

Lateral Field Excited Acoustic Wave Devices: A new Approach to Bio-Interface Sensing

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Abstract— 10 MHz AT-cut quartz crystals have been excited by placing two electrodes on one crystal surface. An electric field is generated, which is not only laterally confined in the plane of the quartz crystal disk, but also penetrates into the medium adjacent to the opposite bare sensor surface. Due to the mechanical and electrical nature of medium-sensor-interaction, access to additional physical material parameters is anticipated. Up to now the influence of an adjacent medium's electrical and mechanical property changes on the sensor response has not been examined in detail for Lateral Field Excited (LFE) sensors. In order to determine these influences, the sensor impedance spectrum has been investigated while the sensor was exposed to liquid acoustic loads exhibiting varying permittivity and conductivity. The associated results will be discussed in terms of reorganization of the electric field distribution within the quartz.

I. INTRODUCTION

Devices based on piezoelectric materials, which allow the transduction between electric and acoustic energy, have been developed in a number of configurations for sensor applications. Quartz crystal resonators (QCRs) which are excited via gold electrodes on both main surfaces and operated in Thickness Shear Mode (TSM) are one important representative. The basic working principle is the decrease of resonance frequency by an added mass in the ng-range. In (bio)chemical analyses, however, this simple mass-assumption has a limited validity. When operated in liquid environment, viscoelastic properties of the medium contribute not only to a change in resonance frequency, but also the influence of acoustic energy dissipation on the sensor signal must be considered. The signal generation is additionally determined by density ρ and viscoelasticity η of the adjacent medium.

In contrast to these Thickness Field Excited (TFE) sensors, the resonant behavior of so-called Lateral Field Excited (LFE) acoustic wave devices is influenced not only by mechanical but also by electrical medium properties. Both electrodes are

placed only on one main surface (reference surface) of the quartz crystal, thereby creating a major electric field component in the plane of the disk. This also causes a TSM excitation as it is known from common QCRs. The aspect ratio between electrode gap distance and crystal thickness is about 3-6. The electric field is not completely confined between the electrodes and thus to the quartz crystal. The electric field also penetrates partly into the medium adjacent to the bare side of the crystal (sensing surface) giving rise to an extended sensitivity. This feature can provide access to additional relevant physical material parameters (permittivity ϵ , conductivity σ along with density, viscoelasticity, shear modulus G) of a target measurand.

Although LFE devices are known for a long time, their application as a sensor has started just recently and up to now no functional relation is available, which correlates mechanical and electrical properties of the medium with properties of the impedance spectrum. In order to improve the understanding of LFE transduction mechanisms, different liquid loads with varying mechanical and electrical properties have been applied to the LFE sensor.

II. EXPERIMENTAL

A. Acoustic Sensor

LFE sensors have been self-fabricated on the basis of 10 MHz AT-cut quartz crystal blanks (Lap-Tech, Canada). In order to establish the piezoelectric excitation, electrodes have been sputtered onto one surface of the blank. In our case, the electrodes are semi-circled, have a diameter of 10 mm and are separated by a gap of 0.5 mm. The electrode structure has been fabricated by cutting the desired shape into sheet plates of 0.2 mm thickness. These shadow masks then have been used for the sputtering process, where electrodes consisting of circa 500 Å of chromium followed by circa 2000 Å of gold have been deposited on the quartz blanks. Fig. 1a) depicts a schematic of the electrode setup.

