



Sezawa wave acoustic humidity sensor based on graphene oxide sensitive film with enhanced sensitivity



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ABSTRACT

The measurement of humidity is very important for air control in ambient, industry, cars, houses, closed apartments, museums, atomic power stations, etc. In the present work the theoretical analysis of the surface acoustic wave propagation in "graphene oxide (GO) film/ZnO film/Si substrate" layered structure has been performed. The change of GO film conductivity due to humidity has been taken into account during the calculations. Based on the obtained results an improved microwave acoustic humidity sensor has been developed. The sensor has enhanced sensitivity of about 91 kHz/% and linear response vs relative humidity in the range 20–98%RH. It is based on the mode belonging to Sezawa wave family that is shown to be more sensitive towards electric conductivity variations in GO film produced by adsorbed water molecules than the Rayleigh counterpart.

1. Introduction

In recent years the microwave acoustic humidity sensors have been developed very extensively [1–26]. These sensors are successfully used for air control in ambient, industry, cars, houses, closed apartments, museums, atomic power stations, etc. [27,28]. For development of the humidity sensors various types of acoustic waves (surface (SAW) [1–11], Love [12–17], bulk [18–24], and Lamb [25,26]) have been used. These waves propagated either in homogeneous crystal substrates or in multi-layered structures containing different sorbent films sensitive towards water vapor adsorption. Usually, the sensitive films are made of ceramic, semiconducting and polymer materials [28,29]. In recent years they are fabricated from graphene-based layers [5–7,17,20–23,25,26,30–32] or special materials doped by nanoparticles, fullerenes, and carbon nano-tubes [8–11,14,17,24]. For any sorbent material adsorption of water molecules produces, in general, the changes in film density, elasticity, electric conductivity, dielectric permittivity, and temperature, which are detected at the output of the sensor as the changes in acoustic wave velocity, frequency, phase and/or amplitude. However, usually, designers use the film with one dominant sensing mechanism. For example, the GO layer with dominant electric conductivity variations, allowed to develop recently the

super-high-sensitive humidity sensor based on high-order (symmetric) Lamb mode in $128Y,X + 90^0 - \text{LiNbO}_3$ plate [26].

In spite of large amount of paper devoted to humidity sensors, further improvements in the sensors performance are still strongly required making new researches and developments in the field actual.

The goal of the present paper is to develop improved humidity sensor basing on theoretical analysis and experimental verification of the acoustic waves propagation in "GO film – ZnO film – Si substrate" layered structure.

2. Theoretical analysis

Propagation of the acoustic waves is studied in two dimensional structures with x_1 - axis parallel to the propagation direction and x_3 - axis perpendicular to the structure surface (Fig. 1). On Fig. 1,a the regions $x_3 < -h_1$, $-h_1 < x_3 < 0$ and $x_3 > 0$ are occupied, respectively, by air, ZnO film and Si half-space. On Fig. 1,b air is for $x_3 < -h_2$, while $-h_2 < x_3 < -h_1$ is for the GO film. Mechanical and electrical variables are assumed to be constant in the x_2 -axis direction.

To solve the problem we write motion Eq. (1), Laplace (2) or Poisson (3) equations, continuity equation for electric charge (4), and material Eqs. (5)–(8) for GO, ZnO and Si medium using the quasi-

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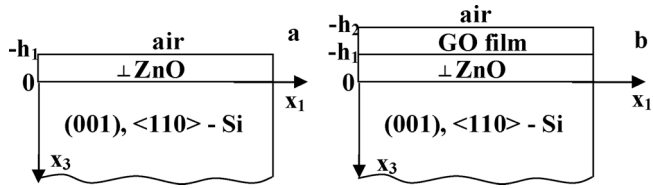


Fig. 1. Geometry of the problems. (a) ZnO/Si structure, (b) GO/ZnO/Si structure.

electrostatic approximation $E_i = \partial\Phi/\partial x_i$ [33]:

$$\rho^{Si,GO,ZnO} \partial^2 U_i / \partial t^2 = \partial T_{ij} / \partial x_j, \quad (1)$$

$$\partial D_j^{Si,ZnO} / \partial x_j = 0, \quad (2)$$

$$\partial D_j^{GO} / \partial x_j = -\delta_v^{GO}, \quad (3)$$

$$\partial J_i^{GO} / \partial x_i + \partial \delta_v^{GO} / \partial t = 0, \quad (4)$$

$$T_{ij}^{Si,GO} = C_{ijkl}^{Si,GO} \partial U_l / \partial x_k, \quad D_j^{Si,GO} = -\epsilon_{jk}^{Si,GO} \partial \Phi / \partial x_k, \quad (5)$$

$$J_i^{GO} = -\sigma_v^{GO} \partial \Phi / \partial x_i + d^{GO} \partial \sigma_v^{GO} / \partial x_i, \quad (6)$$

$$T_{ij}^{ZnO} = C_{ijkl}^{ZnO} \partial U_l / \partial x_k + e_{kij}^{ZnO} \partial \Phi / \partial x_k, \quad (7)$$

$$D_j^{ZnO} = -\epsilon_{jk}^{ZnO} \partial \Phi / \partial x_k + e_{jik}^{ZnO} \partial U_l / \partial x_k \quad (8)$$

