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# GaAs based on bulk acoustic wave sensor for biological molecules detection

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## Abstract

This paper investigates a high sensitive piezoelectric sensor in Gallium Arsenide (GaAs) crystal for biological molecules detection in liquid environment. The lateral field excitation was used to generate bulk acoustic waves (BAW) through (001) GaAs membranes. The crystallographic plane and the electric field orientation were chosen to obtain the highest electromechanical coupling coefficient for shear waves. Gallium Arsenide presents interesting alternative to quartz crystal concerning resonant biosensors thanks to its piezoelectric, acoustic properties, its ability to be directly bio functionalized and its common micro-fabrication processes. This offers an opportunity to combine a highly sensitive transducer fabricated using a batch process with a specific bio-interface which constitutes two essential parts of a biosensor. Analytical calculations and 3D finite element method (3D-FEM) analysis using Comsol Multiphysics® software were carried out to determine static, modal and vibration behaviors of the GaAs membrane. Typical numerical simulations and experimental results of the transducer in air were presented and discussed. The experimental results of GaAs-BAW in liquid for biological detection are also given in this study.

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**Keywords:** Gallium Arsenide; BAW; microfabrication; Biosensor

## 1. Introduction

Piezoelectric MEMS (Piezo-MEMS) technology can enable integrated solutions for a wide variety of applications including biomedical [1] and consumer electronics [2]. Piezo-MEMS actuators potential were studied for a long

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time, particularly in the field of sensors and actuators. In this context, the membranes are widely used together in various engineering applications such as the design of MEMS [3-7], Packaging [8, 9], ultrasonic [10], transducer and other acoustical applications (eg SAW and BAW) [11-13]. Thickness Shear Mode sensors (TSM) are widely used to monitor biological compounds in liquid environment which requires highly sensitive sensors. The commonly investigated bulk acoustic wave device is the Quartz Crystal Microbalance (QCM) commercially available. The Q-Sense E4 [14] of Q-Sense company is probably the most representative example of the effectiveness of that instrument. Poitras et al.[15] detects *Escherichia coli* bacteria with QCM-D technology of Q-Sense until  $3 \times 10^5$  bacteria per mL. However, quartz crystal cannot be processed by common microfabrication techniques as Silicon which is a major constraint for fabrication complex devices. Among the different piezoelectric materials, GaAs is the candidate that fulfills most of the requirements, from the integration through the functionalization [16] and including good mechanical piezoelectric properties [17, 18]. The integration of actuators and micromechanical systems with GaAs-based materials expands the design possibilities for integrated circuits and devices suitable for biosensing applications. In this study, a rapid and low-cost process was investigated [19] to enable large-scale development of piezo-MEMS. The transducer is intended for immunologic biorecognition thanks to the surface functionalization ability of GaAs. Indeed alkanethiols based architectures are widely used to immobilize antibodies on GaAs [20, 21].

In the present work, we propose a BAW sensor device for biological molecule detection which can provide a sufficiently high quality factor in air in large bandwidth resonant frequencies. This work gives new opportunities of cost-effective micro-sensors for biodetection applications.

## 2. GaAs structure for transducer

### 2.1. Principle of the device

The biosensor based on GaAs was designed to make it easier analysis in liquid environment. The device is composed of a resonant membrane where Thickness Shear Mode (TSM) of vibration was excited by electrodes placed on one face of the membrane (see Fig. 1). This is known as Lateral Field Excitation (LFE) which, unlike Thickness Field Excitation (TFE) is not affected by changes to the liquid electrical properties [22]. A low cost process based on wet chemical etching was investigated to enable widespread implementation of this kind of BioMEMS. A specific bio-interface was simultaneously investigated to achieve a specific bacterial detection.

