QCR-SENSORS –MODELS AND APPLICATIONS

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Abstract

Acoustic waves can be employed to measure physical or chemical values. Several kinds of devices have been used for generation and detection of acoustic waves and to pick up the relevant information. Examples are QCR-, SAW-, Lamb wave- or TFBAR-devices. The common aspect for all these devices is their sensitivity to any change of the acoustic properties of themselves or at the device surface. The quartz crystal resonator (QCR) is the most common device used as acoustic-wave based sensor. The CQR-sensor response is very influenced by the coating at sensor surface and its interaction with the environment. Models for describing this sensor behavior are introduced. Examples for sensor applications are shown.

Introduction

Bulk acoustic wave sensors are sensitive, to varying degrees, to perturbation of many different physical parameters, both intrinsic to the device and extrinsic. Thus they are used for the detection of mechanical quantities such as force, pressure or acceleration. Chemical/biochemical sensitivity is typically imparted by attaching a thin film to an acoustically active region, e.g. a region having significant acoustic amplitude, of the acoustic device. Acoustic sensing is only possible when this film or the adjacent medium interacts with the acoustic modes. Commonly, the increased mass density of the film, arising from species accumulation, is relied upon for a sensor response. Such behavior occurs in the so-called mass-sensitive devices. Changes in other parameters, including elastic, electrical, mechanical or other properties, can also considerably contribute to the sensor response. They can be described by the non-gravimetric effects. In last years different models were developed to understand these effects and their influence on the sensor response. The understanding of the transduction mechanisms of acoustic-wave-based sensors is crucial for a pronounced progress in their industrial and scientific applications.

The use of acoustic waves for sensor applications requires a device to generate the waves, a circuitry or instrument to operate the device, and methods to measure wave characteristics. Changes in wave velocity, frequency and/or amplitude indicate physical or chemical property changes occurring at the device surface. The typical acoustic wave device consists of a piezoelectric material with one or more metal transducers on its surfaces. These transducers launch acoustic waves into the material at ultrasonic frequencies, which may range from one to hundreds of MHz. The most commonly utilized acoustic wave resonator sensors are, the quartz crystal (QC)- or thickness shear mode (TSM)-resonator, the surface acoustic wave (SAW)-device, the acoustic plate mode (APM)-sensor and the flexural plate mode (FPM)-device. The acoustic waves generated in TSM-resonators are bulk transverse waves. The resonant frequency of the fundamental mode is typically between 5 and 100 MHz. SAW-devices use IDTs to generate Rayleigh waves whose energies are confined largely to a zone at the surface approximately one acoustic wavelength thick. Typical SAW sensor frequencies are 30-500 MHz. On thin plates, IDTs can generate two types of plate waves: Lamb waves and shear horizontal acoustic plate (SH-AP) waves. In a plate of finite thickness Lamb waves generated by an IDT give rise to a series of symmetric and asymmetric plate modes. Typical frequencies are 2-7 MHz. Characteristic APM-device frequencies range from 25 to 200 MHz. SAW-sensors of surface wave type can be used only in gas whereas the other may be applied in gas as well as in liquids. The sensitivity of all these acoustic wave sensors is strongly influenced by the resonance frequency. They are very sensitive sensors in comparison to many other sensor types.

In the following QCRs (quartz crystal resonator)s are considered more in detail as an example for manifold used bulk acoustic wave resonator sensors. Different models for describing QCRs are introduced. Selected application examples will be presented. Future trends for new sensor applications of bulk acoustic wave sensors are discussed.

QCR- general description

The quartz crystal resonator (QCR) is the most common device used as acoustic-wave based sensor. The simple geometry of the device and the predominant thickness-shear mode of the propagating wave are propitious conditions for a comprehensive derivation of the acoustic-electrical behavior of quartz crystal devices. Acoustic wave generation and propagation is most concise, therefore a coated QCR is used here as example to demonstrate the physical background of acoustic-wave-based sensors. Other acoustic microsensors have a more complicated wave propagation pattern; the concepts of coated piezoelectric resonator modeling are the same.

The analytical approach to describe acoustic wave generation and propagation in a QCR sensor is based on the linear piezoelectric equations together with Newton's equations of motion and Maxwell's equations. A chemical or biochemical QCR sensor can be understood as a multilayer structure of one piezoelectric layer and a certain number of non-piezoelectric. Different cases can be distinguished, e.g. a quartz crystal with a single film as the simplest example of a QCR gas sensor, or a quartz crystal coated with a single film, which is in contact to a liquid as a typical biosensor arrangement. The penetration depth of an acoustic shear wave into a liquid is very small, e.g. ≈250 nm in water at 5 MHz, therefore the acoustic wave decays before reaching the surface of the liquid and almost any liquid film can be treated as semi-infinite.

Acoustic waves are generated in the piezoelectric layer and travel back and forth in normal direction through the layers. Amplitude and phase of the traveling waves are defined by a set of acoustically relevant parameters. This set of characteristic parameters contains a geometric value, the film thickness, and material properties, for example film density and the (complex) shear modulus. The electrical response of such a composite QCR-sensor is however governed by the resulting wave in the quartz crystal. This wave is the superposition of the wave reflected off the boundary to the coating and the wave transmitted into the quartz crystal through this interface.