Here, E_b , U_b , J_b , t , T_{ij} , x_j , D_j , Φ , ρ , C_{ijkl} , e_{ikl} , ϵ_{jk} , δ_v , σ_v and d are the components of the electric field, mechanical particle displacement, electrical current, time, the components of mechanical stress tensor, coordinates, the components of electric displacements, electrical potential, and density, as well elastic, piezoelectric and dielectric constants, bulk charge, bulk conductivity, and diffusion coefficient of a material, respectively. Indexes ZnO, Si, and GO are attributed to different materials.

Outside the plate ($x_3 < -h_1$, Fig. 1,a; $x_3 < -h_2$, Fig. 1,b) electric displacement of the waves satisfies the Laplace equation:

$$\partial D_j^{ar} / \partial x_j = 0, \quad (9)$$

where $D_j^{ar} = -\epsilon^{ar} \partial \Phi^{ar} / \partial x_j$, index ar denotes air, ϵ^{ar} is the air dielectric constant.

Additionally, the waves satisfy mechanical and electrical boundary conditions:

– for structure on Fig. 1a they are

$$x_3 = 0: T_{3j}^{Si} = T_{3j}^{ZnO}, \quad \Phi^{Si} = \Phi^{ZnO}; \quad D_3^{Si} = D_3^{ZnO}. \quad (10)$$

$$x_3 = -h_1: T_{3j}^{ZnO} = 0, \quad \Phi^{ZnO} = \Phi^{ar}; \quad D_3^{ZnO} = D_3^{ar}. \quad (11)$$

– for structure on Fig. 1b they are

$$x_3 = -h_1: T_{3j}^{ZnO} = T_{3j}^{GO}, \quad \Phi^{ZnO} = \Phi^{GO}, \quad D_3^{ZnO} = D_3^{GO}, \quad J_3^{GO} = 0 \quad (12)$$

$$x_3 = -h_2: T_{3j}^{GO} = 0, \quad \Phi^{GO} = \Phi^{ar}, \quad D_3^{GO} = D_3^{ar}, \quad J_3^{GO} = 0. \quad (13)$$

Using relevant equations and boundary conditions, the problem of acoustic wave propagation is solved as follows. The solution is presented in the form of a set of plane inhomogeneous waves [34,35]:

$$Y_i(x_1, x_3, t) = Y_i(x_3) \exp[j\omega(t - x_1/V_{ph})], \quad (14)$$

where $i = 1-8$ for the ZnO, $i = 1-6$ for the mechanical part of task for Si and GO, and $i = 1, 2$ for air and electrical part of task for Si and GO, V_{ph} is phase velocity, and ω – is the angular frequency of an acoustic wave.

Then, the normalization is introduced as follows:

$$Y_i = \omega C_{11}^* U_i / V_{ph}, \quad Y_4 = T_{13}, \quad Y_5 = T_{23}, \quad Y_6 = T_{33}, \quad Y_7 = \omega e^* \Phi / V_{ph}, \quad Y_8 = e^* D_3 / \epsilon_{11}^*, \quad (15)$$

where $i = 1,2,3$, C_{11}^* , ϵ_{11}^* are the normalized material constants of ZnO, Si and GO in crystallographic coordinate system; $e^* = 1$ and it has the dimensional representation of piezoelectric constant.

Substituting (14) in (1)–(8) yields a system of 8, 6, 6, 4 and 2 conventional differential linear equations for ZnO, Si (mechanical part), GO (mechanical part), Si (electrical part), GO (electrical part) and air, respectively, where each system can be written in the matrix form:

$$[A][dY/dx_3] = [B][Y]. \quad (16)$$

Here $[dY/dx_3]$ and $[Y]$ are 8, 6, 6, 4 and 2 dimensional vectors for ZnO, Si (mechanical part), GO (mechanical part), Si (electrical part), GO (electrical part) and air, respectively, whose components are defined according the formulae (15). Matrixes $[A]$ and $[B]$ appeared to be squared and have dimensions of 8 x 8 for ZnO, 6 x 6 for Si and GO (mechanical part), 4 x 4 GO (electrical part), and 2 x 2 for Si and air.