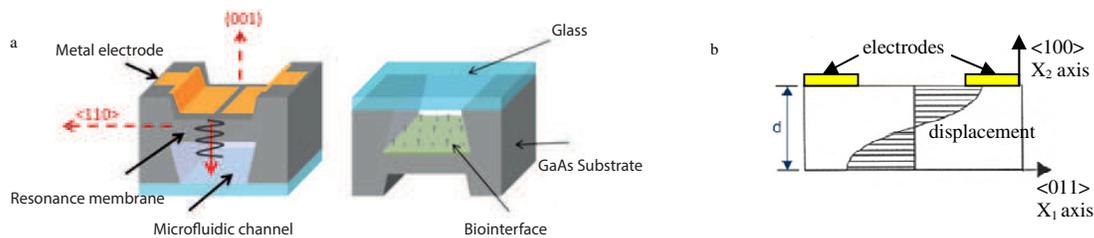


Fig. 1. (a) Experimental set-up based on biosensor in liquid environment, (b) thickness shear wave in the membrane.

With this structure the resonant frequency  $f$  is given by the following equation:

$$f = n \frac{V}{2d} \quad (1)$$

Where  $n$ ,  $V$  and  $d$  are the partial rank, the quasi-transverse mode velocity and the membrane thickness respectively.

The velocity is given by equation (2):

$$V = \sqrt{\frac{\lambda}{\rho}} \text{ with } \lambda \approx \bar{c}_{44} = c_{44} + \frac{e_{24}^2}{\epsilon_{22}} \quad (2)$$

$C_{44}$  and  $e_{24}$  are the elastic and the piezoelectric coefficients respectively.

Table 1 shows the properties of the GaAs piezoelectric substrate. The characteristics of this crystal are a dielectric constant of 12.9 ( $\epsilon_r$ ) and a loss tangent of 0.003 ( $\tan\delta$ ). These attractive properties make GaAs an increasingly popular choice in building BioMEMS devices.

Table 1. Properties of GaAs piezoelectric substrate [22].

Parameters	Symbol	GaAs
Loss dielectric	$\tan \delta$	0.003
Elastic value	$C_{44}$ (GPa)	59.4
Piezoelectric value	$e_{24}$ (C/m <sup>2</sup> )	-0.16
Dielectric relative constant	$\epsilon_r$	12.9
Density	$\rho$ (kg/m <sup>3</sup> )	3156

## 2.2. Process of fabrication

SI-GaAs (100) substrate was first cleaned in organic solvents: sequentially acetone and ethanol baths under sonication to remove most of organic contaminants. The wafer was immersed in ammonium hydroxide solutions (NH<sub>4</sub>OH, 30%) to remove native surface oxides and eliminate greasy residues and metal ion impurities [23]. This precleaning step produces a much more uniform surface and helps to make the future wet etching step more reproducible [24]. Ammonium hydroxide was preferred as other acid and base because it preserves a more stoichiometric surface [24]. The resonant element of the device is a thin membrane directly integrated inside the bulk crystal. The  $625 \pm 25 \mu\text{m}$  thick GaAs wafer was consequently etched to reach a membrane thickness which is comprised between 50 to 100  $\mu\text{m}$ . In order to obtain a flat and undamaged surface, an anisotropic wet etching was done by using orthophosphoric acid based solution. This solution provides smooth surfaces [19], [25] and gives a good control of etch depths at the level of tens angstroms[24]. According to previous studies in our group where several mixtures were tested [19], 1 H<sub>3</sub>PO<sub>4</sub> : 9 H<sub>2</sub>O<sub>2</sub> : 1 H<sub>2</sub>O solution at 0°C seems to give very smooth and flat membranes even for long etching process (etch rate : 0.95  $\mu\text{m}\cdot\text{min}^{-1}$ ). Acid solution was stirred during the process with a magnetic stirrer at 200 rpm to get an uniform etching on the whole wafer. A thick spincoated photoresist film (4  $\mu\text{m}$ ) was used as mask for this process. After etching, a positive photoresist was sprayed (thickness of 7  $\mu\text{m}$ ) to pattern Cr/Au electrodes and wires (400nm) deposited by sputtering. One of the delicate steps in the process is to ensure a reliable wiring of electrodes on the inclined side-wall of the membrane. Another longer etching step was performed on the bottom side using again spray coated photoresist. A membrane topside view and cross section was imaged by Scanning Electron Microscope (SEM) to measure accurately angles of planes involved in the cross sectional plane (100) (Fig. 2). And finally GaAs wafer is then bonded onto the Glass substrate into an EVG wafer bonding machine. For our experiment, this biological sensor was connected on PCB substrate by using the flip-chip techniques which are the most appropriate approach in set up testing device.

