_l \int_0^\infty \cos\left(\frac{z}{\delta_v}\right)e^{-z/\delta_v} \, dz = \frac{\delta_v \rho_l}{2} = \left(\frac{\rho_l \eta}{2\omega}\right)^{1/2}. \]

(3.39)

Inserting the effective mass in the plate wave velocity expression of Eqn. (3.36) yields

\[ c = \left(\frac{D}{\rho_l^2 k + \rho_l \delta_v + 0.5 \rho_l \delta_v^2}\right)^{1/2}. \]

(3.40)

The contribution of the viscous mass loading to the effective plate mass results in an alteration of the AW velocity for high viscous fluids.

In a viscous fluid the AW propagation is subject to an additional loss mechanism. An attenuation factor \( \alpha \) describing the loss due to the viscous fluid can be derived [56]

\[ \alpha = \left(\frac{\omega}{2}\right)^{3/2} \frac{\delta_v^2 k (\rho_l \eta)^{1/2}}{\rho_l^2 k^2 D + \omega^2 \rho_l \delta_v k^{-2}}, \]

(3.41)

where \( \alpha \) gives the loss in [Neper-m\(^{-1}\)] with 1 Neper = 8.686 dB.
3 Vibrating Sensor Theory

EXCITATION AND SIGNAL READ OUT

To excite and detect the Lamb wave propagating in the membrane, a two port delay line setup with piezoelectric ZnO as transducer material is used. Similar arrangements with PZT are presented in [57]. The recently proposed magnetic excitation [30] offers a new approach to circumvent the drawbacks of piezoelectric thin films, provides CMOS process compatibility, but requires an externally applied, strong magnetic field. Thermal actuation principles are not suited due to the low-pass characteristics of the excitation mechanism [58].

The calculation of the AW filter response [59, 60] is beyond the scope of this thesis. An approximate response of a Lamb wave sensor in a delay line setup, as shown in Fig. 3.7, can be calculated with the delta-function model [60, 61] or the impulse model [62]. Although these models are over-simplified they are very convenient for a straightforward approximate analysis. The basic assumptions are that the transducer electrodes are strongly localized and can be idealized by lines. Furthermore, AW reflections are neglected and the material is taken to be linear.

From the impulse response \( h(t) \) of the delay line, the frequency transmission characteristic is calculated using the Fourier transformation. The resulting theoretical frequency transfer characteristics \( H(f) \) for a Lamb wave delay line with periodicity \( \lambda = 80 \mu m \) and center frequency \( f_0 = 14 \) MHz featuring two identical, uniform transducers with 40 finger pairs is shown in Fig. 3.10. The center frequency \( f_0 = c \lambda^{-1} \) is determined by the periodicity \( \lambda \) of the fingers and the phase velocity \( c \) of the AW, which is itself a function of the membrane thickness. The width of the transmission peak or the pass bandwidth is determined by the number of finger pairs \( N \), the IDT design, and the group velocity \( c_{gr} \), which is about twice the phase velocity \( c \). For the example shown in Fig. 3.10 the minimal insertion loss is 6 dB because the emitting IDT radiates equally in both directions and the detecting IDT receives only half of the radiated energy. The minimal side lobe suppression is 26 dB.

CHOICE OF PIEZOELECTRIC MATERIAL.

Only a few of the numerous piezoelectric materials are suitable for thin film applications. While no efficient thin film deposition method exists for some piezoelectric materials, the piezoelectric coupling coefficient \( \kappa^2 \) as a function of the normalized film thickness \( h \lambda^{-1} \) becomes too small for other materials [63, 64].
3.1 Acoustic Wave Sensors

The piezoelectric coupling coefficient $\kappa^2$ is a measure of the efficiency of electrical to acoustic energy conversion. In physical terms, $\kappa^2$ is the ratio of the stored electrical energy to the stored elastic energy. The coupling coefficient $\kappa^2$ of a layered structure depends on the material properties and the normalized layer thicknesses $h \cdot \kappa^{-1}$. $\kappa^2$ can be calculated from the phase velocity change $\Delta c$ that occurs when an electrically perfectly conducting, massless plate is placed at the position of the IDTs and is approximately [65]

$$\kappa^2 = \frac{1}{2} \left| \frac{\Delta c}{c} \right|. \tag{3.42}$$

The coupling coefficient $\kappa^2$ was calculated with a dedicated software [53, 66] for ZnO, CdS, AlN and the layer geometries as given in Tab. 3.5. The material properties for CdS and AlN were obtained from [63], and the ones for sputtered ZnO from [67]. The software package assumes that the applied surface charge distribution varies sinusoidal with the periodicity of the AW. This is not the case for the surface charge distribution generated by real electrode configurations [65]. Therefore, the effective coupling based on a real electrode geometry somewhat
3 Vibrating Sensor Theory

Tab. 3.5: Layer geometries used for the calculation of the coupling coefficient $\kappa^2$. The thicknesses of the non piezoelectric layers Si, SiO$_2$, and Al are given by the IC process. The acoustic wavelength used to calculate the normalized thickness $h/\lambda_1$ is 80 μm.

<table>
<thead>
<tr>
<th>Material</th>
<th>Normalized thickness</th>
<th>Thickness [μm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZnO, CdS, AlN</td>
<td>0.00125 - 0.1</td>
<td>0.1 - 8</td>
</tr>
<tr>
<td>Al</td>
<td>0.0075</td>
<td>0.6</td>
</tr>
<tr>
<td>SiO$_2$</td>
<td>0.00125</td>
<td>0.1</td>
</tr>
<tr>
<td>Si</td>
<td>0.0575</td>
<td>4.6</td>
</tr>
</tbody>
</table>

differs from the calculated one. In addition, non-ideal material properties will influence the coupling coefficient $\kappa^2$. Fig. 3.11 shows the calculated coupling coefficient $\kappa^2$ as a function of the normalized piezoelectric layer thickness $h/\lambda_1$.

Fig. 3.11: Calculated piezoelectric coupling coefficient $\kappa^2$ for different piezoelectric materials as a function of the normalized piezoelectric layer thickness $h/\lambda_1$. The calculation was done for a configuration with ground electrode below and IDTs on top of the ZnO according to Tab. 3.5.
Among the investigated piezoelectric thin film materials CdS, AlN, and sputtered ZnO, the latter shows the highest piezoelectric coupling with $k_{\text{max}}^2 = 0.037$ at a normalized layer thickness of $h \cdot \lambda^{-1} = 0.03$. With an AW length of $\lambda = 80 \mu m$ this translates into an optimal ZnO film thickness of $h = 2.4 \mu m$. Calculated results for PZT are found in [64] and indicate a lower $k^2$ than ZnO for $h \cdot \lambda^{-1} < 0.15$.

Besides the coupling coefficient $k^2$, the electrical conductivity of the piezoelectric material has to be considered. The relatively low resistivity of ZnO, with repeated values ranging from $10^6 - 10^7 \Omega \cdot \text{cm}$ [68] up to $10^{11} \Omega \cdot \text{cm}$ [69], restricts its application to high frequencies. In contrast, the ceramic PZT films with resistivities of about $10^{11} - 10^{13} \Omega \cdot \text{cm}$ are suitable for low frequency operation [70]. The lower frequency limit $f_{\text{min}}$ of operation can be estimated from the characteristic discharge time $\tau$ of a capacitance $C$ over a resistance $R$. The resulting frequency limit $f_{\text{min}}$ is given by

$$f_{\text{min}} = \frac{1}{2 \pi \rho_{\text{ZnO}} \varepsilon_r \varepsilon_0},$$

where $\varepsilon_0$ and $\varepsilon_r$ are the electric permittivity of the vacuum and dielectric constant of ZnO, respectively. A resistivity of $10^6 - 10^7 \Omega \cdot \text{cm}$ results in a frequency limit $f_{\text{min}}$ of about 100 - 200 kHz. As a result, piezoelectric materials with higher electrical resistivity, such as PZT or AlN [71], are used for low frequency applications.

**Choice of Electrode Geometry**

For AW generation using thin piezoelectric films, the four basic electrode configurations shown in Fig. 3.12 can be considered for the fabrication of the IDTs.

Configuration A and B feature an additional metal ground plate on the bottom or top of the piezoelectric layer. These ground plates confine the potential drop to the ZnO layer leading to strong electric fields

$$E = -\nabla \phi$$

within the ZnO with a dominant electric field direction perpendicular to the plate. Hence, the cross field model situation, where the dominating electric field component is perpendicular to the AW propagation direction, is enforced [59]. Since
the sputtered ZnO grows with its piezoelectric (001) axis perpendicular to the surface [72], the two configurations with Al ground plate show better piezoelectric coupling coefficients compared to configuration C and D. The electrical potential for the four electrode configurations in air was simulated with the finite element method (FEM) software package ANSYS. The results of the simulated 2-D model are shown in Fig. 3.13. For the simulations, a split finger arrangement with a periodicity of one wavelength and corresponding periodic boundary conditions were used. The metal ground plate in configuration A prevents electric field lines from penetration into the sample fluids on the back of the membrane. Thus, the electric properties of the fluid do not influence the device performance. The calculated piezoelectric coupling coefficient $k^2$ for sputtered ZnO for the four electrode configurations versus the normalized layer thickness $h\cdot\lambda^{-1}$ of ZnO is shown in Fig. 3.14.

For the given layer sandwich, the optimal choice is electrode configuration A with a normalized ZnO layer thickness of 0.03, i.e., $2.4 \mu m$ ZnO for $\lambda = 80 \mu m$ with a maximal coupling coefficient $k^2_{max} = 0.037$.

**INTERDIGITAL ELECTRODE GEOMETRY**

A detailed description on how to design AW devices is beyond the scope of this thesis. The interested reader is referred to [60, 65, 73] where the theory is explained in great detail. A few important considerations necessary to improve the device performance are discussed in the following.

The AW propagation path is mainly restricted to the width of the electrode finger overlap. A differential interference contrast method [74] can be used to visualize
3.1 Acoustic Wave Sensors

Fig. 3.13: Simulated electric potential for the four electrode configurations, shown in Fig. 3.12, assuming a split finger arrangement. Only one repeating unit of the IDT is simulated and symmetric boundary conditions are applied. Structures with ground plate confine the electric field to the piezoelectric layer with an electric field orientation parallel to the piezoelectric c-axis of ZnO.

the AW. The IDT fingers in the AW propagation path lead to AW reflections due to two reasons:

- The IDT fingers represent a periodically changing surface mass load which leads to AW reflections due to changing acoustic impedance.
Fig. 3.14: Piezoelectric coupling coefficient $\kappa^2$ as a function of the normalized ZnO thickness $h\cdot \lambda^{-1}$ for the four different electrode configurations shown in Fig. 3.12.

- Electric impedance mismatch between IDTs and the connected circuitry causes electric energy reflection. The reflected electric energy restores part of the incoming AW and radiates it in both directions.

Reflections at the fingers lead to the so-called triple transit signals, as explained in Fig. 3.15. AW reflections within the IDTs are efficiently reduced with split finger electrode structures [75] which lead to destructive interference of the reflected waves. To reduce reflections at the membrane edge, the IDTs are slightly slanted by an angle $\alpha$ determined by

$$\tan \alpha = \frac{\lambda}{2w}, \quad i \geq 1. \quad (3.45)$$

For $i = 1$, the difference in propagation path distance for the waves at the edges of the propagation area is one wavelength, as shown in Fig. 3.15. Therefore, the wave parts in the right half and the left half interfere destructively. Higher order $\alpha$'s ($i > 1$) lead to even better reflection suppression.
It is important to use two identical IDTs to avoid small differences in the phase velocity within the two IDT areas due to different mass load distributions on the surface. This would cause a transmission mismatch between the two IDTs and reduce the device performance.

3.2 Membrane and Beam Resonator

The application of resonant sensors for viscosity sensing has been investigated for oscillating fibers [20] and resonant membranes [27]. In the following, a brief summary of the theory of resonant beams and membranes under transverse vibrations is given.

3.2.1 Piezoelectric Cantilever Beam

The piezoelectric cantilever beam used in this work is clamped along one edge and oscillates in a bending mode, as shown in Fig. 3.16.

The free transverse vibration of a homogeneous beam is described by the Euler-Bernoulli differential equation [76] for maximal vibrational amplitudes $u_{\text{max}}$ small compared to the beam thickness $h$ ($u_{\text{max}} \ll h$). Rotary inertia, shear
deformations, and nonlinearities due to large deflections are not included. The one dimensional Euler-Bernoulli differential equation describing the transverse deflection \( u(x,t) \) of a beam subject to an axial force \( N \) and a driving force per unit length \( F(x,t) \) is

\[
EI\frac{\partial^4 u(x,t)}{\partial x^4} - N\frac{\partial^2 u(x,t)}{\partial x^2} + \rho_b A \frac{\partial^2 u(x,t)}{\partial t^2} = F(x,t),
\]

where \( E, I, A \) and \( \rho_b \) denote apparent Young’s modulus, moment of inertia, cross sectional area, and specific mass density of the beam, respectively. The solutions of Eqn. (3.46) are discussed in detail in [76].

Immersing the beam in a liquid or gaseous media leads to two additional effects. The effective mass of the beam is increased due to an additional mass \( m_f \) that has to be accelerated and the media viscosity leads to oscillation clamping. The damping force is assumed to be proportional to the oscillation velocity with the damping coefficient \( \xi \) as proportionality factor.

For a free vibrating beam \((F=0)\) without axial load \((N=0)\), the modified Euler-Bernoulli differential equation is given by

\[
EI\frac{\partial^4 u(x,t)}{\partial x^4} - \xi \frac{\partial}{\partial t} u(x,t) + (\rho_b A + m_f) \frac{\partial^2 u(x,t)}{\partial t^2} = 0.
\]
Separation of variables $u(x,t) = \alpha(x)\beta(t)$ splits Eqn. (3.47) into two separate differential equations for the time and spatial coordinates. The resulting differential equation for the time coordinate is equivalent to that of a damped harmonic oscillator with quality factor [27]

$$Q = \frac{\omega_0 (\rho_p A + m_f)}{\xi^2}$$  (3.48)

and angular resonance frequency

$$\omega^2 = \omega_0^2 - \frac{1}{4} \left( \frac{\xi}{\rho_p A + m_f} \right)^2 = \omega_0^2 \left( 1 + \frac{1}{4Q^2} \right)^{-1}.$$  (3.49)

$\omega_0$ is the fundamental angular resonance frequency of the undamped ($\xi = 0$) oscillation and can be calculated from the differential equation for the spatial coordinates. Assuming $\rho_p A \gg m_f$, the undamped resonance frequency decreases linearly with the accelerated mass, i.e., with the fluid density [77]

$$\omega_0 \approx \omega_{0,\text{vac}} \left( 1 - \frac{m_f}{2\rho_p A} \right).$$  (3.50)

where $\omega_{0,\text{vac}}$ denotes the angular resonance frequency without fluid loading.

