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Coupled resonance in SH-SAW resonator with S1813 micro-ridges for high mass sensitivity biosensing applications

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ABSTRACT

The paper presents 3D finite element simulation and fabrication of SH-SAW resonator to investigate coupled resonance in the device with S1813 micro-ridges and its potential use as a sensitive biosensor. Polymer ridges of fixed width and various heights are designed along the wave propagation direction on the surface of the resonator. The coupled resonance effect showing a sharp decrease and an increase in the resonance frequency of the device is attained at a critical ridge height of 2000 nm and the same configuration is used for specific detection of biotin. The mass sensitivity of the device becomes maximum at coupled resonance along with an increase in the area-averaged stress at the interface between the ridge and the substrate. The S1813 ridges are silanized by APTES and the subsequent protein attachment is confirmed by confocal microscopy. Different concentrations of biotin solution are applied to the sensing and reference devices immobilized with avidin and BSA, respectively. The device offers high mass sensitivity of 6.4 kHz/(μg/mL) which is about three times greater than the mass sensitivity offered by a layered SH-SAW device.

1. Introduction

A surface acoustic wave (SAW) device consists of interleaved-electrodes known as interdigital transducer (IDT), lithographically patterned on the piezoelectric layer or a substrate. Application of an alternating voltage to the electrodes generates a periodic strain field at the surface because of the inverse piezoelectric effect that causes emission of an acoustic wave from either side of the IDT [1]. SAW devices are microelectromechanical systems (MEMS) that are widely used in the design of compact and inexpensive RF filters, correlators, analog signal processing units, and electronic communications systems [2,3]. SAW biosensor is an analytical device that comprises IDT as transducers and a chemically functionalized active area that selectivity and quantitatively detects a specific biological analyte [4]. SAW biosensors offer real-time, label-free detection with high sensitivity and selectivity, and are a promising low-cost alternative to the conventional fluoroimmunoassay, radioimmunoassay and surface plasmon based optical biosensing techniques [5]. SAW biosensors work on the principle of mass loading. Perturbation caused due to specific attachment of the desired bio-analyte on the active region of SAW sensor results in a shift in the frequency and phase of the device which is measured by a frequency counter or network analyzer [6].

Rayleigh wave devices which are mostly used for gas sensing applications are not preferred for biosensing in liquid media as Rayleigh waves due to their shear vertical particle displacements launch compressional wave into the liquid and get attenuated [7,8]. Thus, for biosensing in liquid, piezoelectric substrates such as 36°-YX $LiTaO_3$, 41°-YX $LiNbO_3$, and 90°-ST/AT quartz that generates shear-horizontal (SH) vibrations are required [9]. The SH wave consists of particle displacements parallel to the surface and perpendicular to the direction of wave propagation. Of all the SAW-based biosensing systems, the SH-SAW and the layer-guided Love wave (LW) devices are the most promising choices for biosensor design as they offer high sensitivity to mass loading on the surface and have been successfully used for the detection of various biological analytes [9–11]. Experiments related to biosensing with LW and SH-SAW devices for the detection of immunoglobulins [12,13], bacteria [14,15], viruses [16], and DNA [17,18] have also been reported. Various attempts to increases the mass sensitivity of SAW biosensors have been made by designing nano or microstructures on the active area of the sensor to increase the surface area for the analyte to attach. Gold nanoparticles, graphene oxide, nanostructured PMMA and ZnO nanorods have been reportedly used on the LW sensors for detection of various proteins and disease-causing pathogens [19–22].

The coupled resonance phenomenon was first modeled and explained by Dybwad using a composite system of QCM and micro-sphere of gold present on its surface [23]. Coupled resonance occurs when the resonance frequency of the sphere matches with the resonance

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frequency of the underlying substrate causing an unexpected increase in the resonance frequency of the device upon mass loading. The frequency shift due to the presence of spheres on the substrate surface is given by [24]

$$\frac{\Delta f^*}{f_0} \approx -\frac{\mathrm{n}m_s\omega}{A\pi Z_c}\frac{1}{\left(1-\frac{\omega^2}{\omega_s^2}\right)} \quad (1)$$

where $\Delta f^* = \Delta f + i\Delta\Gamma$ is the complex frequency shift, Γ is the half bandwidth at half-maximum, f_0 is the unperturbed frequency of the device, and Z_c is the acoustic impedance of substrate. The symbol n denotes the number of spheres present over an area A, and m_s is the mass of each sphere attached to the crystal with spring constant k_s. When spheres of small size are present on the surface of the substrate, their resonance frequencies are higher than the substrate ($\omega_s > \omega$) which causes a negative shift in frequency of the device due to inertial loading. However, if the size of the sphere is increased, a critical size is reached when the resonance frequency of the sphere becomes less than the resonance frequency of the substrate ($\omega_s < \omega$) which increases the contact stiffness, resulting in a positive shift in the frequency of the device due to elastic loading. Coupled resonance has been demonstrated in a QCM/PMMA micro-pillar configuration for biological detection in the presence and in the absence of liquid media [25]. The high mass sensitivity at coupled resonance is attributed to the increase in contact stiffness between nano/microstructure and the underlying substrate [24,26]. Investigation of coupled resonance by FE simulation of SAW device containing high aspect ratio palladium nano-pillars for hydrogen detection and experiments concerning coupled resonance with SU-8 micro-pillars designed on the surface of SAW resonators have also been reported [26,27].