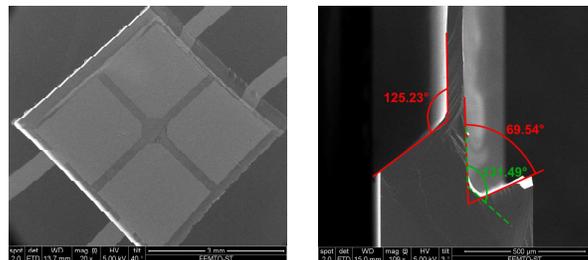


Fig. 2. SEM images of the micromachined GaAs membrane.

### 3. Experiments

We have tested the GaAs device with a vector network analyzer (ZNB 8, Rohde&Schwarz). At room temperature, the transmission coefficient ( $S_{12}$ ) of the fabricated GasAs device was measured to characterize the resonant characteristics. We have calibrated the network analyzer using a SOLT (short-open-50  $\Omega$  load-through). These experiments were performed in air environment. After that we performed some experiments in liquid media: water and a water glycerol-water mixture. Two volumes of the mixture were deposited on the membrane: 100 $\mu$ l and 200 $\mu$ l. The resonance frequency and the impedance value near the resonance were measured with the network analyzer HP 4395A with the kit 43961A.

#### 3.1. Measurements in air

In order to perform measurements in air, the GaAs structure was connected to a PCB as seen in Fig. 3. The GaAs devices have been tested and the results compared to the simulated ones. As shown in Fig. 4 a multiple resonances appear were simulated in a wide band that is the measured of the GaAs device in a frequency range from 10MHz to 100 MHz.

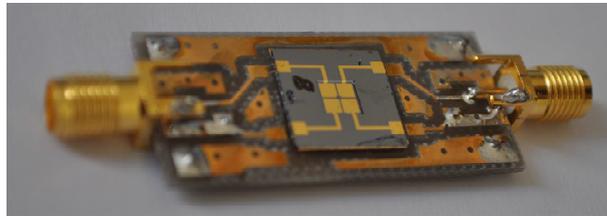


Fig. 3. Photography of the flip chip of GaAs sensor on PCB substrate.

An eigenfrequency analysis was performed to determine the first modes of vibration and the associated mode shapes as shown in Fig.4. The measured resonance frequencies and the associated mode of vibration are in close agreement with the theoretical analysis and finite element simulations (see Fig. 4 and Fig. 5). The frequency-quality factor products reach  $10^{11}$  which proves the potential of these devices to detect very low masses with a high sensitivity (several 100pg/Hz/cm<sup>2</sup> depending on the mode shape).

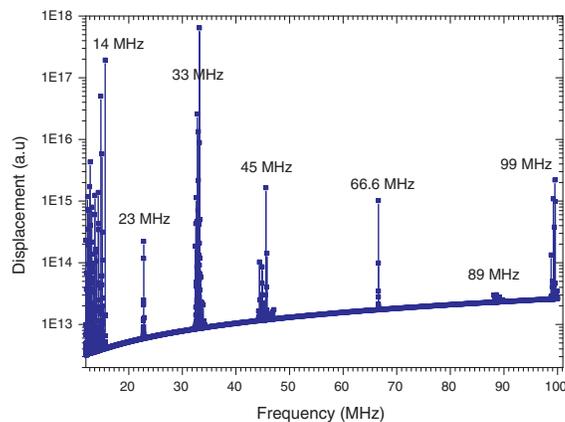


Fig. 4. Resonant frequencies responses of the GaAs membrane: simulated results

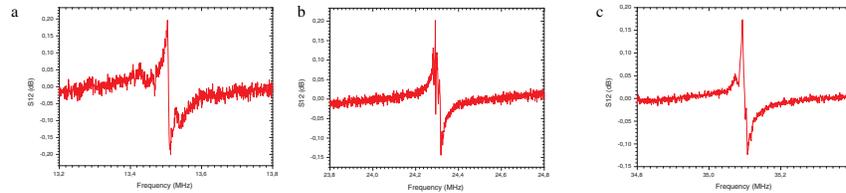


Fig. 5. S-parameter measurement of the resonance frequencies of GaAs structure (a) flexion, (b) torsion, (c) elongation.

