Thermal and conductivity dependence of GaAs based acoustic biosensors

Thérèse Leblois
FEMTO-ST Institute, UMR 6174 CNRS
Université de Bourgogne Franche-Comté
Besançon, France
therese.leblois@femto-st.fr

Vivien Lacour
Interdisciplinary Institute for Technological Innovation
CNRS UMI-3463, Université de Sherbrooke
Sherbrooke, Québec J1K 0A5, Canada

FEMTO-ST Institute, UMR 6174 CNRS
Université de Bourgogne Franche-Comté
Besançon, France
vivien.lacour@femto-st.fr

***Abstract*—GaAs BAW sensors using shear acoustic waves were developed for biological detection and quantification. The sensor consists of a 50-350µm thick membrane where thickness shear acoustic waves were produced with a lateral field excitation (LFE). The LFE electrode arrangement is located on one side of the membrane. The other side is dedicated for proteins or cells capture through a bio-functionalized interface formed on the GaAs surface. Sensitivity of this type of sensor had been evaluated by modeling to 0.1ng.Hz^{-1}. Detection needs to be done in liquid environment with complex biological analytes such as blood or plasma. These analytes can inevitably induce temperature and surface charge variations on the material. Therefore, we have examined in the present work, temperature effect and conductivity influence of surrounding substrate or liquid on the GaAs-BAW sensor. These dependences on the resonant frequency are important features when designing GaAs ultrasensitive sensors. Frequency shift of shear mode induced by temperature deviation is reported at -58.6ppm/°C. Charge effects were measured by placing the resonant structure onto various bulk materials, thin films or liquids with varying permittivity and conductivity. Overall impedance module decreases when the device is on a highly conductive material, in same way resonant frequencies amplitudes are attenuated. The strong dependence of the sensor response on these parameters overcomes the known dependence on mass or viscosity. These studies give essential elements for the use of GaAs as a sensitive acoustic sensor in biological field.**

Keywords—GaAs Biosensors; acoustic transducer; lateral field excitation; temperature dependence; conductivity influence

I. Introduction

The piezoelectric biosensors are known for their high accuracy, low manufacturing cost and high level of miniaturization [1]. Moreover, gallium arsenide is interesting for MEMS applications thanks to its piezoelectric properties, its microfabrication opportunities and the various possibilities to biofunctionalize its surface [2, 3, 4]. For these reasons, we develop a specific GaAs acoustic sensor adapted for cells, vesicles or particles detection in biological media. Thickness shear acoustic waves were generated by lateral field excitation (LFE), this requires that the two electrodes are located on the same surface [5]. These sensors have often been used for detecting various chemical and biological analytes [6, 7]. Adhesion of biological elements can be monitored in terms of measured shifts in resonance frequency. The separated electrical part from the sensing surface makes this structure interesting for the monitoring of mechanical parameters (viscoelasticity, density) as well as electrical parameters (resistivity, permittivity). This multi-parameters sensing requires a thorough understanding of the mechanical, thermal and electrical contributions to the sensor response. Only a few studies on quartz BAW sensors tried to correlate physical properties with the impedance spectrum of the acoustic sensor [8, 9]. Some authors showed that specific designs of electrodes are required to reduce or increase sensitivity to electrical properties of liquids [10]. In this work, we investigate the impedance dependence due to the temperature deviation and the electrical conductivity and permittivity of various known surrounding substrates and liquids. We compare theoretical and experimental results. Finally, we evaluate the influence of electrical charges in liquids on the measured impedance spectrum by changing the concentration of NaCl in water.

II. GaAs transducer

A. Design of GaAs transducer

The geometry of the sensor presented in Fig. 1 was designed to facilitate analysis in liquid medium. The transducer structure consists of a resonant membrane excited by a lateral electric field that allows us to isolate the electrodes from the biological medium. The BAW resonator is excited on a thickness shear mode (TSM). An important advantage of shear vibrations is the limited propagation of waves into liquids, reducing the signal damping. Moreover, relatively high resonant frequencies are obtained with this mode of vibration, increasing the sensitivity of the device. This mode can be generated in the membrane for specific crystallographic orientations of substrate and orientations of electric field. Using an analytical model based on the Christoffel Beckmann method, we determined the best configuration to obtain the highest electromechanical coupling coefficient k. The (100) plane is preferred and the <110> electrode orientation allows us to reach a maximal value of k=6.7% [11]. This result is similar to those presented by Söderkvist [12]. The resonance frequency (f_R) depends on the membrane thickness as seen in (1):

