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# Love Wave Sensor Based on PMMA/ZnO/Glass Structure for Liquids Sensing †

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**Abstract:** PMMA/ZnO/glass structure was investigated to enhance Love mode sensor sentivities. The phase velocities and the attenuation of the acoustic wave propagating along the PMMA/30° tilted c-axis ZnO/glass structure contacting a viscous non-conductive liquid were calculated for different PMMA (Polymethyl methacrylate) and ZnO guiding layer thicknesses, added mass thicknesses, and liquid viscosity and density. The sensor velocity and attenuation sensitivities were also calculated for different environmental parameters. The resulted sensitivities to liquid viscosity and added mass were optimized by adjusting the ZnO and PMMA guiding layer thickness corresponding to a sensitivity peak. The present analysis is of importance in manufacturing and applications of the PMMA-ZnO-glass structure Love wave sensors for the detection of liquids properties, such as viscosity, density and mass anchored to the sensor surface.

**Keywords:** love wave sensor; surface acoustic waves; ZnO; PMMA

## 1. Introduction

The propagation of Love modes along 30° tilted c-axis ZnO/glass-based structures has been modeled and analysed aimed to the design of a sensor able to operate in liquid environment [1]. The sensor velocity and attenuation sensitivities to the changes of liquid viscosity and mass loading were calculated for different ZnO layer thicknesses and the peak sensitivity was achieved at the ZnO thickness to/wavelength ratio $h/\lambda = 0.05$, with electromechanical coupling coefficient is $K^2$ around 0.2% for substrate-transducer-piezolectric film (STF) configuration. $K^2$ is highest at $h/\lambda = 0.3$ but the sensitivies will be reduced significantly. Ideally, the sensor should have higher sensitivities and $K^2$.

The shear velocity of guiding layer must be lower than the substrate to satisfy the condition of existence of love wave. The higher ratio of shear wave velocity, or density, or both between substrate and the guiding layer material, the higher the maximum sensitivity [2]. Polymer material such as polymethyl-methacrylate (PMMA) has been used for Love wave guiding layer due to its low mass density and shear velocity [3]. However, it has high acoustic damping loss due to viscoelastic properties of PMMA. Double guiding layer of PMMA and elastic material such as SiO2 was reported to improve sensitivities without excessive increase in propagation loss [4]. Harding and Du used SiO2 and PMMA as double guiding layer with Quartz piezoelectric substrate. They achieved the highest mass loading sensitivity of 52 $m^2 \cdot kg^{-1}$, which is higher than SiO2 (38 $m^2 \cdot kg^{-1}$) and PMMA (31 $m^2 \cdot kg^{-1}$) single guiding layer mass loading sensitivities.

In this paper, we theoretically investigate the performances of a PMMA/30° tilted c-axis ZnO/glass Love wave sensor for liquid sensings. The organization of the present article begins with wave equations of Love wave when a viscous Newtonian liquid is introduced in contacts with the guiding layer free surface, and a complex wave velocity is defined for the four layers system(substrate-guiding layer 1-guiding layer 2-liquid). The velocity and attenuation sensitivities to viscosity and to an added mass are then calculated in a viscous liquid environment. The aim of the present theoretical calculations is to investigate the influence of the addition of PMMA layer on ZnO/Glass Love wave sensor to obtain a high sensistivity, high $K^2$ love wave sensor.

## 2. Love Wave Sensors in PMMA-ZnO-Glass Layered Structures for Application in Liquid Environments: Statement of the Problem and Basic Equations

The 30° tilted ZnO coordinate system and the PMMA/ZnO/glass layered structure here studied are illustrated in Figure 1a,b. The substrate, that occupies the half-space $x_2 > 0$, is assumed to be an isotropic dielectric elastic medium, glass; the guiding layer 1 is assumed to be a ZnO film with the c-axis tilted 30°. The ZnO layer was considered isotropic but its elastic constants were considered numerically equal to the stiffened elastic constants of the 30° tilted piezoelectric counterpart; the guiding layer 2 PMMA is in contact with the viscous liquid half-space. The wave propagation direction is $x_1$ and no variation of the displacement components in the $x_3$ direction is assumed. The $x_2 = 0$ plane is the interface between the substrate and guiding layer 1, the $x_2 = -h_{gl1}$ and $x_2 = -h_{gl2}$ planes are the guiding layer 1-guiding layer 2 and guiding layer 2-liquid interfaces.