The coated QCR

The linear piezoelectric equations together with the resulting system of differential equations for the unknown mechanical displacement and electrical potential describe in general the behavior of a piezoelectric resonator. The full set of differential equations is, however, difficult to solve for the complete three-dimensional problem, because the unknown displacements and the electrical potential as well as their derivatives with respect to time and location are coupled with each other in these equations. For special geometries certain assumptions can be introduced, which enable an approximate two- or three-dimensional solution [1,2]. These models intend to characterize the *uncoated* quartz crystal but they cannot solve the problem of a *coated* resonator in general. Due to the high ratio between the lateral dimensions and the thickness of a typical quartz resonator it is reasonable to treat the crystal as an infinite plate with a finite thickness and to derive a one-dimensional solution for AT-cut quartz crystal resonators [3]. Consequently, it is assumed that physical properties do not change along the *lateral* directions. The derivatives for these directions vanish, and only the derivatives in normal direction remain.

The solution of the wave equation can be written with two components, namely two waves traveling in positive and negative direction inside the quartz crystal with two unknowns. The solution for the electrical potential, the stress and the electrical displacement introduces two other parameters. The relations for the coating (acoustic load) are similar to those of the quartz crystal. The coating is assumed to be non-piezoelectric, therefore there is no piezoelectric component, and consequently just two further unknown parameters must be introduced. Six boundary conditions provide six equations for the six unknown parameters.

The final solution of the one-dimensional problem can be calculated as follows:

$$Z = \frac{1}{j\omega C_0}\left\{1 - \frac{K^2}{\alpha}\,\frac{2\tan\frac{\alpha}{2} - j\frac{Z_L}{Z_{cq}}}{1 - j\frac{Z_L}{Z_{cq}}\cot\alpha}\right\} \tag{1}$$

Equation (1) uses the following abbreviations:

$$K^2 = \frac{e_q^2}{\varepsilon_q c_q},\ \alpha = \omega h_q\sqrt{\rho_q / c_q},\ Z_{cq} = \sqrt{\rho_q c_q} \qquad \text{(2 a,b,c)}$$

$$C_0 = \varepsilon_q \frac{A}{h_q} \qquad \text{(2 d)}$$

$$e_q \equiv e_{26},\ \ \varepsilon_q \equiv \varepsilon_{22},\ c_q \equiv c_{66} + \frac{e_{26}^2}{\varepsilon_{22}} + j\omega\eta_q \qquad \text{(2 e,f,g)}$$

K^2 is the electromechanical coupling coefficient of quartz, α is the acoustic phase shift inside the quartz crystal and Z_{cq} is the characteristic acoustic impedance of the quartz crystal, whereas Z_L is the acoustic load impedance acting at the surface of the quartz crystal. C_0 is the static quartz crystal capacitance. c_{66}, e_{26}, ε_{22} are the components of the material property tensors for mechanical stiffness, piezoelectric constant, and permittivity, respectively, the index has been replaced by q to denote material properties of the quartz crystal. Furthermore, a viscous term has been included with the phenomenological quartz viscosity, η_q. It is obvious from equations (1) and (2) that the electrical impedance of a coated quartz crystal resonator can be calculated from quartz crystal parameters and the frequency, leaving the acoustic load impedance, Z_L, as the only "foreign parameter".

The transmission line model (TLM) (Fig. 1) describes both the (piezoelectric) transformation between electrical and mechanical vibration and the propagation of acoustic waves in the system acoustic device-coating-medium in analogy to electrical waves [4,5]. This model also assumes a uniform piezoelectric device and isotropic, homogeneous, uniform layers and a sensor configuration, in which lateral dimensions have no effect on the propagation of waves. The model does not have any restrictions to the number of layers, their thickness and their mechanical properties.

One of the representations of the transmission line model is the equivalent circuit from Krimholtz, Leedom, and Matthaei, which is referred to as KLM-model [6]. It is presented in Fig. 1. The elements are defined by eq. (2 d) and eqs. (3).

$$jX = \frac{1}{j\omega C_0}\frac{K^2}{\alpha}\sin\alpha \tag{3 a}$$

$$\frac{1}{N^2} = \frac{1}{\omega C_0}\frac{4K^2}{\alpha}\frac{1}{Z_{cq}}\sin^2\frac{\alpha}{2} \tag{3 b}$$

The equations for the electrical impedance at port AB can be easily derived. The transformer turns the acoustic impedance at port CD into the measurable electrical impedance at port AB:

$$Z_{AB} = \frac{1}{j\omega C_0} + jX + \frac{1}{N^2}Z_{CD} \tag{3 c}$$

In the special case of a single-side coated sensor, i.e. a stress free surface at port GH and an acoustic load impedance, Z_{EF}, acting at port EF, eqs. (3) yield an equation equivalent to eq. (1) but with $Z_{EF} = Z_L$:

