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Viscoelastic Analysis of a Surface Acoustic Wave Gas Sensor Coated by a New Deposition Technique

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An analysis of the response of surface acoustic wave sensors coated with polymer film based on new coating deposition (self-assemble and molecularly imprinted technology) is described and the response formulas are hence deduced. Using the real part of shear modulus, the polymer can be classified into three types: glassy film, glassy-rubbery film and rubbery film. Experimental results show that the attenuation response is in better consistence with the simulation than in Martin's theory, but the velocity response does not accord with the calculation exactly. Maybe it is influenced by the experimental methods and environment. In addition, simulations of gas sorption for polymer films are performed. As for glassy film, the SAW sensor response increases with increasing film thickness, and the relationship between the sensor response and the concentration of gas is pretty linear, while as for glassy-rubbery film and rubbery film, the relationship between the sensor sensitivity and concentration of gas is very complicated. The ultimately calculated results indicate that the relationship between the sensor response and frequency is not always linear due to the viscoelastic properties of the polymer.

Key words: Surface acoustic wave, Gas sensor, Polymer, Viscoelastic property

I. INTRODUCTION

It is known that the detection mechanism of a gas sensor is the sorption of gas onto the coating film on the devices. Tiersten and Wohltjen reported the following equation for a mass loading effect of surface acoustic wave (SAW) [1,2]:

$$\Delta f \propto \rho h f^2 \quad (1)$$

Here, Δf is the frequency shift, f is the operating frequency, ρ and h are the density and thickness of the film respectively. Eq.(1) is based on the assumption that the film is thin and rigid, and the attenuation of the acoustic wave is not influenced by the mass loading effect. Now, a polymer is used for the coating film of the gas sensor due to its good selective detection of gases. However, the characteristics of polymer will be more complex when one considers many special factors, such as the kind of polymer, temperature, thickness, detection frequency, and gas sorption. In such case, it is necessary to consider the viscoelastic effect of the polymer.

If polymer is thin and rigid, there is no film-induced attenuation of acoustic wave during the film-deposition and gas sorption. However, this assumption isn't suitable to polymer film, for example, significant attenuation during gas absorption, and the the nonlinear SAW velocity with gas-phase concentration. In this case, the Tiersten model is inadequacies [3], and Wohltjen's theory and Auld's perturbation theories is not applicable [4].

Furthermore, previous papers usually concern about the viscoelastic properties of the polymer coatings on Rayleigh-SAW or SH-SAW gas sensor by the traditional film-deposition [5,6], such as LB technology, and so on. With the development of the sensitive film film-deposition technology, there are two new film-deposition technologies used for gas sensor, such as self-assemble and molecularly imprinted technology. Different from the traditional polymer film-deposition technology, new technologies need an active surface gold film between the sensitive film and substrate. Martin's theory can not directly be used to analyze the response of this sensor. In this paper a new method will be given concerning the viscoelastic properties of the polymer on an active surface gold film.

This work aims at simulating and analyzing the viscoelastic properties of polymer coatings on an active surface gold film. Originated from the methods of Martin and Auld's perturbation theory, a theoretical analyse is firstly extended to considering perturbation to the SAW by the polymer, and response formulas are deduced. According to the real part of shear horizontal modulus G , polymer films can be classified as glassy film, glassy-rubbery film and rubbery film. Then simulations of gas sorption for polymer films are performed and attention and velocity response are given. The theoretical results indicate that sensor attenuation is not always linear with frequency because of the viscoelastic properties of the polymer. The experimental samples covering gold film and polymer coating were tested. Although the experimental velocity response does not accord with the calculation exactly, the attenuation is better in agreement with simulation than Martin's theory. The deviation may be influenced by the experimental methods and environment. The sensitivity of the sensors covered with glassy film and rubbery film is also simulated when different concentration of gas is

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adopted. It shows that the sensor response is almost proportional to film thickness and the relationship between sensor response and gas concentration is pretty linear for the glass film, while it is very complicated for rubbery polymer film.

II. THEORY

Usually, the response mechanism of the SAW gas sensor by the new film-deposition can be calculated by the Farnell methods [7]. But it is very complicated as for polymer. In this paper, a new method is adopted to analyze the response mechanism. Based on the Martin method and Auld's perturbation theory, the perturbation to SAW by the gold film is analyzed and then extended to perturbation by the polymer coating on gold film respectively. So the process of the analysis becomes simple. Figure 1 shows the coordinate system used in this study. The SAW propagates to the x_1 direction. h is the thickness of gold film, $h_1 - h$ is the thickness of polymer film. Usually, ST-X quartz is used as the substrate for its good frequency temperature properties.