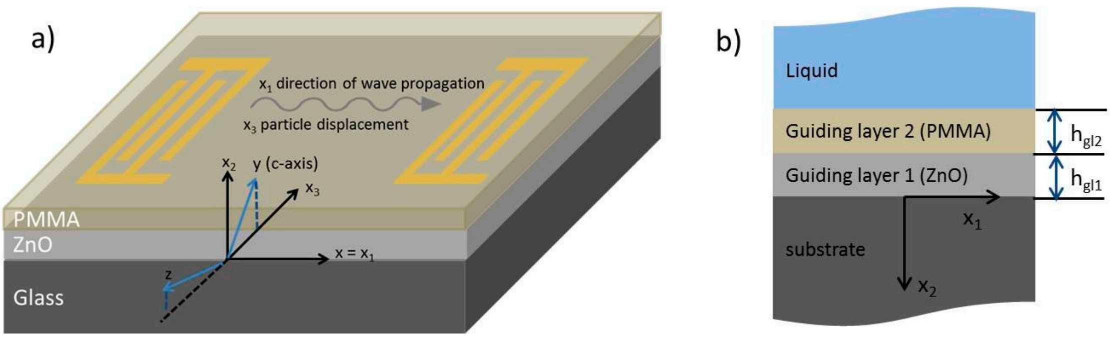


**Figure 1.** (**a**) Love sensor based ob PMMA/ZnO/Glass syructure with the laboratory coordinates system $x_1$ $x_2$ $x_3$ and the ZnO layer coordinate system x y z; (**b**) the geometry of the substrate/guiding layer 1/guiding layer 2/liquid system.

The equations of motion for the four media are the following:

$$\rho_{sub}\frac{\partial^2 u_3^{sub}}{\partial t^2} = c_{44}^{sub}\left(\frac{\partial^2}{\partial x_1^2}+\frac{\partial^2}{\partial x_2^2}\right)u_3^{sub} \quad \text{for } x_2 > 0 \tag{1}$$

$$\rho_{gl1}\frac{\partial^2 u_3^{gl1}}{\partial t^2} = \overline{c_{44}^{gl1}}\left(\frac{\partial^2}{\partial x_1^2}+\frac{\partial^2}{\partial x_2^2}\right)u_3^{gl1} \quad \text{for } 0 < x_2 < -h_{gl1} \tag{2}$$

$$\rho_{gl2}\frac{\partial^2 u_3^{gm}}{\partial t^2} = c_{44}^{gl2}\left(\frac{\partial^2}{\partial x_1^2}+\frac{\partial^2}{\partial x_2^2}\right)u_3^{gl2} \quad \text{for } -h_{gl1} < x_2 < -h_{gl2} \tag{3}$$

$$\frac{\partial v_3}{\partial t} = \frac{\eta}{\rho_l}\left(\frac{\partial^2}{\partial x_1^2}+\frac{\partial^2}{\partial x_2^2}\right)v_3 \quad \text{for } x_2 < -(h_{gl1} + h_{gl2}) \tag{4}$$

The assumed solutions to the Equations (1)–(4) are the following:

$$u_3^{substrate} = [U_{sub}^0 \cdot e^{-qx_2}] \cdot e^{j(kx_1-\omega t)}$$

$$u_3^{gl1} = [U_{gl1}^1 \cdot cos(bx_2) + U_{gl1}^2 \cdot \sin(bx_2)] \cdot e^{j(kx_1-\omega t)}$$

$$u_3^{gl2} = [U_{gl2}^1 \cdot cos(px_2) + U_{gl2}^2 \cdot \sin(px_2)] \cdot e^{j(kx_1-\omega t)}$$

$$u_3^{liquid} = [U_{liq}^1 \cdot e^{\lambda_1 x_2}] \cdot e^{j(kx_1-\omega t)}$$

