Novel Integrated FBAR Sensors: a Universal Technology Platform for Bio- and Gas-Detection

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Abstract

In this paper the feasibility of thin film bulk acoustic resonators (FBAR), for applications in bio- and gas-detection, is shown for the first time. Solidly mounted, ZnO FBARs with frequencies around 2 GHz have been fabricated on silicon substrates. The dependence of the FBAR mass sensitivity on the design of the layer stack has been investigated exhibiting an optimized sensitivity of 2.5 Hz cm^2/pg. Using a common protein assay the capability of detecting bio-molecules has successfully been proved. Gas sensing has been demonstrated by coating the FBAR with a humidity absorbing polymer. A strong non-linear dependence of the humidity sensitivity on the thickness of the polymer coating has been found. When the polymer thickness is far less than the acoustic wavelength, a pure mass dependent response occurs, leading to a negative shift in resonance frequency. Moreover, as the polymer thickness becomes significant, acoustic influences affect the response and the shift becomes large and positive. A sensitivity to humidity of up to two orders of magnitude higher than that of comparably coated quartz crystal micro-balances has been observed.

Keywords

FBAR-Sensor, Gas-Sensor, Bio-Sensor

INTRODUCTION

In the last few years thin film bulk acoustic resonators (FBAR) on silicon substrates have emerged as a possible substitute for surface acoustic wave devices (SAW) in radio frequency (RF) filter applications [6]. FBARS are compact and robust and can be integrated along with CMOS on silicon substrates [1].

Unfortunately compared to other piezoelectric resonator devices like quartz crystals or cantilevers, till now FBARS have not been considered for gravimetric sensor applications. Quartz micro-balances (QCM) have successfully been applied in gas- as well as bio-detection. But they are relatively expensive and suffer from poor integration into arrays. A lot of work has also been done on integrated cantilever based sensors [4]. In spite of being principally suitable for integrated sensors, difficult fabrication technology and the sensitivity to mechanical damage might hinder reliable operation and thus commercialization of this technology.

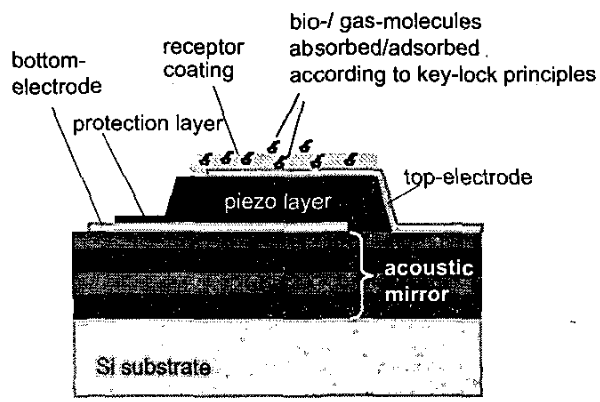

Figure 1. Schematic of the FBAR bio- and gas-sensor. The principle relies on a resonance shift caused by the attachment or absorption of molecules to a receptor coating covering the surface of a thin-film resonator.

Fig. 1 shows a schematic cross section of the novel FBAR sensor proposed in this work. The sensor consists of thin-film bulk acoustic resonators formed by a layer stack of bottom electrode, piezo-electric layer and top-electrode which is integrated onto a silicon substrate. The active vibrating region of the resonator is coated with a receptor layer which is sensitive to the adsorption or absorption of the molecules that are to be detected. The attachment of these molecules leads to an increase of the resonator mass load and thus to a frequency shift of the resonators resonance frequency which can electrically be determined using state of the art RF-circuitry.