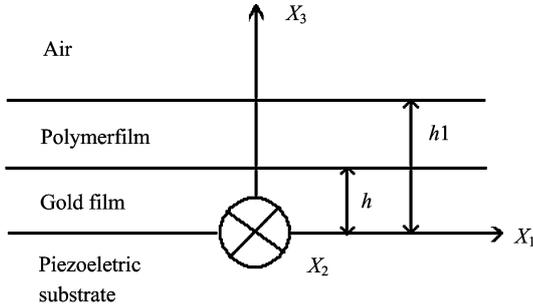


FIG. 1 The coordinate system used in this work

A. Perturbation to the SAW by the gold film

Usually the gold film can be regarded as thin and rigid, so the perturbation to SAW by the gold film can be approximately estimated by the velocity of SAW by the perturbation theory. Isotropic body just has only two compliance constants: c_{11} and c_{44} , which usually can be expressed by Lamé constants: λ and μ . In the perturbation analysis (appendix A), the change in SAW propagation velocity arising in gold film is:

$$V = V_0 \left[1 - \frac{c_1 \beta_1 \mu}{\omega} \tan(\beta_1 h) - \frac{c_2 \beta_2 \mu}{\omega} \tan(\beta_2 h) - \frac{c_3 \beta_3 (2\mu + \lambda)}{\omega} \tan(\beta_3 h) \right] \quad (2)$$

$$\beta_1 = \omega \sqrt{\frac{1}{\mu} \left[\rho - \frac{4\mu(\lambda + \mu)}{\lambda + 2\mu} \cdot \frac{1}{V_0^2} \right]} \quad (3)$$

$$\beta_2 = \omega \sqrt{\frac{1}{\mu} \left(\rho - \frac{\mu}{V_0^2} \right)} \quad (4)$$

$$\beta_3 = \omega \sqrt{\frac{\rho}{\lambda + 2\mu}} \quad (5)$$

V is perturbation velocity, ω is the angular frequency, ρ is the density of the gold film, V_0 is velocity of SAW. h is thickness of gold film, c_i ($i = 1 - 3$) are SAW-film coupling parameters. In addition, the perturbation to the coupling parameters by the gold film can be calculated by

$$c'_i = \frac{V}{\cos^2(\beta_i h) V_0} c_i, \quad (i = 1 - 3) \quad (6)$$

B. Perturbation to SAW by the polymer film

After getting the solution of the perturbation to SAW by the gold film according to Eq.(2) and Eq.(3), the perturbation to SAW by polymer film can be calculated by

$$\begin{aligned} \frac{\Delta\gamma}{k_0} &= \frac{\Delta a}{k_0} - j \frac{\Delta V}{V'} \\ &= j \sum_i^3 c'_i \frac{\alpha_i M^i}{\omega} \tan[\alpha_i (h_1 - h)] \\ \alpha_i &= \omega \sqrt{\frac{\rho - E^i/V'^2}{M^i}} \\ E^1 &= \frac{4G(3K + G)}{3K + G}, \quad E^2 = G, \quad E^3 = 0, \\ M^1 &= M^2 = G, \quad M^3 = K \end{aligned} \quad (7)$$

where $\Delta\gamma/k_0$ expresses fractional perturbation of the complex wave propagation factor, $\Delta a/k_0$ is the change in attenuation per wavenumber, $\Delta V/V'$ is the fractional change in propagation velocity. G and K are the shear horizontal modulus and the body modulus respectively; usually they are both complex, and their real parts represent the storage moduli, whereas the imaginary parts represent the loss moduli. Thus, the perturbation to Rayleigh-SAW by the polymer film can be written as follows:

$$\begin{aligned} \frac{\Delta\gamma}{k_0} &= j c_1 \frac{\alpha_1 M^1}{\omega} \tan[\alpha_1 (h_1 - h)] \\ &\quad + j c_3 \frac{\alpha_3 M^3}{\omega} \tan[\alpha_3 (h_1 - h)] \end{aligned} \quad (8)$$

whereas the perturbation to SH-SAW can be calculated by

$$\frac{\Delta\gamma}{k_0} = j c_2 \frac{\alpha_2 M^2}{\omega} \tan[\alpha_2 (h_1 - h)] \quad (9)$$

III. CALCULATION RESULTS AND DISCUSSIONS

A. Perturbation to SAW and SAW/film coupling parameter by gold film

Figure 2 and Figure 3 show the calculated results according to the Eq.(2) and Eq.(3), respectively. Figure 2 presents the perturbation to SAW velocity by the gold film, and it shows that the velocity shift decreases linearly with film thickness. While Fig.3 shows the perturbation to SAW/Film coupling parameter by the gold film.