The particle displacements and the traction components of stress must be continuous across the substrate/guiding layer, guiding layer/mass layer, and mass layer/liquid interfaces. The continuity conditions are a set of linear homogeneous algebraic equations whose coefficients are function of the wave phase velocity $V_{ph}$: a non trivial solution exists if the determinant of the coefficients vanishes; an optimized procedure was used to find a velocity value that drive the size of the determinant as close to zero as possible. The determinant is:

$$\begin{vmatrix} 1 & -1 & 0 & 0 & 0 & 0 \\ -qc_{44}^{sub} & 0 & -bc_{44}^{gl1} & 0 & 0 & 0 \\ 0 & \cos(bh_{gl1}) & -\sin(bh_{gl1}) & -\cos(ph_{gl1}) & \sin(ph_{gl1}) & 0 \\ 0 & c_{44}^{gl1}b\sin(bh_{gl1}) & c_{44}^{gl1}b\cos(bh_{gl1}) & -c_{44}^{gl2}p\sin(ph_{gl1}) & -c_{44}^{gl2}p\cos(ph_{gl1}) & 0 \\ 0 & 0 & 0 & -j\omega\cos(pH) & j\omega\sin(pH) & -e^{-\lambda_1 H} \\ 0 & 0 & 0 & c_{44}^{gl2}p\sin(pH) & c_{44}^{gl2}p\cos(pH) & -\eta\lambda_1 e^{-\lambda_1 H} \end{vmatrix} = 0$$

where $H = h_{gl1} + h_{gl2}$, $q^2 = k^2 - k_s^2$, $b^2 = k_{gl1}^2 - k^2$, $p^2 = k_{gl2}^2 - k^2$, and $\lambda_1^2 = k^2 - j\omega(\rho_l / \eta)$.

$$k = 2\frac{\pi}{\lambda},\ ks = \left(2\frac{\pi}{\lambda}\right)\left(\frac{Vph}{Vs}\right),\ kgl1 = (2\frac{\pi}{\lambda})(\frac{Vph}{Vgl1}),\ kgl2 = (2\frac{\pi}{\lambda})(\frac{Vph}{Vgl2})$$

$V_s$, $V_{gl1}$, $V_{gl2}$ are shear velocity in substrate, guiding layer 1 and guiding layer 2 respectively. To account viscoelastic properties of PMMA as guiding layer 2, it has complex shear modulus and complex shear velocity.

## 3. Viscosity Sensitivity of the *Liquid-PMMA-ZnO-Glass* Structure

Complex phase velocities $V_{ph}$ were obtained by solving the the determinant of matrix equal to 0 using Levenberg-Marquard-Fletcher method in a Matlab routine. The viscous liquid was supposed to be a mixture of water and glycerol: the fraction of glycerol by volume ranged from 0 (only water) to 0.8 (80% glycerol, 20% water) and the $\sqrt{\rho_l \cdot \eta}$ ranged from 1 to 10.59 $kg{\cdot}m^{-2}s^{0.5}$. The real and imaginary parts of the phase velocity ($V_r$ and $V_i$) of the $L_1$ mode were calculated for different guiding layer thicknesses and for different water/glycerol mixtures. The propagation loss ($\alpha$) is calculated by $\alpha = 2{\cdot}\pi{\cdot}\log(e){\cdot}V_i/V_r$. Figure 2a,b show the real velocity and propagation loss of the $L_1$ mode velocity as a function of the PMMA guiding layer thickness for different water/glycerol mixture. The thickness of ZnO is fixed at 0.5 μm and $\lambda$ = 10 μm, supposing h/λ of ZnO 0.05.