Since matrix $[A]$ is not particular ($\det[A] \neq 0$) we can write the following equations for every contacting medium:

$$[dY/dx_3] = [A - 1][B][Y] = [C][Y] \quad (17)$$

After that, to solve the system of Eq. (17) we need to find the eigenvalues $\beta^{(i)}$ of matrices $[C]$ and corresponding eigenvectors $[Y^{(i)}]$, responsible for the parameters of partial waves, for each of contacting media. General solution would be a linear combination of all partial waves for each medium:

$$Y_k = \sum_{i=1}^N A_i Y_k^{(i)} \exp(\beta^{(i)} x_3) \exp(j\omega[t - x_1/V_{ph}]), \quad (18)$$

where the number of eigenvalues N is 8 for ZnO, $N = 6$ for Si and GO (mechanical part), $N = 4$ for GO (electrical part), $N = 2$ for Si and air. Unknowns A_i and phase velocity V_{ph} can be found using mechanical and electrical boundary conditions (10)–(13) that have also been written in the normalized form (15).

Moreover, as all variables should decay into Si substrate the eigenvalues with positive real parts are eliminated from consideration for nonpiezoelectric half-space. Furthermore, only four eigenvalues with negative real parts are taken into account for the nonpiezoelectric medium, as well as all eigenvalues of corresponding matrix $[C]$ for ZnO film ($0 > x_3 > -h_1$, Fig. 1a,b) and GO film ($-h_1 > x_3 > -h_2$, Fig. 1b).

Finally, since all variables must have decaying amplitudes in air we exclude the eigenvalues with the negative real parts for air ($x_3 < -h_1$, Fig. 1,a; $x_3 < -h_2$, Fig. 1,b). As a result, the described procedure allowed us to calculate the wave phase velocity in structures under the study.

The calculation of the electromechanical coupling coefficient is accomplished using well known formulae [33]:

$$k^2 = 2 \frac{V_{ph} - V_{phm}}{V_{ph}} \times 100\%, \quad (19)$$

where V_{phm} is phase velocity of acoustic waves when the plane $x_3 = -h_1$ is electrically shorted.

3. Experimental

Silicon is well-known as one of the most popular and cheap material in modern electronics. Therefore, design of any, in particular, humidity sensors based on this material is very attractive. On the other hand, Si is not piezoelectric, while acoustic waves are usually generated by exploiting the piezoelectric effect. That is why the silicon substrate should be covered with a piezoelectric layer, say, ZnO film. However, this film itself is not very sensitive towards humidity [14,16]. Therefore, in designing acoustic wave humidity sensor, the other sorbent film above ZnO layer (e.g. GO film) should be additionally applied.

3.1. Preparation of graphene oxide suspension

As in our previous papers [26,36] the graphene oxide (GO) film was fabricated starting from China natural graphite (99.9% C). The material was grinded in a ball crusher 200–300 μm in diameter, oxidized [37], and dried for 6 h at 50–60 $^{\circ}\text{C}$. After that 0.4–0.5 g of graphite oxide was placed in a cylinder chamber (250 ml) together with distilled water (150 ml) and sonicated (20.4 kHz, 0.1–1 W/cm^2) during 15 min. The as-obtained water dispersion with GO particles was centrifuged during 10 min at 8000 rev/min and dried at 70 $^{\circ}\text{C}$ for 6 h [38].

3.2. Design of the humidity sensor

Experimental sample represents common delay line implemented on (001), <110>-Si substrate (4.5 Ω/cm , 500 μm thick) and $\text{C}_6\text{-ZnO}$ film ($h_1 = 3 \mu\text{m}$). Input and output interdigital transducers (IDT) (20 finger pairs) are made on the film by lift-off technique using V-Al film (0.03/0.3 μm , 1.15 Ω/cm). Period of the transducers is $\lambda = 32.1 \mu\text{m}$, the distance between them is $L_{\text{IDT}} = 5 \text{ mm}$, aperture is equal 2 mm and the total phase acquiring an acoustic wave between input and output transducers is $\varphi_0 = 360^{\circ}(L_{\text{IDT}}/\lambda) = 56\,075^{\circ}$.

The fabrication of the c-oriented textured ZnO films with grains of about 0.3 μm is performed in triode sputtering system with dc current, ZnO target, 80% Ar + 20% O_2 gas mixture, and 0.07 Pa pressure. The substrate temperature is 250 $^{\circ}\text{C}$. The rate of the sputtering is 1.2–3 μm per hour.