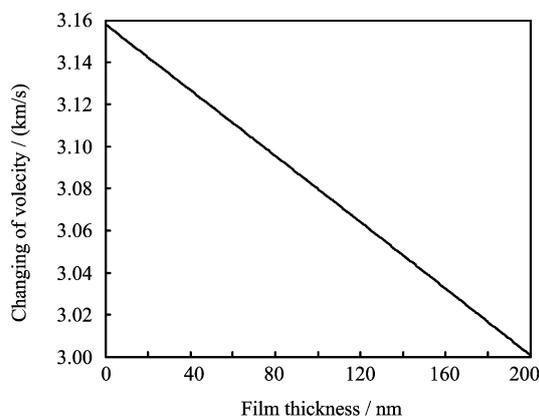


FIG. 2 Perturbation to SAW velocity by gold film

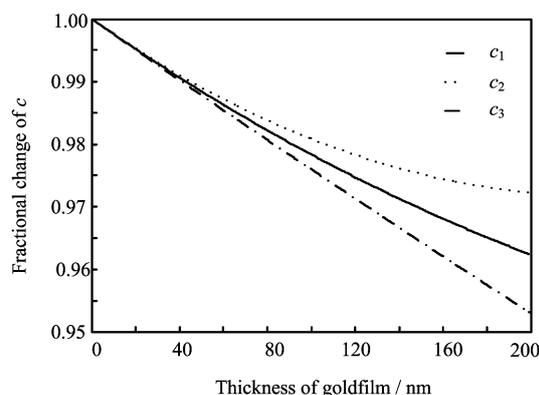


FIG. 3 Perturbation to coupling parameters by gold film

B. Perturbation to SAW by polymer film

According to the Martin theory, a polymer can be divided into three types by the real part of Shear Horizontal modulus of polymer. One is the glassy regime, characterized by $G' \geq 10^9 \text{ N/m}^2$ and $G'' \ll G'$. Its velocity shift decreases and the loss increases monotonously with the film thickness, and its attenuation is very small. The film with $10^7 < G' < 10^9 \text{ N/m}^2$ is called a “glassy-rubbery film”. In this case, the velocity shift is like a sigmoid curve, and the loss has a peak value at a certain film thickness. For $G' \leq 10^7 \text{ N/m}^2$, the velocity shift is small and the loss has a great saturation value. The film is called a “rubbery film”. Calculated results by the Eq.(5) show the perturbation to SAW by the rubbery polymer film as Fig.4. The solid line, dashed line and square dot represent the calculated result according to Eq.(5) and Martin theoretical and experimental result respectively. The rubbery polymer film is assumed to be Poly-epichlorohydrin, its density is about 1.42 g/m^3 , and its modulus can be obtained by method of J.D.Ferry [8]. Experimental testing system includes the Network analyzer, programmable frequency counter and sensor system. The detection frequency of SAW sensor is about 158 MHz. From the Fig.4, it shows that the simulation loss is in better agreement with the experiment than Martin theory. The theoretical velocity by Eq.(5) and Martin theory shows larger errors com-

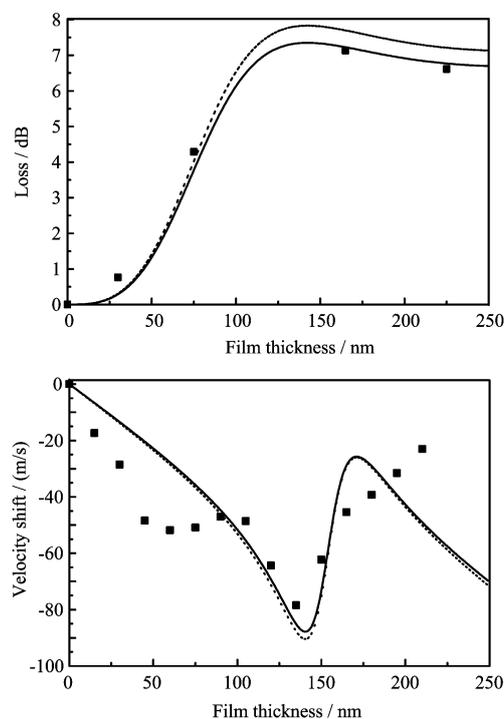


FIG. 4 Calculation result in this work (solid line) and Martin theory (dashed line) respectively, and compared to the experiment (point)

pared to experiment, and the deviation may be influenced by the experimental method and environment.