The dependence of the angular resonance frequency $\omega$ on the $Q$-factor (see Eqn. (3.49)) leads to an additional frequency change with increasing damping $\xi$, e.g., viscosity. Internal damping effects, not considered here, are discussed in [77]. A detailed analysis of the emitted radiation of a beam in air and water is given in [78].

### 3.2.2 Membrane Resonators

The piezoelectric membrane resonator [71] shown in Fig. 3.17 is based on the same excitation principle as the Lamb wave sensor but operates at a lower frequency.

The membrane resonator consists of the same layer sandwich as the Lamb wave device, namely 4.6 $\mu$m Si, 0.1 $\mu$m SiO$_2$, 0.6 $\mu$m Al, 2.4 $\mu$m ZnO, and 0.3 $\mu$m Al.
An electrode in the center of the membrane is used to excite transversal vibrations. At the membrane edge, the deformation causes a piezoelectric signal that is sensed with a second electrode arranged along the membrane edge. Fluids contacting the back of the membrane lead to a shift of the resonance frequency and damping of the oscillation.

**MEMBRANE DEFORMATION**

The equation of motion for a two-dimensional homogenous plate of thickness $h$ in the case of small deflections ($u_{\text{max}} \ll h$) is given by [76]

$$D \Delta^2 u(x, y, t) + \rho \frac{h}{2} \frac{\partial^2}{\partial t^2} u(x, y, t) = F(x, y, t), \quad (3.51)$$

where $\Delta$ is the two-dimensional Laplace operator and $D$ denotes the flexural rigidity. Exact solutions for this differential equation exist only for a few special boundary conditions, e.g., rectangular plates with simply supported edges. Solutions for more complicated boundary conditions can be obtained by using one of the various approximation methods, such as the Rayleigh-Ritz [79] principle or the variational method [80].

The resonance frequencies and vibration modes of the multilayered membrane with patterned electrodes on top were calculated by finite element modeling.
3.2 Membrane and Beam Resonator

Fig. 3.18: FEM simulation of the fundamental resonance mode of a 295 x 295 \( \mu \text{m}^2 \) membrane with 8.6 \( \mu \text{m} \) Si, 0.6 \( \mu \text{m} \) Al, 0.1 \( \mu \text{m} \) SiO\(_2\), 2.4 \( \mu \text{m} \) ZnO, and 0.3 \( \mu \text{m} \) thick Al electrodes. The resonance frequency for a stress-free membrane is 381 kHz. For symmetry reasons only 1/4 of the membrane is simulated.

(FEM) using ANSYS [81]. Fig. 3.18 shows the simulated fundamental mode shape for a 295 x 295 \( \mu \text{m}^2 \) membrane with 8.6 \( \mu \text{m} \) Si, 0.6 \( \mu \text{m} \) Al, 0.1 \( \mu \text{m} \) SiO\(_2\), 2.4 \( \mu \text{m} \) ZnO, and 0.3 \( \mu \text{m} \) thick Al electrodes. The resonance frequency for a stress-free membrane is 381.8 kHz.

The vibration amplitude of the fundamental mode shape can be described with the polynomial function

\[
u(x, y) = u_0 \left( \frac{2}{a} \right)^4 \left( \left( \frac{a}{2} \right)^2 - x^2 \right)^2 \left( \left( \frac{a}{2} \right)^2 - y^2 \right)^2, \quad -\frac{a}{2} \leq x, y \leq \frac{a}{2}, \tag{3.52}\]

where \( u_0 \) is the maximal amplitude in the membrane center.

**Electrode Geometry Design**

The deformation of the membrane causes mechanical stress within the thin film layers. The stress pattern within the ZnO layer is crucial for the excitation and detection of the membrane vibrations. To avoid cancellation effects, the mechanical stress within the ZnO underneath the detection electrode should have the same sign at a given time. The same holds for the ZnO underneath the excitation.
3 Vibrating Sensor Theory

electrode. From the membrane deformation \( u(x,y) \) the in plane stress components \( \sigma_{xx} \) and \( \sigma_{yy} \) of a homogenous membrane can be derived as [51]

\[
\sigma_{xx}(x,y) = \frac{12Dz}{h^3} \left( \frac{\partial^2}{\partial x^2} u(x,y) + v \frac{\partial^2}{\partial y^2} u(x,y) \right), \quad (3.53)
\]

\[
\sigma_{yy}(x,y) = \frac{12Dz}{h^3} \left( \frac{\partial^2}{\partial y^2} u(x,y) + v \frac{\partial^2}{\partial x^2} u(x,y) \right). \quad (3.54)
\]

\( z \) denotes the vertical distance from the neutral plane, i.e., the plane parallel to the membrane surface which does not experience any stress. The electric potential \( \Phi \) at the surface of a piezoelectric layer can be calculated from the stress distribution in the layer using the constitutive equations of piezoelectricity given in Eqn. (3.2) and Eqn. (3.3). For the given mechanical deformation \( u(x,y) \) shear stresses \( \sigma_{yx}, \sigma_{xy}, \sigma_{yy}, \) and the normal stress \( \sigma_{zz} \) in \( z \)-direction can be neglected because they are much smaller than the in-plane normal stresses \( \sigma_{xx} \) and \( \sigma_{yy} \). The relevant material property parameters are the dielectric constant \( \varepsilon_{33} \) and the piezoelectric strain constants \( d_{31} \) and \( d_{32} \). Furthermore, the material properties for sputtered piezoelectric ZnO are isotropic in the \( x-y \) plane such that \( d_{31} = d_{32} \). As a result, only the transverse piezoelectric effect has to be considered, i.e., only the electric field component \( E_z \) contributes to the electric potential \( \Phi \). In the case of shorted electrodes the dielectric displacement is \( D_z = 0 \) and the electric potential \( \Phi \) over the ZnO layer follows from Eqn. (3.3) as

\[
\Phi(x,y) = \int_{h_{ZnO}} E_z(x,y) dz = \frac{d_{31}}{\varepsilon_{33} h_{ZnO}} \int \left( \sigma_{xx}(x,y) + \sigma_{yy}(x,y) \right) dz, \quad (3.55)
\]

The normalized in plane stress

\[
\sigma_n(x,y) = \frac{\sigma_{xx}(x,y) + \sigma_{yy}(x,y)}{\sigma_{xx}(0,0) + \sigma_{yy}(0,0)} \quad (3.56)
\]
3.2 Membrane and Beam Resonator

Normalized stress $\sigma_{n}(x,y)$

Optimal electrode pattern

Fig. 3.19: Normalized in plane stress $\sigma_{n}(x,y)$ on top of the ZnO layer for a quarter of the membrane and the derived optimal electrode pattern. The electrode boundary corresponds to the lines where the stress is 10% of the maximal stress value.

at the surface of the membrane, which is proportional to the electric potential $\Phi$, is shown in Fig. 3.19. From this an optimized electrode geometry can be derived. The boundary of the electrodes are chosen to coincide with the lines of 10% of the maximal in-plane stress. The electrodes should not cross the inflexion lines of the fundamental mode. A schematic of the resulting electrode shape is shown in Fig. 3.19.

However, a simplified electrode geometry with square center electrode and a surrounding second electrode, as shown in Fig. 3.17, is sufficient for a good resonator performance.

**Acoustic Power Radiation**

Contrary to the other three sensor types, where acoustic radiation into the liquid can be neglected, the membrane resonator radiates acoustic power into the liquid. The liquid acts as an impedance to the vibrating membrane. The so-called acoustic impedance is given by [82]
3 Vibrating Sensor Theory

\[ Z_0 = \frac{p}{v}, \]  
\[ (3.57) \]

where \( p \) and \( v \) denote the sound pressure and the fluid velocity, respectively. From the linearized sound field equations for a fluid, damping-free medium, one can derive the differential equation for the propagation of sound [82]

\[ \Delta p(x,t) = c_f^2 \frac{\partial^2}{\partial t^2} p(x,t), \]
\[ (3.58) \]

in a medium with sound velocity \( c_f \). To calculate the sound field generated by a vibrating membrane, Eqn. (3.58) has to be solved with the appropriate boundary condition for the velocity component perpendicular to the vibrating surface. The radiation characteristics of a clamped square membrane in the Fraunhofer far-field approximation [82] is shown in Fig. 3.20 assuming a membrane deflection according to Eqn. (3.52). The frequency parameter \( k/a \) determines the directional characteristic of the sound to be emitted by a transducer with side length \( a \). \( k \) denotes the AW number \( k = 2\pi k^{-1} \). For a 295 x 295 \( \mu \)m membrane in air \( k/a \) is about 2, i.e., sound is emitted almost isotropic into the half space.

The total power \( P \) radiated by a harmonic oscillating surface with arbitrary surface velocity \( v(x,t) \) in a half space can be calculated from the sound pressure \( p(x,t) \) and the fluid velocity \( v(x,t) \) using

\[ P = \iiint_S p(x,t)v(x,t)\,dS, \]
\[ (3.59) \]

where \( S \) designates the surface of half a sphere with the sound emitter in the center. The power radiated from a harmonic oscillating, circular piston with area \( A \) and mean surface velocity \( v_0 \) is given by

\[ P = \frac{1}{2}Z_0A v_0^2 \left( 1 - \frac{2J_1(2ka)}{2ka} \right), \]
\[ (3.60) \]
The radiated acoustic power is proportional to the AW impedance $Z_0$, the oscillation amplitude, and square of the mean surface velocity $v_0$. The AW impedance in the investigated fluids is about a factor of $10^3$ larger than in air (see Tab. 3.6). Therefore, the acoustic power radiated into the liquids is distinctively larger than in air. This effect is partly compensated by the decreasing resonance frequency and vibration amplitude in the liquids. Nevertheless, the net power radiation increases with increasing AW impedance of the liquid. The power loss due to acoustic radiation decreases the $Q$-factor of the membrane.

Tab. 3.6: Comparison of density, sound velocity, and acoustic impedance for some of the test liquids used in this work at 20 °C.

<table>
<thead>
<tr>
<th>Medium</th>
<th>Density [g·cm$^{-3}$]</th>
<th>Sound velocity [m·s$^{-1}$]</th>
<th>Acoustic impedance $Z_0$ [N·s·m$^{-3}$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>0.00129</td>
<td>343</td>
<td>442</td>
</tr>
<tr>
<td>Acetone</td>
<td>0.794</td>
<td>1189</td>
<td>0.94·10$^6$</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>0.998</td>
<td>1483</td>
<td>1.48·10$^6$</td>
</tr>
<tr>
<td>Glycerol</td>
<td>1.228</td>
<td>1895</td>
<td>2.33·10$^6$</td>
</tr>
</tbody>
</table>
3 Vibrating Sensor Theory
4 SENSOR FABRICATION AND PACKAGING

In the first part of this chapter, the fabrication of the piezoelectric microsensors, namely the Lamb wave sensor and the membrane resonator, is explained in detail. The other two sensors, the AT-cut quartz TSMR and the bimorph piezoelectric cantilever, are based on commercially available transducer elements. Therefore, their fabrication is not further discussed in this work.

A good piezoelectric transducer material is a key point for AW devices. For this reason, the second part discusses the piezoelectric ZnO deposition and the ZnO material characterization. Stress, plane strain modulus, growth conditions, and ZnO layer composition were investigated to improve and optimize the piezoelectric layer quality.

In the third part of this chapter, the chosen packaging solutions for the different sensors types are discussed in detail. The packaging requirements for sensors exposed to liquids are very demanding. Only chemically and mechanically stable interfaces should be exposed to the liquid. For this reason, the packaging has to be considered from the very beginning of the sensor development.

4.1 PIEZOELECTRIC MICROSENSOR FABRICATION

4.1.1 PROCESS DESCRIPTION

The fabrication of the Lamb wave sensors and membrane resonators was done at the DIMES Technology Center, Delft, and is based on the DIMES-01 process [83], a 2 μm bipolar IC-process on 4” wafers. The complete process is composed of 15 mask steps including a ZnO and bulk micromachining (BMM) module. Two separate process modules, the ZnO and the BMM module, are used to deposit the piezoelectric transducer material and release the membranes [84]. The BMM module is set up for an array of 6 x 6 chips with a chip size of 1 x 1 cm², as shown in Fig. 4.4. The edge of the wafer is spared for the sealing rings of the wafer holder used in the electrochemical etch (ECE) step.
Starting material are 4", double-side polished, p-type, <100> silicon wafers. In a first step an either 4 μm or 8 μm thick n-doped epitaxial layer is grown. The thickness of the epitaxial layer determines the membrane thickness, i. e., the ECE uses the p-n junction between p-doped substrate and n-epi as etch stop. The wafers undergo the standard IC-process steps if required by the design. Since no circuitry was integrated on the design presented in this work only a few of the standard DIMES-01 process steps were required. The main process steps are briefly reviewed in the following. A complete process description can be found in [83]. To avoid short circuits between the n-epitaxial layer and the substrate at the wafer edge during the ECE step, a deep p+-diffusion (DP) is created around the 6 x 6 chip matrix and, therefore, isolates the n-epi. Ohmic contacts to the n-epitaxial layer, necessary to apply the etching potential, are made with a deep n+-diffusion (DN) in the saw lines. Then, a 1200 - 1300 Å thick oxide layer is grown in a dry oxidation step. A 0.3 μm LPCVD low stress nitride and a 0.2 μm LPCVD low stress poly are deposited on the back side of the wafer as KOH etch mask. The poly serves as a scratch protection for the nitride mask. The next process steps are the etching of the contact openings (CO) and the deposition of the interconnect metal layer (ICM). Subsequently, the ICM layer is patterned and ICM alignment marks are formed. The ICM alignment marks are required for the alignment of the KOH etch mask on the back side. The wafer edge is cleared from ICM to prevent short circuits to the p-substrate during the ECE step. Afterwards, the etch mask on the back side of the wafer is patterned. The wafers are now ready for the ZnO module. A cross section of the wafer before the ZnO module is shown in Fig. 4.1.