The GO-based dispersion was deposited onto ZnO film between IDTs by airgraph and dried at the room temperature for 24 h. The film was $2 \times 2 \text{ mm}^2$ in square and 0.45 μm in thickness.

The scheme (a) and photo of the designed humidity sensor in a holder (b) are shown on Fig. 2.

The as-fabricated sample is tested by network analyser (Keysight E5061B). The type of the wave (Rayleigh/Sezawa) is identified from comparison of the calculated phase velocity V_{ph} with the experimental value determined as $V_{\text{ph}} = f \times \lambda$, where f is the wave central frequency and λ is the transducer period equal to the acoustic wavelength.

3.3. Measurement of humidity effect

The changes in characteristics of the Rayleigh and Sezawa waves produced by humidity are measured using experimental setup presented in Fig. 3 [26]. The delay line (DL) is fixed into the chamber (750 ml) and forced by laminar air flux (100 ml/min). Humidity in the flux is varied in the range $\text{RH} = 3.6\text{--}98\%$ by ratio between dry and humid air injected together. The value of the humidity is also controlled by commercial thermo-hygrometer IVTM-7 (threshold 0.1%) located near the tested sensor. The measurements are carried out at room temperature (22 $^{\circ}\text{C}$).

The steps of the measurements are as follows. First, to avoid interaction of humid air with uncontrollable gaseous species preadsorbed on the surface and in the volume, the graphene oxide film was preliminary cleaned by dry nitrogen for about 5 min. Then, the nitrogen was

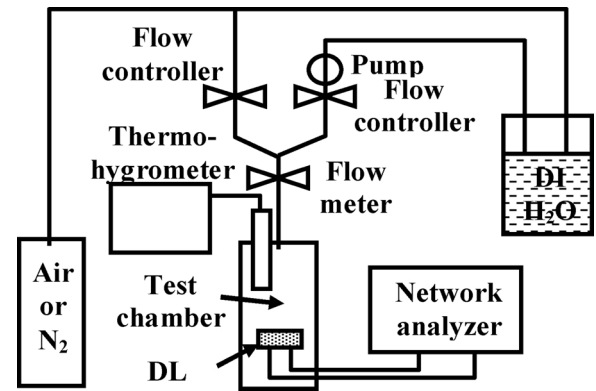
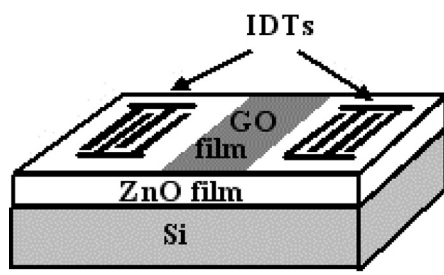


Fig. 3. Experimental setup for testing humidity sensor based on acoustic waves in GO/ZnO/Si structure.

switched off and dry air was introduced into the chamber as reference gas for about 2 min. Finally, the dry air was switched off, the humid air was switched on, and the acoustic response was measured towards dry air for as-cleaned film.

The sensitivity of Rayleigh and Sezawa waves towards humidity is evaluated from the change in the phases $\Delta\varphi$ measured at central frequency. In order to avoid dependence of the measurements on the length of the GO film (L_{GO}) the values of $\Delta\varphi$ are normalized to the total phase $\varphi_0 = 360^{\circ}(L_{\text{IDT}}/\lambda) = 56\,075^{\circ}$ acquiring the wave between input and output transducers and multiplied to enhance factor ($L_{\text{IDT}}/L_{\text{GO}}$). As a result, the normalized responses $R = (\Delta\varphi/\varphi_0)(L_{\text{IDT}}/L_{\text{GO}})$ of the different waves could be compared with each other at identical experimental conditions.

3.4. Measurement of GO film conductivity

The measurement of the electric conductivity of the graphene oxide film was carried out using two-contact method and RF electric bridge (1 kHz) in hermetic box at room temperature and humidity ranged from 5 to 98%. The test sample ($10 \times 3 \times 0.1 \text{ mm}^3$) was placed onto the glass substrate with its surface resistance $\Omega > > 10^{14} \Omega$. At the ends of the sample two silver paste contacts ($\Omega < < 10^{-1} \Omega$) were fabricated. The results of the measurements were recalculated to specific resistance.

It was found that the conductivity of the GO film is increased from 10^{-4} S/m up to 5 S/m. Like in [39] this property may be attributed to the presence of hydroxyl and epoxy groups on the surface of graphene sheets and to absorption of the water molecules by carbonyl and carboxyl groups existing at the edges of sheets or defects.