Commercially available polymer photoresists such as PMMA, AZ 5214E, SU-8, and S1813 are often used for microfabrication process for designing required patterns on the substrate. Their surfaces can be chemically modified for biosensing by various methods [28,29]. The present work concerns with FE simulation and experiments of SH-SAW resonator composed of S1813 polymer micro-ridges designed on the surface of the device. The height of the ridges is varied by spin coating to attain coupled resonance in the device. The ridges not only act as guiding layer but also cause coupled resonance at a critical height. Such a configuration can be very sensitive to mass attachments and can become a highly sensitive biosensing platform.

2. Materials and methods

2.1. Device fabrication

The device fabrication process flow of the conventional layered SH-SAW device and the ridge-based device is illustrated in Fig. 1. A 36°-YX $LiTaO_3$ wafer is cleaned with acetone, IPA and DI water followed by spin coating with AZ 5214E positive resist (Microchemicals GmbH). The wafer is soft baked at 110 °C for 1 min and given a UV-exposure of 45 mJ cm^{-2} using EVG 620 mask aligner. The substrate is then developed in MF-26A solution (Dow Chemicals) to design the IDT pattern on the resist. A thin film of chromium (20 nm) followed by gold (100 nm) is RF sputtered on the wafer and lift off is performed in acetone to realize metal electrodes on the substrate. Fig. 2a shows the design layout of the resonator with gold electrodes of a width of about 5 μm. The device is expected to resonate at 200 MHz with λ = 20 μm. The electrode width and aperture of the IDT are kept at λ/4 and 120λ, respectively. IDT with 20 electrode pairs and reflector grating with 251 electrodes are used in the design. Fig. 2b shows the field emission scanning electron microscope (FESEM) image of the S1813 ridges designed on the resonator surface. The ridge pattern is made by keeping the S1813 resist coated wafer in Dilase 250 laser writer (KLOE, France). The devices are exposed to UV energy dose of 15%. S1813 micro-ridges of 10 μm width are made along the wave propagation direction on the device surface by developing with MF-26A solution. The variation in height of S1813 ridges is attained by varying the spin speed during spin-coating the resist, as per datasheet of the photoresist [30]. A width of 10 μm is selected considering the ease of fabrication, repeatability and size control of the ridges on the device surface using the laser writer.

2.2. Surface preparation and characterization

The surface modification and biofunctionalization process used is similar to as described in [29]. Once the ridges are designed, both the sensing and reference devices are cleaned using the UV-ozone technique to enrich the surface with –OH bonds. The devices are silanized by immersing them in 3% solution of APTES in toluene for 1 h. The devices are rinsed with toluene to remove any unattached APTES and dried on the hot plate. The reference and sensing devices were incubated with a 30 μL solution (1 mg/ml) of BSA and avidin, respectively for 1 h and rinsed with PBS. The protein attachment test on the device surface was done by attaching FITC-BSA on the sample and observing the fluorescence in confocal microscopy and surface morphology in FESEM. AFM images of the bare sample, after silanization and after protein attachment were also taken. Proteins like bovine serum albumin (BSA), fluorescein isothiocyanate (FITC)-conjugated BSA, biotin, and avidin from egg white were all procured from Sigma – Aldrich.

The devices after biofunctionalization were connected to a vector network analyzer VNA (ZVA24, Rohde & Schwarz) via an LC matching circuit using a variable series capacitor of 3–20 pF and a variable parallel inductor of 40–60 nH (Coilcraft Inc.). The matching circuit was used to match the input impedance of SAW device with the characteristic impedance of 50 Ω and minimize the input port reflections. The active area of both the sensing and the reference devices containing ridges were incubated with 30 μL of biotin solution of different concentrations (10, 50, 100, 200 and 400 μg/mL), and then washed with PBS and dried. Biotin selectively attaches on the avidin-functionalized sensing device and not on the BSA coated reference device, producing a frequency shift between the two devices. The slope of the graph of frequency shift versus concentration gives the mass sensitivity S while the limit of detection LOD = $3\sigma/S$, where σ is the standard deviation obtained in the frequency measurement of the reference device under stable conditions.

2.3. Simulation methodology

3D FE simulation of an SH-SAW resonator comprising S1813 ridges, operating at λ = 20 μm is carried out using COMSOL Multiphysics. Fig. 2c shows the simulation geometry of the resonator composed of 36°-YX $LiTaO_3$ substrate and S1813 ridges of width λ/2 made along the direction of wave propagation on the device surface. The ridges are equidistant with their centers λ apart from each other. Massless IDT electrode of width λ/4 is defined on the substrate surface. Periodic boundary conditions are applied along the x and z-axis to simulate one-full wavelength of the SH wave. The bottom surface is kept fixed. A rotated coordinate system with Euler angles (0°, −54°, 0°) is used to simulate the crystal with the correct orientation. An extra-fine mesh of tetrahedral elements is used to mesh the geometry that generates about 20,000 elements and over 120,000 solvable degrees of freedom during simulation. The variation in the frequency of the resonator with ridge height h_t is calculated by using the Eigenmode analysis as ridge height is varied from 100 nm to 3500 nm. The electromechanical coupling coefficient K^2 of the device is calculated from the free surface velocity v_f and metallized surface velocity v_m as per the relation [31]

$$K^2(\%) = 2\left(\frac{v_f - v_m}{v_f}\right) \times 100 \quad (2)$$

Using the small load approximation [24,32], the frequency shift due to coupled resonance can be written in terms of acoustic load impedance