C. Simulation of vapor sorption in polymer by new film-deposition technology

In this section, the theoretical analyse focuses on vapor sorption of the three types of films by new film-deposition technology (Self Assemble technology). In these calculations, it has been assumed that the shear horizontal modulus of the polymer are not influenced by gas sorption, and the bulk modulus is a constant, approximately $1 \times 10^9 + i \times 10^9$; i is an imaginary unit. The thickness and density of the polymer are taken as a function of the gas sorption concentration. If the concentration of sorption species is expressed by C , the thickness and density can be represented by the following equations:

$$\begin{aligned} \rho(C) &= \rho_0 + \frac{CM}{1+CU} \\ h(C) &= h_0(1+CU) \end{aligned} \quad (10)$$

where h_0 and ρ_0 are the thickness and density of the polymer film before gas sorption, respectively without gas sorption and M is the molecular weight of the absorbed vapor. U is also the specific volume of the absorbed vapor obtained from

$$U = \frac{M}{\rho_v} \quad (11)$$

here ρ_v is the density of vapor. Substituting Eq.(7) and Eq.(8) into Eq.(5), the velocity shift and attenuation

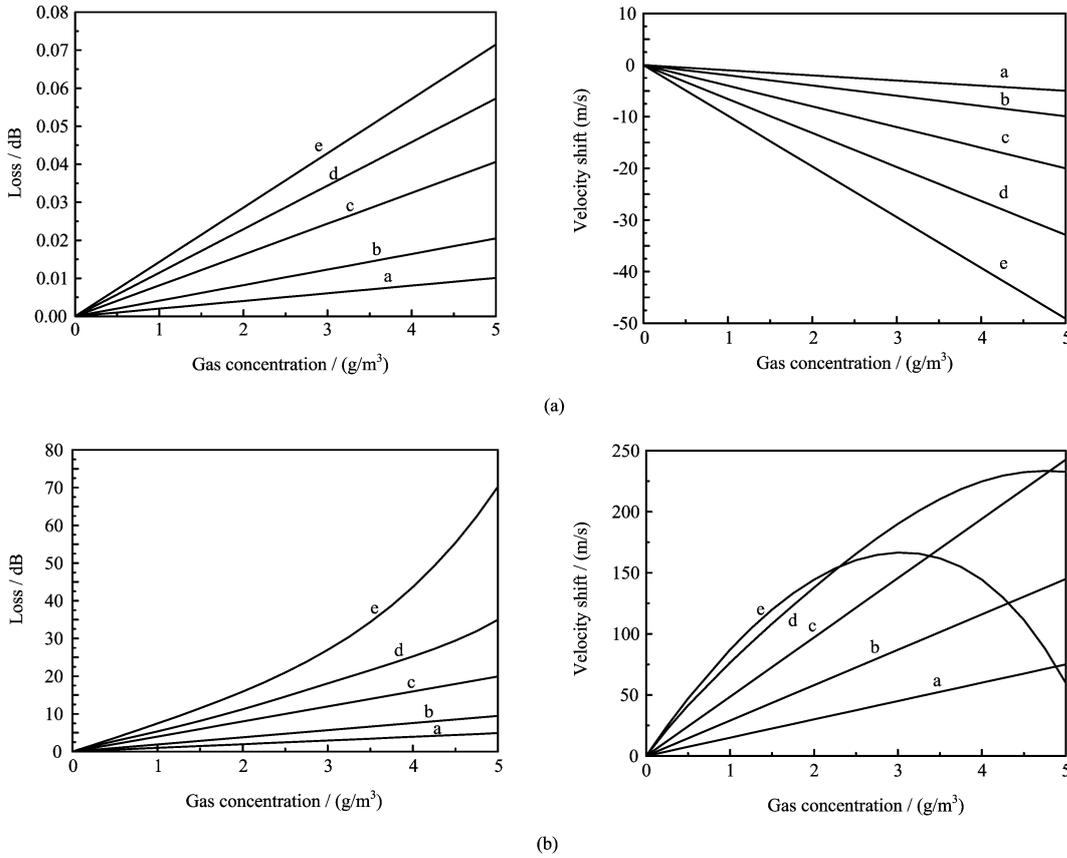


FIG. 5 Calculated results of the velocity shift and attenuation change of SAW as a function of concentration of gas, with the film thickness as a parameter. (a) glassy polymer film, (b) rubbery film. a: 100 nm, b: 200 nm, c: 400 nm, d: 600 nm, e: 800 nm

due to gas sorption are obtained as a function of the concentration of sorption species. C is usually represented as follow:

$$C = \frac{\kappa c_v}{M} \quad (12)$$

where c_v is the concentration of vapor. The partition coefficient, κ is defined by Grate *et al.* [9]. By substituting Eqs.(8) and (9) to into Eq.(7), we can obtain

$$\begin{aligned} h(c_v) &= h_0 \left(1 + \frac{\kappa c_v}{\rho_v} \right) \\ \rho(c_v) &= \frac{\rho_0 + \kappa c_v}{1 + \kappa c_v / \rho_v} \end{aligned} \quad (13)$$

In this calculation, the density of the polymer is assumed to be 1.4 g/cm^3 , the thickness of the gold film is about 140 nm , and the Shear Horizontal modulus of the glassy polymer is $G=1 \times 10^{10} + i \times 10^6 \text{ N/m}^2$, whereas rubbery film is $G=1 \times 10^7 + i \times 10^6 \text{ N/m}^2$. Their bulk modulus are both $K=1 \times 10^9 + i \times 10^9 \text{ N/m}^2$, the constant parameters of sorbed gas are as follows: $\rho_v=0.62 \text{ g/cm}^3$, $\kappa=3 \times 10^6$. The thickness of the calculated polymer films are 100, 200, 400, 600, 800 nm, respectively.