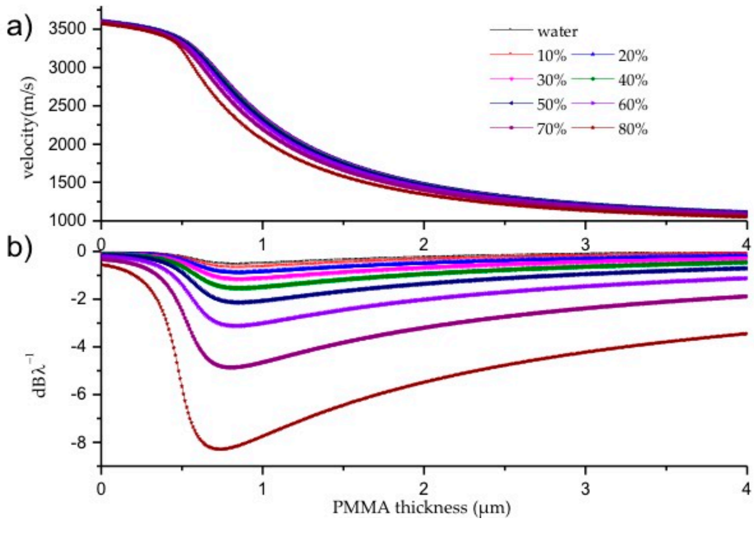


**Figure 2.** The real velocity (**a**) and attenuation (**b**) of the $L_1$ mode phase velocity vs. the PPMA guiding layer 2 thickness for different water/glycerol mixture with 0.5 μm ZnO as guiding layer 1.

The calculated data clearly show that, while the percentage of glycerol in water increases, the wave velocity decreases respect to the velocity along the bare substrate in pure water, and the attenuation increases. Calculations were repeated for different thicknesses of ZnO from 0.5 μm to 3 μm, supposing h/λ of ZnO from 0.05 to 0.3.

The attenuation and velocity relative changes vs. $\sqrt{\rho\cdot\eta}$ were linearly fitted, for each ZnO and PMMA thicknesses, and the slopes, i.e., the phase attenuation sensitivities $S^{v_{ph}}_{\sqrt{\rho\cdot\eta}} = \left(\Delta v_{ph}/v_{hp}^{water}\right)/\sqrt{\rho\cdot\eta}$, $S^{att}_{\sqrt{\rho\cdot\eta}} = \alpha/\sqrt{\rho\cdot\eta}$, were plotted vs. the total thicknes; the calculated sensitivities dispersion curves are shown in Figure 3a,b.

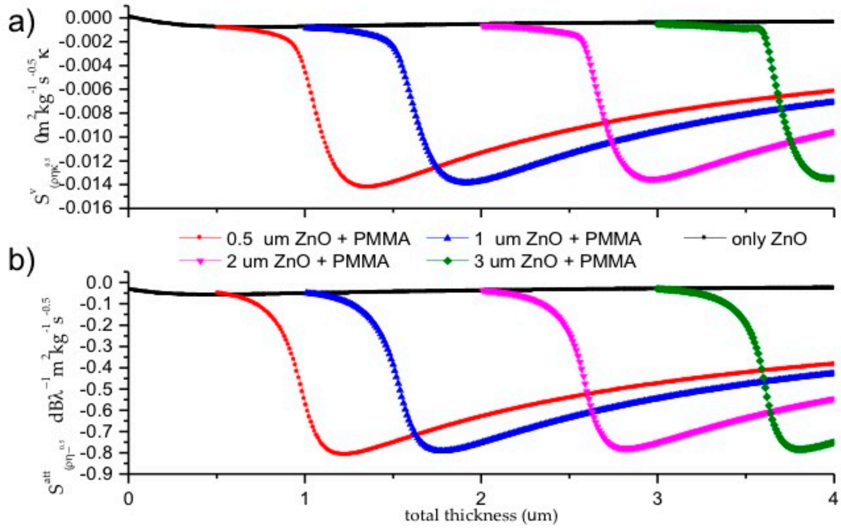


**Figure 3.** The phase velocity sensitivities **(a)** and the attenuation sensitivity **(b)** to added mass in liquid vs. the total guiding layer thickness, for different PMMA/ZnO combination

The calculated sensitivities are significantly increasing when PMMA layers are added onto ZnO layer, with the optimum PMMA thickness of around 0.85 μm. Additional advantage of using PMMA layer is that the sensitivities are less dependant on ZnO thickness so that thicker ZnO can be used to obtain higher $K^2$.