Since the sensitivity i.e. the frequency shift per mass attachment of a single layer piezoelectric resonator is given by [5]

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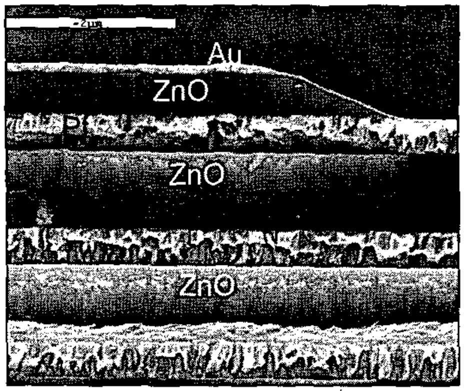

Figure 2. SEM cross section of the ZnO FBAR sensor. For acoustic de-coupling from the substrate a 3-fold ZnO/Pt mirror is employed.

$$s = \frac{\partial f}{\partial m} = \frac{f_0}{M} \propto -f_0^{\,2}, \qquad (1)$$

with f_0 the operation frequency and M the mass of the resonator, for FBARs typically operating in the GHz region, much higher sensitivities than for quartzes can be expected.

The FBAR sensor which can also be arranged into sensor arrays may operate in gaseous as well as liquid environments, making it an attractive device for a variety of applications in gas- as well as bio-sensing. For applications in liquids, to provide high Q-factors and thus low detection limits, the sensor needs to operate in the thickness shear-mode requiring piezo-layers with suitable orientation.

The aim of this paper is a first proof of the applicability of the proposed FBAR sensor for bio- as well as gas sensing.

DEVICE FABRICATION

Solidly mounted FBAR sensors with areas ranging from 50 x 50 to 500 x 500 μm^2 have been fabricated on 4'' silicon substrates (Fig. 2) employing reactive magnetron sputtering. The piezo-electric layer consists of a highly <002> textured reactively sputtered ZnO layer allowing the stimulation of longitudinal oscillation modes.

The 100 nm Au top-electrode provides a low electrical series resistance as well as a common chemical base for the binding of bio-receptor molecules. For the acoustic isolation from the silicon substrate a 3-fold ZnO/Pt mirror is employed. The quarter wavelength thick bottom electrode acts acoustically as an efficient reflection layer and ensures a high mass sensitivity (see results) as well as a low ohmic series resistance.

RESULTS AND DISCUSSIONS

Sensors have been characterized by standard RF s-parameter measurement techniques. All measurements have been done solely on wafer. To provide a determined atmosphere in gas sensing experiments, a plexiglass flow cell which could be hermetically sealed over the wafer in the active sensor region was employed.

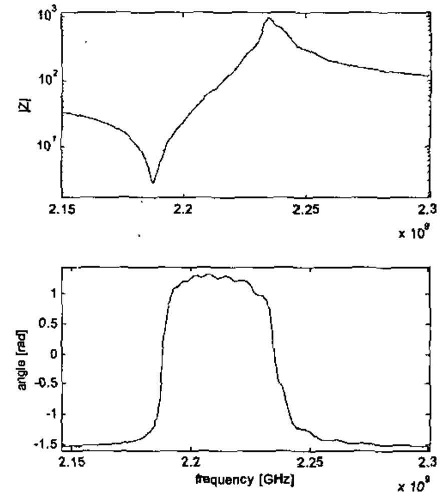

Figure 3. Measured impedance characteristics for a resonator of 70 x 70 μm^2 area exhibiting a Q-factor of more than 400 in both series and parallel resonance.

FBAR Characteristics

Fig. 3 shows the typical measured impedance characteristics for a FBAR of 70 x 70 μm^2 area with resonance frequencies around 2.2 GHz. By fitting the Butterworth van Dyke model, a Q-factor of about 400 has been extracted for both, the series and parallel resonances. Simulations using the Mason model combined with additional lumped elements reveal that the Q-factor is being limited by ohmic losses in the electrodes as well as by mechanical losses in the acoustic layers. Due to the thick bottom electrode the extracted electromechanical coupling coefficient k_{eff} of 19 % is likewise low.

