Topical Review

Guided acoustic wave sensors for liquid environments

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Abstract

Surface acoustic wave (SAW)-based sensors for applications to gaseous environments have been widely investigated since the 1970s. More recently, the SAW-based sensors focus has shifted towards liquid-phase sensing applications: the SAW sensor directly contacts the solution to be tested and can be utilized for characterizing physical and chemical properties of liquids, as well as for biochemical sensor applications. The design of liquid-phase sensors requires the selection of several parameters, such as the acoustic wave polarizations (i.e. elliptical, longitudinal or shear horizontal), the wave-guiding medium composition (i.e. homogeneous or non-homogeneous half-spaces, finite thickness plates or composite suspended membranes), the substrate material type and its crystallographic orientation. This paper provides an overview of different types of SAW sensors suitable for application to liquid environments, and intends to direct the attention of the designers to combinations of materials, wave nature and electrode structures that affect the sensor performances.

Keywords: liquid environment, Love waves, SHAPMs, Lamb waves, HVPSAWs, PSAWs, SAW sensors

(Some figures may appear in colour only in the online journal)

1. Introduction

The basic structure of a surface acoustic wave (SAW) sensor includes a piezoelectric substrate and a pair of interdigitated transducers (IDTs) photolithographically patterned onto the free surface of the piezoelectric wafer \[1\]. The piezoelectric substrate can be a \textit{bulk} single crystal substrate (such as quartz, LiNbO\textsubscript{3} or LiTaO\textsubscript{3}), with thickness \(h\) larger than that of the acoustic wavelength \(\lambda\) (\(h \gg \lambda\)), or a \textit{thin film} (such as ZnO or AlN) with thickness \(h < \lambda\), grown onto a non-piezoelectric bulk substrate (such as silicon, sapphire, glass or diamond). In SAW devices, the acoustic wave travels at the surface of the propagating medium and its energy is confined within one wavelength of depth; it follows that the SAW properties (wave velocity and amplitude) are highly affected by any physical and chemical changes that occur at the surface of the propagating medium and/or at the adjacent medium, when the SAW device interacts with an external environmental stimuli (such as humidity, temperature and pressure). In the presence of a liquid environment, the wave properties can be perturbed by the changes in the electrical (conductivity and permittivity) and mechanical (mass density and viscosity) properties of the liquid contacting the sensor surface, or by the anchorage of a mass onto the sensor surface.

The environment can interact with the SAW directly or by means of a thin \textit{sensing} membrane that, for some specific applications, covers the wave propagation path between the two IDTs and is in direct contact with the environment to be tested. If the membrane is an insulating material, it can cover the entire SAW device surface, including the IDTs, while if it...
is conductive, it is positioned in between the two IDTs. As a consequence, part of the SAW energy is distributed into the sensing membrane and any change in its physical properties affects either or both the wave velocity and propagation loss, giving rise to a detectable output signal (a frequency and/or insertion loss shift) that represents the sensor response. The affinity of the membrane towards a specific target analyte is a fundamental prerequisite as it can drive the sensor selectivity towards a specific application. In gas sensing applications, the sensing membrane can be a thin Pd film [2, 3], a thin lead phthalocyanine (PbPc) film [4], a graphene-like nanosheet [5], a calixarene layer [6] or a polyethylene-fluorenol layer [7], to cite just a few examples, to detect H2, NO2, carbon monoxide, organic vapors or simply the relative humidity of the surrounding environment. In liquid-phase sensing applications, the membrane can be poly(isobutylene) (PIB), poly(epichlorohydrin) (PECH) or poly(ethyl acrylate) (PEA) to test toluene, xylene and ethyl benzene solutions [8], or polysiloxane film containing acidic functional groups for detection of organic amines in aqueous phase [9], or macrocyclic calixarenes for the detection of organic pollutants in drinking water [10]. The SAW sensors described in [11] were fabricated and derivatized with a rabbit polyclonal IgG antibody, which selectively binds to Escherichia coli O157:H7. A dual channel SAW biosensor for the simultaneous detection of Legionella and E. coli was fabricated using a novel protocol of coating bacteria on the sensor surface prior to addition of the antibody [12]. Länge et al. [13] presents an overview of 20 years-worldwide developments in the field of SAW-based biosensors for the detection of biorelevant molecules in liquid media.

Whatever the sensing membrane is, it is important to underline that the design of the device can significantly affect the performance of the sensor since its sensitivity is also dependent on the wave type and polarization, on the electroacoustic coupling configuration, on the material thickness and crystallographic orientation.

SAW devices are manufactured with semiconductor integrated circuit technologies that include the metal and piezoelectric layer deposition techniques (such as sputtering, pulsed laser deposition or thermal evaporation), optical or electronic lithography and the lift-off process to pattern the IDTs and the sensing membrane. The selection of the substrate (the material type, crystallographic cut and thickness), the IDT’s design (metal fingers width and spacing, number of fingers pairs, aperture, IDT’s centre-to-centre spacing, to cite just a few), as well as the selection of the electroacoustic coupling configuration affect the characteristics of the electroacoustic devices (such as the operating frequency, quality factor, insertion loss) in such a way as to enhance the sensitivity towards the environmental parameter changes, regardless of the type of the adopted sensing membrane.

The SAW sensors are implemented on piezoelectric substrates showing high electroacoustic coupling, such as ZnO, LiNbO3 and LiTaO3, that ensure a high sensor sensitivity, sometimes at the cost of moderate temperature stability. It can happen that the thermal gradient can lead to frequency shifts, which are comparable to the sensor response to the measurand. The selection of temperature stable cuts of the piezoelectric substrates, such as the quartz ST-cut, is one way to improve the temperature-frequency stability of the SAW sensors. A dual sensor configuration, which includes the active sensor and a reference sensor [14–15], can be designed to compensate common mode influences other than temperature, such as humidity or aging, that affect both the active device, coated, for example, with a selectively adsorbing membrane, and the uncoated device that acts as a reference device. Alternatively, a temperature-compensated configuration, i.e. a multi-layered structure including opposite-sign temperature coefficients of delay (TCD) materials, with the appropriate thicknesses [16], can be designed to cancel the device’s spurious responses due to thermal drift [17, 18].

The present paper presents a survey of the electroacoustic devices based on the propagation of elastic waves travelling at the plane surface of half-spaces and within finite thickness plates; the common characteristic of these waves is the ability to travel in a medium contacting a liquid environment, without suffering excessive energy loss. The paper is structured as follows: in section 2, we consider features of the pseudo surface acoustic waves (PSAWs) and high velocity PSAWs (HVPSAWs), showing in-plane polarization and travelling at the surface of piezoelectric substrates; the results of numerical calculations and finite element modelling of the PSAW and HVPSAW-based sensors for liquid environments are presented. In section 3, the features of Love wave-based sensors are studied for different combinations of substrate and guiding layer materials, with varying layer thicknesses. In section 4, the features of shear horizontal acoustic plate mode (SHAPM)-based sensors are studied for different plate thicknesses, material types and mode order. In section 5, Lamb wave-based sensors are studied, including the quasi-longitudinally polarized fundamental and higher order modes, the fundamental symmetric mode and the quasi-Scholte wave. Section 6 concludes our paper.

2. Pseudo SAW and high velocity PSAW sensors

SAWs are elastic waves that travel at the free surface of a half-space and are confined within one acoustic wavelength λ in depth. The physical motion of the SAW is mechanically associated with an elliptical displacement of the surface that is characterized by one out-of-plane particle displacement component, U3, and two in-plane components, U2 and U1, normal and parallel to the wave vector k = 2π/λ. When travelling in a piezoelectric medium, the SAW strain field is accompanied by a travelling electric potential wave: the linear electromechanical coupling effect in piezoelectric materials enables the inter-conversion between electrical and acoustic signals. As a consequence, the excitation and detection of SAWs, as well as of any types of plate waves on piezoelectric substrates, is accomplished by means of metal IDTs, as first reported by White and Vollmer [19]. Figure 1 shows the schematic of a single-electrode-type IDT with uniform finger spacing and constant overlap: several metal strips are aligned and connected to the bus bars with a periodicity corresponding to the
The fingers’ width and spacing are equal to \( \lambda/4 \); the total length of the IDT is \( L = (N - \frac{1}{4}) \cdot \lambda \), being \( N \) the number of finger pairs; \( W \) is the electrodes overlapping (the IDT aperture); \( d = W/\lambda \) is the IDT directivity. The frequency of the propagating wave is \( f = v/\lambda \), where \( v \) is the velocity of sound in the half-space material. The SAWs propagate in both directions away from the IDT, along the propagation axis; thus, the inherent loss of the two IDTs is equal to 3 dB.