Thus calculation results shown in Fig.5(a) and (b) is given by Eq.(10), where velocity shift and the attenuation versus the concentration of vapor, c_v , dependence. The detection frequency, 158 MHz is employed here. As

for the glassy film, seen as Fig.5(a), the velocity shift and attenuation increase with the increasing amount of absorbed gas, and the velocity is quite linear with concentration of vapor. But for rubbery film, the linear property is not distinct, shown as Fig.5(b), where the polymer is assumed to very thin. With increasing concentration of vapor, the velocity shift increases, while the linear regime between the velocity shift and concentration of vapor is deteriorated. Therefore, based on the calculated results, we can conclude that the sensitivity doesn't always increase with the thickness.

D. Frequency characteristics of gas sorption

Eq.(1) shows that the sensor response is proportional to both film thickness and frequency. However, the calculated results of previous section show that the sensor responses are a function of film thickness. Therefore, it is necessary to study the frequency characteristics of gas sorption. For the calculation purpose, we assume that c_v is 3.0 g/m^3 , the modulus parameters of polymer are not influenced by the detection frequency, and the thickness of polymer films are assumed as 100, 200, 300, 400, 500 nm, respectively. Figure 6 shows the simulated results of the frequency properties for response of SAW gas sensor coated with different polymer. For the glassy film, as shown in Fig.6 (a), the sensor ve-

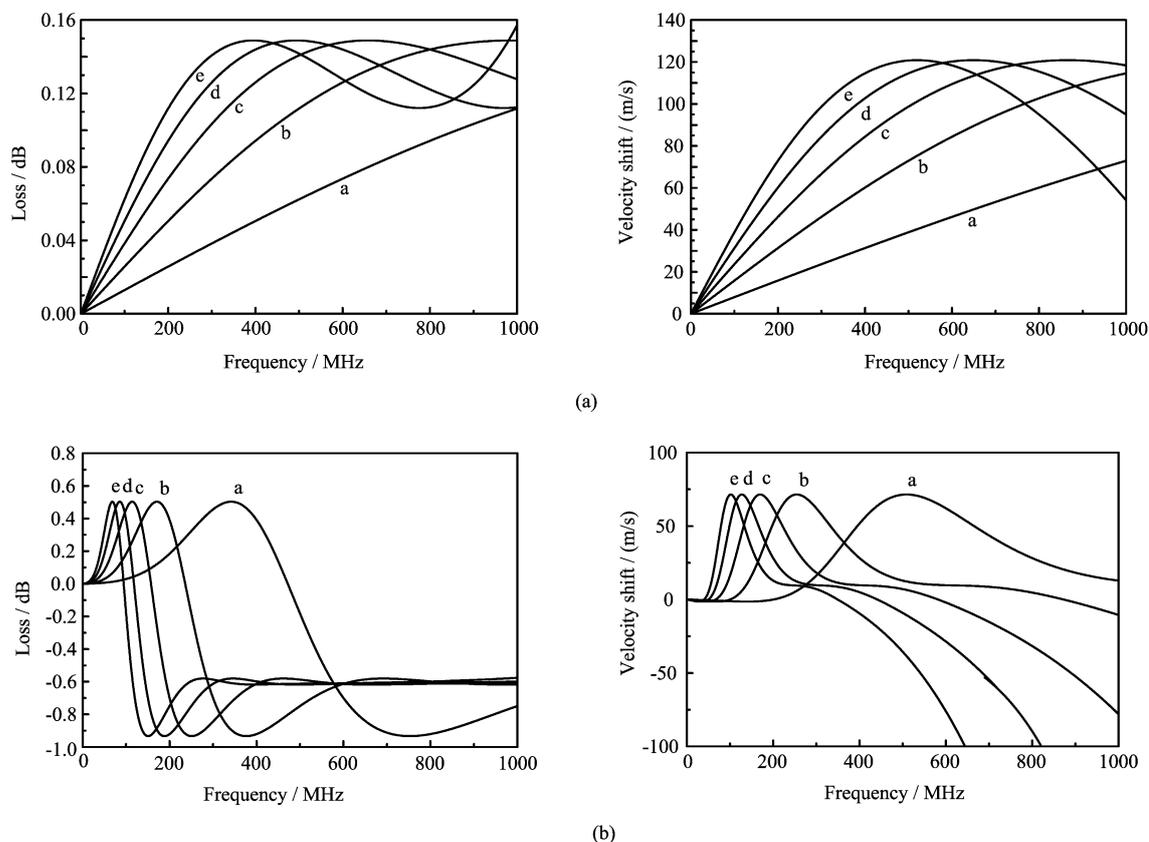


FIG. 6 Calculated results of the velocity shift and loss of SAW as a function of detection frequency, with the film thickness as a parameter. (a) glassy film, (b) rubbery film, a: 100 nm, b: 200 nm, c: 300 nm, d: 400 nm, e: 500 nm