The standard procedure for ZnO deposition is described in [29]. To improve the quality of the Lamb wave devices, a new process sequence for the ZnO deposition was developed in this work and is described in detail in Section 4.1.2. Following the ZnO module, the wafers were electrochemically etched in a 6 molar KOH etch solution at 85 - 90 °C for about 5.5 - 6 hours. The ECE setup used at DIMES is a two electrode setup [85] with a platinum electrode in the KOH etch solution and an anodic contact at the n-epitaxial layer which serves as etch-stop layer for the ECE. The etching stops automatically at the reverse biased p-n junction when a bias voltage of more than 0.6 V is applied. Due to the well established ECE setup at DIMES, a process yield of 100 % was achieved for the ECE step, even for large 8 x 3 mm² membranes with a thickness of 4 μm. The nitride mask underetching was about 40 - 50 μm. Finally, the wafers were diced.
4.1 Piezoelectric Microsensor Fabrication

**Fig. 4.1:** Schematic cross section of a wafer before the ZnO module. A p⁺ diffusion layer (DP) along the wafer edge isolates the n-epitaxial layer (n-epi) used for the ECE stop. The ICM layer is removed at the wafer edge to prevent short contacts to the substrate (p-wafer). A thin polysilicon layer serves as scratch protection for the nitride KOH mask on the wafer back.

and cleaned chipwise. Special precaution had to be taken to avoid breaking of the large membranes due to the water pressure during dicing.

The key technology for high performance acoustic devices is the deposition of the piezoelectric ZnO film. ZnO is a widely used piezoelectric transducer material and unifies a number of advantages such as high coupling coefficient, low deposition temperature [86], excellent adhesion on various substrate materials, and the possibility of monolithic integration with electronic circuitry [87]. However, its drawbacks such as high chemical reactivity, high intrinsic stress, and poor reproducibility still hamper its widespread use in sensor fabrication. Moreover, Zn can affect the IC performance [29]. Structure, orientation, stress, and quality of the ZnO layers strongly depend on the substrate and deposition parameters [88].

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For this reason, the mechanical properties of ZnO as a function of its deposition parameters were studied in detail to determine optimal growth conditions for ZnO films with low tensile stress.

4.1.2 NEW PROCESS SEQUENCE FOR ZNO DEPOSITION

Deposition of ZnO based on the standard process sequence described in [29], i.e., on structured Al ground plates, resulted in ZnO films with large compressive stress. Deposition of ZnO on substrate materials other than Al, such as Si3N4, results in large compressive stress of the ZnO film, as shown in Section 4.2.3. Due to large compressive stress and the limited adhesion of the ZnO films on the ICM layer the ZnO layers peel-off and crack. Moreover, DC sputter deposition of ZnO on floating Al structures can lead to punch through and electric discharge due to charging effects during the sputtering process.

Therefore, a new process sequence for the Lamb wave fabrication was developed to ensure optimal growth conditions for the ZnO films on an all Al layer, as shown in Fig. 4.2. First, the Al layer for the ground plate was deposited, then the ZnO, and finally the Al for the IDTs, as seen in steps 1.-3. in Fig. 4.2. Afterwards, the layers were patterned in reverse order from top to bottom as shown in steps 4.-6. of Fig. 4.2. In a final step, the membranes were released using anisotropic KOH etching. The resulting ZnO films were almost stress-free or had slightly tensile stress, according to the results in Section 4.2.3. Even 3 x 8 mm² mem-

![Diagram of process sequence](image)

Fig. 4.2: Schematic of the new ZnO process sequence. After deposition of the required layers (1.-3.), they are patterned in reverse order (4.-6.).
branes of 8 μm thickness exhibited almost no buckling. Moreover, no further punch through due to electric discharge were observed.

ZnO is a reactive material and attacked by all common acids and bases including standard Al etch solutions. To etch the Al in the presence of ZnO, a special etch solution made of KOH, K₃Fe(CN)₆ and H₂O (1 g : 10 g : 100 ml) was used [29]. ZnO is inert to this solution and Al is etched at a rate of about 0.3 μm-min⁻¹ at 20 °C. ZnO itself is etched with a mixture of H₃PO₄, CH₃COOH, and H₂O (10 ml : 10 ml : 100 ml) which yields typical etch rates of 0.5 μm-min⁻¹ [29]. Fig. 4.3 shows photographs of a finished membrane resonator and a Lamb wave sensor.

Fig. 4.4 shows a wafer after the final Al etch. Before the anisotropic ECE and dicing, an additional photoresist coating is spun on top to provide additional protection. After dicing, the chips were subsequently cleaned with acetone, toluene, dimethylsulfoxide (DMSO), and a short DI water rinse [29].

A cross section of a finished Lamb wave device is shown in Fig. 4.5. The individual layers can be clearly distinguished.

4.2 Optimization of ZnO Deposition

4.2.1 ZnO Deposition

There are numerous methods to deposit high quality piezoelectric ZnO layers [64]. The fastest and most common are the various sputter techniques. The piezoelectric ZnO layers presented here were sputtered in a Varian 3119 R&D DC sputter-coater, as shown in Fig. 4.6.

Fig. 4.3: Close-up photograph of a membrane resonator (left) and a part of a Lamb wave sensor (right).
6 x 6 chip matrix  
Alignment marks

Lamb wave sensors  
Anode contact

Fig. 4.4:  *Picture of a wafer after finishing the ZnO module.*

Fig. 4.5:  *SEM picture of a cross section of a Lamb wave device.*

The pre-processed substrate wafer is placed in a single wafer substrate holder 11 cm above the Zn target. A dummy wafer is placed between the substrate holder and the substrate wafer to provide further scratch protection against the thermal heater block mounted on top of that stack. The films were sputtered from the Zn target in a pure O₂ atmosphere. The adjustable sputter parameters, namely the O₂ gas pressure, the substrate temperature \( T \), and the sputter current \( I \), were manually controlled. A set of typical deposition parameters is listed in Tab. 4.1.
4.2 Optimization of ZnO Deposition

Fig. 4.6: Photograph of Varian 3119 R&D DC sputter-coater.

| Tab. 4.1: Typical set of sputter parameters for the deposition of piezoelectric ZnO films. |
|-------------------------------------------------|---------------------------------|
| Target                                         | Zn (99.999%)                    |
| Sputter gas                                    | O$_2$ (100%)                    |
| Oxygen pressure                                | 6 mTorr                         |
| Substrate-target spacing                       | 11 cm                           |
| Substrate temperature                          | 400 °C                          |
| DC sputter power                               | 1610 W (520 V, 3.1 A)           |
| Sputter rate                                   | 75 nm-min$^{-1}$                |

The resulting ZnO layers are polycrystalline with hexagonal 6mm structure. The piezoelectric c-axis grows preferentially perpendicular to the substrate surface.
Fig. 4.7 shows SEM pictures of 2.4 μm thick ZnO layers deposited with optimized process parameters. Close to the substrate surface, the polycrystalline structure is not yet fully developed. With increasing thickness, the column structure becomes more pronounced and the preferential orientation of the ZnO columns with their c-axis perpendicular to the substrate can be clearly seen.

Fig. 4.7: SEM photographs of 2.4 μm thick ZnO layers. Close to the substrate the polycrystalline structure is not yet fully developed. With increasing thickness the column structure and their orientation perpendicular to the wafer surface can be seen clearly.

4.2.2 ZnO Stress

The residual stress of the ZnO films on the Al-SiO2-Si substrate sandwich strongly depends on the sputter parameters. For the application in Lamb wave devices, the deposition of ZnO films with low tensile stress is desirable in order to avoid excessive buckling of the large membranes. To investigate the influence of the sputter parameters on the film stress, a series of nine Al-SiO2-Si wafers was sputter coated with 1.4 μm thick ZnO layers at varying sputter current $I$ and substrate temperature $T$. The deposition of a stressed, thin film on a wafer leads to a measurable change of the wafer curvature. From the change of the wafer curvature before and after thin film deposition, the residual stress $\sigma_f$ of the added layer can be calculated using Stoney's formula [89]

$$\sigma_f = \frac{E_f t_s^2}{6(1-v_s)t_f} (K_f - K_s),$$  \hspace{1cm} (4.1)
where $E_s$, $t_s$, $v_s$ are the Young's modulus, the thickness, and the Poisson's ratio of the substrate, respectively. $t_f$ is the thickness of the film, $K_s$ and $K_f$ denote the curvature before and after deposition of the layer, respectively. The stress measurements were performed with a Tencor FLX 230 which allows contactless wafer profile measurements using a laser. The thin film stress is automatically calculated according to Stoney's formula Eqn. (4.1). The resulting film stresses are displayed in Fig. 4.8 as a function of the substrate temperature and the sputter current. The residual stress changes from compressive to tensile with increasing sputter current and increasing substrate temperature. For a fixed substrate temperature of $T = 375$ °C, $\sigma_f$ becomes tensile for $I \geq 3.0$ A. Likewise, $\sigma_f$ becomes tensile for $T \geq 410$ °C and a fixed sputter current of $I = 2.8$ A. Combining these results indicates that optimal ZnO deposition conditions can be achieved with $I \geq 3$ A and $T \geq 400$ °C. These results are in good agreement with earlier results [29].

Two mechanisms are responsible for temperature induced stress changes. At low substrate temperature, the adsorbed atoms have too little energy to arrange themselves in their lowest energy state and intrinsic stress can build up. An additional thermal stress component arises from the mismatch of the thermal expansion coefficients of ZnO ($\alpha_{ZnO} = 4\cdot10^{-6}$ K$^{-1}$) and silicon ($\alpha_{Si} = 2.33\cdot10^{-6}$ K$^{-1}$) [72] due to the difference between deposition temperature and measurement temperature. This effect partly explains the increasing tensile stress for increasing substrate temperatures.

Fig. 4.8: Residual stress $\sigma_f$ of ZnO films as a function of (left) the substrate temperatures $T$ at a constant sputter current $I = 2.8$ A and (right) the sputter currents $I$ at a constant substrate temperature $T = 375$ °C. All samples were deposited under otherwise identical conditions.
4.2.3 ZnO Plane Strain Modulus

In addition to residual stress measurements, the Young's modulus of the ZnO layers was measured with the load-deflection method [90, 91, 92]. From the deflection $u_{zf}(x,p)$ of a rectangular, multilayered plate with layer thicknesses $t_i$, width $a$, and length $b$ under an applied differential pressure $p$, the residual stresses $\sigma_i$ and the plane strain moduli $S_i$

$$S_i = \frac{E_i}{1 - \nu_i^2},$$

(4.2)

can be determined, respectively [93]. The plate is clamped to a rigid support and the residual stresses $\sigma_i$, the Young's moduli $E_i$, and Poisson's ratios $\nu_i$ are assumed to be homogenous and isotropic. A schematic of the setup is shown in Fig. 4.9.

![Schematic deformation of a membrane under an applied differential pressure $p$. The wafer is glued to the wafer chuck with pressure supply with a double-sided adhesive tape. The membrane deformation $u_z(x,p)$ is scanned in x-direction with an optical profilometer.](image)
4.2 Optimization of ZnO Deposition

For aspect ratios \( \beta \alpha > 4 \), the middle section of the structure responds to the pressure difference \( p \) by a plane strain deformation with negligible dependence on the \( y \)-coordinate and, therefore, behaves like a section of an infinitely long plate [94, 95]. A detailed description of the load deflection theory for rectangular membranes and sample preparation is given in [93].

Although the sputter parameters were optimized for growth of high quality piezoelectric ZnO on Al-SiO\(_2\)-Si substrates, SiN\(_x\)-Si wafers had to be used as substrates for the load-deflection experiments because the thin membrane approximation was not valid for the thicker Al-SiO\(_2\)-Si membranes. The SiN\(_x\) films were deposited in a PECVD 310 PC system from STS. A film thickness of 360 ±4 nm was measured with an MCS 400 spectrometer from Zeiss. Using the sputter conditions listed in Tab. 4.1, a 0.7 ± 0.04 μm thick ZnO film was deposited. The ZnO films on SiN\(_x\)-Si substrates are highly compressive with residual stress values up to -980 MPa. The large compressive stress resulted in buckling and cracking of the membranes during the anisotropic KOH etch. To avoid breaking of the ZnO-SiN\(_x\) membranes, a test wafer was annealed at 400 °C for 16 hours before the anisotropic etching. As a result a stress reduction of around 900 MPa in the ZnO layer was achieved. Subsequently, a protective polymer coating was spun on the wafer front to provide additional stability and front side protection during the anisotropic etching step [96]. The wafer was etched at 90 °C in a 6 molar KOH etching solution with the SiN\(_x\) serving as an etch stop layer. Finally, the polymer layer was removed with hot acetone. The resulting ZnO covered SiN\(_x\) membranes had widths ranging from 562 - 969 μm.

The computer controlled load-deflection test setup consists of an optical profilometer from UBM with Microfocus\textsuperscript{®} measurement head with a spatial resolution of 1 μm in horizontal and ±10 nm in vertical direction and a pressure controller DPI 520 from Druck Ltd. The pressure control accuracy is ±0.4 mbar. The samples were glued with double-sided adhesive tape on a wafer chuck with pressure supply. The wafer chuck was placed on the \( x-y \) table of the optical profilometer and aligned to be scanned in \( x \)-direction, as shown in Fig. 4.9. Then, the membranes were loaded with a differential pressure ranging from -350 to +350 mbar in steps of 10 mbar. For each stabilized pressure level, a deflection profile in \( x \)-direction across the membrane was recorded. The center deflection \( u_0 \) was automatically determined and the full pressure-deflection curve \( p(u_0) \) was stored at the end of each experiment. Eight ZnO-SiN\(_x\) membranes were characterized. After measuring the pressure-deflection curves of the ZnO covered SiN\(_x\) mem-
branes, the ZnO was removed and the pressure-deflection curves of the remaining SiNx membranes were measured. Subsequently, the \( p(u_0) \) data of SiNx were fitted according to [93] which yields the residual stress and plane strain modulus of the SiNx film. Finally, the \( p(u_0) \) data of ZnO-SiNx sandwich was fitted using the results from the SiNx layer and the two layer theory [93]. Fig. 4.10 shows the measured pressure-deflection curves \( p(u_0) \) including the calculated fits for a sample membrane.

The average plane strain moduli of SiNx and ZnO are 145.5 ± 3.8 GPa and 114.6 ± 5.9 GPa, respectively. The analyzed membranes were distributed over the entire wafer, i.e., the plane strain moduli of SiNx and ZnO are almost homogeneous across the wafer with a deviation of less than 6%. The measured residual stress \( \sigma_{SiNx} \) of the SiNx layer is 111.1 ± 18.0 MPa, whereas the ZnO layer experienced a stress gradient ranging from 135 MPa in the center to -16 MPa at the edge of the wafer.

Load-deflection experiments were also carried out on thick ZnO-Al-SiO\(_2\)-Si membranes. The layer thicknesses of ZnO, Al, SiO\(_2\), and Si were 1.4 \( \mu \)m, 0.83 \( \mu \)m, 0.16 \( \mu \)m, and 4.7 \( \mu \)m, respectively. The thick membranes lead to an asymmetric pressure-deflection curve for equivalent negative and positive differential pressures. Therefore, the fit quality was poor resulting in large errors for
4.2 Optimization of ZnO Deposition

The plane strain modulus. This indicates that the theory cannot be applied to the 8 µm thick ZnO-Al-SiO₂-Si membranes. Similar results were found for homogenous, 4 µm thick SiNx membranes [97].