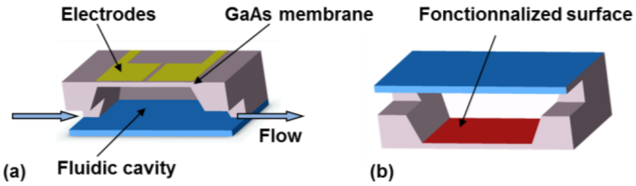

Fig. 1. Design of the lateral field excited GaAs structure: (a) top view (b) bottom view

$$f_R = \frac{n}{2t} v_b \left(1 - \frac{8k^2}{n^2\pi^2}\right)^2 \quad (1)$$

where n is the overtone, t the membrane thickness and v_b the velocity of the fast quasi transverse shear mode b.

The velocity v_b is a function of the elastic (c_{ij}), piezoelectric (e_{ijk}) constants and dielectric permittivity (ε_{ij}) as shown in (2):

$$v_b = \frac{\sqrt{c_{44} + \frac{e_{24}^2}{\varepsilon_{22}}}}{\rho} \quad (2)$$

where ρ is the density of GaAs crystal.

The physical properties of gallium arsenide are given in table I. Using piezoelectric equations and based on transmission lines and acoustic impedances theories, the Butterworth Van Dyke (BVD) model is used to transcribe the behavior of the mechanical resonator to an electrical model. Added mass on the surface shifts resonances (f_R) to lower frequencies and decreases also their magnitude. The mass-load dependence of the frequency can be used for molecules detection (such as quartz crystal microbalance). In that case, molecules of interest are immobilized on the sensing surface by a specific interface and quantified by frequency shift. For a solid film which has elastic properties close to the piezoelectric material, an additional mass induces only a shift of the resonance frequency. The magnitude is given by the Sauerbrey equation [13]. For a viscous mass, in addition to the frequency shift induced by the added mass Δm, a supplementary frequency shift occurs and the quality factor decreases. The value of the frequency shift Δf_R induced by viscosity is given in (3) by the Kanazawa and Gordon relation [14].

$$\Delta f_R = -f_R^2 \left(\frac{\Delta m}{A\rho v_b} + \frac{\sqrt{\eta \rho_L}}{\rho v_b \sqrt{\pi f_R}}\right) \quad (3)$$

where A is the area between electrodes, η the viscosity of the liquid and ρ_L the density of the liquid media.

The mass addition and viscosity modification are respectively represented in BVD model as an inductance and an inductance plus a resistance in series in the motional arm. As seen in table I, the material constants of GaAs are temperature T dependent. So, we can expect that the resonant frequency f_R which is expressed in terms of material constants and viscosity varies with T.

Moreover the modification of the conductivity of the liquid can affect the stability of the impedance spectrum. The aspect ratio between electrode gap distance (200 µm) and crystal thickness (70-350µm) is about 0.5-3. Smaller gap/crystal thickness ratio results in a higher electric field when TSM are generated.

TABLE I. PARAMETERS USED IN OUR CALCULATIONS

Properties		**Gallium Arsenide**	
		Values	***References***
Stiffness constants [GPa]	C_{22}	12.17-1.44 10^{-3}T	[15]
	C_{12}	5.46-0.63 10^{-3}T	[15]
	C_{44}	6.16-0.70 10^{-3}T	[15]
Piezoelectric constant [C/m^2]	e_{14}	-0.16	[16]
Permittivity [F/m^3]	ε_{22}	9.73 10^{-11}	[16]
Density [kg/m^3]	ρ	5.307	[16]
Velocity [m/s] propagation direction [100]	v_b	3347	[16]
Coupling coefficient	k	6.7%	[11]

Therefore the electric field is not completely confined between the electrodes. The electric field penetrates partly into the medium adjacent to the sensing surface of the crystal. This feature can provide access to additional relevant physical properties of the material under investigation, namely the electrical parameters permittivity and conductivity. As explained by Hempel et al the sensor response to electrical properties can be much larger than that to density-viscosity [8] or to an added mass.

Considering the fluidic system and modeling the force applied on membrane, we calculated that the highest sensitivity is equal to 0.1ng.Hz^{-1}.cm^{-2} for a 50µm membrane thick. The associated resonant frequency is f_R=28MHz (fundamental rank of vibration).