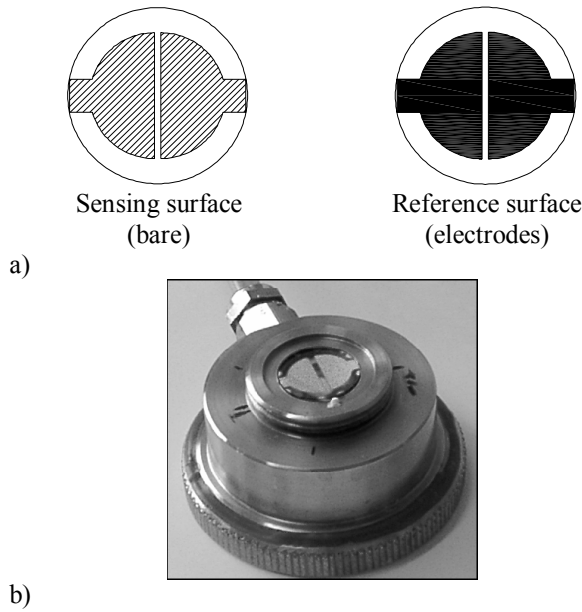


Figure 1: a) Electrode layout for LFE sensors
b) 10 MHz LFE sensor in the measurement cell

The acoustic device has been assembled in a measurement cell providing pin contact to RF source and ground potential at the reference surface. Fig. 1b) shows the measurement cell with a lateral field excited 10 MHz AT-cut quartz crystal sensor. In the figure the bare sensing surface is on the top and can be exposed to different media of interest whereas the electrode covered surface is at the back and protected from the media via O-ring sealing. The blank sensing surface can be advantageous for the long-term stability of the sensors, since cleaning the electrodes is always accompanied with abrasion of gold and thus a degradation of the sensor performance.

The data acquisition for all experiments has been performed at fundamental frequency at room temperature with an in-house developed fast digital sensor-interface based on impedance-analysis [1]. The single-board RF impedance spectrum analyzer (RF-ISA), capable of operating up to 150 MHz, is specifically interfaced to the LFE sensors and provides a higher frequency resolution as well as a faster data acquisition compared to commercial network analyzers.

In order to completely determine the influence of the medium property changes on the sensor response, the entire impedance spectrum of the LFE sensor has been measured. The critical impedance parameters, namely the series resonance frequency f and resistance R , can be obtained on-line from the sensor's conductance maximum. Note that compared to TFE, the lateral field excited main resonance in the impedance spectrum is less pronounced, therefore so-called spurious modes become more visible. More attention in terms of the experimental setup is hence required.

B. Materials

The influence of non-polar, non-conductive liquids on the acoustic devices has been studied by applying different alkanes to the bare sensor surface. Hexane, Heptane, Decane

and Dodecane (>99%, Merck, Germany), Octane, Nonane (99,5%, Fluka, Germany), as well as cyclohexane (Sigma-Aldrich, Germany), as a representative of cycloalkanes, have been used.

Electrolytic solutions consisting of KCl (Fluka, Germany) and de-ionized (DI) water have been applied to analyze the influence of varying electric liquid properties on the sensor signal. According to concentration levels of 0 wt%, 0.01, 0.02 ... 0.05 wt%, 0.1 wt%, 0.3 wt% KCl in water, different conductivity levels can be achieved.

Prior to every measurement the sensor has been cleaned. We recommend using a mixture of concentrated sulfuric acid (Applchem, Germany) with an amount of 1-5 mg/ml potassium permanganate (Riedel-de Haën, Germany) for 30 to 300 seconds, followed by rinsing with DI water and drying with a stream of nitrogen.

III. THEORY

The thickness of rotated Y-cut quartz crystal plates is usually defined parallel to the Cartesian y' -axis, while the crystallographic x - and z' -axis are located in the plane of the crystal disk (the primed variables describe the rotated axes while the unprimed one expresses the unrotated axis). Unlike the Thickness Field Excitation, where the electric field is applied parallel to the quartz plate normal (y'), a lateral excitation applies the exciting electric field in the plane of the disk. In this configuration the electrodes should be deposited at the edges of the crystal blank. Due to the fact, that the thickness of such crystal disks is typically in the sub-millimeter range, a more practical realization applies semi-circled electrodes on the main surfaces of the blank crystal leaving a gap between them. For sensor purposes we use electrodes only on the bottom surface. The alignment of the gap can be done in parallel to the crystallographic x -axis, z' -axis or any orientation consisting of a vector combination of both. However, for the excitation of the pure Thickness Shear Mode a z' -directed electric field, $E_{z'}$, in the crystal plate is required. As a consequence the gap must be arranged parallel to the x -axis [2, 3].