When an RF voltage is applied between the two bus bars of the transmitting IDT, a periodic strain is generated. As a travelling electric field is associated with the SAW, the metal strips of the receiving IDT will detect the SAW-induced charges. By changing the overlapping of the fingers, the IDT aperture, \( N \) and the spacing of the metallic fingers, the SAW bandwidth and the directivity can be changed. A complete description of the theory and modelling of IDTs employed for bandwidth and the directivity can be found in [20]. The electromechanical coupling factor \( K^2 \) is a measure of the electric-to-acoustic energy conversion efficiency; it is represented by the fractional change in wave velocity due to surface metalization, \( K^2 = 2 \cdot \left[ (v_{\text{free}} - v_{\text{met}}) / v_{\text{free}} \right] \), where \( v_{\text{free}} \) is the free surface wave velocity and \( v_{\text{met}} \) is the velocity on the metalized surface. The \( K^2 \) values of the SAWs travelling along common piezoelectric substrates are equal to 0.16, 0.75, 4.8 and 5.31\% for ST-quartz, 112°-x-y LiTaO\(_3\), x- and 128°-y-x LiNbO\(_3\), respectively [21]. Due to the anisotropic properties of the piezoelectric materials, the \( K^2 \) depends on the substrate crystallographic cut and wave propagation direction. The electrical impedance of the IDT depends on several factors, such as \( K^2 \), the dielectric permittivity of the substrate and the geometry of the IDT; it must match as closely as possible to that of external components (50 \( \Omega \)) and be resistive. IDT apertures of less than approximately 30\( \lambda \)- are inadvisable, as the transducer can diffract the acoustic beam resulting in an acoustic beam considerably diverging before reaching the output IDT [22]. The metal used to fabricate the IDTs is generally a low-resistance material highly adhesive to the substrate surface, with thickness around 0.1 \( \mu \)m, to make the transduction process more efficient; Al, Au, Cu and Mo are a few examples of commonly used metals. Some of these materials require the presence of a thin adhesive inter-layer (Ti or Cr) to improve the adhesion to the substrate or to avoid the diffusion of the metal into the substrate. The cost is also an important factor that drives the choice of the IDT metal type: Cr/Al appears to be the best choice with a good balance of relatively low resistivity, low cost and good surface adhesion. In harsh environment applications (such as high temperature or corrosive environments), the metal used to fabricate the IDTs should exhibit a high electrical conductivity, high melting temperature, a good resistance to oxidation and chemical inertness. Iridium, rhodium and platinum are a few examples of metals suitable for harsh environment applications [23–26].

In contrast to SAWs, which are polarized perpendicularly to the surface, the PSAWs and HVPSAWs are predominantly in-plane polarized; that makes them suitable for low loss propagation in contact with a liquid environment. The three SAW displacement components decay exponentially with the depth, while the PSAWs and HVPSAWs have both decaying and radiating components; the latter component radiates power into the half-space, thus resulting in an attenuation of the field amplitudes as the wave propagates. If the contribution from the radiating terms is sufficiently small, these two pseudo waves are observed as in standard SAW devices. The PSAW usually has the \( U_2 \) component as the dominant one \( U_2 \gg U_1, U_3 \) at the half-space surface), while the HVPSAW usually has the longitudinal component \( U_1 \) as the dominant term \( U_1 \gg U_2, U_3 \) at the half-space surface) [27]. For specific crystallographic cuts and wave propagation directions in the most common piezoelectric substrates, piezoelectrically-active PSAWs and HVPSAWs travel with minimum propagation loss and, since they are both in-plane polarized, they are suitable to work in contact with liquid. Three dimensional (3D) eigenfrequency finite element method (FEM) analysis was performed using COMSOL Multiphysics® Version 5.2 to explore the field shape of the SAW, PSAW and HVPSAW travelling at the surface of an ST-x quartz substrate (Euler angles 0° 132.75° 0°). Figure 2 shows the 3D primitive SAW cell considered in the analysis: beneath the single wavelength cell is a perfectly matched layer (PML) at the bottom for capturing losses related to bulk wave radiation.
the half-space. The primitive SAW cell has two periodic and two continuity boundary conditions applied on the sidewalls. The Al electrodes are 0.1 µm thick, with a pitch of \( p = 5 \mu m \) (\( \lambda = 20 \mu m \)). The IDT fingers width-to-spacing ratio was set to 1. The base material is ST-x quartz.

From figure 2 it can be clearly observed that, unlike what happens for the SAWs, the particle motion of the PSAW and HVPSAW is contained in the surface plane of the propagating medium, since the shear vertical displacement component is very small for both waves.

Attractive properties of PSAWs and HVPSAWs are the high velocity (close to that of the transverse and longitudinal bulk acoustic wave, respectively), low propagation loss and high electromechanical coupling coefficient [27].

The sensors based on PSAWs and HVPSAWs can measure the mechanical (mass density and viscosity) and electrical (conductivity and relative permittivity) property changes of the liquid that contacts the wave path directly, without covering the sensor surface with any selective film. The sensor response is caused by the mechanical and electrical boundary condition changes resulting from the perturbations the adjacent medium undergoes. If the wave propagation path is metallized and electrically shorted, only the liquid mechanical properties will affect the sensor response, as only the particle displacement component interacts with the adjacent liquid. If the bare acoustic path is in direct contact with the liquid, the sensor will also be sensitive to the electrical properties of the liquid as both the wave electrostatic potential and particle displacement interact with the liquid, and two perturbations occur. The electrical perturbation can be discriminated by detecting differential signals between two delay lines.

The absolute value of the admittance \( Y \) versus frequency curves for the three modes, propagating in ST-x quartz, in air and in water, were calculated by a frequency domain study, and the curves are shown in figure 3. Three peaks are clearly visible when the surface contacts the air (black curve): they correspond to the SAW, PSAW and HVPSAW whose velocities \( v = f \cdot \lambda \) are equal to 3167, 5083 and 5751 m s\(^{-1}\), respectively. The PSAW has a propagation loss higher than that of the SAW and HVPSAW [28]. The red curve of figure 3 corresponds to the ST-quartz half-space contacting the water. The SAW, which has a large displacement component normal to the substrate, is almost totally damped by the water as expected; the PSAW shows nearly no damping while the HVPSAW is affected by a small attenuation. As the vertical displacement component \( U_{3\text{surf}}/U_{1\text{surf}} \) of the HVPSAW, normalized to the \( U_1 \) at the surface, is about four times that of the PSAW, the former wave is more dampened by the water than the latter.

A two dimensional (2D) COMSOL simulation of the HVPSAW displacement profile inside the ST-x quartz is plotted in figure 4(a); the blue and the green curves represent the \( U_1 \) and \( U_3 \) HVPSAW displacement components; the abscissa is the normalized depth (for \( \lambda = 20 \mu m \)). Figure 4(b) shows the 2D representation of the HVPSAW total displacement. The substrate and liquid depths (120 µm) are equal: the ST-x quartz extends from −120 to 0 µm of the abscissa values, while the liquid half-space extends from 0 to 120 µm abscissa value. The liquid was modelled as a linear isotropic viscoelastic material with independent elastic constants; the bulk modulus and the dynamic viscosity were extracted from [29]. When the quartz is contacted by the water, the surface-normal displacement component of the HVPSAW (the blue line) generates compressional waves in the liquid phase: the power dissipated leads to just a small attenuation since the wave is predominantly in-plane polarized.

In an attempt to design a sensor packaging able to protect the IDTs from the liquid environment and to confine the measurand in the device sensing area, the electroacoustic device can be integrated with different types of microchannels. Typically, the test cell that localizes the liquid to the surface of the device consists of a pre-molded polydimethylsiloxane
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or an SU-8 cell [31] that is mechanically pressed against the surface of the sensor to avoid any contact between the IDTs and the liquid to be tested. The cell can be positioned in three different configurations: (1) in between the IDTs, as shown in figure 5(a), to prevent the presence of liquid on the IDTs; (2) it can occupy the entire device surface if the IDTs are shielded by a proper layer, as shown in figure 5(b), to avoid conduction through the liquid between the electrodes; (3) it can be positioned in between the IDTs and integrated with two air cavities that protect the IDTs. A vertical structure shaped as walls is positioned around the IDTs to separate them from the liquid-filled container, as shown in figure 5(c).

In the first and third configuration, the cell adds some disturbance effect to the propagation of the wave due to the mechanical stresses induced on the surface by the packaging, thus resulting in an additional improvement in damping losses. The second configuration ensures maximum sensitivity, as the physical size of the liquid cell includes the total wave propagation path, but requires a careful selection of the type and thickness of the protective surface layer to minimize potential perturbations to the device insertion loss, thermal stability, sensitivity, to cite just a few. The thin layer can even be a sensitive coating layer that exhibits high sensitivity towards a certain analyte but low sensitivity towards other species: thus, the layer drives the sensor selectivity towards a specific target analyte. A liquid cell integrated with the sensor during the fabrication process is preferred as it reduces the device’s complexity and enhances its performance. References [27–33] provide a very useful list of substrate types and crystallographic orientations along which low-attenuated, strongly-coupled PSAW and HVPSAW travel (such as 64°YX LiNbO₃, 36°YX LiTaO₃, quartz ST-X, LiNbO₃ with Euler angles (90° 90° 36°), (90° 90° 31°) LiTaO₃ and (0° 45° 90°) Li₂B₄O₇). For sake of completeness, the same references list other information that is fundamental for the design of the device parameters (operating frequency, IDT’s centre-to-centre distance, number of IDT finger pairs, IDT’s aperture, etc), such as the phase velocity, propagation loss, particle displacement components, electric-to-acoustic energy conversion efficiency, $K^2$ and power flow angle of the SAW, PSAW and HVPSAW. Moreover, these references also describe the wave’s behaviour with increasing the thickness of a metallic layer covering the wave propagation path: depending on
the layer thickness, the wave diffraction into the bulk can be prevented, while the transitions from HVPSAWs to higher order PSAW modes, and from PSAW to the SAWs can also be observed. The piezoelectric substrates of the LGX-family group have been studied in [34–36] for application to temperature stable, high coupling, low loss sensors for liquid environments. Several applications of PSAW sensors are presented in the available literature for the measurement of viscosity, electrical properties and mass loading of an adjacent liquid. In [37] a sensor for liquid viscosity and conductivity measurement is described that is based on a PSAW dual delay line on 41°-Y LiNbO3 and covered by a SiO2 protective layer. In [38] a PSAW sensor is implemented on 36°-Y LiTaO3 substrate for methanol concentration measurement. In [39] the PSAW trapping efficiency of the free, metallized and grating paths in YX-36° and YX-42° LiTaO3 and YX-64° LiNbO3 are compared at fundamental and harmonic frequencies. A high frequency (>500 MHz) PSAW sensor for a liquid environment is designed that shows high coupling, low loss, high operating frequency and high resistance to surface contamination. In [40] the propagation of PSAWs along bare 41° and 64°-Y LiNbO3, and 36°-Y LiTaO3 substrates is studied and compared with that along the same substrates covered by thin sputtered glass films to increase the resistance to surface contamination. It was demonstrated experimentally and theoretically that the glass layer does not affect the wave loss but it lowers the temperature stability. Film thickness-to-wavelength ratio regions were found, which represent a good trade-off between a lowered temperature coefficient of frequency and a preserved high coupling factor. Sensors of liquid viscosity and mass loading have been demonstrated utilizing the present mode on 36°-Y LiTaO3 [41].