4. Results and discussion

4.1. Theoretical results

At first step the SAW properties in ZnO/Si structure supporting

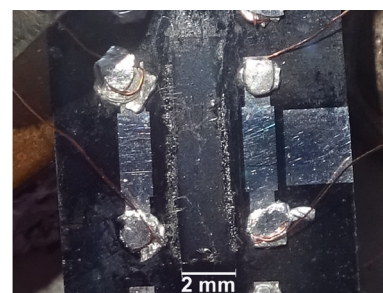


Fig. 2. (a) The scheme and (b) photo of the designed humidity sensor in a holder.

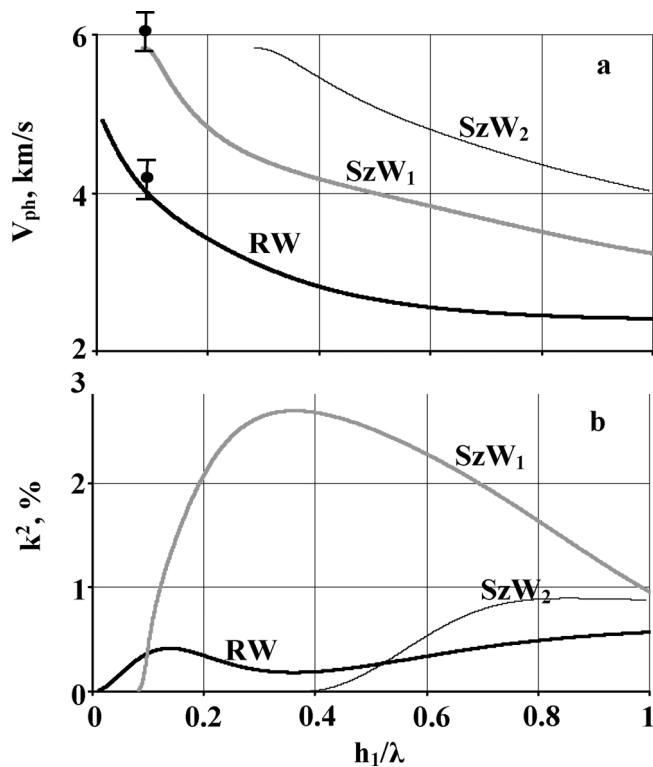


Fig. 4. (a) The phase velocity and (b) electromechanical coupling coefficient versus the thickness of ZnO film h_1/λ (λ is wavelength) in ZnO/Si structure. RW – Rayleigh wave; SzW₁, SzW₂–1st and 2nd Sezawa modes.

Rayleigh and Sezawa waves [40–42] are analyzed using material constants from [43] (Si) and [40,41] (ZnO) (Fig. 3). The crystal orientations are (001), <110> for Si (Eugler angles 0°, 0°, 45°) and (001), <100> for ZnO (0°, 0°, 0°).

Fig. 4 shows that for $h_1/\lambda > 0.1$ the k^2 of the 1st Sezawa mode is larger than that is for the Rayleigh wave. It is just this property that is exploited in present paper to improve sensitivity of the SAW humidity sensor based on GO sorbent film whose dominant sensing mechanism is related to the change in film conductivity as approved in [26] by 2 independent experiments. The first one showed that the response calculated from preliminary measured density, elastic moduli, and their changes is much lower than the value measured for the same GO film and substrate material – so that the contribution of the mechanical properties of the film is not the main. Second, the SAW humidity response was measured for one and the same GO film deposited on strong (LiNbO₃) and weak (quartz) piezoelectric substrates. It turned to be out that the former substrate ensured much higher response than the latter one – so that the dominant contribution is originated from electric properties of the film and electric fields of the SAW.

At second step the properties of the Rayleigh and Sezawa waves are examined for the same structure with additional GO film. The film is supposed to be isotropic. The material constants of the film at different relative humidities RH are taken from [26].

The calculations show that before humid air exposure (“clean” GO film) the velocities and k^2 of the waves at h_1 (ZnO) = 3 μm, h_2 (GO) = 0.45 μm, and $h_2/h_1 = 0.15$ are $V_{ph}^{RW} = 3521.5$ m/s, $k^2 = 0.27\%$, and $V_{ph}^{SzW_1} = 5644.6$ m/s, $k^2 = 0.52\%$. After exposure (the film with water molecules), when the thickness of the GO film is doubled [26] (h_2 (GO) = 0.9 μm, $h_2/h_1 = 0.3$), the same velocities become larger: $V_{ph}^{RW} = 3546.4$ m/s and $V_{ph}^{SzW_1} = 5657.4$ m/s.