FBAR Mass Sensitivity

Beneath the Q-factor one of the most important parameters determining the resolution of the FBAR sensor is the mass sensitivity s. Since the FBAR consists of an acoustic multilayer stack, the dependence of the mass sensitivity on the FBAR design i.e. the electrode thickness and electrode material has been investigated experimentally as well as by 1D-acoustic simulations using a chain matrix formalism.

Fig. 4 shows the variation of simulated and measured data for the mass sensitivity of 2 GHz FBARs with varying thickness of the bottom electrode as well as different top electrode material. For resonators with Pt top electrode the sensitivity has been determined by re-sputtering some material of the top-electrode and determining the change of resonance frequency versus the change in sheet resistance.

Results reveal good agreement between measurement and simulations. It is found that generally the sensitivity in-

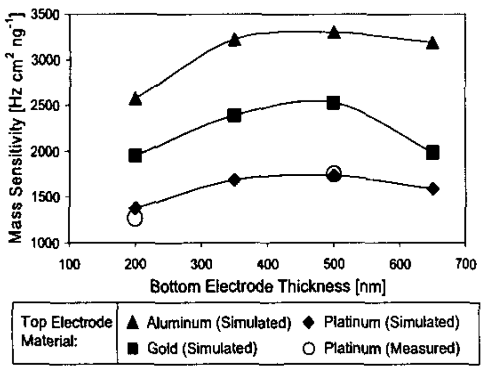

Figure 4. Variation of mass sensitivity with bottom electrode thickness and material of the 100 nm thick top electrode. Simulated and measured values for devices with Pt top electrodes show good correlation.

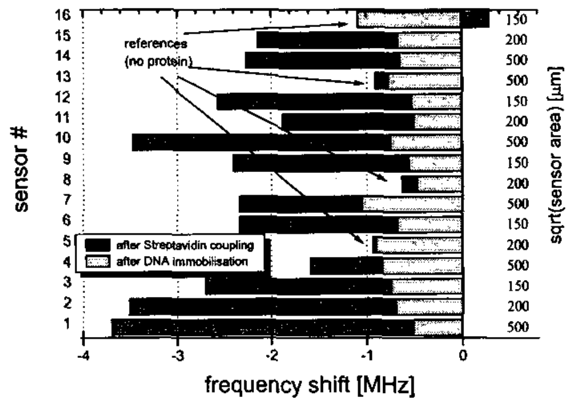

Figure 5. Measured frequency shifts at 25°C caused by the attachment of bio-molecules to FBAR sensors of different area, using biotin-labeled DNA oligos as a receptor and streptavidin as the target protein.

creases with decreasing density of the top electrode material which reduces the mass of the overall resonator.

With respect to the bottom electrode, the sensitivity reaches a maximum at a thickness of a quarter wavelength. We can qualitatively understand this behavior if we consider that the thickness of the bottom electrode changes the acoustic reflection at the interface between the piezo-layer and the electrode. Thus at a quarter wavelength, due to higher reflection, the acoustic wave is more confined within the piezo-layer region. Formally this is equivalent to a thinner resonator with reduced mass and thus resulting in a higher sensitivity (equ. 1).

For the FBARS investigated, simulations show a mass sensitivity of 2.5 Hz cm^2/pg, a value which is 2500 times larger compared with typical 20 MHz quartzes [3]. As long as the resonators are homogeneously coated, in a first order approach the sensitivity is independent of the sensor area. This shows up potential for further integration. Nevertheless for resonators much smaller than 20 x 20 µm^2 spurious modes appear. These modes leading to multi resonance characteristics of the FBARs may hinder the exact determination of the resonance frequency.

Protein Detection

To verify its use in bio-sensing, a receptor assay of Biotin-labeled DNA oligos coupled to the gold surface and Streptavidin as the target molecule, were used.

