## Surface Acoustic Wave Studies for Chemical and Biological Sensors

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Abstract: Surface Acoustic Waves on piezoelectric substrates are very sensitive to any external modulation of the mechanical and/or electrical boundary conditions at the surface on which they propagate. This makes them a perfect tool for sensor applications. In this manuscript, we demonstrate that a sophisticated transducer design allows for a spatial resolution of the interaction of SAW and local modulation of the electrical and mechanical boundary condition. If such local disturbances of parts of the functionalized sample surface are due to a chemical or optical interaction, a single chip with many different 'pixels' can act as a novel type of sensor.

Keywords: surface acoustic waves, biosensors, biochips

Modern sensors are nowadays also meant to act as the 'interfacing link' between high performance electronic circuitry and the 'outside world'. More and more electronic systems are equipped with a whole variety of sensing abilities to make them able to react to and possibly interact with the environment. A good example is the automobile industry. Modern cars have the ability to "sense" their environment and to react accordingly. For instance, the windshield wipers turn on and off automatically depending on whether it is raining or not, the lights are automatically switched on if it gets dark, and the air conditioning system is able to not only adjust the right temperature within the car, it also "smells" the environmental air and reacts by adjusting the outside air supply accordingly. The engine and exhaust system is a very complex feed back mechanism, these days. Many different

parameters of the environment, driving behavior and street conditions are the input for an 'intelligent' modern (though at least upper middle class) car. Hence, sensor technology is not only a niche market. In the recent past, unfortunately, we constantly hear about the need for sensors that are able to detect chemical warfare reagents, biological substances, bacteria and other frightening material. Fortunately, however, this kind of sensors still remains a relatively small market. Much more important are those uncountable sensors and smart systems out there, of their ambiguous presence we sometimes not even know.

Exactly for this reason, it would be desirable to have a sensor system available, where many different environmental parameters can be determined at the same time. Let us compare it to the sensing abilities of a living organism: Most of them are able to "see", "hear", "smell", "taste", and "feel". These five senses have been developed during evolution and – apart from some species living under special environmental conditions – seem to be sufficient to satisfactorily react to the environment. If we try to categorize the senses into the framework of "sensors", we thus have an optical sensors (eyes), two types of sensors being sensitive to mechanical quantities (tactile sense and ears), and two sensors being sensitive to basically chemical reactions (nose and tongue). All the five sensor systems have in common that they not only consist of a single element but a usually large number of "channels" being able to differentiate different colors, sound frequencies, and chemicals.

In this article, we wish to describe the fundamentals of a sensor system that in principle is able to act as a simplified eye, an artificial nose, and even a tactile sensor for smallest forces employing the exact same basis technology in all cases. The sensor is electrically addressable and hence also fulfils the requirements to act as a link between an electronic circuit and the environment. The sensor principle is based on the interaction between surface acoustic waves (SAW) and an externally induced change of the boundary conditions which determine their wave equation. SAW are modes of elastic energy propagating at the surface of a solid. They usually have two components of particle displacement in certain directions with respect to the surface. Two of the simplest modes a called "Rayleigh-Wave", where the wave particle displacement as compared to the unperturbed surface is elliptically polarized with the two axes in the direction parallel to the propagation direction, and the one normal to the surface. Another simple mode is the "Shear-Wave", where the particle displacement is polarized along the two directions in the plane of the surface [1]. In Fig. 1, we schematically depict a snapshot of a Rayleigh mode. This decay gives the wave the name "surface-wave". The energy



flux in such waves is usually confined to a layer of approximately one wavelength thickness, and the decay is more or less exponential.

**Fig. 1.** Sketch of a Rayleigh-wave at the surface of an elastic solid. Note the decay of the wave amplitude into the depth of the substrate.

In contrast to the case of an isotropic solid, where all properties of a SAW are independent of the choice of the surface and propagation direction of the SAW, for anisotropic solids like semiconductor crystals, one has to include this anisotropy into the description of the SAW itself. For crystals with the lack of inversion symmetry (like the zinc blende lattice as in GaAs), additional effects arise from the piezoelectricity of such materials. Regarding such a piezoelectric crystal and ignoring free charges for a moment, the wave equation for a Rayleigh SAW is usually written in terms of a modified elastic constant  $c^*$ , taking into account the effect of piezoelectricity [2].

$$\rho \frac{\partial^2 u}{\partial t^2} - c^* \frac{\partial^2 u}{\partial r^2} = 0 ; \qquad c^* = c \left( 1 + \frac{p^2}{c \cdot \varepsilon} \right) \cong c \left( 1 + K^2 \right). \tag{1}$$