The increase of the velocities in this case is attributed to the large increase in the GO film thickness. On the other hand, for analyses accounting the increase in the GO film conductivity the effect of water vapor results to the decrease in the wave velocities: $V_{ph}^{RW} = 3511.4$ m/s

Table 1
Results of the theoretical analysis.

h_2^{GO} , μm	h_2/h_1	RH,%	Taking into account only GO film thickness changes		Taking into account only GO conductivity changes ($\sigma_V = 5$ S/m)	
			V_{ph}^{RW} , m/s	$V_{ph}^{SzW_1}$, m/s	V_{ph}^{RW} , m/s	$V_{ph}^{SzW_1}$, m/s
0.45	0.15	0.47	3521.5	5644.6	3511.4	5614.7
0.6	0.2	26	3566.1	5651.5	3556.2	5629.2
0.84	0.28	56	3546.0	5650.1	3538.6	5628.1
0.9	0.3	67	3459.3	5641.5	3453.4	5611.5

and $V_{ph}^{SzW_1} = 5614.7$ m/s at $h_2/h_1 = 0.15$. Therefore, the responses of the waves towards humidity predicted by our calculations may be estimated as 0.3% and 0.53% at RH = 67%, respectively. The corresponding theoretical data are presented in Table 1.

The same calculation for the Lamb wave sensor and GO film/128Y-X + 90° structure [26] gives lower humidity response (0.05%) though the coupling coefficient k^2 of the wave is higher (10.3%). Such sort of contradiction may be explained by stronger energy concentration of the Rayleigh and Sezawa SAWs into GO film than that is for the Lamb wave distributed over the whole film/plate structure.

As a result, theoretical analysis accomplished in this chapter predicts that humidity sensor based on Sezawa wave and GO/ZnO/Si structure should provide better sensitivity than it was achieved before.

4.2. Experimental results

The measured transfer function $|S_{21}|$ of Rayleigh wave (1) and Sezawa family waves (2) in GO film/ZnO film/Si substrate layered structure is shown in Fig. 5. The central frequencies of the waves measured in experiment are, respectively, 134.75 MHz for Rayleigh wave and between 95 and 270 MHz for Sezawa waves. The measured velocities of the waves determined as described in Section 3.2 are 4300 ± 430 m/s (Rayleigh wave 1) and 6600 ± 660 m/s (Sezawa wave 2), where the experimental errors are estimated from precision of the frequency measurements suffered of $|S_{21}|$ ripples and of the averaged period of IDT perturbed by etching process. The family of Sezawa waves consists of a number of modes with various characteristics each.

As an example Fig. 6 demonstrates humidity responses of the Rayleigh (a, c) and Sezawa (b, d) waves measured in ZnO/Si (a, b) and GO/ZnO/Si (c, d) structures at RH ≈ 26%. Selected Sezawa wave has lowest insertion loss among other modes of the family. It is seen that i) responses of both waves much higher with GO film (c, d) than without it (a, b), ii) responses of Sezawa wave ($\Delta\phi_{Sezawa} = -4.46^\circ, -161.42^\circ$) are larger than those are for Rayleigh wave ($\Delta\phi_{Rayleigh} = -2.28^\circ, -16.48^\circ$)

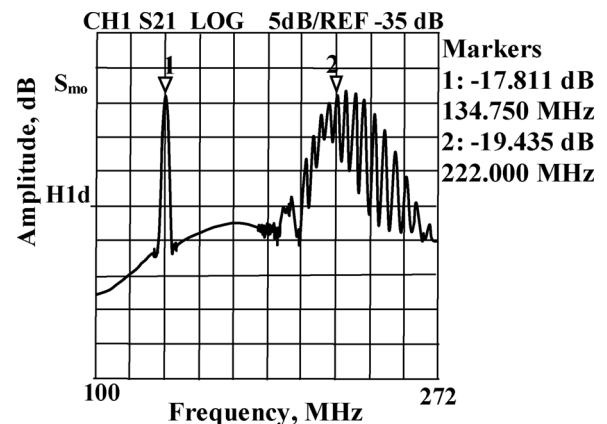


Fig. 5. Transfer function $|S_{21}|$ of Rayleigh wave (1) and one of Sezawa wave (2) in GO film/ZnO film/Si substrate layered structure.