One way to enhance the sensor sensitivity (i.e. the frequency change per unit incremental change in the measured quantity) is to raise the working frequency: this effect can be obtained by reducing the size of the IDT metal strips or by utilizing SAW devices based on high velocity acoustic wave modes. The HVPSAWs are attractive for this potential application as they travel at velocity close to the longitudinal bulk acoustic wave (BAW) velocity. In [42] the propagation of HVPSAWs in LiNbO3 has been studied in the range of Euler angles (0°, 124°, 90°); the corresponding theoretical velocities are between 6700 m s⁻¹ and 7400 m s⁻¹, about twice that of normal surface waves, but the K² varies between about 0.14% to 0.5%, much less than that of surface waves. In [43] some experiments show that the HVPSAW phase velocity in (0°, 124°, 50°) quartz reaches 6992 m s⁻¹ and the propagation attenuation is as low as less than 1 · 10⁻⁴ dB a⁻¹, thus it is suitable for liquid sensing applications. In [44] the propagation of HVPSAW along semi-insulating Fe-doped GaN films grown on sapphire substrates is experimentally studied and the small propagation attenuation of the mode when travelling at a liquid/solid interface is demonstrated in glycerol solutions. In [45] the propagation loss due to bulk wave radiation of a HVPSAW is reduced by loading the 36°Y- LiNbO3 substrate with a dielectric amorphous AlN thin film with a higher velocity than the substrate. The amorphous AlN layer plays a double role to protect the IDT's patterned onto the substrate surface and to enhance the device’s performance. Table 1 lists some examples of PSAW-based sensors and just two examples related to the HVPSAW; it is evident that, despite the long-time-recognized suitability of the HVPSAWs for liquid sensing applications [46] there is a lack of experimental validation.

### Table 1. Some practical examples of SAW sensors for application to a liquid environment test.

| Wave type | Substrate | Layer | Application | Reference |
|-----------|-----------|------|-------------|-----------|
| PSAW      | 41°-Y LiNbO3 | SiO₂ | Liquid viscosity and conductivity | [37] |
| PSAW      | 36°-Y LiTaO3 | | Methanol concentration | [38] |
| PSAW      | 36°-Y LiTaO3 | | Liquid viscosity and mass loading | [41] |
| HVPSAW    | Fe-doped GaN/sapphire | | Viscosity-density product | [44] |
| PSAW      | 36°Y-LiTaO3 | Poly(methyl methacrylate) (PMMA) or cyanoethylcellulose (CEC) | Rabbit anti-goat IgG | [47] |
| PSAW      | 64°Y-LiNbO₃ | 1,10-phenanthroline | Heavy metal compounds: PbNO₃ and CdNO₃ | [48] |
| PSAW      | 36°Y-LiTaO3 | | Tiny particles mixed with distilled water | [49] |
| PSAW      | 36°Y-LiTaO3 | | Detection and discrimination of various detergents | [50] |
| PSAW      | 36°Y-LiTaO3 | Parylene thin film | Urea biosensing | [51] |
| PSAW      | 36°Y-LiTaO3 | Parylene thin film | Water loading | [46] |
| HVPSAW    | Quartz (0° 124° 50°) | | | |

3. Love wave sensors

Love waves are a type of SAW characterized by a shear horizontal particle displacement component $U_2$ dominant over the vertical and longitudinal ones ($U_2 \gg U_1, U_3$). The propagation of the Love waves is excited and revealed by means of a couple of IDTs, as for the SAW-based devices. Due to the in-plane polarization, the Love waves, as well as the PSAW and HVPSAWs, are suitable for travel at a surface contacting a liquid environment. In the most general sense, Love waves propagate along the surface of a piezoelectric half-space covered by a thin layer: the substrate is responsible for the excitation of a surface skinning bulk wave (SSBW) that propagates below the substrate surface; the thin overlayer traps the acoustic energy and slows down the wave propagation velocity, thus reducing the loss from radiation into the bulk. As a result,
the SSBW is converted into a shear surface wave, the Love wave. Figure 6 shows the field profile of the SSBW travelling at the surface of a bare ST-quartz substrate, and of the Love wave travelling in the same substrate covered by a thin SiO$_2$ trapping layer. As can be seen, the strain associated with the SSBW penetrates deep within the bulk of the bare quartz substrate; the strain associated with the Love wave remains close to the surface of the SiO$_2$/quartz substrate.

The number of Love modes (LMs) that can propagate in the layer/substrate medium depends on the layer thickness, but the essential condition for the propagation of the Love waves is that the shear bulk wave velocity (SHBAW) of the half-space, $v_{\text{SHBAW}}^\text{sub}$, is larger than the shear bulk wave velocity of the layer, $v_{\text{SHBAW}}^\text{layer}$, as the velocity of the Love wave lies in between the $v_{\text{SHBAW}}^\text{sub}$ and the $v_{\text{SHBAW}}^\text{layer}$ [52]. Higher order LMs develop at their respective cut-off frequencies, which are related to the thickness of the layer: they are dispersive as their velocity depends on the layer thickness, other than on the substrate and the layer’s material properties. As the LM’s acoustic energy is mostly concentrated inside the guiding layer, the LM-based devices show good performances in terms of sensitivity to any disturbance loading the surface of the guiding layer. A comprehensive review of Love wave sensors can be found in [53]. As an example, figure 7 shows the phase velocity dispersion curves of the first five modes (LM1, LM2, LM3, LM4 and LM5) travelling along the ST 90°-x quartz half-space, bare and covered by a SiO$_2$ guiding layer, 2 µm thick, being $\lambda = 20$ µm.

When the guiding layer is very thin, the velocity of LM1 tends to the half-space SHBAW velocity; with increasing the layer thickness, the velocity of both the fundamental and higher order modes asymptotically reaches the layer SHBAW.
velocity. Love waves vanish if the frequency is lower than the cut-off frequency.

Figure 8(a) shows the delay line on top of the quartz half-space with the two IDTs located at a distance equal to 3-λ (λ = 20 µm); the depth of the substrate is 10 λ. The figure represents the total displacement 1 ns after the electric signal is applied at the transmitting IDT. Figure 8(b) shows the time evolution of the SSBW and LM1 total displacement propagating at the surface of the ST 90°-x quartz half-space, bare and covered by a SiO₂ trapping layer, 2 µm thick. The total displacement of the SSBW and LM1 was calculated by 3D COMSOL simulation: the time domain analysis was carried for 20 ns and the total displacement of the propagating medium was recorded at an interval of 1 ns. The transmitting bidirectional IDT launches two waves in opposite directions, as indicated by the arrows in figure 8(b); the signal applied at the transmitting IDT was a 10 V peak-to-peak sinusoidal signal at 231 and 217 MHz, for SSBW and LM1. The plot clearly shows that the acoustical displacement propagates into the depth of the substrate for the SSBW, while it is more confined to the surface for the LM1.