velocity and attenuation are proportional appreciatively to the detection frequency which agrees with Eq.(1). If the coating polymer is a glassy film, a high-frequency device and a thick film are preferable to highly sensitive detection, and the attenuation caused by the film is small. On the other hand, the sensor response for the rubbery film, as shown in Fig.6 (b), is not proportional to the frequency. Maximum sensitivity depends on both the detection frequency and the thickness. Therefore, a high-frequency device can not yield high sensitivity, as described in Eq.(1).

IV. CONCLUSION

A method to analyze the response mechanism of SAW gas sensor coated with polymer film by new film-deposition, Self-Assemble technology, was described and the relative response equations were deduced. The simulated attenuation with polymer film thickness was in good agreement with the experimental results, although the velocity shift was not. Maybe it was influenced by the experimental methods and experimental environment. In addition, vapor sorption of the different types of films was described. For the glassy film, the relationship between the sensitivity and the concentration of vapor is linear, but for the rubbery film, the linear property was limited in a small range of concentration. The simulation shows that the sensitivity

of SAW sensor depends on not only the detection frequency but also the viscoelastic properties of the polymer coatings. This is because viscoelastic properties of polymer films depend on temperature, frequency and gas sorption, proper selection of the thickness and detection frequency are required. Thus a high-frequency device can't always yield high sensitivity.

V. ACKNOWLEDGMENT

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APPENDIX

APPENDIX A: Perturbation to SAW by the gold film

Figure 1 shows the coordinate system used in this study. The SAW propagates to the x_1 direction. A complex propagating factor β is defined by the wave number k and attenuation α as

$$\beta = k - j\alpha \quad (\text{A1})$$

From Eq.(A1), the variation of the complex propagating factor can be derived as

$$\frac{\Delta\beta}{k} = \frac{\Delta\alpha}{k} - j\frac{\Delta V}{V} \quad (\text{A2})$$

where, V is the phase velocity of SAW, and $\Delta\beta$, ΔV and $\Delta\alpha$ are the fractional changes of β , V and α , respectively. Hence, the variation of the complex propagating factor can be decomposed into the variations of the velocity and attenuation. Usually, the frequency shift is considered approximatively as the velocity shift. The attenuation (ΔL , dB) of SAW can be represented by

$$\Delta L = 54.6 \times \frac{\Delta\alpha}{k} N_\lambda \quad (\text{A3})$$

N_λ is the acoustic path length.

A perturbation method can be applied to approximate the changes in SAW propagation velocity and attenuation contributed by the viscoelastic polymer film: changes in the complex propagation factor are related to the surface mechanical impedances Z_i :

$$\frac{\Delta\beta}{k} = \frac{\Delta\alpha}{k} - j\frac{\Delta V}{V} = \sum_{i=1}^3 c_i Z_i \quad (\text{A4})$$

where c_i are the normalized surface particle velocities and the surface mechanical impedances are

$$Z_i = -\frac{T_{i3}}{v_i} \Big|_{y=0} \quad (\text{A5})$$

where T_{i3} and v_i , evaluated at $y=0$, denote interfacial stress and particle velocity in the i -direction, respectively.

As the SAW is assumed to be a plane wave, the differentiation with respect to x_2 is zero. From the equation of motion ($\nabla \cdot T = \rho(\partial v/\partial t)$), the following equations are obtained:

$$\begin{aligned} \frac{\partial T_{11}}{\partial x_1} + \frac{\partial T_{13}}{\partial x_3} &= -\omega^2 \rho u_1 \\ \frac{\partial T_{21}}{\partial x_1} + \frac{\partial T_{23}}{\partial x_3} &= -\omega^2 \rho u_2 \\ \frac{\partial T_{31}}{\partial x_1} + \frac{\partial T_{33}}{\partial x_3} &= -\omega^2 \rho u_3 \end{aligned} \quad (\text{A6})$$

here $\partial/\partial t = j\omega$. The relationships between stress T_{ij} ($ij=1, 2, 3$), and strain S_{ij} , for the isotropic film are expressed as