## 4. Mass Sensitivity of the *Liquid-PMMA-ZnO-Glass* Structure

Figure 4a,b show the calculated relative change of phase velocity when a mass loaded onto the sensor. The data show an order increase of velocity change for PMMA/ZnO double guiding layer. Similarly to viscosity sensitivities, the mass loading sensitivities for PMMA/ZnO double guiding layer are less dependant on ZnO thickness. The mass loading sensitivities were calculated from the slope of the graphs. They are 55 $m^2 \cdot kg^{-1}$ for 0.5 μm and 29 $m^2 \cdot kg^{-1}$ for 3 μm ZnO layer only. For PMMA/ZnO double guiding layer with 0.85 um PMMA, the mass loading sensitivities are 683 $m^2 \cdot kg^{-1}$ for 0.5 μm and 553 $m^2 \cdot kg^{-1}$ for 3 μm ZnO layer.

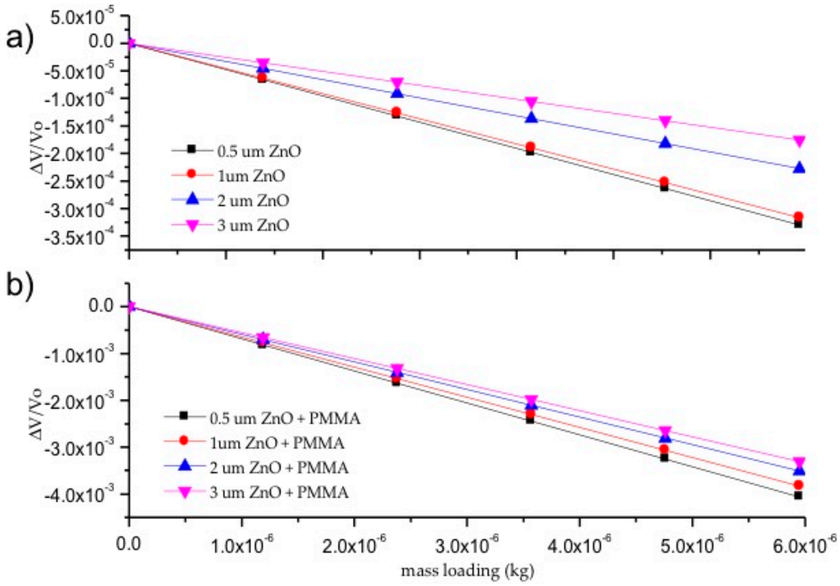


**Figure 4.** The relative change of phase velocity of (**a**) ZnO/Glass and (**b**) PMMA/ZnO/Glass structure vs. the mass loading for different PMMA/ZnO combination.

## 5. Conclusions

The theoretical sensitivity of Love wave sensors, based on PMMA/ZnO/Glass structure, to the properties of a liquid environment, and to a mass deposited from a viscous Newtonian liquid phase is derived. The calculation of the real and imaginary parts of the phase velocity allowed the estimation of both the phase velocity and attenuation sensitivities to the liquid viscosity and density, and to a mass loaded onto guiding layer surface for different ZnO and PMMA thickness. It resulted that the sensitivities are increasing by adding PMMA layer on ZnO layer. Moreover, the sensitivies of PMMA/ZNO double guiding layer are less dependent on ZnO thickness.

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**Author Contributions:** M.H. and C.C. derived the equation shown in Section 2. All three authors performed the calculations shown in Sections 3 and 4 and wrote the manuscript

**Conflicts of Interest:** The authors declare no conflict of interest

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