The time and frequency domain response analysis of a delay line based on the SSBW and LM1 propagation was performed by 3D COMSOL simulation assuming a pair of Al IDTs, 0.1 µm thick, positioned onto the quartz surface (see figure 9). The distance between the transmitting and receiving IDT was set equal to 3-λ (active gap region) and the fingers overlap W = 1-λ; the substrate propagation loss was not accounted for in the calculation. The IDT number of finger pairs N was assumed to be equal to 2 for both the SSBW and LM1 devices. The electrical voltage at the receiver electrode was recorded for 30 ns in the time domain analysis, as shown in figure 9(a) where the ratio V_{out}/V_{in} of the voltage at the receiver and transmitter IDT in the time domain is shown. The insertion loss of the delay line is calculated by applying a unit impulse at the input IDT: the Fourier transformation of the device impulse response allowed the calculation of the scattering parameter S_{21} versus frequency, for the SSBW- and LM1-based delay line onto the ST-90°-x quartz substrate, bare and covered by a SiO₂ guiding layer, 2 µm thick. The presence of the guiding layer is fundamental to trap the Love wave energy, and only a well-defined thickness guarantees enhanced device performances (such as minimum delay line insertion loss and maximum gravimetric sensitivity). Figure 10 shows the K² (black curve) and the derivative of the phase and group velocity versus the normalized SiO₂ layer thickness, h_{SiO₂}/λ, of the fundamental LM1 travelling along the ST 90°-x/SiO₂ substrate.
the maximum mass sensitivity of the phase and group velocity of the LM1, as the derivative of the \( v_{gr} \) and \( v_{ph} \) is proportional to the gravimetric sensitivity \( S_{grav} = \frac{\Delta v}{v_0} / (\rho \cdot h_{\text{SiO}_2}) \), where \( \rho \) and \( h_{\text{SiO}_2} \) are the layer mass density and thickness, \( \Delta v = v - v_0 \), \( v_0 \) and \( v \) are the wave velocity along the bare and covered half-space [55]. The \( K^2 \) values calculated at the abscissa values corresponding to the \( v_{gr} \) and \( v_{ph} \) maximum sensitivity are quite similar (0.230% and 0.204%), while the expected \( v_{gr} \) sensitivity is predicted to be about twice that of the \( v_{ph} \).

Figure 11 shows the derivative of the \( v_{ph} \) and \( v_{gr} \) of the first five LMs in ST 90°-x quartz/SiO2 with respect to the layer normalized thickness. As can be seen, the magnitude of the gravimetric sensitivity \( S_{grav} \) increases with increasing the layer thickness and reaches a peak after which, with increasing the guiding layer thickness, it decreases. The peak of the \( v_{ph} \) sensitivity decreases with increasing the mode order and the highest value corresponds to the LM1 mode; the \( v_{gr} \) mass sensitivity increases rapidly with the layer thickness, and it can be larger than the former as its peak can be sharper.

Both the LM’s group and phase velocity can represent a sensor response [56]; the phase velocity can be experimentally estimated by measuring the operating frequency \( f = v_{ph} / \lambda \) of the sensing device at the minimum insertion loss of the scattering parameter \( S_{12} \). The group velocity can be estimated by measuring the group time delay \( \tau = L / v_{gr} \) of the sensing device at the minimum insertion loss of the scattering parameter \( S_{12} \) in the time domain, where \( L \) is the acoustic wave delay path (the IDT’s centre-to-centre distance).

In the most general cases the LM devices consist of a semi-infinite piezoelectric substrate (for example, 41°YX LiNbO3, 36°YX LiTaO3 and ST-90°X quartz) [57] covered by a thin slowing layer (for example ZnO, Au, PMMA or SiO2), which traps the propagating wave to the surface of the substrate. The IDTs can be located only onto the piezoelectric substrate surface, under the overlayer, and thus they are isolated from the

![Figure 11](image-url). The derivative of the phase and group velocity versus the normalized layer thickness for the first five LMs travelling along the ST 90°-x quartz/SiO2 substrate.

**Table 2.** Some practical examples of LM sensors for application to liquid environments.

| Substrate      | Layer          | Application                        | Reference |
|----------------|----------------|------------------------------------|-----------|
| ST-90° quartz  | SiO2           | Mass sensing in liquids            | [58]      |
| ST-90° quartz  | PMMA           | Mass sensing in liquids            | [59]      |
| LiTaO3         | SiO2, ZnO, gold, SU-8 and parylene C | Comparison of electromechanical coupling coefficient, displacement profile and mass sensitivity | [60]      |
| ST-90° quartz  | ZnO            | Liquid viscosity and conductivity  | [61]      |
| ST-90° quartz  | PMMA           | Detection of high molecular weight targets in liquid samples | [62]      |
| 36°-YX LiTaO3  | ZnO            | Methanol in water                  | [63]      |
| 36°-YX LiTaO3  | ZnO            | Antibody-antigen immunoreactions in aqueous solutions | [64]      |
liquid environment. Table 2 lists some practical examples of LM sensors for application to liquid environments.

The LMs also propagate along a non-piezoelectric half-space (such as Si, glass, BN, a-SiC,...) covered by a piezoelectric layer (such as c-axis tilted ZnO or AlN) [56, 65–67].

For example, when the hexagonal ZnO film has its c-axis parallel to the substrate free surface, it is effective in the electroacoustic transduction of LMs in glass/ZnO substrates; when the c-axis is tilted at an angle $\mu$ with respect to the normal to the substrate surface, for wave propagation along the (100) direction, two types of surface modes propagate, the LM with predominant shear horizontal polarization, and the Rayleigh-like, with a prevailing sagittal polarization. Both modes are coupled to the electric field via the effective piezoelectric constants of the ZnO film. The LM and the SAW play different roles on the same sensing platform: the former is suitable for liquid environment characterization, while the latter is suitable for mixing and pumping small liquid volumes. LM sensors implemented on silicon or glass substrate materials offer the great advantage of the sensor’s integration with the surrounding electronic circuits.

For biosensing applications, a sensitive layer can be positioned along the acoustic wave propagation path: in this case the thickness of the sensing membrane must be properly designed so as not to perturb the wave propagation characteristics. Figure 12 shows the schematic representation of the Love wave sensor. The membrane can only cover the path between the IDTs or the entire wave path if the IDTs are buried under the guiding layer. In the latter case, the trapping layer must also satisfy the requirement of good chemical and mechanical resistance [53] as it has the additional role of shielding the IDTs.

The Love wave trapping layer can be a thin polymeric film such as polymethylmethacrylate (PMMA) [68], polyimide, SU-8 or polyethylene. C. In [59], the adsorption of h-IgG on the polymer surface was investigated by using a 1.23 $\mu$m thick PMMA guiding layer onto a quartz Love wave device, and finally the potential of the device as a biosensor was investigated by detecting the binding of anti-IgG.

The LM sensor based on a piezoelectric layer/non-piezoelectric substrate has a remarkable advantage over its counterpart based on the piezoelectric half-space/non-piezoelectric layer, as well as over PSAW and HVPSAW-based sensors: four coupling configurations can be investigated to enhance the $K^2$ and to take advantage of the protecting role of the guiding layer if the IDTs are buried under it. The IDTs can be positioned at the layer/substrate interface (substrate/transducer/film, STF) or at the layer free surface (substrate/film/transducer, SFT), with or without a floating metal layer onto the opposite surface of the layer (substrate/transducer/film/metal, STFM, or substrate/metal/film/transducer, SMFT), as shown in figure 13.

The $K^2$ of the LM device is affected by the mode order, the crystallographic orientation of both the half-space and layer, the layer thickness and also the coupling configuration (the electrical boundary conditions). As an example, figure 14 shows the $K^2$ dispersion curves for the first Love mode (LM1) in ZnO/glass for the four coupling configurations and different $K^2$ values for $h/\lambda = 0.3$ (0.4) for large tilt angles; the STF and STFM configurations reach their maximum $K^2$ values (~1.6% and 1.1%) for the 50° tilt angle.

As an example, figure 15 shows the $K^2$ dispersion curves for the first four Love modes (LM1, LM2, LM3 and LM4) in 90°-tilted c-axis ZnO/wBN, for the four coupling configurations.

With increasing the Love mode order, ever decreasing $K^2$ values can be reached by the four coupling configurations. The remarkable advantage of the LM-based sensors fabricated onto silicon is the possibility to integrate the sensor with other devices.

4. Shear horizontal acoustic plate mode sensors

SHAPMs are waveguide modes that propagate in finite thickness plates with energy distributed throughout the bulk of the waveguide. The SHAPMs are shear horizontally polarized ($U_1; U_3 = 0$), hence the absence of the out-of-plane displacement component allows each mode to propagate in contact with a liquid without coupling excessive amounts of acoustic energy into the liquid. Here, 3D eigenfrequency FE analysis was performed using COMSOL Multiphysics® Version 5.2 to explore the field shape of the SH0, SH1, SH2 and SH3 traveling along a GaPO4 piezoelectric plate, 150 $\mu$m thick, with Euler angles ($0^0, 5^0, 90^0$) and thickness-to-wavelength ratio $h/\lambda = 0.6$, as shown in figure 16. The GaPO4 material constants are those provided by Piezocrystal Advanced Sensors GmbH, which is an European GaPO4 wafers supplier [69, 70]. The SHAPM-based device employs input and output IDTs to launch and receive the acoustic wave, as for the SAW-based devices. The energy of these modes is distributed between the two plate surfaces as for a standing wave in a BAW sensor but the SHAPMs travel along the plate like a SAW. The continuous change of energy between the two plate surfaces allows the signal between the two IDTs to be affected by any changes in the surrounding environment that the opposite plate sides undergo. For liquid sensing applications the plate itself can be employed as a physical barrier between the electronics and the liquid environment to be sensed. The IDTs may be placed onto the surface opposite to the one in contact with the liquid solution, as shown in figure 17: the IDTs are naturally isolated.
Figure 13. The four coupling configurations for the non-piezoelectric half-space/piezoelectric layer structure. Reproduced from [56]. CC BY 4.0.