$$T_{ij} = \begin{cases} \lambda(S_{11} + S_{22} + S_{33}) + 2\mu S_i & (i=j) \\ 2\mu S_i & (i \neq j) \end{cases} \quad (\text{A7})$$

here, λ and μ are the Lamé constants. The Lamé constants and the bulk modulus K and the shear horizontal G are related by the following equations:

$$\lambda = K - 2/3G, \quad \mu = G, \quad (\text{A8})$$

where

$$K = K' + jK'', \quad G = G' + jG'' \quad (\text{A9})$$

Their real parts mean storage modulus and imaginary parts are loss moduli. These parameters are utilized as variances to change the polymer characteristics. The displacements are assumed as:

$$u_i = (Ae^{i\beta_i y} + Be^{-i\beta_i y})e^{i(\omega t - kz)} \quad (0 \leq y \leq h) \quad (\text{A10})$$

where h is gold film thickness, and A and B are constants. The factors β_i are propagation constants describing wave propagation across the gold film. Using Eq.(A7) and Eq.(A10), Martin's method and Auld's perturbation theory, surface stresses $T_{3j}|_{x_3=0}$ ($j=1-3$) are derived.

$$\begin{aligned} T_{13}|_{x_3=0} &= -j\frac{\beta_1 M_1}{\omega} \tan(\beta_1 h) v_1|_{x_3=0} \\ T_{23}|_{x_3=0} &= -j\frac{\beta_2 M_2}{\omega} \tan(\beta_2 h) v_2|_{x_3=0} \\ T_{33}|_{x_3=0} &= -j\frac{\beta_3 M_3}{\omega} \tan(\beta_3 h) v_3|_{x_3=0} \end{aligned} \quad (\text{A11})$$

where $M_1 = M_2 = G$, $M_3 = K$,

$$\begin{aligned} \beta_1 &= \omega \sqrt{\frac{1}{G} \left[\rho - \frac{1}{V^2} \cdot \frac{4G(3K+G)}{3K+4G} \right]} \\ \beta_2 &= \omega \sqrt{\frac{1}{G} \left(\rho - \frac{G}{V^2} \right)} \\ \beta_3 &= \omega \sqrt{\frac{\rho}{K + 4/3G}} \end{aligned} \quad (\text{A12})$$

Using Eq.(A5), the surface mechanical impedance can be obtained:

$$Z_i = j\frac{\beta_i M_i}{\omega} \tan(\beta_i h), \quad i = 1-3 \quad (\text{A13})$$

Then substituting Eq.(A13) into Eq.(A4), the fractional change of the complex propagating factor can be expressed as:

$$\begin{aligned} \frac{\Delta\beta}{k} &= \frac{\Delta\alpha}{k} - j\frac{\Delta V}{V} \\ &= jc_1 \frac{\beta_1 G}{\omega} \tan(\beta_1 h) + jc_2 \frac{\beta_2 G}{\omega} \tan(\beta_2 h) \\ &\quad + jc_3 \frac{\beta_3 K}{\omega} \tan(\beta_3 h) \end{aligned} \quad (\text{A14})$$

From Eq.(A14) and Eq.(A2), the velocity shift of SAW can be expressed by

$$\begin{aligned} \frac{\Delta V}{V} &= -c_1 \frac{\beta_1 G}{\omega} \tan(\beta_1 h) - c_2 \frac{\beta_2 G}{\omega} \tan(\beta_2 h) \\ &\quad - c_3 \frac{\beta_3 K}{\omega} \tan(\beta_3 h) \end{aligned} \quad (\text{A15})$$

APPENDIX B: Perturbation toe coupling parameters by the gold film

From the appendix A, the gold film surface displacements can be calculated by

$$u_i|_{y=h} = u_{i0}/\cos(\beta_i h) \quad (\text{B1})$$

And the particle velocity is as follows:

$$\begin{aligned} v_{ih} &= v_i|_{y=h} = j\omega u_i|_{y=h} \\ &= j\omega u_{i0}/\cos(\beta_i h) \end{aligned} \quad (\text{B2})$$

where $v_{i0} = v_i|_{y=0} = j\omega u_{i0}$, $v_{ih} = v_{i0}/\cos(\beta_i h)$.

It is assumed that the perturbation to the attenuation of SAW by the gold film can be ignored, and the power flow approximates a constant. The coupling parameter c_i is defined as $v_{i0}^2/(4k_0P)$, where v_{i0} is surface particle velocity, P is the SAW acoustic power density (assumed as a constant). From Eq.(B3), the coupling parameters perturbed by the gold film c'_i can be calculated in the following

$$c'_i = \frac{c_i V'}{\cos^2(\beta_i h) V_0} \quad (\text{B3})$$

APPENDIX C: Perturbation toe SAW by the polymer film based on new film-deposition

It is assumed that the surface of gold film is used as the substrate surface, with Martin's method, the perturbation to the SAW by the polymer film can be calculated.