To investigate the plane strain modulus of ZnO films deposited on the Al substrate, nano indentation experiments [98] were performed on a ZnO-Al-SiO₂-Si wafer without membranes. Four indentations were made in equidistant steps of 1 cm from the center to the wafer edge. The calculated plane strain modulus $S_{ZnO}$ versus the indentation depth is shown in Fig. 4.11.

The large error bars at the surface are due to an insufficiently defined surface contact of the indenter. To exclude poorly defined surface contact and the influence of the underlying material, the plane strain modulus is evaluated in the range from 0.1 - 0.3 µm. The average plane strain modulus $S_{ZnO}$ of the polycrystalline ZnO in c-axis direction is 121.8 ± 2.7 GPa and corresponds well to the value for ZnO on SiNx. However, the effects of the polycrystalline and anisotropic structure of the ZnO are not considered [98, 99]. The corresponding values for crystalline ZnO in a and c-axis direction are 157 GPa and 186 GPa, respectively [100].

![Fig. 4.11](image)

**Fig. 4.11:** Nano indentation profile for a ZnO layer of 1.4 µm. The plane strain modulus $S_{ZnO}$ in c-axis is shown as a function of indentation depth. The measurements were performed at Nano Instruments, Tennessee, USA.
4 Sensor Fabrication and Packaging

4.2.4 ZnO Composition

To investigate the chemical composition of the sputtered ZnO films glow discharge optical electron spectroscopy (GDOES) analysis were carried out at the EMPA Thun, Switzerland. Fig. 4.12 shows the relative composition of a ZnO film on an all Al substrate wafer as a function of the distance from the surface. The film was deposited with the optimized sputter parameters $I = 3.1 \, \text{A}$ and $T = 390 \, \text{°C}$.

The ZnO film composition corresponds very well to the expected ratio of Zn:O = 1:1. At the surface of the ZnO film there is less Zn and more O compared to the composition within the film. This behavior is observed for various film thicknesses and samples and is due to the surface roughness and the sputter conditions at the beginning of the GDOES analysis.

The gradient in the O content of the film may be due to drifting sputter conditions, such as substrate temperature or sputter pressure. The sharp rise of the Al content at the ZnO-Al boundary indicates a well defined interface between ZnO and Al. The tail of Zn and O into the Al is due to the curved topology of the sputtered area (ø 4 mm). Material in the center of the sputtered area is removed ear-

![Fig. 4.12: Profile of the atomic composition of a ZnO covered wafer as a function of depth made with GDOES. The four main components are Zn, O, Al, and Si. The contribution of other elements are below 1% and can be neglected.](image-url)
lier than material near the edge of the sputter area. A similar effect can be observed at the Al-Si interface. However, the change of slope of the Al curve at 3.2 and 3.5 μm depth indicates that Al is also present in Si. Al probably diffuses into the Si during the ZnO deposition at 400 °C. Finally, the contribution of other elements are below 1% and can be neglected.

The presence of other materials in the piezoelectric ZnO layer most likely decreases the film quality. On one hand, the piezoelectric coupling coefficient is modified, on the other hand, the electric resistivity can change. Decreasing resistivity increases the lower limit of the operation frequency and can lead to sensor failure. Otherwise selective addition of impurities, such as Cu [101] or Li [102], have been reported to increase the ZnO resistivity.

4.3 SENSOR PACKAGING

Appropriate sensor packaging is one of the major challenges in microsensor development. The ambient conditions to which an on-line viscosity sensor is exposed can be extremely harsh. Abrasion, aggressive chemicals, as well as temperature and pressure cycles can damage the sensor or cause aging effects and subsequent degradation of the device performance. Therefore, a high degree of mechanical and chemical robustness is required. Appropriate packaging and design of the sensor must be chosen to meet the high quality standards of foodstuff industry. Nevertheless, the packaging has to be transparent for the liquid property to be measured. For this reason, the packaging has to be considered from the very beginning of the sensor development.

The influence of the environment on the system performance should be minimized. Hence, a solution is favored where neither the electronic circuitry nor the transducer are in contact with the liquid. Only a chemically and mechanically stable interface should be exposed to the liquid. In the case of the AW sensors investigated in this work, the AW are generated and detected on the front side of the chip, whereas the interface to the liquid is on the back side of the chip. Thus, the sensor packaging ensures the separation of circuitry and transducer from the sensing domain. If needed, an additional thin film protection layer can be easily deposited onto the silicon back surface. In the following the packaging of the TSMR, Lamb wave sensor, membrane resonator, and piezoelectric cantilever is presented in detail.
4 Sensor Fabrication and Packaging

4.3.1 **Thickness Shear Mode Resonator**

The TSMR used in this work is a 5 MHz overtone polished AT-cut quartz with 2.54 cm diameter obtained from Maxtek, Torrance, USA. The two Cr/Au electrodes have different radius \( r_u = 6.4 \text{ mm}, \ r_d = 3.2 \text{ mm} \). The electrical contacts for both electrodes are on the same side, as shown in Fig. 4.13.

The quartz was mounted in a Voltaite/Teflon housing and contacted with two spring-loaded contacts from the back side, as shown in Fig. 4.14. Only the front side of the quartz is in contact with the liquid. The back side electrode and the contacts are protected from direct exposure to the liquid. In order to reduce field fringing effects, the large front electrode exposed to the liquid is held at ground potential. Using a Teflon spacer, the quartz is pressed against an O-ring seal to achieve controlled clamping conditions. Since the oscillating quartz volume is restricted to the electrode area, the quartz can be clamped near the edge without affecting its performance. The quartz can be replaced fast and easy.

The liquid sample is applied through the aperture at the top. The system is placed in a temperature chamber and held at constant temperature during the measurement cycle.

4.3.2 **Lamb Wave Sensor and Membrane Resonator**

The Lamb wave sensors and membrane resonators are mounted in cavity-down ceramic pin grid arrays (CPGA) with laser-drilled quadratic holes of

![Diagram of 5 MHz AT-cut quartz TSMR featuring single-sided electric contacts. Both quartz electrodes are contacted on the back side. The active oscillating quartz volume is confined by the circular electrodes.](image)

**Fig. 4.13:** Photograph of the 5 MHz AT-cut quartz TSMR featuring single-sided electric contacts. Both quartz electrodes are contacted on the back side. The active oscillating quartz volume is confined by the circular electrodes.
12 x 12 mm$^2$. The back side of the chip is flush-mounted to the top surface of the CPGA. The gap between chip and CPGA is filled with the polyurethane based compound VU 4453/71 HE-T from Lackwerke Peters, Germany. The compound glues the chip to the CPGA and seals the liquid exposed back side of the sensor from the front side with the electric contacts and the delicate ZnO layer. A schematic cross-section of the mounted chip is shown in Fig. 4.15. The elastic polyurethane compound ensures stress-free mounting of the sensor chip. The CPGA is mounted on a zero-insertion-force connector to provide input and output con-

Fig. 4.14: Disassembled and assembled quartz package. The quartz is clamped between a Teflon spacer and an O-ring. Electric contacts are provided with two spring-loaded contacts from the back side. The sample is applied on the front side of the quartz.

Fig. 4.15: Schematic cross section of a packaged chip. The epoxy fills the gap between sensor chip and CPGA and provides a tight sealing.
4 Sensor Fabrication and Packaging

Measurements. During measurement cycles the whole setup is placed in a temperature controlled chamber. The liquid is applied through a cuvette with a lid.

Fig. 4.16 shows a photograph of a mounted chip with two identical Lamb wave devices. The cavity of the CPGA can be closed with a lid to completely seal the device from the ambient.

4.3.3 Piezoelectric Cantilever

The piezoelectric cantilevers investigated in this work are commercially available, bimorph PZT based cantilevers from Physical Instruments, Waldhorn, Germany. The devices consists of two PZT layers with metal electrodes on both sides and a protection coating, as shown in a schematic cross-section view in Fig. 4.17. During operation the center electrode is held at ground potential while the outer electrodes are used to apply an AC voltage to excite transverse cantilever vibrations and to detect the piezoelectric response signal. At the maximum static driving voltage of 60 V, the tip deflection is around 150 μm.

Front side Back side

Fig. 4.16: Photograph of a chip with two identical Lamb wave sensors mounted in a cavity-down CPGA. The liquid is applied on the front side of the CPGA and, therefore, contacts only the back side of the membranes.
4.3 Sensor Packaging

Fig. 4.17: Schematic cross-view of the piezoelectric cantilever sandwich. One piezoelectric PZT layer is used to drive the cantilever, while the other is used to monitor the beam deflection.

The cantilever is glued to a brass rod with a compound of Styca 2651 MM and Catalyst 9 from Emerson & Cuming. The electrical contacts to the cantilever are made through the inside of the rod. A polyurethane compound encapsulates the wires and prevents them from liquid contact. The beam is either 2.4 or 4.8 cm long, 1 cm wide, and about 1 mm thick. Its robustness, compactness, and simplicity make it very easy to handle. Fig. 4.18 shows a photograph of a piezoelectric cantilever mounted on a brass rod. The rod is screwed in a supporting structure. The cantilever is then immersed vertically into the fluid until the fluid meniscus reaches the tip of the brass rod.

Fig. 4.18: Photograph of a piezoelectric cantilever mounted on the tip of a brass rod. The electric contacts are protected with a polyurethane compound.
4 Sensor Fabrication and Packaging
5 SENSOR CHARACTERISTIC IN AIR

Before the sensors were tested in liquids, they were characterized in air to provide a reference for the liquid loaded state. The various measurement techniques used to test the sensors are established in the following chapter. The measurement results are compared to the theories presented in Chapter 3, the data analysis is explained, and reference values for liquid data evaluation are calculated.

5.1 THICKNESS SHEAR MODE RESONATOR

An HP 4195A network/spectrum analyzer with an HP 41951A impedance test kit was used for impedance measurements in the vicinity of the fundamental resonance frequency of the quartz based TSMR. The calibration reference plane for the impedance measurements was chosen as close as possible to the quartz, i.e., at the tips of the spring contacts. The circuit parameters of the equivalent electric circuit presented in Section 3.1.1 were automatically calculated by the HP 4195A from the measured impedance curve.

Fig. 5.1 shows a typical impedance curve for a 5 MHz AT-cut quartz TSMR in air at room temperature. \( f_s \) and \( f_p \) signify the series and parallel resonance frequency, respectively. \( f_s \) and \( f_p \) are defined as the frequencies where the phase \( \phi = 0 \) and the absolute impedance \( |Z| \) is minimal or maximal, respectively.

Series and parallel resonance frequency can be extracted from the expression for the total equivalent circuit impedance given in Eqn. (3.17) using the resonance condition \( \phi = 0 \), i.e., \( \text{Im} \, Z = 0 \). Assuming that the influence of the resistance \( R_1 \) is negligible, i.e., assuming large quality factor \( Q \), leads to the analytical expressions [103]

\[
 f_s = \frac{1}{2\pi} \sqrt{\frac{1}{L_1 C_1}},
\]

(5.1)
Fig. 5.1: Measured impedance of a 5 MHz AT-cut quartz TSMR in the vicinity of the series and parallel resonance frequencies \( f_s \) and \( f_p \), respectively. The frequency difference between \( f_s \) and \( f_p \) is determined by the static capacitance \( C_0 \) of the electrodes.

and

\[
f_p = \frac{1}{2\pi} \sqrt{\frac{1}{L_1 C_1} + \frac{1}{L_1 C_0}}. \tag{5.2}
\]

The series resonance frequency \( f_s \) is independent of the static capacitance \( C_0 \), while \( f_p \) depends on \( C_0 \) and consequently on all other parasitic capacitances too. Therefore, the parallel resonance frequency \( f_p \) also depends on a connected electric circuit, e.g., a driving oscillator. The mechanical \( Q \)-factor of the TSMR can be calculated from the equivalent circuit parameters with \[104]\n
\[
Q = \frac{1}{2\pi f_p R_1 C_1}. \tag{5.3}
\]

Tab. 5.1 compares the calculated theoretical equivalent circuit parameters, see Section 3.1.1, of a 5 MHz AT-cut quartz with two identical electrodes \((r_u = r_d = 3.2 \text{ mm})\) with measured values for two different types of TSMR: (A) a
quartz with a big upper \((r_u = 6.4 \text{ mm})\) and a small lower electrode \((r_d = 3.2 \text{ mm})\), as shown in Fig. 4.13, and (B) a quartz with two identical small electrodes \((r_u = r_d = 3.2 \text{ mm})\). The measured values for \(C_0\) are higher than the calculated ones because the electric contacts cause an additional contribution to the static capacitance. As expected, the TSFR with the larger top electrode has a larger capacitance \(C_0\). The measured resistance \(R_1\), representing the losses in the quartz, is higher than expected due to material imperfections and the influence of the clamping of the quartz. The higher \(Q\)-factor of the quartz with two identical small electrodes compared to the quartz with larger upper electrode is due to the better localization of the quartz oscillation to the center, the so-called energy-trapping. The smaller electrodes confine the electric field, and, therefore, the quartz deformation, more to the center such that the influence of the quartz clamping is reduced. Other quartz geometries, such as plano-convex geometries [45], can be used to enhance the energy-trapping effect and further reduce the influence of the quartz clamping. The mass load due to the Cr/Au electrodes explains the initial frequency difference from the expected 5 MHz. The measured equivalent circuit parameters \(C_1\) and \(L_1\) match the theoretical values for the quartz with identical electrodes very well. The values for the other quartz type deviate due to the different electric field distribution in the quartz caused by the larger upper electrode.

<table>
<thead>
<tr>
<th>Geometry</th>
<th>(C_0) [pF]</th>
<th>(C_1) [pF]</th>
<th>(R_1) [Ω]</th>
<th>(L_1) [mH]</th>
<th>(f_s) [MHz]</th>
<th>(Q)</th>
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<tbody>
<tr>
<td>Theory</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(r_u = 3.2 \text{ mm}) (r_d = r_u)</td>
<td>3.85</td>
<td>24.05</td>
<td>2.5</td>
<td>42.12</td>
<td>5</td>
<td>5.6 \times 10^5</td>
</tr>
<tr>
<td>A</td>
<td>(r_u = 6.4 \text{ mm}) (r_d = r_u/2)</td>
<td>6.83</td>
<td>28.64</td>
<td>6.19</td>
<td>35.52</td>
<td>4.9899</td>
</tr>
<tr>
<td>B</td>
<td>(r_u = 3.2 \text{ mm}) (r_d = r_u)</td>
<td>5.39</td>
<td>24.32</td>
<td>7.52</td>
<td>41.7</td>
<td>4.9981</td>
</tr>
</tbody>
</table>

Tab. 5.1: Comparison of theoretical equivalent circuit parameters for a 5 MHz AT-cut quartz with electrodes of radius \(r_u = r_d = 3.2 \text{ mm}\) and measured values for two different electrode geometries.
For continuous on-line liquid property sensing, the quartz is often used as frequency determining element in a feedback oscillator [105]. Two modes of operation can be chosen. The oscillator can be operated either at the parallel resonance frequency $f_p$ or the series resonance frequency $f_s$. In both cases, the closed loop amplifying condition with gain $> 1$ and phase shift $\phi = 0$ has to be fulfilled. $f_p$ offers the advantage of better frequency resolution, whereas $f_s$ allows oscillator operation at higher loads, i.e., allows to measure fluids with higher viscosity or sense thicker material layers [106].