Under real conditions the applied $E_{z'}$ cannot be expected to be ideal and homogeneously distributed within the entire quartz plate between the electrodes. The electric field penetrates into the quartz plate and a parallelism of the field lines in the electrode gap can only be assumed near the interface between the quartz disk and electrodes. With increasing distance from this location the electrical field lines will exhibit also a y' -directed component of varying but rather small amplitude. Because these y' -directed components are comparable to the typical exciting electric field known from TSM sensors a similar mechanical displacement must be assumed. In order to verify the presence of such normal orientated field component, recent studies analyzed the impedance spectrum by rotating a blank quartz crystal disk relative to the orientation of the LFE electrode gap [4]. As a result, neither of the major resonance peaks in the spectrum, also recorded at a larger frequency span, remained unchanged in air. From this one can exclude a noticeable contribution

from E_y field component and thus a corresponding resonance peak in the spectrum in the acoustically unloaded case.

The impedance spectrum of a laterally excited resonator in air does not show one main resonance, as known from common QCRs. It shows at least two resonances with significant amplitudes. This feature has not only been found for 10 MHz resonators. Also in previous studies a couple of strong resonance peaks appeared when using 5 MHz crystals in combination with a comparable electrode setup [4]. Evaluating this phenomenon is focus of current work and is assumed to be closely connected to the geometry of the quartz resonator. In all studies the strongest resonances has been considered.

Comparable to common TFE acoustic devices, the resonance peak of LFE resonators can electrically be described by two characteristic frequencies - the resonance (f_r) and antiresonance (f_a) frequency [3, 5]. Table I summarizes the resonance conditions for both excitation forms, TFE and LFE, based on the acoustic wave velocity v_a , resonator thickness d , and coupling constant k of the particular excitation. It is obvious from the table, that the LFE resonator features a resonance frequency, which is higher for a given resonator thickness compared to the TFE mode. Moreover the antiresonance frequency of a crystal excited in thickness direction coincides with the resonance frequency of the same resonator in lateral excitation.

IV. RESULTS AND DISCUSSION

The application of LFE sensors in liquid environments is the focus of current research, since here they evolve their full capabilities. The acoustic sensor response of commercial TFE sensors (AT cut quartz with fundamental frequency of 10 MHz) to Newtonian liquids can be predicted from the equation of Kanazawa and Gordon and is in case of DI water approximately -2 kHz for the series resonance frequency shift, Δf , and +150 Ω for the resistance change ΔR [6]. Exposing bare LFE sensors of the same fundamental resonance frequency to DI water however results in changes of -18...-19 kHz accompanied by a significant decrease of the resistance at the conductance maximum [7]. Moreover, the entire impedance magnitude spectrum outside the mechanical resonance declines significantly indicating a significant change of the parallel capacitance of the sensor setup. Fig. 2 exemplarily depicts these results in a semi-logarithmic plot of a LFE sensor impedance spectrum.

As a consequence of that extraordinary sensor response, we suppose that the piezoelectric excitation and transduction mechanism within the quartz plate has been changed by the presence of the adjacent liquid. The property of the interface of bare sensor surface and a surrounding medium is hereby of main importance. As long as the sensor faces a medium which features a relative permittivity ϵ_r lower than that of the quartz crystal ($\epsilon_r \approx 4.6$), the electrical field is distributed mainly in lateral direction between the electrodes. For a medium featuring a dielectric permittivity higher than that of quartz (e.g. DI water $\epsilon_r \approx 80$) the internal lateral electric field component decreases in strength and components of the TFE will be amplified. In this case the adjacent medium can be seen as a virtual electrode opposite to the metallic LFE

TABLE I.
RESONANCE CONDITIONS FOR TFE AND LFE RESONATORS IN
FUNDAMENTAL MODE [5].