First from the appendix A, the SAW displacements can be obtained:

$$u_i(y) = \frac{u_{i0} \cos[\beta_i(y-h)]}{\cos(\beta_i h)} \quad (\text{C1})$$

Also using the motion Eq.(A6), this is

$$\frac{\partial T_{3i}}{\partial y} + \frac{\partial T_{1i}}{\partial z} = -\rho\omega^2 u_i \quad (\text{C2})$$

where

$$T_{3i} = \frac{M^i \partial u_i}{\partial y}, \quad T_{1i} = \frac{E^i \partial u_i}{\partial z} \quad (\text{C3})$$

Substituting the Eq.(C3) into Eq.(C2)

$$\frac{\partial^2 u_i}{\partial^2 y} + \alpha_i^2 u_i = 0 \quad (\text{C4})$$

where $\alpha_i^2 = \omega^2(\rho - E^i/V'^2)/M^i$, V' is velocity perturbed by the gold film and polymer film, E^i are strain moduli,

$$\begin{aligned} E^2 &= G, \quad E^3 = 0 \\ E^1 &= 4G \frac{3K+G}{3K+4G} \\ M^1 &= M^2 = G, \quad M^3 = K \end{aligned} \quad (\text{C5})$$

The solution to Eq.(C4) for $h \leq y \leq h_1$ is

$$u_i = C e^{i\alpha_i y} + D e^{-i\alpha_i y} \quad (\text{C6})$$

The constants C and D in Eq.(C6) can be determined from the boundary conditions that are applied at the upper and lower film surface. The first boundary condition stipulates that the displacement at the film/substrate interface be continuous, i.e., $u_i(h^+) = u_{ih}|_{y=h}$, u_{ih} is the displacement at the lower film surface. Eq.(C6) and Eq.(C1) give

$$C e^{i\alpha_i h} + D e^{-i\alpha_i h} = u_{i0}/\cos(\beta_i h)|_{y=h} \quad (\text{C7})$$

The second boundary condition stipulates that the upper film surface (film/air interface) be stress-free: $T_{3i}|_{y=h_1} = 0$, evaluating Eq.(C3) at $y = h_1$, gives

$$C e^{i\alpha_i h_1} - D e^{-i\alpha_i h_1} = 0 \quad (\text{C8})$$

Solving Eqs.(C7) and (C8) simultaneously determines C and D :

$$\begin{aligned} C &= \frac{u_{i0}}{\cos(\beta_i h)} \frac{e^{-j\alpha_i h_1}}{e^{i\alpha_i(h-h_1)} + e^{-i\alpha_i(h-h_1)}} \\ D &= \frac{u_{i0}}{\cos(\beta_i h)} \frac{e^{j\alpha_i h_1}}{e^{i\alpha_i(h-h_1)} + e^{-i\alpha_i(h-h_1)}} \end{aligned} \quad (\text{C9})$$

Substituting Eq.(C9) into (C3) determines the displacements, then, using Eq.(C3), the interfacial shear stress can be calculated:

$$\begin{aligned} T_{3i}(h) &= M^i \frac{\partial u_i}{\partial y} \Big|_{y=h} \\ &= j\alpha_i M^i (C e^{j\alpha_i h} - D e^{-j\alpha_i h}) \end{aligned} \quad (\text{C10})$$

And the interfacial particle velocity is found from Eq.(C6)

$$v_i(h) = j\omega u_i(h) = j\omega (C e^{j\alpha_i h} + D e^{-j\alpha_i h}) \quad (\text{C11})$$

Substituting (C9) and (C10) into Eq.(C9) and using Eq.(A4) for C and D gives the surface mechanical impedance associated with each displacement

$$Z_i = j \frac{\alpha_i M^i}{\omega} \tan[\alpha_i(h_1 - h)] \quad (\text{C12})$$

From Eq.(A4), the perturbation in Rayleigh SAW propagation arising from viscoelastic film based on new film-deposition technology is determined from the surface mechanical impedance

$$\begin{aligned} \frac{\Delta\gamma}{k_0} &= \frac{\Delta a}{k_0} - j \frac{\Delta V}{V} \\ &= j \sum_i^3 c'_i \frac{\alpha_i M^i}{\omega} \tan[\alpha_i(h_1 - h)] \end{aligned} \quad (\text{C13})$$

For the SH-SAW, Eq.(C13) becomes

$$\frac{\Delta\gamma}{k_0} = j c_2 \frac{\alpha_2 M^2}{\omega} \tan[\alpha_2(h_1 - h)] \quad (\text{C14})$$

The velocity shift and attenuation change are obtained by substituting Eq.(C13) and Eq.(C14) into Eq.(A4).