5.2 LAMB WAVE SENSOR

The fabricated Lamb wave sensors were characterized by measuring the $S_{21}$ scattering parameter with an HP 4195A network/spectrum analyzer. $S_{21}$ in [dB] is defined as the ratio of the power $P_{out}$ delivered by the output of a network to the power $P_{in}$ delivered to the input of the network, if the network is terminated by a non-reflecting load and exited by a non-reflecting generator [107].

$$S_{21}(f) = 20 \log \left( \frac{P_{in}(f)}{P_{out}(f)} \right) \text{ [dB]}.$$  \hspace{1cm} (5.4)

To reduce the cross-talk signal due to electromagnetic feedthrough between emitting and receiving IDT, the input and output signals of the Lamb wave delay line are balanced using low loss transformers, as shown schematically in Fig. 5.2.

![Schematic representation of a balanced Lamb wave delay line using low loss transformers.](image)

Fig. 5.2: Schematic representation of a balanced Lamb wave delay line using low loss transformers.
5.2 Lamb Wave Sensor

Fig. 5.3 shows the measured and calculated $S_{21}$ transfer characteristic of a Lamb wave device with 40 finger pairs, wavelength $\lambda = 80 \mu m$, 8 $\mu m$ membrane thickness, and a center-to-center distance of $48 \lambda$ of the two IDTs. The measured Lamb wave has a resonance frequency of 13.6 MHz and less than 20 dB insertion loss (IL). Assuming perfectly matched input and output impedance, a IL of only 6 dB is expected from theory, because half of the acoustic energy is radiated in the opposite direction and lost. However, the impedances are not matched in the test setup. Most of the initial IL is, therefore, due to electric energy reflection at the unmatched input and output impedance of the IDTs.

The measured Lamb wave devices show excellent transfer characteristics with clearly distinctable side lobes. The side lobe suppression is almost 26 dB as expected from theory. The noise level is more than 60 dB below the main lobe. Time domain analysis shows that the main acoustic signal is more than 10 dB above the AW reflections and electric feedthrough signal (see Fig. 5.5).

The theoretical $S_{21}$ transfer characteristic (see Fig. 3.10) was shifted to match the experimentally measured minimal IL in air in order to allow comparison with the measured data. The difference between experiment and theory away from the

![Image](https://via.placeholder.com/150)

Fig. 5.3: Measured $S_{21}$ transfer characteristic of an unloaded Lamb wave device with 1.4 x 8 mm$^2$ membrane area, a total membrane thickness of 8 $\mu m$, 40 finger pairs, and 1 mm aperture. The theoretical transfer characteristic based on the impulse model is shown for comparison. The jagged peaks are mainly due to the triple transit signal.
Sensor Characteristic in Air

center frequency $f_0$ is caused by the simplifications made in the impulse model, which does not account for AE dispersion of the Lamb waves. The jagged peaks in the transfer signal of Fig. 5.3 are mainly due to AE reflections within the IDTs and at the membrane edge, e.g., the so-called triple transit signal.

Electromagnetic (EM) feedthrough and AE reflections distort the transfer characteristic, as can be seen in Fig. 5.4. However, these distortions can be filtered with a time gate in the time domain of the Lamb wave response, as shown in Fig. 5.5. To this end, the impulse response in the time domain is calculated with fast Fourier transformation (FFT) from the recorded transfer characteristic. In the time domain, the electric feedthrough signal, the acoustic response, and the triple transit signal are clearly separated. After filtering the electric feedthrough and AE reflections, the inverse FFT (IFFT) returns the time-gated transfer characteristic in the frequency domain without perturbations.

The temperature sensitivity of the center frequency $f_0$, shown in Fig. 5.6, is $-0.74 \text{ kHz/K}$ and $-1.5 \text{ kHz/K}$ with and without time gating, respectively. The jagged peaks close to the center frequency $f_0$ can slightly shift the maximum of the transmission characteristic. The limited frequency resolution of 15 kHz of the data in Fig. 5.6 is due to the frequency resolution of the network/spectrum analyzer for a 10 MHz frequency span.

The minimal IL decreases with increasing temperature at a rate of $0.03 \text{ dB/K}$. Again, the influence of the electromagnetic feedthrough and AE reflections leads to deviations from the otherwise linear insertion loss change, as can be seen in Fig. 5.7.

### 5.3 Piezoelectric Cantilever

The piezoelectric cantilever is excited by applying a burst signal from a HP 33120A arbitrary wave form generator to the upper electrode, with the center electrode held at ground potential, as shown in Fig. 4.17. The output signal, i.e., the potential of the lower electrode, is directly monitored with an oscilloscope LeCroy 9450. No further amplification is required. An excitation burst signal with an amplitude of 4 V$_p$ and a frequency close to the resonance frequency of the cantilever as well as the corresponding piezoelectric response signal are shown in Fig. 5.8. The resonance frequency $f_0$ and quality factor $Q$ are extracted.
Fig. 5.4: **Comparison between the measured and the time-gated transfer characteristic after filtering the EM feedthrough and AW reflections.** The jagged peaks in the transmission characteristics are strongly suppressed in the filtered signal.

Fig. 5.5: **Typical impulse response calculated with an FFT from the measured transmission characteristic.** In the time domain the different signal contributions can be separated and filtered with a time gate.
5 Sensor Characteristic in Air

Fig. 5.6: Temperature drift of the Lamb wave resonance frequency as obtained from the gated and non-gated transfer characteristics. The discrete steps of the frequency change are due to the frequency resolution of the HP 4195A for a 10 MHz frequency span.

Fig. 5.7: Temperature drift of the minimal IL for gated and non-gated $S_{21}$ transfer characteristics.
from the free oscillation of the cantilever after switching off the excitation burst. The exponential amplitude decay function of a damped harmonic oscillator

\[ A(t) = A_0 \sin(\omega_0 t + \phi)e^{-t/\tau} \]  \hspace{1cm} (5.5)

was used to determine \( f_0 = \omega_0/(2\pi) \) and \( Q = \pi f_0 \tau \). The results for the two investigated cantilevers from *Physical Instruments* are summarized in Tab. 5.2.

Fig. 5.8: *Response of a piezoelectric cantilever P-801.10 to an excitation burst signal with an amplitude of 4 Vp.* The exponential decay of the output signal was used to extract \( Q \) and \( f_0 \) of the cantilever. The sinusoidal pattern of the data points is due to the sampling rate of the data acquisition software.
5 Sensor Characteristic in Air

Tab. 5.2: Resonance frequency $f_0$ and quality factor $Q$ of the oscillating piezoelectric cantilevers in air.

<table>
<thead>
<tr>
<th>Type</th>
<th>Geometry [mm$^3$]</th>
<th>$f_0$ [Hz]</th>
<th>$Q$</th>
</tr>
</thead>
<tbody>
<tr>
<td>P-801.10</td>
<td>23 x 10.5 x 0.65</td>
<td>735</td>
<td>128</td>
</tr>
<tr>
<td>P-801.50</td>
<td>48 x 12.5 x 0.8</td>
<td>263</td>
<td>79</td>
</tr>
</tbody>
</table>

5.4 Membrane Resonator

The vibration amplitude in the center of the piezoelectric membrane resonators was measured optically with a laser heterodyne interferometer from Polytec, Germany, in combination with a HP 4194A gain/phase analyzer. The output of the laser vibrometer is proportional to the membrane velocity and can be converted to the vibration amplitude by integration. Accurate positioning of the membrane center with respect to the laser beam focus was done with microtranslation stages. Transverse membrane vibrations are excited by applying an AC voltage to the center electrode, shown in Fig. 3.17. The vibration amplitude of a 295 x 295 μm$^2$ membrane with 4.6 μm epi thickness as a function of the excitation frequency is shown in Fig. 5.9. The membrane has a fundamental resonance frequency of 196 kHz with a $Q$-factor of 71 in air. An applied AC voltage of 0.77 V results in a maximal vibration amplitude of 0.67 μm. An identical membrane with 8.6 μm epi thickness has a fundamental resonance frequency around 381 kHz and a $Q$-factor of 91.

Due to the piezoelectric actuation principle, a linear change of the vibration amplitude with the applied AC voltage is expected. Fig. 5.10 shows the vibration amplitude as a function of the applied voltage. The oscillation amplitude changes linearly with the applied AC voltage at a rate of 0.9 μm·V$^{-1}$. Compared to thermally actuated membranes [77, 108], where the vibration amplitude increases linearly with the applied power, the piezoelectric actuation is more efficient. An excitation power of 10 mW applied to a 8 μm thick, 295 x 295 μm$^2$ square membrane with electrode geometry shown in Fig. 3.17 leads to a vibration amplitude of 1.1 μm. In contrast, a thermally actuated, 4-5 μm thick, 925 x 925 μm$^2$ square membrane deflects about 0.35 μm at an applied dynamic heating power of 42 mW.
5.4 Membrane Resonator

Fig. 5.9: Vibration amplitude in the center of a 295 x 295 μm$^2$ membrane with 8 μm thickness around its fundamental resonance frequency. The membrane was excited at the center electrode using an AC voltage of 0.77 V.

Fig. 5.10: Vibration amplitude at the resonance frequency as a function of the applied AC voltage. The amplitude increases linearly with increasing driving voltage. An applied AC voltage of 1.4 V corresponds to a power of 10 mW and result in a vibration amplitude of about 1.1 μm.
The electrical resistivity \( \rho_{ZnO} \) of ZnO was measured to estimate the lower frequency operation limit \( f_{\text{min}} \) of the membrane resonators. Resistivity values up to \( 4 \times 10^6 \, \Omega \cdot \text{cm} \) result in a lower frequency operation limit around 50 kHz. The corresponding sensors could be operated without problems. However, most devices showed resistivity values in the order of \( 10^5 \, \Omega \cdot \text{cm} \) with a corresponding frequency limit of 2 MHz. The corresponding resonators could not be excited at their fundamental resonance frequency. To increase the yield of working devices either the ZnO resistance has to be improved or the resonance frequency has to be increased by reducing the membrane size or increasing the membrane thickness.

Fig. 5.11 shows the amplified (with gain 100) response of a 295 x 295 \( \mu \text{m}^2 \) membrane resonator of 8 \( \mu \text{m} \) thickness to an applied burst signal with an amplitude of 0.5 \( V_p \). The test equipment is the same as for the piezoelectric cantilever except for an additional differential amplifier Tektronix AM 502 used to amplify the output signal by a factor of 100. From the exponential decay of the piezoelectric output signal, a quality factor \( Q \) of 78 and a resonance frequency of 197.7 kHz were calculated for the free vibrating membrane.

![Amplified piezoelectric response of a membrane resonator excited with a burst signal of 0.5 Vp. From the exponential decay after switching off the burst, the Q-factor and the resonance frequency of the oscillating membrane can be extracted.](image-url)
6 Viscosity Sensing

As already mentioned in Section 1.3, macro- and microsensors are distinctively different regarding the operation frequency and probed sample volume. Taking these differences into account, the sample liquids investigated in this work are divided into three categories.

The first category are homogenous, Newtonian liquids where the same results are expected for micro- and macrosensors. Per definition the viscosity of a Newtonian liquid does not depend on frequency and the liquid can be treated as homogenous even for small sample volumes.

The second category are homogenous, non-Newtonian liquids. Similar to the liquids in the first category, the sample volume plays a minor role due to the homogenous characteristics of the liquids. Due to the non-Newtonian liquid behavior, however, the measured viscosity is strongly frequency dependent. Therefore, increasingly diverging results must be expected with increasing operation frequency.

A third, more complex category includes inhomogeneous liquids, whereby only suspensions and no dispersions are considered here. If the probed sample volume is of the order of the particle dimension or less, the liquid cannot be regarded as continuum anymore and the question rises what does the microsensor actually measure? Does it measure the carrier matrix only or does it also sense the presence of the particles and to what extend? The complex rheological behavior of suspensions involves also demixing processes which makes an exact control of the experimental conditions more demanding. Natural demixing processes, such as sedimentation, can be reduced with an appropriate choice of the test liquid, whereas measurement induced demixing processes, e. g., due to a shear rate gradient [8], depend on the sensor. Demixing can lead to a change of the particle concentration in the measured sample volume and, therefore, to deceptive results.

The main issue that arises when evaluating and interpreting the measurement results is the large gap between the operation frequency of the microsensors and the conventional rheometers, used to characterize the sample liquids in order to obtain reliable reference data. Conventional rheometers must be used to charac-
terize the liquids because there is no method in the high frequency domain that can be used as reference and calibration method to verify the microsensor results. Therefore, the results of the microsensors operating in the high frequency domain have to be compared with the results obtained through conventional methods. One way to circumvent this discrepancy is to use the same or similar sensors operating at different frequencies to cover an entire frequency range. This would allow to subsequently fill the frequency gap, similar to conventional rheometers operating in an entire low frequency range.

In this section, the liquid property sensing capabilities of the four viscosity sensors were tested with the various sample liquids mentioned in Section 2.3. In a first stage Newtonian liquids, such as the $\text{H}_2\text{O}/\text{glycerol}$ mixtures, were tested and compared with theory or used as reference for further experiments. In a second stage other homogenous samples, with Newtonian viscosity characteristics in the low frequency range, were measured to investigate the impact of the high operation frequencies. Finally, first tests with inhomogeneous liquids were carried out.

### 6.1 Thickness Shear Mode Resonator

The liquid property sensing was carried out with a 5 MHz AT-cut TSMR with large upper and small lower electrode, as described in Section 4.3.1. The large front electrode, held at ground potential reduces field fringing effects and, therefore, reduces the influence of the dielectric constant of the liquid on the sensor characteristics. The quartz was loaded with liquid samples of 2 ml through the aperture on top of the holder. The temperature was held at 25 °C during the measurement cycle. The impedance of the TSMR was measured with a HP4195A network/spectrum analyzer as described in Section 5.1. As an example, the impedance curves for various Newtonian $\text{H}_2\text{O}/\text{glycerol}$ mixtures are shown in Fig. 6.1 and Fig. 6.2. The corresponding steady shear viscosity $\eta$ as a function of the glycerol volume concentration $\Phi$ is given in Fig. 2.4.