### 3.2. Measurements in liquid media

Preliminary tests of mass measurements were then engaged with liquid to validate the functionality of the devices in these media. The liquid was deposited using a pipette to control the volume deposited on the sensitive membrane. Table 2 summary the main results obtained with this set-up.

Table 2. Results in liquid media compared to results in air.

Liquid composition	$\Delta f/f$ (ppm)	$\Delta C_0/C_0$ (ppm)	$\Delta R_m/R_m$ (ppm)	Q value
DI Water (200 $\mu$ l)	-12.1	-6.4	243	1200
Water+glycerol (100 $\mu$ l)	-4.2	-5.7	378	1100
Water+glycerol (200 $\mu$ l)	-14.7	-7.6	580	730

In this table,  $R_m$  is the motional resistance and  $C_0$  is the capacitance far from the resonance frequency. The experimental values seem consistent. The evolution of resonance frequency, motional parameter and capacitance are convenient with the expected values. The deposition of a thin film of glycerol on the sensitive membrane induced a large decrease in the admittance magnitude represented here by the resistance. The admittance amplitude at the resonance compared to the value in air (not presented here), is divided by a factor ten approximately. Similarly, the quality factor is strongly reduced. Considering now a thicker layer of the same glycerol solution, we can observe furthermore a decrease of the admittance amplitude and an increase of the peak width. These results are consistent with the theoretical considerations of the behavior of resonant bulk acoustic wave sensor when we put viscous layer on the surface.

### 4. Conclusion

This study presents provides a low cost manufacturing method of BAW biosensor using a piezoelectric substrate. These biosensors were developed and validated together by comparing to measured resonance frequency and the ones obtained by finite element simulation. The resonant frequencies of the fabricated sensor based on GaAs were measured in air and liquid. We have shown that the results given by simulation and experimental ones are in a relative good agreement. The next step of this work will be the validation of this device with biological liquid to detect and measure the concentration of specific biomolecules. With these features piezoelectric and this biocompatibility, the GaAs devices open large area applications. It is anticipated that the results presented here are useful and inspiring for bioengineers interested in the biomedical analysis.