	TFE	LFE
resonance frequency f_r	$\frac{v_a}{d} \sqrt{(\pi^2 - 8k_{TFE}^2)}$	$\frac{v_a \pi}{d}$
antiresonance frequency f_a	$\frac{v_a \pi}{d}$	$\frac{v_a}{d} \sqrt{(\pi^2 + 8k_{LFE}^2)}$
frequency separation	$\frac{4k_{TFE}^2}{\pi^2}$	$\frac{4k_{LFE}^2}{\pi^2}$

electrodes. Consequently the electric field distribution within the quartz plate is changed and the strength of the electric field performs a modification to the thickness direction of the quartz disk. Nevertheless, this enhancement of the 'traditional' kind of piezoelectric excitation in normal direction is not necessarily correlated to the typical shifts in frequencies or resistances defined by the medium's density and viscosity.

Further it should be noted that the resistance shift of common QCRs is defined by the medium properties only, as long as the geometric and material properties of the resonator remain constant. An altered excitation mechanism however can also influence these parameters. Since, e.g. the acoustically active area of the resonator in contact to different loads can not exactly be determined, we concentrate in the following on investigating the changes in the series resonance frequency only.

To further verify the assumption, that electrical medium property changes (i.e. the liquid's permittivity) can influence the piezoelectric excitation mechanism, several non-polar liquids have been investigated. For this purpose different (cyclo-)alkanes have been applied to the sensor since their relative permittivity ϵ_r is much lower than that of water and

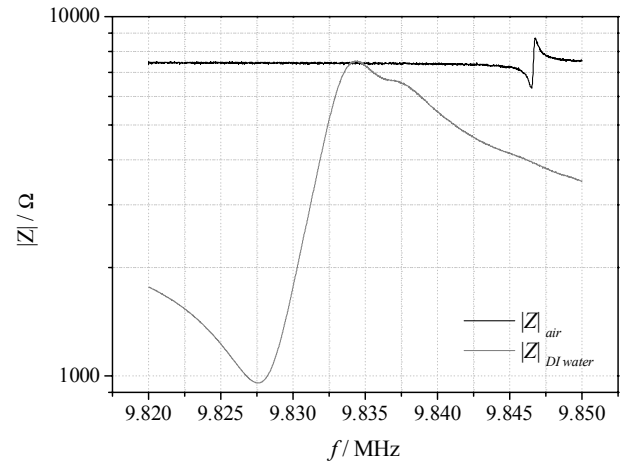


Figure 2: Semi-logarithmic plot of the impedance spectrum of a 10 MHz LFE sensor exposed to air and DI water.

slightly lower than ε_r of the quartz crystal. The frequency shifts of LFE sensors that have been exposed to these liquids show a dependency as it is displayed in Fig. 3. Additionally the frequency shifts for a 10 MHz QCR sensor in contact to these liquids have been calculated based on Kanazawa's equation. Since the QCR is sensitive to mechanical medium property changes only, a frequency shift dependency determined by the medium's density and viscosity can be found in the graph. The data in Fig. 3 have been sorted at the abscissa according to the relative permittivity of the substances (material data are summarized in Table II). The parallelism of the theoretical and the experimental curve depict the known dependence on density viscosity for all LFE sensors. However, an absolute difference of more than 500 Hz remains. A closer inspection shows that the frequency shift differences increase with permittivity of the applied liquid. Fig. 3 shows the difference between the averaged measured frequency shifts and theoretically expected ones.

We cannot exclude that the applied substances are entirely free of water and thus being fully non-polar. The impurities may contribute to a change in the excitation mechanism as already described in case of pure DI water.