The quartz is cleaned by rinsing it with an appropriate solvent and finally with deionized water. Due to the very thin probed liquid layer it is important to remove any surface films. Remaining liquid films can shield the following sample from being sensed by the quartz. Mechanical contact with the quartz should be avoided to prevent scratches on the surface. Liquid trapped in scratches causes excess frequency shifts [50].

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6.1 Thickness Shear Mode Resonator

Fig. 6.1: Absolute value $|Z|$ of the measured quartz impedance. The quartz is loaded with different $H_2O$/glycerol solutions. Increasing liquid shear viscosity $\eta$ causes a decrease of the series and parallel resonance frequencies $f_s$ and $f_p$ as well as broadening of the resonance peaks.

Fig. 6.2: Phase angle $\phi$ of the measured quartz impedance $Z$ in $H_2O$/glycerol mixtures. With increasing glycerol concentration, i.e., increasing viscosity, the phase change is reduced and becomes broader.
With increasing glycerol concentration, i.e., increasing viscosity, the series and parallel resonance frequency, $f_s$ and $f_p$, shift to lower values. The frequency shift is accompanied by a broadening of the resonance peaks. Additionally, the absolute impedance $|Z|$ at $f_s$ increases, while $|Z|$ at $f_p$ decreases. Finally, the phase changes at series and parallel resonance frequency are less than 180°. As a result it becomes more and more difficult to sustain oscillation with increasing liquid viscosity in a continuous feedback oscillator setup. The application of continuously operating oscillators for on-line viscosity monitoring is limited to approximately 200 mPa-s \[109\]. Viscosity monitoring up to 1.5 Pa-s requires impedance analysis methods as demonstrated here, i.e., measurement of the impedance transfer characteristics and analysis through equivalent circuit models.

The extracted viscous loss parameter $R_2$, shown in the equivalent circuit schematic in Fig. 3.6, is shown in Fig. 6.3 as a function the liquid density viscosity product $\rho \eta$ for liquids at 25°C and compared to the theoretical values from Eqn. (3.25). The $R_2$ values of the H$_2$O/glycerol solutions are smaller than the values predicted by Eqn. (3.25) for a quartz with identical electrodes with radius $a = 3.76$ mm and $r_{he} = 3.2$ mm.

![Fig. 6.3: Equivalent circuit resistance $R_2$ (see Section 3.1.1) representing the viscous energy loss of a TSRR as a function of the density viscosity product $\rho \eta$ for different test liquids. The gray and black line represent the theory for a quartz with equal electrodes of 3.2 mm and 3.76 mm radius, respectively. For H$_2$O/glycerol mixtures, $R_2$ increases according to the theory of a purely Newtonian liquid.](image)

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6.1 Thickness Shear Mode Resonator

$r_{ele} = 3.2 \text{ mm}$ on both surfaces. This discrepancy is due to the simplifying assumption that the area of the smaller electrode also defines the oscillating quartz volume and the electrode capacitance $C_0$. Considering the real electrode geometry with a larger front electrode, the exciting electric field extends over an area larger than the area of the smaller electrode and, therefore, the active quartz area and electrode capacitance $C_0$ are increased. An effective electrode size in between the smaller and larger electrode area can be used to compensate for this effect. With an estimated effective electrode radius of $r_{eff} = 3.76 \text{ mm}$, good agreement with the measured $R_2$ values for H$_2$O/glycerol solutions is achieved. For low viscosities $\eta < 10 \text{ mPa-s}$, most test liquids correspond to theory, whereas PDMS samples with higher viscosity saturate at about $R_2 = 2500 \text{ } \Omega$, indicating viscoelastic behavior.

From the measured equivalent circuit parameters, the $Q$-factor was calculated according to Eqn. (3.27) and compared with theoretical results for Newtonian liquids, as shown in Fig. 6.4. The H$_2$O/glycerol mixtures behave according to theory, even at a probing frequency of 5 MHz. PDMS with shear viscosity $\eta < 60 \text{ mPa-s}$ also behave Newtonian. The saturation effects observed for PDMS with $\eta > 60 \text{ mPas}$ indicate viscoelastic behavior. More energy is stored elasti-

![Graph](image_url)

**Fig. 6.4:** $Q$-factor of a TSMR loaded with different sample liquids. H$_2$O/glycerol mixtures coincide with the theory for Newtonian liquids (solid line). PDMS with $\eta > 60 \text{ mPas}$ show viscoelastic behavior.
6 Viscosity Sensing

cally within the fluid and less is dissipated viscously. This explains why the $Q$-factor does not decrease anymore.

As far as homogenous liquids are concerned, a stable result, i.e., a stable impedance curve, is obtained almost immediately. Contrary, the measured impedance curves for suspensions slowly drifted towards a more or less stable value. The higher the volume concentration of solids, the longer it took to get to an almost stable situation. The resulting $Q$-factor indicates a lower viscosity than expected from the shear viscosity measurements. Whether this is because of shear thinning, sedimentation, or particle migration due to the shear rate gradient in the measured liquid layer is not understood. A clear distinction between $\text{H}_2\text{O}$ based and solvent based desk jet inks can be made. $\text{H}_2\text{O}$ based samples closely match with Newtonian theory, whereas solvent based inks indicate a higher viscosity value than expected. Although the sample volume was tightly sealed, solvent evaporation may contribute to this deviation.

6.2 Lamb Wave Sensor

The Lamb wave sensor with its $3 \times 8 \text{ mm}^2$ membrane is quite large and care must be taken when it is loaded or cleaned. A sample volume of 2 ml was applied through an opening in the test chamber. The test setup was temperature controlled at 20 °C and the $S_{11}$ parameter was measured with an HP 4195A network/spectrum analyzer as described in Section 5.2. The Lamb wave devices were cleaned after each measurement by rinsing with appropriate solvents and deionized water.

Fig. 6.5 shows the transmission characteristic of a Lamb wave device loaded with 60% $\text{H}_2\text{O}$/glycerol solution. The theoretical transfer characteristic shown in Fig. 6.5 is calculated with the impulse model (see Section 3.1.2). The AW velocity required in the theory was derived from the measured center frequency $f_0$ and the damping coefficient was chosen such that the resulting IL change matches the additional IL introduced by the liquid load. Hereby, it is assumed that the IDT impedance remains the same for the unloaded and the loaded Lamb wave device such that no additional IL occurs due to power reflection at the IDTs. The resulting liquid induced IL is therefore the IL difference between the loaded and unloaded state. Loading with a 60% $\text{H}_2\text{O}$/glycerol solution resulted in a center frequency of 10.7 MHz and an IL of 25.5 dB.
The AW reflections causing the jagged peaks in the transfer characteristic of the unloaded device (see Fig. 5.3) are strongly damped. AW reflections, such as the triple transit signal, have longer propagation paths than the main acoustic signal and are, therefore, more damped. The influence of a liquid load on the transmission characteristic becomes even more clear in the time domain response shown in Fig. 6.6. The EM feedthrough is not affected by the liquid load. However, with increasing liquid viscosity, the AW reflections are strongly damped, and the ratio between main AW signal and AW reflections continuously increases. Moreover, the width of the main AW signal becomes smaller with increasing viscosity. The width of the main AW signal in air is smaller because the AW velocity $c$ in air is much higher.

The pass band of the transmission characteristics becomes broader with increasing viscosity. This can be explained by a decrease of the effective IDT length. The more viscous the test liquid is, the more damped is the AW. With increasing damping, the finger pairs at the far end of the transducer sense less of the AW than those close to the emitting transducer. Therefore, the effective length of the transducer is shorter, which consequently leads to a shorter impulse response of
the Lamb wave (see Fig. 6.6) and to a broader pass band in the frequency domain. Neglecting finger pairs at the remote ends of the transducers also results in a shorter center-to-center distance of the IDTs. Hence, for high viscous liquids the IDTs of the sensor seem to be shorter and lie closer together.

The center frequency $f_0$ and the minimal IL are extracted from the transfer characteristics and related to liquid density $\rho_l$ and shear viscosity $\eta$. Fig. 6.7 shows the center frequency $f_0$ as a function of the liquid density $\rho_l$ for a series of different test liquids. The theoretical frequency shift (solid line in Fig. 6.7) was calculated for $\text{H}_2\text{O}/\text{glycerol}$ mixtures according to Eqn. (3.40) and is based on measured material properties. The shear viscosity $\eta$ was measured (see Section 2.3), the density $\rho_l$ and sound velocity $c_l$ data were obtained from [110] and [111], respectively. The results for $\text{H}_2\text{O}/\text{glycerol}$ mixtures closely match the theoretically calculated values. Good agreement between experiment and theory was also found for the measured solvents and desk jet inks as can be seen from Tab. 6.1.

For low shear viscosity, the thickness of the viscously entrained mass layer $\delta_\eta$ is much smaller than the skin depth $\delta_p$ of the evanescent pressure wave and, as a
6.2 Lamb Wave Sensor

Fig. 6.7: Center frequency $f_0$ of the Lamb wave device as a function of the liquid density $\rho_f$ for different lidded loadings. For highly viscous H$_2$O/glycerol an additional frequency drop due to the increasing effective mass of the viscously entrained liquid layer is observed. The solid line shows the theoretical result for H$_2$O/glycerol mixtures.

result, the resonance frequency shift is dominated by the liquid density and sound velocity, as shown in Eqn. (3.40). Typical values for the skin depth of evanescent pressure wave are $\delta_p = 16 \mu m$ for $\lambda = 80 \mu m$ in H$_2$O. With increasing shear viscosity $\eta$, the effective mass contribution of the viscously entrained liquid layer increases and leads to an additional frequency shift for $\eta > 60$ mPa-s.

The frequency shift measured in PDMS is considerably larger than that of liquids with comparable density and viscosity. The lower sound velocity $c_l$ of PDMS leads to an increasing penetration depth $\delta_p$ of the evanescent pressure wave, as can be verified with Eqn. (3.37). Hence, the resulting increase of the effective mass load leads to a considerably larger frequency shift for PDMS, in correspondence with theory (see Fig. 6.7 and Tab. 6.1).

For the suspensions the sound velocity $c_l$ is almost constant with 1480 - 1500 m·s$^{-1}$ and the viscosity for the measured samples is $\eta < 10$ mPa-s. Therefore, the frequency changes predominantly as a function of density $\rho_f$ at a rate of about 3.1 MHz·g$^{-1}·$cm$^3$. 

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Fig. 6.8: Center frequency $f_0$ of a Lamb wave sensor loaded with acetone/ak10 mixtures as a function of the liquid density $\rho_l$. The theoretical behavior according to Eqn. (3.40) is represented by the solid line.

Fig. 6.8 shows the center frequency as a function of the density $\rho_l$ for a series of acetone/ak10 mixtures. These liquids have low viscosities ($\eta < 11.3 \text{ mPa s}$ at $20^\circ \text{C}$), i.e., only changes in density and sound velocity contribute to the frequency shift. The measured data closely matches the calculated values according to Eqn. (3.40). Again, the theoretical frequency data were calculated using measured and tabulated material properties.

The measured minimal IL (see Fig. 6.9) is related to the product of the liquid density and the shear viscosity $\rho_l \eta$. For H$_2$O/glycerol mixtures, the IL decreases linearly with $(\rho_l \eta)^{1/2}$, as expected from Eqn. (3.41). However, the measured rate of change is smaller than calculated from theory (see solid line in Fig. 6.9). The deviation can be explained by the simplified model used to calculate the IL and partly by the shorter effective center-to-center distance of the IDTs with increasing liquid viscosity. Since AW damping is proportional to the propagation distance, a shorter center-to-center distance of the IDTs reduces the IL. The theoretical curve (solid line in Fig. 6.9) is not exactly linear because the IL calculated with Eqn. (3.41) also depends on the material properties, for which tabulated and measured values have been used.
6.2 Lamb Wave Sensor

![Diagram](image)

**Fig. 6.9:** Minimal IL for the Lamb wave sensor loaded with different sample liquids as a function of \((\rho \cdot \eta)^{1/2}\). The black line represents the theory for the H$_2$O/glycerol mixtures. The gray line is a linear fit to the H$_2$O/glycerol mixtures. The saturation behavior of PDMS for \(\eta > 60 \text{ mPa}.\text{s} \) indicates viscoelastic behavior.

The minimal IL of PDMS loaded Lamb wave devices saturates with increasing shear viscosity similar to the Q-factor behavior in the case of the TSMR (see Fig. 6.4). Again, the saturation effect can be explained with viscoelastic behavior of the fluid at the relatively high probing frequencies. All sample liquids with low viscosity, including suspensions, show a linear dependence as expected from theory.

Tab. 6.1 summarizes the calculated and measured center frequency \(f_0\) and IL for a number of sample liquids. The center frequency and IL of the Lamb wave sensor in air were used as reference values.

The viscosity of a liquid can be easily changed by varying its temperature (see Section 2.3), while the density remains almost constant. To test the Lamb wave devices, the viscosity of pure glycerol was measured in a temperature range from 10 - 70 °C and the results were compared with theory. Fig. 6.10 shows the measured and calculated IL change as a function of temperature. To compensate for the temperature drift of the Lamb wave sensor, the transfer characteristic of the
6 Viscosity Sensing

**Tab. 6.1:** Comparison of the theoretical and experimental center frequency $f_0$ and minimal IL of the Lamb wave sensor loaded with different liquids. Air is used as reference for the IL.

<table>
<thead>
<tr>
<th>Liquids</th>
<th>Theory</th>
<th>Experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$f_0$ [MHz]</td>
<td>$\Delta$IL [dB]</td>
</tr>
<tr>
<td>Air</td>
<td>13.64</td>
<td>0</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>10.73</td>
<td>1.74</td>
</tr>
<tr>
<td>Acetone</td>
<td>10.79</td>
<td>1.31</td>
</tr>
<tr>
<td>PDMS (AK10)</td>
<td>10.10</td>
<td>3.12</td>
</tr>
<tr>
<td>88% glycerol</td>
<td>10.35</td>
<td>19.09</td>
</tr>
</tbody>
</table>

Unloaded device was monitored over temperature and subtracted from the measured data, yielding the IL change due to the viscous liquid. The measured data qualitatively corresponds to theory.

![Graph](image)

**Fig. 6.10:** IL change of the Lamb wave device immersed in glycerol with respect to the unloaded device as a function of temperature. Circles and solid line represent measured data and theory, respectively.
6.3 Piezoelectric Cantilever

The piezoelectric cantilevers are the simplest and most convenient devices investigated in this work. Their size and robustness make the handling very easy. For liquid sensing the cantilever is vertically immersed in the test liquid. All experiments were carried out at room temperature. Drift effects were not observed and the output signal was almost instantaneously stable. After finishing a measurement, it is sufficient to clean or dry the sensor with a napkin. Small scratches do not effect the performance.