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## References

- [1] G.-T. Hwang, M. Byun, C.K. Jeong, K.J. Lee, flexible piezoelectric thin-film energy Harvesters and nanosensors for biomedical Applications, *Adv. Healthcare Mater.* 4 (2015) 646–658.
- [2] O. Mareschal, S. Loiseau, A. Fougerat, L. Valbin, G. Lissorgues, S. Saez, C. Dolabdjian, R. Bouregba, G. Poullain, Piezoelectric aluminum nitride resonator for oscillator, *IEEE Trans on Ultrasonics, Ferroelectrics, and Frequency Control.* 57 (2010) 513–517.
- [3] E. Algre, G. Gaudin, A. Bsiesy, J.-P. Nozieres, Improved patterned media for probe-based heat assisted magnetic recording, *IEEE Trans Magn.* 41(10) (2015) 2857–2859.
- [4] G. Guisbiers, E. Herth, B. Legrand, N. Rolland, T. Lasri, L. Buchaillet, Materials selection procedure for RF-MEMS, *Microelectronic Engineering.* 87(9) (2010) 1792–1795.
- [5] E. Algre, B. Legrand, M. Faucher, B. Walter, L. Buchaillet, Surface microscopy with laserless MEMS based AFM probes, 23<sup>rd</sup> IEEE Int. Conf. on Micro Electro Mechanical Systems (MEMS) (2010) 292–295.
- [6] E. Herth, E. Algré, B. Legrand, L. Buchaillet, Optimization of ohmic contact and adhesion on polysilicon in MEMS–NEMS wet etching process. *Microelectronic Engineering* 88 (2011), pp.724–728.
- [7] A. Bongrain, E. Scorsone, L. Rousseau, G. Lissorgues, C. Gesset, S. Saada, P. Bergonzo, Selective nucleation in silicon moulds for diamond MEMS fabrication, *J Micromech Microeng.* 2009 Jul 1; 19(7):074015.
- [8] E. Herth, N. Rolland, T. Lasri, Circularly polarized millimeter-wave antenna using 0-level packaging, *IEEE Antennas and Wireless Propagation Letters.* 9 (2010) 934–937.
- [9] E. Herth, S. Seok, N. Rolland, T. Lasri, Wafer level packaging compatible with millimeter-wave antenna, *Sensors and Actuators A: Physical.* 173(1) (2012) 238–243.
- [10] L. Valbin, L. Sevely, Piezoelectric aluminum nitride thin film for ultrasonic transducer, MEMS components applications for industry automobiles aerospace and communication, *Proc. SPIE.* 4559 (2001) 95–102.
- [11] M. Vanotti, V. Blondeau-Patissier, J.-Y. Rauch, D. Rabus, B. Mazal, S. Ballandras, SAW sensor exploiting palladium layer properties for selective detection of hydrogen, in *European Frequency and Time Forum International Frequency Control Symposium (EFTF/IFC)*, 2013 Joint, 2013, pp. 775–778.
- [12] R. Tabrizian, M. Pardo, F. Ayazi, A 27 MHz temperature compensated MEMS oscillator with sub-ppm instability, *Proc. MEMS Conf.* (2012) 23–26.
- [13] O. Mareschal, S. Loiseau, F. Verjus, L. Valbin, G. Lissorgues, R. Bouregba, G. Poullain, S. Saez, C. Dolabdjian, Modeling and fabrication of piezoelectric Aluminum Nitride resonator and its application in oscillators, *Int. Solid-State Sensors, Actuators and Microsystems Conference.* (2009) 565–568.
- [14] Q-Sense, “Q-Sense E4.” [Online]. Available: <http://www.q-sense.com/products/q-sense-e4>. [Accessed: 02-Jul-2015].
- [15] C. Poitras and N. Tufenkji, A QCM-D-based biosensor for *E. coli* O157:H7 highlighting the relevance of the dissipation slope as a transduction signal., *Biosens. Bioelectron.* 24(7) (2009) 2137–42.
- [16] J.J. Dubowski, O. Voznyy, G.M. Marshall, Molecular self-assembly and passivation of GaAs (001) with alkanethiol monolayers: A view towards bio-functionalization, *Appl. Surf. Sci.* 256 (2010) 5714–5721.
- [17] K. Hjort, Gallium Arsenide micromechanics, A comparison to Silicon and Quartz, *Proc. GAAS symp.* (1994) 65–72
- [18] K. Hjort, J. Soderkvist, J. Schweitz, Gallium arsenide as a mechanical material, *J. Micromechanics Microengineering,* 4 (1994) 1–13.
- [19] A. Bienaimé, C. Elie-Caille, T. Leblois, Microstructuration of GaAs surface by wet etching: towards a specific surface behavior, *JNN,* 50 (2012) 6855–63.
- [20] A. Bienaimé, T. Leblois, G. Lucchi, V. Blondeau, P. Ducoroy, W. Boireau, C. Elie-Caille, Reconstitution of a protein monolayer on thiolates functionalized GaAs surface, *Int. J. Nanosci,* 404(2) (2012) 1240018.
- [21] V. Duplan, E. Frost, J.J. Dubowski, A photoluminescence-based quantum semiconductor biosensor for rapid in situ detection of *Escherichia coli*, *Sensors Actuators B Chem.* 160 (2011) 46–51.
- [22] A. Bienaimé, Microcapteur en arséniure de gallium pour la détection de molécules dans un fluide biologique, PhD report, UFC, 2012.
- [23] J.S. Song, Y. Choi, S.H. Seo, D.C. Oh, M.W. Cho, T. Yao, M.H. Oh, Wet chemical cleaning process of GaAs substrate for ready-to-use, *J. Cryst. Growth.* 264 (2004) 98–103.
- [24] A.G. Baca, C.I.H. Ashby, *Fabrication of GaAs Devices*, London: Institution of Electrical Engineers, 2005.