Here, *p*, *c*, and  $\varepsilon$  denote the components of the piezoelectric, the elastic, and the dielectric tensor. Usually, these material constants are combined into a single constant  $K^2 = p^2/c\varepsilon$ , describing the amount of piezoelectricity of the respective substrate. The effect of piezoelectricity hence slightly stiffens the substrate, leading to a somewhat higher sound velocity  $v = v_0 + \Delta v/v_0$ , being connected to the bulk coupling coefficient  $K^2$  via [2]

$$\frac{\Delta v}{v_0} = \frac{K_{eff}^2}{2} \cong \frac{K^2}{2}$$
(2)

To distinguish between the constant  $K^2$  used in eq. 2, and defining the piezoelectric stiffening in bulk material, the index *eff* is introduced for the effective electromechanical coupling to surface waves. Here, it is interesting to note that eqs. (1) and (2) basically describe the possibility to use SAW as a sensing element. All the quantities defining  $K^2$ , namely the piezoelectric, the dielectric, and the elastic tensor components can be slightly modified by the interaction with an external source, and hence modify the SAW propagation parameters. Usually, these are the attenuation  $\Gamma$ , and the renormalization  $\Delta v/v_0$  of the sound velocity. Both quantities can be read out and hence provide the sensor signal.



**Fig. 2.** Simple SAW delay line. In between the inter-digital SAW transducers, a functionalized and sensitized thin film is responsible for the interaction with an external parameter to be sensed. This interaction is detected by a change of the propagation parameters of the SAW.

Moreover, as the sensitivity usually strongly increases with increasing frequency, SAW are usually regarded to be superior to bulk crystal resonators like quartz micro balances, for example. The reason is that SAW can be excited employing planar metal electrode arrays, whose lateral spacing determines the resonance frequencies. Bulk resonators, on the other hand, rely on thickness vibration modes. Apart from some modern implications like "FBARs", fabrication processes and reproducibility restrict their application to rather low frequencies [3]. In a SAW, however, only a thin layer of the order of a wavelength is effectively oscillating and hence sensitive to external changes of the boundary conditions.

The simplest SAW sensor hence consists of a so-called delay line, where one transducer is used to excite a SAW, and another is used to detect the transmitted SAW after passing a sensitized area in between the

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**Fig. 3.** The principle of the tapered SAW transducer for sensing purposes. A varying distance between adjacent fingers provides a frequency dependent position of the launched SAW beam. In (b), we depict the frequency response of such a tapered transducer. In this case a split-4 design was used, resulting at four different bandpass regions at odd harmonics of the SAW mode.

two transducers. In Fig. 2, we depict such a simple sensor element. The sensitized area needs to be a functionalized region of the sensor chip, changing some of its properties under the influence of a sensor signal. This could be for instance a change of the conductivity [4] under illumination [5] or accumulation of a reagent, a change of the mass loading the chip, a change of the dielectric properties and alike. Based on this concept, a variety of sensors have already been described and even commercialized. Usually, each "channel" in these cases consists of a single SAW delay line, being more or less sensitive to a single ingredient of the analyte. To gain specificity, at least of order ten different sensors have to be combined to result in reliable, specific analysis of, say, a gas mixture. A more sophisticated SAW sensor scheme relies on the combination of gas

chromatography and a mass sensitive SAW delay line [6]. Here, the specificity of the chromatographic process acts as the different "channels". The SAW delay line only detects unspecific mass loading of the different ingredients of the analyte mixture, and the time sequence of the sensor signal results in the specific signal.



reconstructed position of a laser spot on the sample

**Fig. 4.** Spatially resolved perturbation of the electrical boundary conditions for SAW propagation on a semiconductor thin film. In this case, a laser was used to locally excite free carriers in the film which locally altered the sheet conductivity. This local conductivity change can be monitored by the spatially resolved SAW – thin film interaction as described in the text.

Here, we wish to describe a sensor element, which by a special design of the sound transducers allows for the parallel detection of many different ingredients in a gas mixture at the same chip. We therefore use so-called "tapered" transducers, where the applied high frequency signal is converted into a narrow SAW beam, propagating at different sound paths for different frequencies [7]. The basic idea behind such a "tapered transducer" is shown in Fig. 3, where we show a two-dimensional version of our sensor element. Both sets of transducers each define a bandpass filter, as shown in Fig. 3b. Once an external perturbation of the boundary conditions for SAW propagation is present on part of the active sensor area, a signal in either  $\Gamma$ 

(b) 10 µm 0.0 -1.0x10<sup>-2</sup> -2.0x10 4 i-Butan 333 K -3.0x10<sup>-2</sup> -4.0x10<sup>-2</sup> -20 0 20 40 60 80 100 120 140 160 time [s]

or  $\Delta v/v_0$  is observed in the respective bandpass, at a specific frequency which can be easily converted into the real space coordinate on the chip.