As described in Section 5.3, the piezoelectric cantilevers were excited at their resonance frequency using a burst signal. The characteristics of the resonator, namely the resonance frequency $f_0$ and the quality factor $Q$, were extracted from the free oscillation decay of the beam after switching off the excitation signal. The piezoelectric response signal of a 2.4 cm long cantilever immersed in PDMS with $\eta = 290$ mPa·s after switching off the excitation is shown in Fig. 6.11. The damped oscillations are fitted with the amplitude decay function of a linearly damped, harmonic oscillator. From the fit parameters, the resonance frequency $f_0$.

![Free oscillation](image)

**Fig. 6.11:** Piezoelectric output signal of a 2.4 cm long cantilever immersed in PDMS with a shear viscosity of $\eta = 290$ mPa·s after switching off the excitation burst signal. The gray line is a fit function describing the amplitude decay of a damped harmonic oscillator with resonance frequency $f_0 = 459$ Hz and quality factor $Q = 6.3$.  

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and the $Q$-factor are calculated. Hence, the immersed cantilever can be described as a damped harmonic oscillator with resonance frequency $f_0 = 459$ Hz and quality factor $Q = 6.3$.

The transverse vibration of the cantilever generates a fluid flow field around the cantilever. The generation of this fluid flow is equal to an additional mass that has to be accelerated, whereby the mass is proportional to the product of the liquid density and the effective flow volume. For liquids with low viscosity, the flow volume is almost constant because the viscosity has little effect on the fluid flow field (potential flow). Therefore, the effective mass that has to be accelerated scales mainly with the density of the liquid and the resonance frequency decreases with increasing density.

Increasing liquid viscosity reduces the $Q$-factor and leads to an additional frequency decrease according to Eqn. (3.49). Furthermore, it is assumed that acoustic radiation can be neglected because back and front side of the cantilever are acoustically short circuited such that no net energy flow takes place.

Fig. 6.12 depicts the resonance frequency $f_0$ as a function of the liquid density $\rho_l$. For low viscous sample liquids a linear relation between frequency and density is found. For PDMS with almost constant density and low viscosity, in this case $\eta < 20$ mPa·s, the resonance frequency is independent of viscosity and remains constant. For the suspensions the frequency changes linearly with liquid density at a slope of $130$ Hz·g$^{-1}$·cm$^3$, while the relatively small shear viscosity of $\eta < 15$ mPa·s has no detectable influence on the resonance frequency $f_0$. The low viscous H$_2$O/glycerol solutions coincide with the suspensions.

High viscous PDMS and H$_2$O/glycerol solutions exhibit a strong additional frequency decrease. Since the fluid density changes are small, the additional frequency change is due the low $Q$ mentioned above. Liquids with similar density and shear viscosity exhibit a similar frequency shift, independent of the type of fluid. Due to the transverse beam motion and the large deflection, the probed fluid volume is larger than that of the microsensors and all sample fluids appear homogenous.

The corresponding results for the $Q$-factor versus the shear viscosity $\eta$ are shown in Fig. 6.13. The $Q$-factors of all sample liquids follow a single trend line. This is a further indication that the investigated liquids behave purely viscous at frequencies around 400 Hz and can be related to the steady shear viscosity $\eta$ measured with conventional rheometers as introduced in Section 2.3. For high shear viscous
6.4 Membrane resonator

The membrane size of the piezoelectrically excited resonators (see Fig. 4.3) is small compared to the size of the Lamb wave membranes. This makes the devices much more stable and easier to handle. Nevertheless, the loading with a liquid has to be done very carefully to avoid air bubbles in the small cavity of the etch groove. Trapped air bubbles impede liquid contact with the membrane and lead to wrong results. The measurement setup was temperature controlled and held at 20 °C. The sensors were loaded with a constant sample volume of 2 ml.

The transverse membrane motion leads, as for the Lamb wave and piezoelectric cantilever, to a pressure driven fluid flow. In contrast to the piezoelectric cantile-
Fig. 6.13: Q-factor of a 2.4 cm piezoelectric cantilever as a function of the liquid shear viscosity $\eta$. The Q-factor continuously decreases with increasing shear viscosity. The Q-factors for all sample liquids coincide on a single trend line indicating Newtonian behavior in the investigated frequency range.

ver, only one side of the membrane is in contact with the liquid, while the other side is exposed to air. Since there is no acoustic short circuit between the back and front side of the membrane, acoustic energy is radiated from the vibrating membrane, causing an additional clamping mechanism. As described in Section 3.2.2, the emitted energy is proportional to the product of liquid density and sound velocity $\rho c$. 

Fig. 6.14 shows the piezoelectric response of a membrane resonator loaded with 48% H$_2$O/glycerol to an input burst of 1 V$_p$ at 130 kHz. The response signal of the resonator to its mechanical vibration is superimposed with a low frequency capacitive cross talk signal at the start and end of the excitation burst signal. The capacitive crosstalk causes a distortion in the piezoelectric output signal while switching on and off the excitation. The cross talk signal was filtered by applying an FFT and removing the low frequency part of the frequency spectrum. With an inverse FFT the response signal without cross talk was obtained. The filtered amplitude decay after switching off the excitation burst was then used to extract the Q-factor and the resonance frequency of the liquid loaded resonator. For
48% H₂O/glycerol, a quality factor $Q = 18.9$ and a resonance frequency $f_0 = 129.8$ kHz were found.

The resonance frequency $f_0$ of a 295 x 295 μm² membrane as a function of the liquid density $\rho_l$ is shown in Fig. 6.15 for a series of homogenous sample liquids. Due to the additional accelerated mass, the resonance frequency decreases linearly with the fluid density with a slope of $-63.0$ kHz·g⁻¹·cm³⁻¹.

As for the piezoelectric cantilever, the $Q$-factor was calculated from the amplitude decay after switching off the burst signal and related to the liquid shear viscosity $\eta$. The results are depicted in Fig. 6.16. A common trend for PDMS and the acetone/ak10 mixtures seems to exists. Both fluid types have similar density $\rho_l$ and sound velocity $c_l$ and, therefore, about the same acoustic radiation losses. However, the measurement points scatter considerably because of errors due to the data evaluation. AW reflections at the solid/liquid interface, or trapped air in the recessed cavity. Compared to PDMS and acetone/ak10 solutions, H₂O/glycerol mixtures have higher densities and sound velocities (see Tab. 2.2). There-
Fig. 6.15: Resonance frequency of a 295 x 295 μm² membrane resonator loaded with different sample liquids as a function of the liquid density $\rho_l$. The resonance frequency decreases linearly with liquid density at a rate of $-63.0 \text{ kHz} \cdot g^{-1} \cdot \text{cm}^3$.

Fig. 6.16: $Q$-factor of the membrane resonator as a function of the liquid shear viscosity $\eta$. Higher acoustic radiation losses in H$_2$O/glycerol due to higher acoustic wave impedance cause a lower $Q$. 

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fore, more energy is radiated into the liquid and a reduced $Q$-factor is expected. Above 1 Pa·s the low $Q$-factor limits the measurable viscosity range.

6.5 **SENSOR COMPARISON**

Each of the four investigated sensor systems has its advantages and disadvantages. In the following the different aspects of liquid property sensing with the four presented sensor systems are discussed. The performance of the different sensors are compared and specific topics for future on-line applications are addressed.

6.5.1 **SENSITIVITY**

All sensors are sensitive to liquid density and shear viscosity changes, as seen in the precedent sections. In the case of the Lamb wave sensor and the membrane resonator, the response also depends on the liquid sound velocity $c_f$. The sensitivity, as defined in Eqn. (3.1), varies with the liquid properties in a three-dimensional parameter space. To compare the performance of the four sensors the relative density sensitivity $S_{f_0}^{p_j}$ of the resonance frequency $f_0$ and the relative viscosity sensitivity $S_Q^\eta$ of the quality factor $Q$ are calculated from the measured data of $H_2O$ and 48% $H_2O$/glycerol mixture according to

$$S_{f_0}^{p_j} = \frac{1}{f_0} \frac{f_0(H_2O) - f_0(48\% \text{ glycerol})}{\rho_f(H_2O) - \rho_f(48\% \text{ glycerol})}$$

and

$$S_Q^\eta = \frac{1}{Q} \frac{Q(H_2O) - Q(48\% \text{ glycerol})}{\eta(H_2O) - \eta(48\% \text{ glycerol})}.$$  

Tab. 6.2 compares the relative density and viscosity sensitivity of the resonance frequency and the $Q$-factor for the different sensors.

The relative density sensitivity of the different devices has the same order of magnitude. However, the membrane resonator is about twice as sensitive to density as the piezoelectric cantilever which is about twice as sensitive as the Lamb
### Tab. 6.2: Comparison of the relative density $S_{f}^{R}$ and viscosity sensitivity $S_{Q}^{R}$ of the resonance frequency $f_{r}$ and quality factor $Q$, respectively.

<table>
<thead>
<tr>
<th></th>
<th>H₂O</th>
<th>TSMR</th>
<th>Lamb wave</th>
<th>Cantilever</th>
<th>Membrane resonator</th>
</tr>
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<td>$S_{f}^{R}$ [g⁻¹·cm³]</td>
<td>-</td>
<td>-0.09</td>
<td>-0.28</td>
<td>-0.43</td>
<td></td>
</tr>
<tr>
<td>$S_{Q}^{R}$ [mPa⁻¹·s⁻¹]</td>
<td>-0.13</td>
<td>-</td>
<td>-0.09</td>
<td>-0.15</td>
<td></td>
</tr>
</tbody>
</table>

wave sensor. The mass sensitivity of the sensors depends on the ratio of vibrating sensor mass to the accelerated liquid mass. While the Lamb wave sensor moves only a thin liquid layer given by the skin depth of the evanescent pressure wave or the viscously entrained liquid layer, the membrane resonator has a much larger liquid volume to move, leading to a larger liquid density sensitivity.

The $Q$-factor sensitivity to viscosity is about the same for all sensors, but large errors must be expected due to the errors of the measured values. The viscosity sensitivity of the $Q$-factor is the highest for the membrane resonator, but this is partly due to the acoustic radiation losses. The highest sensitivity would be expected from the TSMR where the liquid is subject to a pure viscous shear deformation.

#### 6.5.2 Measurable Viscosity Range

The operation frequency range of the investigated sensors covers five orders of magnitude, from $2 \cdot 10^2$ - $2 \cdot 10^7$ Hz. Moreover, the measurement principles vary from sensor to sensor and pure shear deformations compete with predominantly pressure driven flows. Considering the importance of these issues to viscosity sensing, as pointed out in Chapter 2, it becomes clear that the methods must be compared on the base of liquids largely insensitive to sensor specific issues. For this reason only Newtonian liquids, such as the H₂O/glycerol mixtures with an upper shear viscosity of 1.5 Pa·s for pure glycerol at 20 °C, are considered in this Section. All sensor types are capable of measuring up to the 1.5 Pa·s. Accounting for the resolution and measurement technique, there are, however, differences in the measurable viscosity range. Using impedance analysis methods, the TSMR can be operated over the full viscosity range (0 - 1.5 Pa·s). The equivalent circuit resistance $R_2$ as a measure of $Q$ provides sufficient resolution even in the high
6.5 Sensor Comparison

viscosity region. For continuous operation methods, the upper viscosity limit is around 200 mPa·s in the case of a TSMR [109]. A similar picture is found for the Lamb wave device. With a network/spectrum analyzer, the limit of detection is given by the noise level of the transmission characteristic. For the best devices this also resulted in an upper viscosity limit of 1-2 Pa·s. In a self-oscillating setup, upper values of about 200 mPa·s have been reported [29, 112]. The piezoelectric cantilever can be operated in liquids with viscosity up to 5 Pa·s. Low Q-factors for high viscosity solutions, however, restrict the practical application range to about 0.5 Pa·s. The main advantage of the piezoelectric cantilever beam is its low operation frequency, which increases the range of measurable liquids tremendously. Finally, the membrane resonator can be operated in fluids up to 1 Pa·s, but similar to the piezoelectric cantilever low Q-factors reduce the practical measurement range to less than 200 mPa·s, depending on the type of liquid. Fluids with viscosities η ≥ 1 Pa·s cannot be probed by the sensors investigated in this work. However, the ultrasound Doppler velocity profile measurement technique, as indicated in Section 1.2.2, provides an advanced solution to circumvent this limitation.

Monitoring non-Newtonian liquids with the presented sensors requires a detailed analysis of the relevance of the measured parameters in comparison to low frequency liquids properties. Care must be taken when choosing a high frequency microviscometer for viscosity sensing, as seen from the results for the TSMR and the Lamb wave in Section 6.1 and Section 6.2. Viscoelastic properties influence the measurement results and comparison with steady shear viscosity measurements is hardly possible. However, the high frequency data might provide other crucial information about the sensed fluid. Ultimately, an array of resonant sensors covering a large frequency range should be used to probe the liquid properties at different operating frequencies at the same time.

6.5.3 Packaging

Packaging has to address a number of issues, especially regarding the on-line applications in, e.g., the foodstuff industry. The packaging solutions presented in Section 4.3 take some of these issues into account, but are primarily optimized for fast sample and sensor exchange under laboratory conditions. Adaptation for future on-line applications is a must. In the following, some of the packaging key points, namely chemical and mechanical stability, ease of replacement, on-line application, and cleaning, are discussed for the individual sensors.
6 Viscosity Sensing

The sensor with the best chemical stability is the AT-cut TSMR. Quartz as a piezoelectric material and gold electrodes are hardly attacked by most common liquids. The housing, made of Voltalef and Teflon, is also chemically inert. In contrast to its chemical resistivity, the quartz is mechanically very sensitive. Abrasion, wear, scratches, or deposits cause frequency drifts. A thin hard coating layer might be useful for improved mechanical stability. The TSMR holder provides the most economic packaging solution. In the case of a defect, the quartz disc with the electrodes can be replaced in a matter of half a minute. All the electronics can be reused and the spring contacts offer a fast and reliable solution for contacting the TSMR. Sensor cleaning is made be rinsing the sensor with solvents or water. Mechanical cleaning methods can damage the sensor.