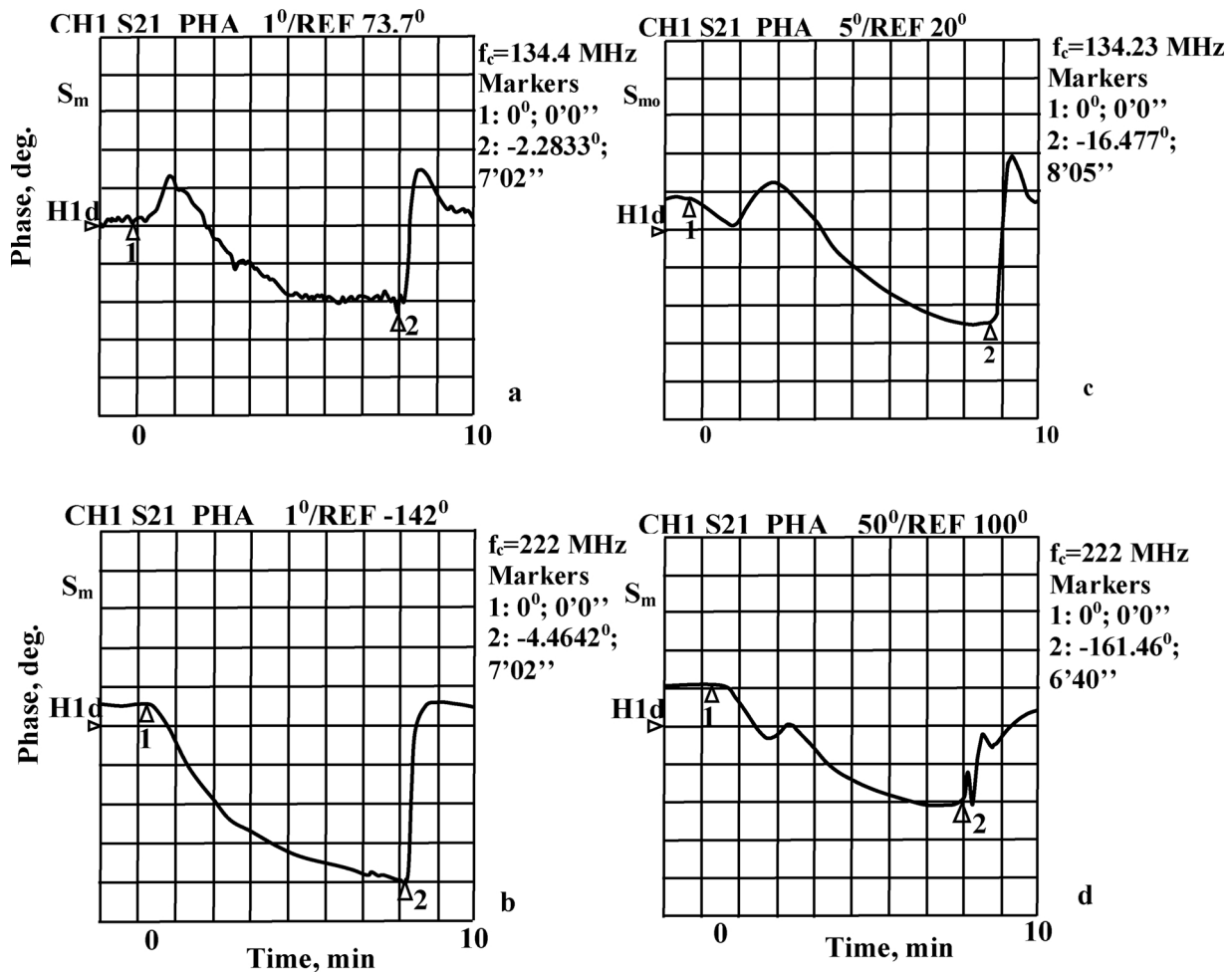


Fig. 6. Humidity responses vs time for (a, c) Rayleigh and (b, d) Sezawa waves measured in (a, b) ZnO/Si and (c, d) GO/ZnO/Si structures at RH \approx 26%. f_c is the wave central frequency. 1 – dry air off, humid air on, 2–humid air off, dry air on.

for both test structures, iii) response times (\approx 600 s) are much longer than those are for recovery (\approx 6 s) for both structures and both waves. Analysis of the same values for the other humidity has shown that the values of the response/recovery times are increased from 500s/5 s at RH = 5% to 700s/18 s at RH = 98%. Like for hydrogen detection with polycrystalline Pd film [4] the large times may be attributed to the porous nature of the sensitive GO film making penetration of the adsorbed water species into the film volume quite difficult, iv) after switching humidity off the output signal restores completely, v) Rayleigh wave response is positive at the beginning and negative at its saturation indicating that the response of the wave is a combination of various sensing mechanisms.

Possible explanation for the initial increase in the Rayleigh wave velocity produced by water vapor adsorption (Fig. 6a,c) is the presence of two opposite sensing mechanisms [44]. On the one hand, the water molecules change mechanical properties of the GO film [26] and ZnO layer [44], i.e. the first mechanism is the elastic loading effect resulting to increase in the Rayleigh wave velocity. On the other hand, the water vapor adsorption increases the GO film conductivity and the mass loading [44] resulting to decrease in the wave velocity. As a result, at initial stage the first sensing mechanism is larger than the second one, while after some time the second mechanism becomes dominant.