Figure 14. The $K^2$ dispersion curves for the first Love mode in ZnO/glass for the four coupling configurations and different ZnO $c$-axis tilt angles (from 10° to 90°). The colour of each curve represents a different $c$-axis tilt angle.
from the (potentially corrosive) liquid environment without adding any protective layer to the device surface, as for the SAW-based sensors, thus taking advantage of the entire sensor surface to maximize the interaction of the wave with the analyte. A metal film can be placed between the input and output IDTs to cancel any direct electromagnetic feedthrough. A sensing membrane may be attached to the upper side of the plate that is selectively sensitive to a specific measurand contained in the test liquid solution contacting the sensor. Any interaction (mechanical and/or electrical) between the measurand and the sensing membrane will cause a shift in the attenuation and/or velocity of the wave, which represents the sensor response.

The number of modes that propagate along the plate is dependent on the normalized thickness \( h/\lambda \) of the plate; the modes are excited at frequency \( f_n = v_n/\lambda \) where \( v_n \) is the velocity of the \( n \)th mode corresponding to the selected \( h \) value. As an example, figure 18 shows the phase velocity dispersion curves of the first six SHAPMs travelling along a \( \gamma \)-rotated GaPO4 plate with Euler angles (0° 1° 90°); the data were obtained using McGill software [54]. As can be seen, the fundamental mode, SH0, is a low-dispersive symmetric mode that travels at velocity equal to the transverse BAW velocity. The higher order modes can be symmetric and anti-symmetric. They are highly dispersive and their velocity asymptotically reaches the shear BAW velocity with increasing the plate thickness. They have a cut-off thickness: below the cut-off frequency, the mode becomes evanescent, i.e. the wavenumber is imaginary. Higher order modes can reach very high velocity as near the cut-off the slope of the dispersion curves is near to being infinite.

SHAPMs have maximum displacements that occur on the top and bottom surfaces of the plate, with sinusoidal variation between the two plate sides. The field profile of the first four SHAPMs (SH0, SH1, SH2 and SH3) in ZnO (0° 90° 0°) with \( h/\lambda = 0.5 \) are shown in figure 19. SHAPMs are divided into symmetric and anti-symmetric modes: for each mode the maximum displacement occurs on the top and bottom surfaces of the plate, allowing the use of either side of the plate for liquid sensing applications; and the number of zeros is equal to the order of the mode. The fundamental symmetric mode (SH0 in figure 19(a)) differs from the others in that the acoustic field is uniformly distributed along the plate depth.

Excitation of shear plate modes, showing dominant shear horizontal polarization, can be accomplished by tilting the \( c \)-axis away from the vertical by an angle \( \alpha \). Pure SHAPMs

![Figure 15. The \( K^2 \) dispersion curves for the (a) LM1, (b) LM2, (c) LM3 and (d) LM4 modes in 90°-tilted \( c \)-axis ZnO/wBN, for the four coupling configurations. Reproduced from [66]. CC BY 4.0.](image)
exist on 90°-x-propagating rotated y-cuts of trigonal class 32 group crystals, which include the GaPO₄ and the quartz crystals [72, 73], and in x-propagating rotated y-cut hexagonal plates, such as AlN or ZnO. Figure 20 shows the rotated crystallographic system of the piezoelectric plate.

The tilt angle $\alpha$, as well the plate thickness, affects the phase velocity and hence the $K^2$. As an example, figures 21(a) and (b) show the $K^2$ of the two coupling configurations, ST and MST, on ZnO versus the normalized plate thickness, where the tilt angle $\alpha$ is the running parameter: the data were calculated with McGill software [54]. ST stands for substrate/transducer and refers to the normal case where the IDTs are placed on one plate side, while MST stands for metal/substrate/transducer and refers to the previous configuration with the opposite surface covered by a floating mass-less, infinitesimally thin metal layer.

The viscosity sensitivity of the SHAPM sensors as well as the relative surface displacement (and particle velocity) increase with increasing mode order [1, 74, 75]. Figures 22(a)–(c) show the attenuation of the SH1, SH2 and SH3 modes versus the frequency thickness product for a glass plate immersed in ethylic alcohol ($\rho = 790$ kg m⁻³, $v_l = 1238$ m s⁻¹, dynamic viscosity = $1.2 \cdot 10^{-3}$ Ns m⁻², kinematic viscosity = $1.52 \cdot 10^{-6}$ m² s⁻¹), benzene ($\rho = 881$ kg m⁻³, $\eta = 1117$ m s⁻¹, dynamic viscosity = $0.65 \cdot 10^{-3}$ Ns m⁻², kinematic viscosity = $7.38 \cdot 10^{-7}$ m² s⁻¹) and kerosene.
(\(\rho = 822\, \text{kg}\, \text{m}^{-3}\), \(v_1 = 1319\, \text{m}\, \text{s}^{-1}\), dynamic viscosity = \(1.5 \cdot 10^{-3}\, \text{Ns}\, \text{m}^{-2}\), kinematic viscosity = \(1.82 \cdot 10^{-6}\, \text{m}^2\, \text{s}^{-1}\)). The modes are sensitive to the viscosity of the liquids, even when the mass density \(\rho\) and/or the velocity \(v_1\) are quite similar.

The sensitivity also increases as the device is thinned: the lower limit of the plate thickness is limited by production processes and plate fragility. Figures 23(a) and (b) show the attenuation and the phase velocity of the SH1 mode versus frequency for three different thicknesses (1, 1.2 and 1.4\(\text{mm}\)) for a glass plate immersed in kerosene: the curves move towards higher attenuation and velocity values with decreasing plate thickness. The data of figures 22 and 23 were obtained using Disperse software [76].

Martin et al [77] were the first to use the SHAPM device as a fluid phase sensor in 42.75° rotated Y-cut (RYC) quartz (ST-quartz): they experimentally verified the ability of the sensor to monitor the conditions at the solid/liquid interface. A bare quartz plate was used to measure the viscosity of water/glycerol mixtures, while the plate with the sensing surface chemically modified by ethylenediamine ligands was used to detect low concentrations of \(\text{Cu}^{2+}\) ions in solution. Following this paper, SHAPM sensors have been successfully investigated for many applications. Some non-exhaustive examples of applications include the detection of mercury contamination in water, with (sub)-nanogram sensitivity, by using ZLNO and −65° Y-rotated quartz plates covered by a gold sensitive membrane to accumulate the mercury via surface amalgamation [78]; the detection of potassium ion concentration in water with a relative frequency shift per unit potassium ion concentration was found equal to \(-8.37 \cdot 10^{-4}\) for the fundamental mode, using an ST-90° x quartz plate covered with a polyvinyl-chloride-valinomycin membrane [74]; the detection of the concentration of NaCl and tris(hydroxymethyl) aminomethane (Tris) in aqueous solution [79]; or to analyse the surface density changes associated with cell adhesion and proliferation in \textit{in vitro} conditions, using an STx quartz plate [80].

In [75] experimental results with various SHAPMs in an ST 90°-x quartz plate concerning the influence of temperature, viscosity and the concentration of NaCl and tris(hydroxymethyl) aminomethane (Tris) in aqueous solution are presented: the
higher order modes appeared to be more sensitive than the first ones, although they had more transmission losses.

5. Lamb wave sensors

Lamb waves (LWs) are elastic guided waves that travel in finite thickness plates, between stress-free planes and parallel boundaries; they are elliptically polarized in that they show in-plane and out-of-plane particle displacement components, \( U_1 \) and \( U_3 \). We remind the reader of the book by Victorov [81] for LW propagation details. LWs are divided into symmetric (\( S_n \)) and anti-symmetric (\( A_n \)) modes (where \( n \) is the mode order). The former modes have the longitudinal displacement component \( U_1 \) that is symmetric with respect to the mid plane of the plate while \( U_3 \) is anti-symmetric; the opposite happens in the case of the anti-symmetric modes. Figure 24 shows the total field profile and the single displacement components of the fundamental mode travelling along a Si plate.

The velocity of the modes depends on the plate characteristics (material type, thickness, crystallographic cut and wave propagation direction); the thicker the plate is, the more LW modes exist. LWs are highly dispersive: as an example, figure 25 shows the phase velocity \( v_{ph} \) versus the plate...
thickness-to-wavelength ratio curves of the symmetric $S_n$ (red curves) and anti-symmetric $A_n$ (blue curves) LWs travelling in a Si(001) plate of thickness $h$. The shape of the modes, the displacement components’ variation across the cross section of the plate, changes considerably with the plate thickness and with the mode order [82]. The insets of figure 25 show the mode shape of the first six modes travelling along the plate with fixed thickness ($h/\lambda = 0.5$); figure 26 shows the same $v_{ph}$ dispersion curves as in figure 25 but the insets are related to the shape of one mode ($S_2$) at different $h/\lambda$ (0.4, 1.0 and 1.8). The data of figures 25 and 26 were calculated using Disperse software [76].