B. *Microfabrication of GaAs transducer*

Only wet etching and photolithographic techniques were used in the following process to provide highly reproducible way to fabricate the GaAs device without excessive costs. The semi-insulating GaAs(100) wafer (625±25µm thick) was etched to reach a membrane thickness which is comprised between 70 to 350µm. The anisotropic etching was performed by using orthophosphoric acid based solution. In accordance with previous reported results [17], 1 H_3PO_4 : 9 H_2O_2 : 1 H_2O solution at 0°C was used to provide flat and smooth surfaces. Electrodes were deposited on one face by sputtering chromium and gold layers. The details of this process are reported in [11] and the choice of wet etching baths and the etching conditions are detailed in [17]. Fig. 2 gives optical views of the membrane and electrodes.

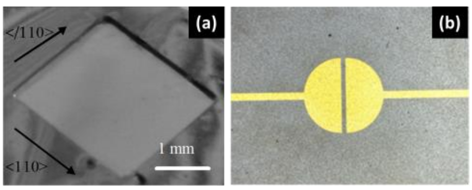

Fig. 2. Optical images of GaAs membrane (a) and electrodes (b)

III. Modelisation of temperature dependence

To determine the temperature sensitivity of our GaAs structures, we used numerical simulation. This was performed using a FEM analysis with COMSOL Multiphysics® software.

The geometrical parameters introduced in the model are indicated in Fig. 3a. The model uses a 70µm-thick membrane and a gap of 200µm between electrodes. GaAs crystal coordinates were rotated with α=45°, β=-90° and γ=-90° Euler angles. The model incorporated also mechanical (η_s=0.001) and electrical ($\eta_{\varepsilon S}$=0.01) losses in the piezoelectric material. Two perfectly matched layer (PML) domains were added to each sides of the resonator and were used to simulate the effect of propagation and absorption of elastic waves in these regions (which were not resolved in the true geometric scale).

Simulations of the GaAs-BAW transducer were considered in air. We know that the stiffness constants of the material are dependent to the temperature [15]. A parametric sweep simulation was performed to observe the impact of temperature over the fundamental mode. As shown in the absolute impedance versus frequency plot (Fig. 3b), the increase of the temperature shifts the resonant frequency to the lower values. The temperature shift is almost linear over the simulated range (20°C – 110°C). We can evaluate by numerical calculation, a frequency deviation of -59.3ppm/°C.

IV. Experimental results and discussion

The impedance measurements were carried out using an impedance analyzer HP4194 in Z probe mode and collected on a PC with a GPIB interface. The two electrodes of the transducer were connected to the analyzer through an electrical interface on PCB board with contact springs and SMA connectors. For testing, several structures with membranes of various thicknesses (from 70µm and 350µm) were made and controlled with an optical microscope. Electrical characterizations were done in air and in liquids with the impedance analyzer. All experiments have been performed at fundamental or third overtone frequencies.

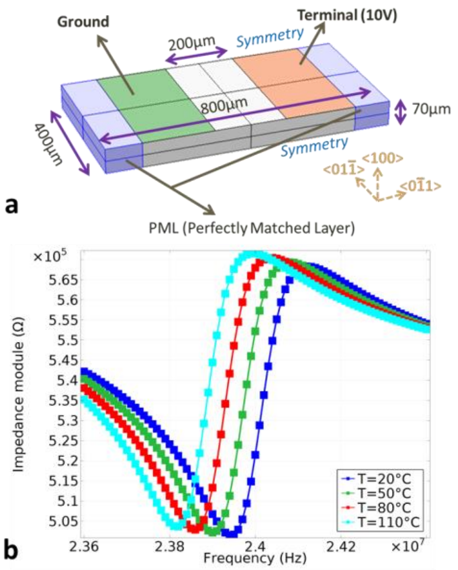

Fig. 3. (a) COMSOL's model geometries used for the GaAs-BAW modelisation and (b) absolute impedance (Ω) versus frequency (Hz) at various temperatures

A. Frequency-Temperature results

The samples were introduced in a regulated chamber. Fig. 4a shows the impedance amplitude and phase spectra when the temperature was fixed at 23.9°C. The experimental resonance frequency is f_R=20.13 MHz which is associated to the third overtone of the quasi transverse shear mode. The amplitude at the resonance frequency is 503Ω and the phase shift is 17°.

For frequency-Temperature measurements a linear temperature ramp is applied from 21°C to 62°C with a step of 4°C /min. Fig. 4b shows a linear behavior of the resonance frequency with the temperature. The slope is reported at -1179Hz/°C giving -58.6ppm/°C which is in fair agreement with simulations.

B. Experimental results on conductivity dependence in air

Experiments were conducted on two identical samples at a constant temperature of 21°C. The samples were placed onto various materials or thin films with different surface electrical conductivities. These substrates were previously characterized with a four probes conductivity meter. The substrates conductivity range was: 10^{-23} to 10^{7}S/m. Fig. 5a gives the evolution of the amplitude at the resonance with the conductivity. This curve confirms the results given by Hempel et al [8] which have disclosed a reduction of Q with an increase of conductivity at the interface of the active surface. We can see a marked attenuation of peak amplitude and followed by a decrease of the Q factor when the conductivity exceeds 10 S/m.