From these experiments we conclude, that LFE sensors in case of non-polar liquid media clearly respond similar to TSM sensors. The electric field distribution remains almost in the plane of the acoustic device as long as ε_r does not exceed the one of the crystal. Therefore the sensor response Δf to these type of media is describable with traditional models of QCRs. At low values of liquid permittivity, changes in this value provide an additional contribution to the frequency shift of about 600-700 Hz per permittivity increase with respect to air.

Another important liquid property is conductivity σ . This can be significant e.g. in biochemical applications, where buffer solutions containing different amounts of salts are used as liquid matrix for cells, proteins, liposomes, etc. . The influence of such electrolytic solutions to LFE sensors has also been focus of the investigation.

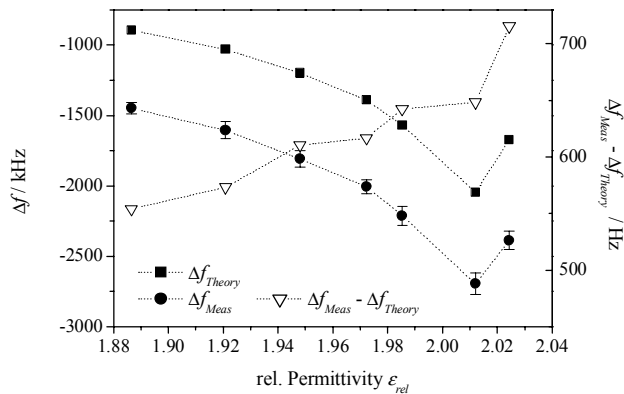


Figure 3: Measured frequency shifts of LFE sensors together with theoretical values valid for standard QCRs as a function of the relative permittivity. Additionally the difference of measured and theoretical data is shown.

TABLE II.
DENSITY , VISCOSITY AND RELATIVE PERMITTIVITY OF SELECTED
(CYCLO-)ALKANES [8]

Substance	Density ρ (g/cm ³)	Viscosity η (mPas)	rel. Permittivity ε_r
Hexane	0.6606	0.3	1.8865
Heptane	0.6795	0.387	1.9209
Octane	0.6986	0.508	1.948
Nonane	0.7192	0.665	1.9722
Decane	0.7266	0.838	1.9853
Dodecane	0.7495	1.383	2.012
Cyclohexane	0.7739	0.894	2.0243

Electrolytic solutions have been prepared by adding varying amounts of KCl to DI water. Constant volumes of 300 μ l have been applied to the sensor. Fig. 4 exemplarily summarizes the measured frequency shifts of two LFE sensors as a function of the KCl concentration. The frequency shift caused by pure DI water has been subtracted previously in the plot. As can be seen, the frequencies decrease significantly for low concentrations, saturate above 0.1 wt% and approach a constant value for KCl concentrations above 0.5 wt% (this part not shown), so that the entire frequency shift drops up to 16 kHz. Note that the density-viscosity product of the solutions stays almost constant. The influence of these acoustically relevant properties on the resonance frequency can be neglected for a concentration up to 0.5 wt% because it is just 4 Hz according to QCR theory. The significant change in resonance frequency therefore must have been caused by the liquid's electrical properties only. Concerning the relative permittivity and conductivity, a comparison to theoretical QCR models is not feasible since common QCRs are insensitive to these parameters. The inset in Fig. 4 shows the relative changes of both parameters in a semi-logarithmic plot

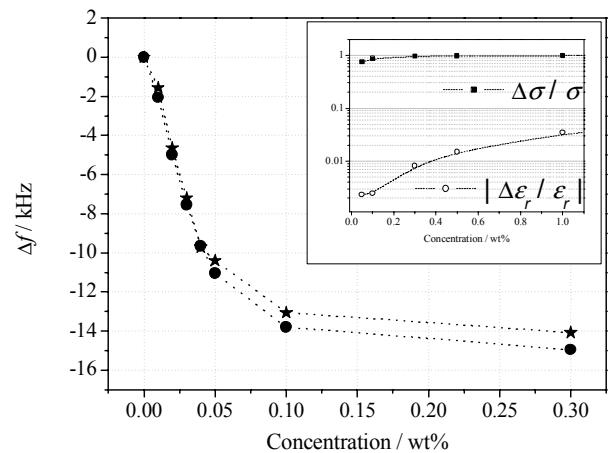


Figure 4: Frequency shifts of two bare LFE sensors as a function of KCl concentration in DI water. The influence of pure DI water (approx. -19 kHz.) has been subtracted previously.