Fig. 5. SEM micrographs of a zeolite thin film (silicatlite-1), deposited on a sensor chip (top). In the bottom picture, we show the sensor response (SAW phase shift) for different i-butane partial pressures in a carrier gas.

To prove the concept of such a sensing element, we depict in Fig. 4 sensor being sensitive to illumination. This is accomplished be depositing a semiconducting thin film on the piezoelectric chip providing the SAW. Illumination creates free electron and holes in the semiconductor layer, thus increasing its conductivity, locally. This change in conductivity returns a



SAW signal according to eqs. (1), and (2), respectively, which can be used to reconstruct the position and intensity of the illuminated pattern on the chip (see Fig. 4). Even complex optical images can be reconstructed this way, by employing a tomographic technique [8].



Fig. 6. Sensitivity of eight different functionalized thin films for different gas mixtures. Note that basically each pixel is sensitive to all the three gases, the degree of sensitivity, however, strongly varies [10].

For chemical or biological applications, sensitivity to mass loading is sometimes an appropriate tool to detect specific substances. There are many different approaches for molecular specific capture functionalizations on such sensors [9], all of which have some pros and some cons. The major disadvantage of most of them, however, is the fact that only monolayer mass loading can be detected. Here, we wish to describe a novel type of mass loading functionalization, being molecular specific, and at the same time provide a large mass loading capacity. We functionalize the active surface of our chip by monolayers of nanocrystalline zeolithes with chemically adjustable pore size.

In Fig. 5, we show the micrograph of such a thin zeolite layer used for sensor purposes on our spatially resolving SAW chip. In this case, a silicalite-1 system has been used in which the pore size can be adjusted to a diameter of about 0.55 nm. In the lower panel of the figure, we show the response of the sensor for different butane-1 partial pressure in a carrier gas at room temperature. Many different sensitized functionalized thin films are presently under investigation, according to their specificity with respect to different gases. If such sensor "pixels" are deposited within the active area of a two-dimensional spatially resolving SAW sensor chip with two sets of tapered transducers, like the one described in Fig. 4, a very specific and

highly sensitive sensor for different gas mixtures and/or contents can be devised. For this purpose, an array of differently sensitized pixels (single sensors) is deposited in a checkerboard like manner in between the four senor transducers. The different pixels aught to have a different response for a given gas or gas mixture (see Fig. 6). The spatially resolving SAW chip employing the tapered transducers is the used to read out the accumulated sensor signal for a set of pixels in either a row or a column.



Fig. 7. Spatially resolving SAW sensor employing tapered SAW transducers as described in the text. The active area consists of an array of different sensitized pixels, each having a specific response (numbers on squares) to a given reagent. The SAW can read out the accumulated signal ( $\Sigma$ ) of, in this case three pixels at a time.

In Fig. 7, we depict the idea of such a sensor chip with many different pixels. Each pixel exhibits a specific sensitivity for a given gas mixture. This sensitivity results in a SAW sensor signal like attenuation and/or phase change. In the figure, we have denoted the sensor signal for a given gas mixture by the numbers superimposed to the pixels. The read-out SAW signal is then given by the accumulated signals for a single row or column, respectively. In the figure, we have denoted these accumulated signals by the sum sign and the arithmetic sum of the different rows and columns.

In Fig. 8, finally, we propose a display technique for such sensors, especially well suited for human inspectors. Humans are very good in pattern recognition, hence we convert the sum signals of figure 7 into a polar diagram, for instance, resulting in an easily recognizable pattern for a given gas mixture. A similar technique had been described in [6]. There, however, a gas chromatograph has been used as a sensor.



**Fig. 8.** Proposed readout scheme for the sensor depicted in Fig. 7. The accumulated sensor signals (S) are plotted in a polar-type diagram, providing an easily recognizable pattern for a human inspector.

In summary, we have described a highly sensitive sensor scheme for different external parameters. We use surface acoustic waves which can interact with such external parameters, altering the propagation parameters of the SAW. Such parameters may be conductivity changes due to illumination, adsorption, intercalation etc., or more direct measurements like mass loading of the surface. To increase the sensitivity for gas adsorption, we have used functionalized mesoporous zeolite thin films, which can be used as sensor pixels on our chip.

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