C. Experimental results on conductivity dependence in liquid

We evaluate the effect of liquid conductivity on the impedance spectra by injecting water with different NaCl concentrations at the vicinity of the sensing surface through a Teflon fluidic chamber. The reference frequency was obtained with DI water and salted water with added NaCl ranging from 0 to 1mol/l were used to evaluate conductivity impact of a liquid solution. According to Hasted et al [18], at low concentration C of the

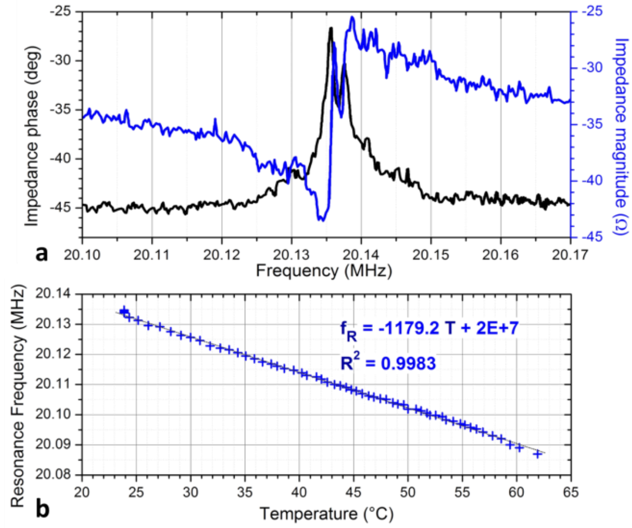

Fig. 4. (a) Impedance amplitude (red) and phase (blue) vs frequency; (b) Resonance frequency vs temperature plot

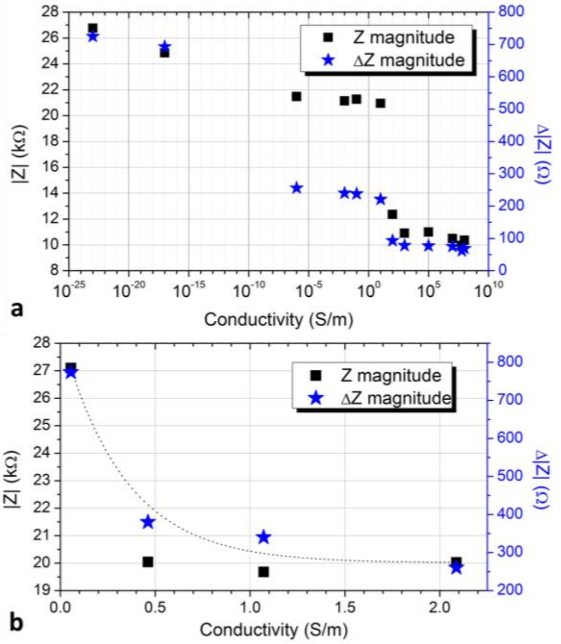

Fig. 5. Impedance values (black square) and peak amplitudes (blue star) at the resonance versus conductivity when sensing surface is in contact with (a) various substrates or (b) liquids

solution the conductivity σ follows the linear relationship σ=0.55+95C. The permittivity of the liquid doesn't change significantly. The results are given in Fig. 5b. The impedance decreases with σ and as measured in the case of the experiments in air on thin films samples the marked impact on the impedance and Q factor is reached for a few S/m conductivities.

V. CONCLUSION

We designed and fabricated GaAs BAW sensors using shear acoustic waves for detection and quantification of bacteria or vesicles present in low quantities in biological liquids. The sensitivity and specificity required for this detection involve a comprehensive knowledge of the sensor behavior. Indeed, the surface of the sensor may be in contact with liquid having different electrical conductivities and temperature. This study has proved that the frequency shift due to temperature changes is linear and then can be easily compensated using electronic circuits. However, we have shown that the change in conductivity of the liquid or the film in contact with the active surface induces a major attenuation of the impedance amplitude and a decrease of the impedance phase variation at the resonance. For liquid, the attenuation of the resonance peak increases with ion concentration which reduces the sensitivity of the device. The impact of electrical and mechanical properties of the liquid interfaces on the resonance has to be reduced for enhanced mass-sensing measurement. Differential methods of measurements may be conducted and new design and geometry of electrodes may be investigated in future work.

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