Resonators of different areas ranging from 150 x 150 µm^2 to 500 x 500 µm^2 have been coated on-wafer by dispensing 25 base-oligos with thiolalkyl groups in the 5'-position and Biotin in the 3'-position (5'-Thiolalkyl-ACC TCT TCT GGC TCA AAA AGA GAA T-3'-Biotin). Millipore-water was used as a solvent. The chemical reaction between the thiolalkyl groups and the gold surface was completed in a conditioning chamber. The wafer with the immobilised oligos was thoroughly washed with Millipore-water and dried with nitrogen.

In a next step Streptavidin, as the target molecule diluted in a saline sodium citrate (SSC) buffer solution, was dispensed on the oligo layer in nanoliter scale. The formation of the complex between Biotin and Streptavidin was completed in the conditioning chamber. The wafer was washed with SSC-buffer followed by SSC-buffer including detergent. Finally it was washed with Millipore-water and dried with nitrogen.

In Fig. 5 the frequency shifts with respect to the initial resonance frequency are shown. Both DNA attachment and even more so protein coupling result in clear signals. As expected, the resonator size was found to have no significant influence on signals. The large scattering of measurement data results mainly from variations in the various biochemical process steps. Furthermore intentionally untreated resonators show a small response, since experiments were performed on-wafer and sensors had to undergo some common treatment (washing steps).

Due to the oscillation mode of the resonator, these very first results have been achieved with dried sensors. Though these conditions are rather impractical for a real bio-sensor, they clearly show the capability of these new sensors for protein detection. The development of shear mode FBARs working in liquids will be the major challenge of further developments.

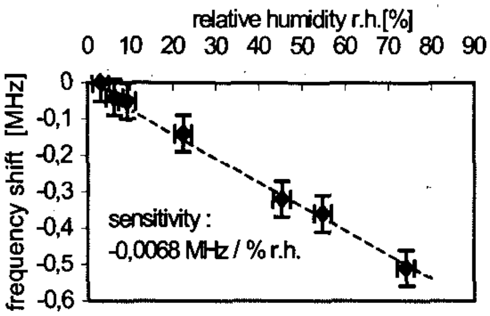

Figure 6. Frequency shift of an FBAR with 200 µm diameter coated with a 50 nm thick humidity absorbing polyimide.

Table I

Sensitivity, relative to resonance frequency, of the FBAR humidity sensor for different receptor thicknesses compared to a typical 5 MHz quartz crystal micro-balance.

Sensor Type	Nominal Frequency[GHz]	HC-Polymer Thickness [nm]	Sensitivity Relative to Resonance Freq. [$(\% \text{r.h})^{-1}$]
Quartz	0.005	640	$-4,0 \times 10^{-7}$
FBAR	2.0	650	$+2.7 \times 10^{-5}$
FBAR	1.6	280	$+6.3 \times 10^{-5}$
FBAR	1.6	140	-2.6×10^{-6}
FBAR	1.6	50	-2.4×10^{-6}

Demonstration of Gas Detection

To test the viability of FBARs for gas sensing, a number of wafers were spin-coated with a humidity absorbing polyimide from E+E Elektronik. The thickness was varied from 50 nm to 650 nm to investigate this influence. The polymer was then removed from the electrical contacts by laser to allow for on-wafer probing.

To deliver the gases directly to the sensor, a plexiglass flow-cell was lowered onto the wafer. The flow-cell has a neoprene seal to isolate the environment around the resonator which is being characterized. The coated resonators were each characterized once in a dry N_2 gas environment. A humid N_2 gas flow was then mixed with the dry N_2 to achieve variations in relative humidity, that has been measured at the flow cell exhaust. After letting the resonators stabilize for 15 minutes, they were characterized once again.

Figure 6 shows the frequency shift versus relative humidity for FBARs coated with a 50 nm polyimide layer. A nearly linear decrease in frequency with increasing humidity is observed. Here the resonance frequency shift is caused by an increase of the polymers' effective mass by absorbing water molecules. A sensitivity of 6.8 kHz / % r.h. has been extracted.