As the LWs have velocity higher than that of the surrounding liquid medium and have both in-plane and out-of-plane displacement components, they are not suitable for sensing applications in liquids, except in some special cases. These cases include: (1) a branch of the fundamental symmetric $S_0$ mode dispersion region where the longitudinal particle displacement component, $U_1$, is dominant over the out-of-plane component $U_3$ at both the plate surfaces and in the plate depth (the mode is mostly linearly polarized and propagates at a velocity slightly lower than the velocity of the longitudinal bulk acoustic wave, $v_{LBAW}$); (2) a branch of the higher order symmetric modes dispersion curve, where the modes have $U_3 \approx 0$ at the plate surfaces (but not in the plate depth), and travel at velocity equal to $v_{LBAW}$; (3) a branch of the fundamental anti-symmetric $A_0$ mode dispersion curve, to which a velocity lower than that of the fluid corresponds.
5.1. Quasi-longitudinal symmetric modes

Of great interest are certain points of the symmetric LW dispersion curves where the phase velocity is close to the LBAW velocity of the plate material, \( v_{\text{LBAW}} \), and the field profile has particular characteristics, such as \( U_3 \ll U_1, U_2 \) at the plate surfaces. These waves, named quasi-longitudinal LWs (QL-LWs), are able to travel along the surface of the plate while contacting a liquid environment without suffering large attenuation. Inside a small branch of the \( S_0 \) \( v_{\text{ph}} \) dispersion curve, corresponding to \( h/\lambda \ll 1 \), \( U_3 \) can even have a constant amplitude along the whole depth of the plate, while \( U_3 \) is at least ten times less than \( U_1 \) at any plate depth [83, 84]: the shape of the membrane particle movement is a flat ellipse and its longer axis is parallel to the surface of the plate. The higher order symmetric mode dispersion curves intersect the velocity of the LBAW in the plate material (\( v_{\text{LBAW}} = 8440 \text{ m s}^{-1} \) for Si) and they show equal group velocity (\( v_{\text{gr}} = 7275 \text{ m s}^{-1} \)). Figure 27 highlights the intersection of the LW’s dispersion curves in a Si(0 0 1) \( \langle 1 0 0 \rangle \) plate with the plate material \( v_{\text{LBAW}} \).

As an example, figure 28 shows the field profile of the first four symmetric QL-LWs of figure 27: QL-\( S_0 \) (\( f = 0.441 \text{ MHz} \)), QL-\( S_1 \) (\( f = 5.85 \text{ MHz} \)), QL-\( S_2 \) (\( f = 11.70 \text{ MHz} \)) and QL-\( S_3 \) (\( f = 17.56 \text{ MHz} \)). For the QL-\( S_0 \) to QL-\( S_3 \) modes, \( U_3 \) is null only at the plate surfaces but, while the \( A_n \) curves are highly dispersive, the \( S_n \) modes show a flat dispersion region centred at the intersection point; this region corresponds to a \( h/\lambda \) range where the condition \( U_3 \ll U_1 \) is satisfied, thus preventing the sensor performances from being highly affected by possible errors in the fabrication technology of the sensor device.

As can be seen in figure 28, the through-thicknesses for \( U_1 \) and \( U_3 \) are symmetric and anti-symmetric about the mid plane of the plate, and the number of the minima increases with increasing the mode order. Since the \( U_3 \) component of these higher order modes vanishes on the free surfaces of the plate, these modes are suitable for liquid sensing.

As an example, figure 28 shows the field profile of the first four symmetric QL-LWs of figure 27: QL-\( S_0 \) (\( f = 0.441 \text{ MHz} \)), QL-\( S_1 \) (\( f = 5.85 \text{ MHz} \)), QL-\( S_2 \) (\( f = 11.70 \text{ MHz} \)) and QL-\( S_3 \) (\( f = 17.56 \text{ MHz} \)). For the QL-\( S_1 \) to QL-\( S_3 \) modes, \( U_3 \) is null only at the plate surfaces, but not inside the bulk of the plate; for QL-\( S_0 \) the \( U_3 \) vanishes on the plate surfaces and remains very small, even in the plate depth, while \( U_1 \) is almost constant through the plate thickness.

As can be seen in figure 28, the through-thicknesses for \( U_1 \) and \( U_3 \) are symmetric and anti-symmetric about the mid plane of the plate, and the number of the minima increases with increasing the mode order. Since the \( U_3 \) component of these higher order modes vanishes on the free surfaces of the plate, these modes are suitable for liquid sensing.

Figure 29 shows the \( v_{\text{ph}} \) and attenuation versus \( f \cdot h \) curves for the first four symmetric QL-LWs (\( S_0, S_1, S_2 \) and \( S_3 \)) for a Si plate immersed in water: when the velocity of the higher order modes reaches the \( v_{\text{LBAW}} \) (8440 m s\(^{-1}\)), the attenuation rapidly drops to zero, thus confirming the mode’s suitability to sensing applications in liquid environments. The dispersion curves of figure 29 were normalized with respect to the plate thickness by plotting them against the frequency thickness product. The fundamental mode QL-\( S_0 \) exhibits a plate normalized thickness value, \( h/\lambda_{\text{threshold}} \), beyond which \( U_3 \) is no more...
Figure 28. Cross-sectional normalized distribution of $U_1$ and $U_3$ displacement components in Si for: (a) QL-$S_0$ at $f = 0.441$ MHz, (b) QL-$S_1$ ($f = 5.8219$ MHz), (c) QL-$S_2$ ($f = 11.642$ MHz) and (d) QL-$S_3$ ($f = 17.537$ MHz).

Figure 29. The $v_p$ and attenuation versus $f \cdot h$ curves for the (a) QL-$S_0$, (b) QL-$S_1$, (c) QL-$S_2$ and (d) QL-$S_3$ modes travelling in a Si plate contacting a water half-space on both of the two plate sides.
negligible ($U_3 > 10\% \cdot U_1$ while $U_1 = 1$ at the plate surfaces but it is no longer constant inside the plate). For the $h/\lambda < h/\lambda_{\text{threshold}}$, the wave has $U_1 = 1$ and is constant along the plate depth, and the $U_3$ component is less than 10% of $U_1$.

The $h/\lambda_{\text{threshold}}$ has been calculated for several piezoelectric materials using McGill software [54] and the data are listed in table 3 [84]. For the higher order modes, the $h/\lambda_{\text{range}}$ values listed in table 3 have a different meaning with respect to QL-$S_0$: it is the plate thickness corresponding to the minimum value of $U_3$ at the plate surfaces ($U_3/U_1 \sim 10^{-3}$), for $v_{ph} \sim v_{LBAW}$. By varying the thickness of the plate around the $h/\lambda_{\text{threshold}}$, a $h/\lambda$ range ($h/\lambda_{\text{range}}$) can be found inside which the condition $U_3/U_1 < 0.1$ at the plate surfaces is verified. Table 3 summarizes the $h/\lambda_{\text{threshold}}$ and $h/\lambda_{\text{range}}$ of the fundamental and higher order quasi-longitudinal modes for some piezoelectric materials; the $K^2$ of the LWs have been evaluated for each material at the corresponding $h/\lambda_{\text{threshold}}$ for two coupling configurations.

The $K^2$ of the two configurations are quite different for the QL-$S_0$ modes, but they become very similar with increasing

| Material | QL-$S_0$ | QL-$S_1$ | QL-$S_2$ | QL-$S_3$ |
|----------|----------|----------|----------|----------|
| BN       | $h/\lambda_{\text{threshold}}$ | 0.325     | 1        | 1.97     | 2.95     |
|          | $h/\lambda_{\text{range}}$    | —        | 0.70–1.30| 1.665–2.26| 2.64–3.24|
|          | $K^2_{ST}$ ($K^2_{MST}$) (%)  | 0.09 (0.14)| 0.035 (0.04)| 0.018 (0.020)| 0.012 (0.013)|
| ZnO      | $h/\lambda_{\text{threshold}}$ | 0.07      | 0.65     | 1.24     | 1.86     |
|          | $h/\lambda_{\text{range}}$    | —        | 0.59–0.68| 1.17–1.305| 1.81–1.925|
|          | $K^2_{ST}$ ($K^2_{MST}$) (%)  | 0.47 (8.5)| 0.42 (0.50)| 0.22 (0.24)| 0.15 (0.16)|
| AlN      | $h/\lambda_{\text{threshold}}$ | 0.11      | 0.79     | 1.58     | 2.37     |
|          | $h/\lambda_{\text{range}}$    | —        | 0.75–0.89| 1.5–1.67 | 2.29–2.46|
|          | $K^2_{ST}$ ($K^2_{MST}$) (%)  | 0.35 (3)  | 0.31 (0.37)| 0.17 (0.19)| 0.11 (0.13)|
| GaN      | $h/\lambda_{\text{threshold}}$ | 0.12      | 0.77     | 1.53     | 2.29     |
|          | $h/\lambda_{\text{range}}$    | —        | 0.735–0.86| 1.51–1.62| 2.2–2.38 |
|          | $K^2_{ST}$ ($K^2_{MST}$) (%)  | 0.26 (1.61)| 0.18 (0.20)| 0.095 (0.1)| 0.06 (0.07)|

Figure 30. The field profile of the (a) q$S_0$, (b) q$L_1$ and (c) q$L_2$ modes in air. Reproduced from [85]. CC BY 4.0.
the mode order. The $K^2$ decreases with increasing the mode order, and the MST configuration is always more efficient than the ST: this last effect is particularly evident for the QL-$S_0$ mode, while the $K^2_{ST}$ and $K^2_{MST}$ values become similar with increasing the mode order. Due to the small thickness value ($h/\lambda_{\text{threshold}} \ll 1$) of the QL-$S_0$-based plates, the IDTs’ fingers are quite close to the opposite floating metal electrode for the MST configuration and consequently the electric field is mainly perpendicular to the plate surfaces. This results in a coupling efficiency that is quite larger than that of the ST configuration [84].