The commercially available piezoelectric cantilever has not been designed for liquid sensing applications. The coating that encapsulates the cantilever dissolves in common solvents. No solvent based sample liquids were measured for this reason. A new, chemically inert protection coating, e.g., a thin Teflon coating, has to be applied to provide sufficient protection. The cantilever has to be firmly attached to a rigid support to generate well defined operation conditions. It has to be verified that the combined structure has no resonance frequency in the vicinity of the fundamental cantilever resonance frequency. Otherwise, distortion of the response signal occurs. Tip-mounting of the cantilever on a rod with thread enables comfortable replacement, if needed. A tuning fork arrangement with two cantilevers operating in opposite direction can be used to reduce the energy dissipation into the support. In contrast to the TSMR and the piezoelectric microsensors, the static liquid pressure acts homogeneously on the cantilever allowing for high pressure applications. Moreover, the piezoelectric cantilever is very stable and easy to handle. Between two measurements, the beam is cleaned by wiping it off with a tissue.

The microsensors (Lamb wave and membrane resonator) are the most delicate structures. The mechanical stability of the 3 x 8 mm² membranes is rather small. At the present state the application area is focused at ambient pressure conditions [112]. Moreover, any applied differential pressure changes the membrane tension and the Lamb wave sound velocity accordingly. To replace a broken sensor, the entire packaged device must be replaced. Any electronics on the chip front side is protected as long as the backside of the chip cavity is sealed. The only difference between the Lamb wave and the membrane resonator is the size of the membrane. Due to the smaller membrane size, the membrane resonator withstands
6.5 Sensor Comparison

higher pressure. Sensor cleaning is made by rinsing the sensor with solvents or water. Care must be taken not to break the membrane. Mechanical cleaning methods can damage the sensor.

6.5.4 On-line Application

The investigated devices have been tested in standing liquids at atmospheric pressure and negligible differential pressure. The only liquid motion is generated by the motion of the operating devices themselves. From this point of view, all systems have been proved to operate successfully in non-flow situations. The response of the different sensors to a change of the liquid properties is almost instantaneous, as can be verified from the immediate change of the output signal when the sensor is loaded with a test sample. How fast the liquid in the sample volume actually achieves an equilibrium state (taking into account, e.g. demixing effects) is a different issue.

Moving from laboratory conditions with standing liquids to on-line applications with pressure driven flow introduces a number of new aspects to be taken into account. In a steady state situation with fully developed flow profile, the shear rate depends on the location. Therefore, the viscosity of a non-Newtonian liquid also depends on the location within the flow profile. A detailed introduction to viscometric flows for a Poiseuille flow configuration is given in [8]. To simplify the following discussion it is assumed that:

- The ambient conditions, e.g., pressure and temperature, are within the sensor operation range and do not damage the sensor.
- The liquid can be measured under laboratory conditions.
- A steady state condition, i.e., no pressure pulsations, with a laminar flow profile is established.

Under these conditions, the main difference to the standing liquid situation are the shear rate gradient in the liquid and boundary layer effects at the wall/liquid interface of the tube. The fluid velocity profile and the corresponding shear rate profile develops according to the liquid properties. The interaction of the involved parameters can lead to very different fluid flow profiles [7].

Two effects that influence the measurement results of a wall-mounted sensor are:

- Particle migration in suspensions, particles in a dispersion move to the center of the pipe where shearing is a minimum.
• The occurrence of plug flow [8].

For the standing liquid experiments, a non-slip boundary condition at the solid/liquid interface was assumed. In an in-line application, this condition may not be valid if boundary layer effects occur. Examples of boundary layer effects are:

• Wall-slip effects.

• Large shear rate gradients, e.g., due to particle migration.

Considering all these effects, the question arises whether a wall based or a point-wise measurement method provides enough information to sufficiently characterize the overall liquid property for a sensible in-line control. Depending on the type of process and the liquid, microsensors can still be an alternative.

Fig. 6.17 shows a cross-section view of a Lamb wave sensor or membrane resonator in an on-line set-up. The sensor back is flush-mounted with the tube wall. The etched cavity causes a recess of the active membrane. The cavity has a number of drawbacks. Depending on the cavity size, the liquid flow pattern deviates from the flow pattern at the pipe wall. Eventually, the liquid in the cavity is standing. As a consequence, the speed of the liquid property adjustment in the cavity due to a change in the liquid property in the main flow is reduced and delays the sensor response. Other effects, such as vortices, turbulences, sedimentation, or the formation of plug in the cavity, are possible.

Fig. 6.17: Schematic cross-section of an in-line mounted Lamb wave or resonant membrane sensor. The fluid flow in the etched cavity can be distinctively different from the main flow behavior.
Fig. 6.18 is a schematic picture of an in-line mounted piezoelectric cantilever. The narrow side of the cantilever faces the liquid flow direction and the oscillation is perpendicular to the flow direction. This arrangement minimizes the flow perturbation and the effect of the impact pressure on the cantilever. As illustrated in Fig. 6.18 and mentioned earlier, a shear rate gradient exists along the cantilever and changing liquid properties at different location in the flow can be the consequence.

For on-line applications, the TSMR can be either mounted in the wall, as seen in Fig. 6.17, or in an in-flow position similar to the piezoelectric cantilever in Fig. 6.18. The resulting advantages and drawbacks are similar to the ones mentioned above. Differential pressure effects are circumvented with complete immersion of the TSMR in the liquid. However, this requires either an electric isolation of the two electrode contacts or restriction to non-conductive liquids.

In real on-line applications, a number of other undesired effects have to be mastered. Examples are secondary flows, gas bubbles, and pressure pulsations. On-line applications also require clean-in-place capability. Depending on the specific cleaning cycle, the devices must be able to withstand high temperatures and aggressive detergents. These issues have not been investigated in this thesis and adaptation of the microsensors to real on-line conditions remains a topic for future work.

Fig. 6.18: Schematic of an in-line mounted piezoelectric cantilever. The cantilever and its motion lead to flow disturbances. Furthermore, shear rate and fluid properties can change along the cantilever.
6 Viscosity Sensing
7 Conclusion and Outlook

7.1 Conclusion

In this thesis, the feasibility of micro viscosity sensors for simultaneous liquid density and viscosity sensing has been demonstrated. Four different systems, a TSMR, a Lamb wave sensor, a membrane resonator, and a piezoelectric cantilever, have been successfully tested in various sample liquids with viscosities up to 1.5 Pa·s. The four sensors cover a frequency range from 200 Hz up to 20 MHz.

The TSMR is an AT-cut quartz with 5 MHz resonance frequency featuring single sided electric contacts. Impedance measurements have been carried out and modeled with an equivalent circuit model. The calculated equivalent circuit parameters were related to liquid density and viscosity. Excellent results were achieved for Newtonian H₂O/glycerol solutions with shear viscosities up to 1.5 Pa·s. For non-Newtonian liquids viscoelastic behavior was observed.

The Lamb wave sensor and the membrane resonator are based on the DIMES-01 bipolar process including a bulk-micromachining module and a ZnO module for deposition of thin piezoelectric ZnO films. The material properties of the piezoelectric ZnO layers have been investigated and a new fabrication process has been established to grow ZnO layers with low tensile stress on an all Al substrate layer held at ground potential. Based on a series of wafer curvature measurements the sputter parameters were optimized to deposit almost stress free ZnO. For the used Varian sputter system, ideal sputter parameters are a substrate temperature of 400 °C and a sputter current of 3.0 A in 100% O₂ atmosphere at a pressure of 6 mTorr. The optimized piezoelectric ZnO films show a clear polycrystalline structure with a preferential orientation of the piezoelectric c-axis perpendicular to the substrate. Piezoelectric films with a specific resistance up to 4·10⁶ Ω·cm were deposited, enabling a piezoelectric operation mode down to 50 kHz. GDOES analysis of the sputtered film confirmed an optimal stoichiometric composition of Zn:O = 1:1. Contributions of other elements and molecules
are negligible. With load-deflection and nano indentation experiments the plane-strain modulus of sputtered, polycrystalline ZnO was found to be $114.6 \pm 5.9$ GPa and $121.8 \pm 2.7$ GPa, respectively.

Based on the optimized ZnO films, piezoelectrically actuated Lamb wave sensors and membrane resonators were fabricated. The sensors were mounted in a cavity-down CPGA with a laser drilled hole to allow back side exposure of the membrane to the liquid. The $S_{21}$ parameter of the Lamb wave sensors was measured and excellent transfer characteristics with a strong acoustic signal were observed. An IL of less than 20 dB was measured in air and good agreement with theory was achieved. In air, the main AW signal was more than 10 dB above the AW reflections. A time gate technique was applied to suppress EM feedthrough signals and AW reflections. For liquid loaded Lamb waves, the measured IL and center frequency of the transmission characteristics were related to the liquid properties. Good correspondence to theory was achieved for Newtonian H$_2$O/glycerol solutions and liquid viscosity was measurable up to 1.5 Pa-s. Viscoelastic effects, similar to those found for the TSMR, were observed in non-Newtonian liquids.

Membrane resonators, based on the same fabrication process as the Lamb wave devices, were also tested as liquid property sensors. An AC burst signal applied to an electrode located in the membrane center excites a transverse vibration mode, while an electrode along the edge of the membrane is used to detect the membrane vibration. The resonance frequency $f_0$ and quality factor $Q$ are extracted from the vibration decay after switching off the burst signal. In air, a resonance frequency of 381 kHz and $Q$-factor of 91 was measured for 12 $\mu$m thick, 295 x 295 $\mu$m$^2$ square membranes. Contrary to the other three sensors, where acoustic radiation losses can be neglected, part of the resonator energy is radiated into the liquid, giving rise to an additional energy loss mechanism besides viscous energy dissipation. Therefore, the quality factor $Q$ also depends on the acoustic impedance of the liquid. The resonance frequency decreases linearly with the liquid density at a rate of $-63$ kHz·g$^{-1}$·cm$^{-3}$.

Finally, commercially available bimorph PZT cantilevers with either 2.4 or 4.8 cm length, 1 cm width, and 1 mm thickness were tested as viscosity sensors. After burst excitation of a transverse vibration mode, the resonance frequency $f_0$ and quality factor $Q$ were extracted from the free amplitude decay after switching off the excitation signal. Resonance frequencies of 735 and 263 Hz, as well as quality factors of 128 and 79 were measured in air, respectively. Immersion of the
cantilevers in liquids resulted in a decrease of the resonance frequency and the quality factor. For viscosities $\eta < 20$ mPa·s, the frequency decreases linearly at a rate of $-130 \text{ Hz} \cdot \text{g}^{-1} \cdot \text{cm}^3$. For high viscosities, an additional resonance frequency drop due to low $Q$-factors was observed. The $Q$-factor for all sample liquids followed a common trend line and could be related to the steady shear viscosity $\eta$. This indicates that all liquids respond purely viscous in a frequency range below 500 Hz.

At first sight, one could conclude that the piezoelectric cantilever beam has an edge over the other sensors. Its low operation frequency make it suitable to measure a large range of different types of liquids. The good mechanical stability is advantageous for many sensor applications. Furthermore, the obtained results are relatively easy to interpret.

However, the piezoelectric cantilever requires a relatively large sample volume compared to the microsensors. To gain rheological information about small liquid samples microsensors are a promising alternative.

The experience made with the tested sample liquids indicates that the measurable viscosity range is limited to a few PAs due to heavy viscous damping and resulting insufficient measurement accuracy. Therefore, future sensor development based on resonant sensors should focus on application areas in the low viscosity range.

### 7.2 Outlook

Further investigations in the area of thin film rheology and microstructuring are necessary to gain a deeper understanding of the processes taking place near the liquid/solid interface and how they can be related to high frequency liquid deformation. Especially, the separation from purely viscous to viscoelastic behavior has to be investigated in detail. Moreover, it has to be investigated how the sensed viscosity of a thin liquid layer can be related to the viscosity of the bulk media.

One way to close the gap between conventional rheometers and resonating viscosity sensors may be the use of an array of similar sensors with different operation frequencies to cover an entire range of frequencies, similar to conventional oscillating shear rheometers. Measuring the resonance frequency $f_0$ and the quality factor $Q$ or the IL, respectively, for several similar devices, operating at differ-
Conclusion and Outlook

ent resonance frequencies, allows a better characterization of the investigated fluid.

Further improvement of the piezoelectric cantilever can be achieved by using a tuning fork setup with two identical cantilevers. The potential viscosity sensing range might also be increased with partial immersion of the cantilever into the liquid. A feedback oscillator can be used to automatize the sensor system.

Up to this point, no on-line experiments were carried out. Future work will have to prove the on-line capability of the sensors. However, the experiences made so far with the investigated microsensors are encouraging to continue the development of a low cost viscosity sensor for on-line applications in the low viscosity range.
REFERENCES


References


References


References


References


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[106] A. Hierlemann, ETH Zurich, Zurich, private communication.


### ABBREVIATIONS

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tr>
<td>AW</td>
<td>Acoustic Wave</td>
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<tr>
<td>BMM</td>
<td>Bulk Micromachining</td>
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<td>CMOS</td>
<td>Complementary Metal Oxide Semiconductor</td>
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<tr>
<td>CPGA</td>
<td>Ceramic Pin Grid Array</td>
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<tr>
<td>DMSO</td>
<td>Dimethylsulfoxide</td>
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<td>ECE</td>
<td>Electrochemical Etch</td>
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<td>EM</td>
<td>Electromagnetic</td>
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<td>FEM</td>
<td>Finite Element Method</td>
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<td>FFT</td>
<td>Fast Fourier Transformation</td>
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<td>FPW</td>
<td>Flexural Plate Wave</td>
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<td>GDOES</td>
<td>Glow Discharge Optical Electron Spectroscopy</td>
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<tr>
<td>IC</td>
<td>Integrated Circuitry</td>
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<tr>
<td>IDT</td>
<td>Interdigital Transducer</td>
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<tr>
<td>IFFT</td>
<td>Inverse Fast Fourier Transformation</td>
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<td>IL</td>
<td>Insertion Loss</td>
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<tr>
<td>iMEMS</td>
<td>Integrated Micro Electro Mechanical System</td>
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<tr>
<td>ISA</td>
<td>Integrated Sensors and Actuators</td>
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<tr>
<td>KOH</td>
<td>Potassium Hydroxide</td>
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<tr>
<td>LPCVD</td>
<td>Low Pressure Chemical Vapor Deposition</td>
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<td>LW</td>
<td>Love Wave</td>
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<tr>
<td>NMRI</td>
<td>Nuclear Magnetic Resonance Imaging</td>
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<tr>
<td>PDMS</td>
<td>Poly(dimethylsiloxane)</td>
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<td>Abbreviation</td>
<td>Description</td>
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<tr>
<td>PECVD</td>
<td>Plasma Enhanced Chemical Vapor Deposition</td>
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<td>PZT</td>
<td>Lead Zirconate Titanate</td>
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<td>QCM</td>
<td>Quartz Crystal Microbalance</td>
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<td>SAW</td>
<td>Rayleigh Wave</td>
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<td>SEM</td>
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<td>SH-APM</td>
<td>Shear Horizontal Acoustic Plate Mode</td>
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