Inset: Relative changes of conductivity and permittivity as a function of concentration in a semi-logarithmic plot.

up to 1 wt%. For comparison reasons the absolute value for $(\Delta \varepsilon_r / \varepsilon_r)$ has been given, because the relative permittivity decreases slightly with higher concentration. Instead of applying the values of DI water, data for infinite dilution have been extrapolated for both parameters. As can be seen, the relative change of σ is more pronounced than that of ε_r , however, $\Delta \sigma / \sigma$ is almost constant in the interesting concentration range. Similar experiments with different concentrations of 2-Propanol in DI water, which provide a stronger variation of ε_r and less variation in σ , show shifts in f comparably small to those measured in the KCl studies.

Recapitulating these findings one can suggest that the liquid's permittivity is indeed involved into the process of the rearrangement of the electric field distribution in the plane of the quartz disk. The conductivity however has an even more pronounced influence on the sensor signal. We assume that the potential of the virtual electrode at the interface of quartz and adjacent liquid can be altered with σ and reaches saturation already at low concentrations of KCl in DI water. This saturation effect and thus the almost constant Δf remains for much higher concentrations (up to 16 wt%, not shown here).

V. CONCLUSION

Significant frequency changes have been observed when bare LFE sensors are exposed to a liquid medium which offers a high relative permittivity. It has been suggested that the electric field distribution within the quartz disk is altered by the medium, so that the normal component of the electric field dominates the general excitation and transduction mechanism. Taking Table I into account, an estimation of the correlated resonance frequency shift is possible when changing from lateral to thickness excited mode. The equivalence of the resonance frequency f_r in lateral excitation and the antiresonance frequency f_a in thickness excitation is hereby of importance. Assuming a LFE sensor, oscillating at a resonance frequency of 10 MHz, the corresponding TFE antiresonance frequency is known and allows - by use of the TFE frequency separation

$$\frac{f_{aTFE} - f_{rTFE}}{f_{aTFE}} = \frac{4k_{TFE}^2}{\pi^2} \quad (1)$$

along with the coupling constant $k_{TFE} = 0.088$ [5] - the calculation of the TFE resonance frequency: $f_{rTFE} = 9.968615$ MHz. The separation of both frequencies by 31.385 kHz also describes the shift in the resonance frequency that can be expected in case of a fully modified direction of the electric field. The contribution of the electrodes has been neglected.

Indeed the electrolytic solution caused higher frequency shifts as has been observed for pure DI water, but even in the saturated case the highest observed frequency change is smaller than this value. Therefore one can assume that an

adjacent medium cannot completely alter the electric field direction from the former lateral towards the thickness direction and thus a superposition of both components of excitation is present in the quartz plate.

This extremely high sensitivity of LFE sensors observed for low concentrations of ions should be taken into account when using such type of sensor in biochemical studies, where e.g. the immobilization of biomolecules onto the sensor surface is of main interest. Slight variations in a low concentrated buffer matrix, e.g. when changing from the idle buffer to a slightly different one containing the biomolecule, can influence the sensor signal much more than the reaction of interest.

On the other hand the focus of future work is addressed to such slight variations near the sensor-liquid interface. LFE sensors will be applied in a liquid environment, which additionally contains (solid) nanoparticles. Particle adsorption to the sensor surface is hereby of main interest. It is known that repulsive forces can hinder this process resulting in a much lower than expected frequency change. One can expect supportive information from electrical property changes during adsorption process, especially when these nanoparticles possess a permittivity which differs significantly from the surrounding liquid matrix.

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