When LWs travel along a non-homogenous plate (e.g. a bi-layered composite plate), the symmetry of the particle displacement components with respect to the mid plane of the plate is lost, unlike the homogeneous isotropic and anisotropic plates. The modes can be considered as quasi-$S_0$ and quasi-$A_0$ ($qS_0$ and $qA_0$) for a limited plate thickness range, while all the other modes can be generically labelled as the $i$th mode. Unlike the single material plates, the $U_1$ and $U_3$ displacement components at the free surfaces of the composite plates can be quite different. As an example, figures 30(a)–(c) show the field profile of three quasi-longitudinal modes, $qS_0$, $qL_1$ and $qL_2$, traveling in an AlN(1.4 µm)/SiN(0.2 µm) plate: the corresponding SiN/AlN total thicknesses values $H_{\text{total}}/\lambda = (h_{\text{AlN}} + h_{\text{SiN}})/\lambda$ values are 0.08, 0.80 and 1.6. As can be seen, the condition $U_3 \ll U_1$ is verified on one plate side that is thus the one suitable for contacting a liquid environment [85].

Here, 2D COMSOL Multiphysics software was employed to simulate the $qS_0$, $qS_1$ and $qS_2$ mode propagation along the AlN(1.4 µm)/SiN(0.2 µm) composite plate while contacting the liquid (water) environment from the SiN side of the plate where the $U_3 = 0$ is satisfied, as opposed to the AlN side of the plate. As can be seen in figure 31, the acoustic energy remains confined inside the plate [85].

In [86] the sensitivity to liquid viscosity of the $qS_0$ mode in wz-BN/c-AlN thin composite plates is theoretically predicted.
for different layer thicknesses. As an example, table 4 lists the relative velocity shifts and the attenuation of the qS0 mode in a wBN/c-AlN composite plate contacting a liquid environment (70% of glycerol in water with \( \rho = 1091.6 \text{ kg m}^{-3} \), \( \eta = 0.003 \text{ Pa s} \), \( \lambda = 10 \) and 100 \( \mu \text{m} \)) for four different combinations of wBN and AlN thicknesses.

5.2. Fundamental anti-symmetric mode

Inside the LWs’ dispersion curves of figure 25, the \( A_0 \) mode is clearly identified by its reducing velocity as the plate thickness approaches zero. The \( A_0 \) mode, while being elliptically polarized, with \( U_3 \) not null at the plate surfaces, can travel along thin membranes that are in contact with a liquid if designed to travel at a velocity lower than that of most liquids, which lies in the range from 900 to about 1500 m s\(^{-1}\), by choosing the proper plate thickness. At very small thickness-to-wavelength ratios, the phase velocity of the \( A_0 \) mode approaches zero; as the thickness increases, the velocity increases, and reaches asymptotically from below the SAW velocity of the plate material.

The Scholte mode, not shown in figure 24, is an anti-symmetric mode that propagates at the solid-fluid interface: its name comes from its similarity to the Scholte wave that is widely known in geophysics. The characteristic equation for the dispersion curve of this mode is obtained as a solution to the equations of continuity of stress and displacement at the solid–fluid interfaces to be solved for anti-symmetric modes. In the low frequency limit, one solution is the \( A_0 \) mode and the other solution is the quasi-Scholte (Q-Sch) mode: their velocity dispersion curves have a linear dependence with the frequency thickness product. The dispersion curve of the latter mode is characterized by an asymptotic behaviour of the phase velocity approaching the sound speed in the fluid at high frequencies; this non-dispersive branch of the quasi-Scholte mode dispersion curve is named the Scholte mode. The polarization of the mode is mostly parallel to the interface with a small out-of-plane displacement component.

Figure 32 shows the phase velocity and attenuation curves versus the frequency for the \( S_0, A_0 \) and Q-Sch modes in a glass plate, 0.15 mm thick, immersed in water.

The \( S_0 \) mode attenuation starts from zero with the frequency and reaches a local maximum value at 2.68 MHz-mm. In the limit as \( f \cdot h \) tends to zero, the \( S_0 \) mode behaves basically as a longitudinally polarized wave, which explains its weak attenuation. When \( f \cdot h \) tends to infinity the attenuation increases asymptotically with \( f^2 \); the mode is essentially concentrated at the surfaces of the plate and it radiates energy into the surrounding liquid in the same manner as a Rayleigh wave, which is proportional to \( f^2 \). When the plate thickness becomes comparable with the acoustic wavelength, the \( A_0 \) mode behaves in the same way as the mode \( S_0 \) but it has an important attenuation in the low \( f \cdot h \) limit, caused by its flexural motion normal to the plate surface. The cut-off of the attenuation curve is at \( f \cdot h = 0.25 \text{ MHz-mm} \); below this limit, the phase velocity of the \( A_0 \) mode is smaller than the sound velocity in the liquid. As a result, no radiation of guided waves is allowed by the Snell law. The phase velocity of the Q-Sch plate mode rises with frequency from zero and gradually asymptotes to the velocity of the liquid half-spaces. Its attenuation is affected by the fluid bulk velocity, viscosity and bulk longitudinal attenuation. The Q-Sch mode travels attenuated in the direction of the wave-vector (if the fluid has no longitudinal attenuation); as it travels at velocity lower than the bulk velocity of the fluid, it is consequently evanescent in the direction orthogonal to the interface. The wave amplitude decays in an exponential manner with distance from the interface. The extent to which the wave penetrates into the fluid depends on the frequency, as shown in figure 33, where \( U_1, U_3 \) and the strain energy density (SED) of the Q-Sch mode travelling in a glass plate (1 mm thick) immersed in water at \( f = 107.602 \) and 454.186 kHz are shown.

The Q-Sch mode energy distribution between the fluid and the plate depends on the frequency, as shown in figures 33(a) and (b): the out-of-plane displacement component at 107.602 kHz is almost constant across the section of the plate and the SED indicates that the energy travels predominantly in the plate. At frequency 454.186 kHz a relevant part of the energy is travelling in the fluid. At higher frequencies (>1 MHz-mm) most of the energy travels in the fluid; the displacements decay away from the surfaces and are a minimum at the centre of the plate. The Q-Sch waves can be used to characterize the fluid properties [87] since the wave attenuation, phase and group velocity are affected by the viscosity, longitudinal bulk attenuation and bulk velocity of the fluid. In [88] the influence of the waveguide material (steel, aluminium and brass) on the Q-Sch mode phase and group velocity sensitivities to the liquid parameters (longitudinal velocity and density) is theoretically studied. The study concluded that higher waveguide material density leads to higher sensitivities, and higher waveguide acoustic velocities lead to an extended effective sensing range.

As an example, figure 34 shows the Scholte mode phase and group velocity dispersion curves in an Al plate (1 mm thick) immersed in water (\( \rho = 1000 \text{ kg m}^{-3} \), \( v = 1500 \text{ m} \))
s−1), benzene (ρ = 881 kg m−3, v = 1117 m s−1, dynamic viscosity η = 0.65 · 10−3 Ns m−2) and diesel (ρ = 800 kg m−3, v = 1250 m s−1).

In [89] the sensitivity of the quasi-Scholte mode for fluid characterization was assessed experimentally by measuring the phase velocity values for the quasi-Scholte mode in distilled water and in different ethanol-water concentrations. In [90] a preliminary sensitivity analysis is performed for application to simultaneous multi-sensing physical quantities of liquids, such as temperature, viscosity and density, using interface waves.

Figures 35(a) and (b) show the A0 and S0 mode velocity and attenuation dispersion curves in a Si plate (1 mm thick) immersed in glycerol (ρ = 1258 kg m−3, v = 1860 m s−1, dynamic viscosity η = 1.49 Ns m−2), water (ρ = 1000 kg m−3, v = 1500 m s−1), benzene (ρ = 881 kg m−3, v = 1117 m s−1, dynamic viscosity η = 0.65 · 10−3 Ns m−2) and diesel (ρ = 800 kg m−3, v = 1250 m s−1); the longitudinal attenuation is set to zero.

The A0 and S0 mode attenuation curves related to benzene and diesel are very close and difficult to distinguish; the same applies to the phase velocity curves but for all of the studied liquids. The A0 mode attenuation is larger than that of the S0 mode due to its faster decay over the propagation distance: the maximum attenuation (127.33 Nepers m−1) happens when f · h equals 0.15 MHz mm, when the A0 Lamb wave phase velocity is close to the water sound speed. When the A0 phase velocity is less than the water sound speed, there is still attenuation that approaches zero slowly as h/λ comes to zero.

In the low viscosity range, the amplitude response of the sensor is also affected by other parameters, such as temperature, pressure and density, which can play more important roles than the viscosity. In the case of water and diesel, the sensor responses to these two liquids are well distinguishable and are affected only by the mass density and velocity of the liquids, with the viscosity being assumed to be equal to zero.

On the contrary, the sensor responses to benzene and diesel, which have quite similar ρ and v, but different (and very low) η, are very similar.