With increasing receptor thickness, the FBAR sensor response on humidity changes dramatically. While for polymer coatings up to about 150 nm a negative frequency shift occurs, for thicker layers a large and positive shift of the resonance frequency on humidity is observed. Comparison with typical coated 5 MHz quartzes (TABLE I) reveals a relative sensitivity which is for 280 nm coated FBARs at least two orders of magnitude larger than for the quartz micro-balance.

The observed thickness dependence of the sensitivity can be understood if we consider that a pure mass dependent i.e. gravimetric response of the frequency shift can only be expected for the attachment of a rigid mass or receptor layers with thicknesses much smaller than the wavelength. With a polymer density of 1500 kg m^{-3} supplied by the manufacturer and with values of 7.5 GPa for the E-modulus and a Poisson ratio of 0.35, both taken from literature [2], a wavelength of 1.04 µm has been estimated for the polymer layer at 2 GHz.

With increasing polymer thickness, the sensor and polymer coating have to be treated as an acoustic multi-layer system where the resonance frequency is not only affected by changes in the effective polymer density but also by changes in its acoustic velocity. If we assume that with increasing humidity the polymer becomes stiffer, as a consequence the resonance frequency increases.

With a 1D multi-layer model and fitting a linear increase of 4 % humidity from 0 to 100 % r.h., good agreement with the observed characteristics is achieved (Fig. 7). A linear increase in effective polymer density from 0 to 100 % r.h.

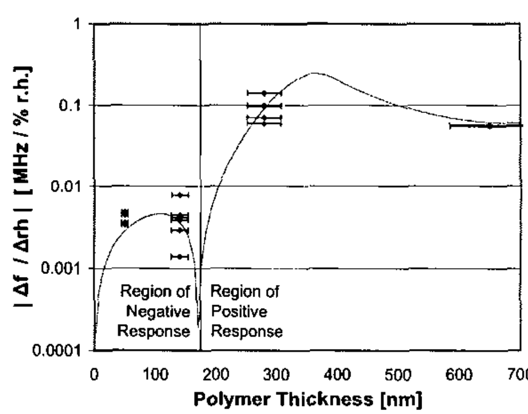

Figure 7. Measured sensitivity to relative humidity versus the thickness of a humidity polymer covering 1.6 GHz FBAR sensors, compared to simulation data (line).

has been modeled and set to 2.3 %, a value which has been determined from experiments on 5 MHz quartzes. Increasing the thickness of the sensitive polymer layer the sensor behavior changes from a pure gravimetric response to an acoustic response. At about 180 nm, the effects of density increase and the increase in acoustic velocity on the resonance frequency, cancel out.

It has to be noticed that for thicker polymer coatings the effect of humidity on resonance frequency is several orders of magnitude larger than would be expected from a pure gravimetric response increasing linearly with the layer thickness. Moreover the strong non-linear dependence on the layer thickness might potentially be used to discriminate between several analyte gases by using FBAR sensors arranged to an array and coated with polymer layers of varying thickness.

CONCLUSION

In this paper, to our knowledge for the first time, the capability of thin film bulk acoustic resonators on silicon for bio- as well as gas detection has been proved. For solidly mounted 2 GHz ZnO FBARs on silicon substrates a mass sensitivity three orders of magnitude larger than for typical quartz micro-balances and being strongly dependent on the design of the resonator stack has been found.

Employing a common Biotin-Streptavidin assay the detection of proteins with the new FBAR sensors has been demonstrated. Here the sensor operates according to gravimetric principles without using a label.

Employing a humidity absorbing polymer the feasibility of FBAR sensors for gas detection has successfully been proved. A strong non-linear dependence of the humidity sensitivity on the thickness of the receptor coating has been found. This is a consequence of the transition from a pure gravimetric response for thin polymer coatings to a more complex acoustic response for thicker polymer layers where the variations of polymer density and acoustic velocity with humidity have to be considered.

Presented results on bio- and gas detection as well as a robust technology promise, the presented universal sensor technology will become a genuine platform for bio- and smart gas sensor applications.

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