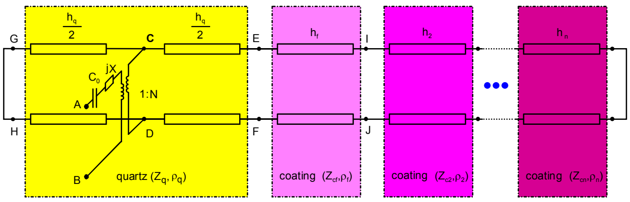

Fig. 1: The transmission line model in its KLM presentation

The transmission line model performs a formal separation of the acoustic wave propagation inside the acoustic device, including the transformation of mechanical displacement into the electrical signal and vice versa, and outside the acoustic device. In this context the acoustic load impedance, Z_L, plays the central role in the understanding of the transduction mechanism of chemical or biochemical acoustic-wave- based sensors. In case of a multilayer loading, the surface acoustic load $Z_L = Z_{EF}$ acting at the port EF of the transmission line representing the piezoelectric quartz crystal, would be the resulting impedance of all layers placed on the quartz surface. The acoustic load summarizes all acoustically relevant information. It does not play any role, if this load is generated by a simple mass, a single viscoelastic coating, a multilayer arrangement, or a semi-infinite material. Consequently, the acoustic load, Z_L, carries all information, which is related to changes in the (bio)-chemically sensitive coating, no matter if it is pure mass accumulation, mass accumulation accompanied by material property changes, or only material property changes induced by chemical (e.g. cross-linking) or physical (e.g. phase transition) effects. A change in the acoustic load impedance results in a change of the electrical impedance of the QCR. Finally these changes are responsible for the frequency shift and damping change of the acoustic device. The acoustic impedance transformation performed with layer i can be written as follows:

$$Z_i = Z_{ci}\frac{Z_{i+1} + j\sqrt{\rho_i G_i}\tan\left(\omega\sqrt{\frac{\rho_i}{G_i}}h_i\right)}{Z_{ci} + jZ_{i+1}\tan\left(\omega\sqrt{\frac{\rho_i}{G_i}}h_i\right)} \tag{4}$$

where ρ_i is the density, G_i is the shear modulus and Z_{ci} is the characteristic impedance of the i-th layer. The complex shear modulus $G = G' + jG''$ has a real part G' which is called shear storage modulus and accounts for acoustic energy storage. The imaginary part G'' is the shear-loss modulus corresponding to acoustic energy dissipation. The characteristic impedance is a material property:

$$Z_{ci} = \sqrt{\rho_i G_i} \tag{5}$$

It must not be mixed up with the acoustic impedance, Z_i, of the i-th layer giving finally the overall acoustic load impedance, Z_L (i = 1).

The equations in this chapter are exact within the one-dimensional assumptions. They should be used in all cases, where highest accuracy of the calculation is required and in all cases, where no information is available about error propagation [7]. Although they are easy to handle with a computer, their comprehensibility is limited. Therefore several approximations are applied to transform these equations into a more convenient form. An overview about the state-of-the-art is given in [8].

Experimental

Two different kinds of systems are used as sensor interface electronics for the determination of frequency, phase and damping changes of the QCR sensor. On the one hand, there are expensive, voluminous network analyzers, which are applicable for laboratory analysis and research. An alternative concept of a miniaturized network analyzer was shown in [9]. On the other hand, there are oscillator circuits with a quasi-digital output signal [10]. The application properties of both systems are compared. Selected examples of using QCRs for chemical and biochemical sensing are presented. Acoustic devices are now established in chemistry as one analytical method. The possibilities and limitations of using this transduction principle for a (bio)chemical analysis are determined by the sensor itself, the sensitive layer or adjacent medium and the sensor interface electronics.

The classical Sauerbrey-equation provided the theoretical base for most successful industrial application of QCRs up to now in the form of the DuPont humidity sensor system and in different monitoring systems for the observation of deposition processes, e.g. metal deposition. In the last years many attempts were undertaken to put new products on the market which use bulk acoustic wave sensors. Thus QCRs were applied in a few "electronic nose" systems, e.g. by the company APPLIED SENSOR (Swe). Fig. 2 shows a commercial QCR system for the investigation of liquids developed at the applied research institute "ifak" (Germany) [11,12].

Fig. 2: Commercial Liqui Lab System

Summary and Outlook

Bulk acoustic wave resonator sensors are widely accepted and used for chemical/ biochemical sensing. The better understanding of the interaction at the boundary sensor/ sensitive layer can enhance the quality of this sensor type and open new applications. That is why the basic theoretical relationships were introduced and discussed. Last years besides QCRs Love wave sensors, surface transverse wave (STW) sensors and guided layer APM sensors are more in the center of interest. Recently, the MARS (Magnetic Acoustic Resonance Sensor) system was introduced. TFBARs (thin film bulk acoustic resonators) are new competitors to the sensors mentioned above. A very interesting concept for using QCRs in biosensing was published in [13]. It was termed rupture event scanning (REVS). For the foreseeable future it is expected that bulk acoustic wave resonators in different versions have an increasing impact to measurement methods in laboratories and turn into industrial products in attractive niches.

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