The devices based on the A0 mode suffer some limitations, such as a low operating frequency (f = vph/λ) due to the vph of the A0 mode, which must be lower than the liquid velocity;
depends on the membrane thickness. The theoretical
lists the threshold plate thicknesses for operation in water
coupling structures: the ST and MST configurations. Table 5
piezoelectric plates (BN, ZnO, InN, AlN and GaN), for two
= 881 kg m
benzene (s
of the acoustic wave. The
structural quality imposes
plate; (2) if the sensor is implemented onto a thin suspended
substrate, the plate thickness must be scaled down together with
sensor is implemented onto a single crystal piezoelectric sub-
phases in between 295–312 m s
and an IDT periodicity of 100 µm. In this configuration, the
membrane normalized thickness is thin enough to obtain an
A
0 phase velocity lower than the sound velocity in water. For
h/λ = 0.06, they obtained A
0 and S
0 modes with velocities of
470 m s
−1 and 7850 m s
−1, respectively. The effect of viscous fluid loading on the A
0 mode is reported, which shows a linear relationship between the attenuation loss and the square root of the product of fluid mass density and viscosity. Moreover, the authors demonstrated that simultaneous measurement of frequency shift and attenuation loss allows a fluid’s viscosity and density to be determined.

In [93] experimental results on an A
0-based sensor on
PZT are described. The device was fabricated by deposition
of low-pressure chemical-vapor deposition (LPCVD) silicon nitride, 1 µm thick, a metal ground plane of Ta/Pt (10 nm/150 nm), and a 750 nm-thick layer of sol-gel-derived PZT on silicon wafer, followed by the lift-off process of Ta/Pt (10 nm/150 nm) IDTs. The KOH was used for anisotropic etching of the back side of the silicon wafer to release the composite membrane’s structure. They obtained A
0 mode phase velocities in between 295–312 m s
−1 and group velocities in between 414–454 m s
−1. A frequency shift of 850 kHz and an insertion loss as low as 3 dB are observed when the back side of the membrane is in contact with a column of 15 mm height of deionized water.

A theoretical study based on Rayleigh’s perturbation
approach to compare the A
0 and S
0 Lamb wave sensors’
sensitivity in liquid is reported in [94]: it is shown that sensitivity of the A
0 mode is much greater than that of the S
0 mode. In
[95] an experimental test of the A
0, S
0 and SH
0 sensors
demonstrates that the A
0 mode frequency shift caused by the
presence of liquid is quite larger than that of the S
0 and SH
0 modes.

The most recent experimental result of the S
0 mode to
measure mechanical and electrical liquid properties is reported by
Miera et al [96]: using two AlN-based S
0 sensor topologies, with and without a floating bottom metallic layer, the influence of mechanical and electrical properties of different aqueous mixtures was experimentally assessed.
6. Discussion and conclusions

All acoustic wave devices behave like sensors since they are highly sensitive to any change in the boundary conditions that result in a wave velocity and/or propagation loss change. If the acoustic wave path is covered by a sensitive coating that is able to absorb only specific chemical vapors or specific biochemicals in liquids, the sensor becomes a chemical sensor or a biosensor. All the acoustic wave sensors are able to work in gaseous environments, but only a subset of them can operate in contact with liquids. The waves that are predominantly in-plane polarized do not radiate appreciable energy into liquids contacting the device surface, as opposed to those waves with a substantial out-of-plane displacement component, which radiates compressive waves into the liquid, thus causing excessive damping. An exception to this rule occurs for devices utilizing waves that propagate at a velocity lower than the sound velocity in the liquid.

The quartz crystal microbalance (QCM) is one of the most common shear horizontal mode sensors: it typically consists of a thin disk of AT-cut quartz with parallel metal electrodes patterned on both sides. When a voltage is applied between these electrodes, the plate undergoes a shear deformation. The QCM resonant frequency, \( f = v_{BAW}/2h \), is inversely proportional to the quartz plate thickness \( h \) (few hundreds of micrometers): its value is typically between 5–30 MHz. When the QCM contacts a Newtonian liquid, it results in a resonance and handle. An increased QCM sensitivity requires a higher resonant frequency: this effect can be achieved by thinning the quartz plate, at the cost of a lack of substrate robustness. These sensors are more sensitive than the QCM, but less sensitive than the other surface-generated acoustic wave sensors since the energy of the wave is distributed throughout the bulk of the substrate and not trapped at the surface where the sensing phenomena take place. An advantageous feature of the SHAPM sensors is that these devices are sensitive on both sides of the substrate so that the back side can be used for sensing while the front is protected from the liquid [83]. This simple technology is opposed to that required by Lamb wave sensors, which requires the thinning of the bulk piezoelectric substrate or the release of a thin suspended membrane, or to that of the Love wave sensors, which requires the growing of a guiding layer on top of the piezoelectric half-space.

Thin piezoelectric suspended membranes are fabricated by employing silicon micromachining techniques. By using thin piezoelectric film technology, high-frequency devices can be designed that offer an important advantage: the surrounding electronic circuitry (amplifier) can be integrated with the acoustic device, thus offering the possibility for on-chip compensation of disturbing effects, such as temperature or pressure variations. The fundamental anti-symmetric Lamb mode travelling along a very thin membrane (thickness typically 5% or less of the wavelength) has a phase velocity lower than the sound velocity of the loading liquid \( v_l \) (typically from 900 to ~1900 m s\(^{-1}\)). Therefore, the plate functions as a wave guide yielding no radiation losses despite the fact that the wave has a non-null shear vertical particle displacement component. Due to the low phase velocity of the \( A_0 \) mode, the operating frequency is typically in the range of 5–20 MHz. The fundamental symmetric Lamb mode, \( S_0 \), is predominantly longitudinally polarized just for small plate thickness values: as its phase velocity is much higher than that of the \( A_0 \) mode, it can reach higher sensitivity.

The higher order quasi-longitudinal Lamb modes are promising candidates for liquid sensing applications: they have high velocity (close to that of the longitudinal BAW in the plate material) and require a fabrication technology simpler than that of the other Lamb wave sensors since they correspond to a thicker plate thickness-to-wavelength ratio. For example, \( h/\lambda = 0.07 \) is the upper limit of the ZnO plate thickness that allows the propagation of longitudinally polarized \( S_0 \) mode, with \( K^2 = 9\% \). Higher order quasi-symmetric modes \( S_1, S_2, S_3 \) and \( S_4 \) propagate for \( h/\lambda \sim 0.65, 1.24, 1.86 \) and 2.48. Their \( K^2 \) is equal to 0.42%, 0.22%, 0.15% and 0.10%, respectively, much lower than that of the \( S_0 \) mode. For a 10 \( \mu \)m thick ZnO membrane, the \( S_1, S_2, S_3 \) and \( S_4 \)-based devices’ operating frequencies are 394 MHz, 754 MHz and 1125 MHz and 1507 MHz, respectively, as opposed to that (38 MHz) estimated for the \( S_0 \) mode [98]. The thin suspended membrane devices may be only a few micrometers thick, thus the mass per unit area of the thin plate can be increased significantly by the mass-loading effect produced by the changes in the density of a fluid, or by the attachment of protein molecules, cells and bacteria from a liquid onto the suspended membrane surface. Moreover, these sensors can be fabricated by techniques compatible with planar integrated circuit technology, thus allowing low cost and small-sized sensor fabrication and the integration of the device with the surrounding electronic circuitry.

Love wave devices include a wave-guiding film deposited on top of a substrate [99]. The sensitivity of the Love wave
sensors depends on the layer and half-space materials type and their crystallographic orientation, and on the layer thickness. There is a relationship between the slope of the dispersion curve to the mass sensitivity of a Love wave sensor that allows one to find the layer thickness corresponding to the optimal sensor sensitivity. If the Love wave sensor consists in a piezoelectric crystal substrate properly rotated, the wave propagation mode changes from an elliptically polarized SAW to in-plane polarized SAWs: these are the PSAW and HVPSAW. This dramatically reduces the losses when liquids contact the propagating medium, thus allowing these devices to operate as biosensors. The PSAW and HVPSAW phase velocities are higher than the generalized SAW phase velocity, thus allowing the fabrication of higher frequency devices for the same photolithographic resolution, which also increases the sensor sensitivity. The PSAW and HVPSAW-based sensors have many advantages, such as a simple structure, highly reproducible large-scale fabrication technology and straightforward integration with microfluidic channels; moreover, their operating frequencies are higher than the QCM counterpart. However, a disadvantage of such devices is that the IDTs are located on the same side of the substrate that contacts the liquid: as a consequence, the test cell must have a reduced size and must be sealed within the propagation path.

Provided that the LWs, SHAPMs, the LMs and the PSAW-based devices are good candidates for biosensing and chemical sensing in liquids, it is worth emphasizing that the design parameters (such as materials type, crystallographic orientation, electrical boundary conditions, layers thickness, etc.) of any electroacoustic device highly affects the sensors performances. It has been proved that numerical calculations and FEM analysis yield an in-depth study of SAW characteristics to obtain a very useful description of the sensor’s performance and a deep assessment of the device sensitivity.

The present study wishes to point the reader’s attention towards the features of different types of acoustic waves and modes, as well as optimal combinations of materials and electrode structures, that can be exploited to design electroacoustic devices suitable for chemical and biochemical sensing applications. A promising application of SAW-based devices consists of a fully integrated platform that uses SAWs to develop several processes, from sample handling to detection and measure. SAW-directed transport of analytes can be coupled to the SAW sensing functionalities. The SAW microfluidic systems can move and remove liquid targets to and from different sensing modules on the same chip; SAW sensors can carry out analysis of fluids as well as separation and fractionation of cells. However, it is important to point out that future developments in acoustic-type chemical and biological sensors are also strongly related to the development of sensitive membranes, which are responsible for the selectivity of the sensors: with this aim, coordinated efforts between researchers in diverse disciplines (such as electrical engineering, physics, chemistry, microbiology and medicine) are needed.

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