As for Sezawa wave, the increase in the GO film thickness produced by water vapor adsorption leads to insignificant increase in the wave velocity (Table 1). This fact is attributed to less localization of the wave near the surface as compared with the Rayleigh wave [45]. As a result, for Sezawa wave in ZnO/Si structure the main sensing mechanism is

every time the mass loading producing decrease in the wave velocity (Fig. 6b) [46], while in GO/ZnO/Si structure the main sensing mechanisms are the mass loading and the electric conductivity decreasing the wave velocity as well (Fig. 6d).

Calibration curves of the humidity sensors based on Rayleigh and Sezawa waves are linear in the range from RH = 20 to 98% (Fig. 7). The slope of the curve for Sezawa wave demonstrates record sensitivity of the sensor as compared both with Rayleigh wave in the same structure (Fig. 7) and with other prototypes known so far (Table 2). On

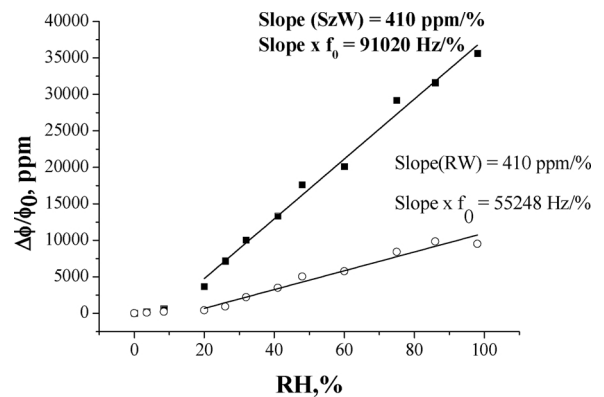


Fig. 7. Calibration curves of the humidity sensors based on Rayleigh(open circles) and Sezawa (black squares) waves in GO/ZnO/Si structure. Solid lines represent the fitting curves obtained by Origin 8,0 Programme.

Table 2
Comparison of the known humidity sensors based on the GO sorbent films.

Wave type	Materials	Humidity range, %RH	Resp./rec. time, s	Sens., kHz/%, RH	Ref.
SAW	Quartz + GO	5–100	n/a	1.54	[5]
SAW	ZnO/glass + GO	0.5–85	19/ < 1	53	[6]
Love wave	Si/SiO ₂ + GO	10–76	n/a	2.4	[17]
QSM	Quartz + GO	6.4–93.5	18/12	2.6	[20]
QSM	Quartz + GO/PEI	11.3–97.3	53/18	27.25	[22]
FBAR	Si/SiO ₂ /ZnO + GO	5–83	n/a	25.5	[23]
A ₀ Lamb wave	Flexible ZnO/PI + GO	10–85	22/5	22	[25]
S ₀ Lamb wave	Flexible ZnO/PI + GO	10–85	16/5	35	[25]
Lamb wave	128Y-X + 90	0.03–75	87/50	80	[26]
Sezawa wave	ZnO/Si + GO	3.6–98	600/6	91	Present paper
Rayleigh wave	ZnO/Si + GO	3.6–98	600/6	6	Present paper

Table 3
Key performance parameters of the humidity sensor based on Sezawa wave propagating in GO/ZnO.Si structure developed in the paper.

Parameter	Values
Central frequency	222 MHz
Insertion loss	19.4 dB
Sensitivity tow. humidity	91020 Hz/%
Sensitivity tow/temperature	9050 Hz/%
Humidity range, RH	3.6–98%
Experimental accuracy	$\Delta RH = \pm 0.1\%$
Resp./rec. times	600 s/6 s
Operation temperature	$20 \pm 5^\circ\text{C}$

the other hand, the temperature instability of the Rayleigh wave sensor estimated from data [47] is lower (4100 Hz/°C) than that is for Sezawa wave device (9050 Hz/°C), while both values are much less than the sensitivity of the same waves towards humidity (55250 Hz/% and 91020 Hz/%, respectively). Therefore, the measurement accuracy for the temperature variations $\pm 1^\circ\text{C}$ is about $\Delta RH = \pm 0.1\%$.

The key performance parameters of the humidity sensor developed in the paper are presented on Table 3.

5. Conclusion

Theoretical predictions and experimental verifications accomplished in the paper showed that Sezawa wave and GO/ZnO/Si structure provide an improved humidity sensor with better performance as compared with other prototypes based on the same film. The sensor has enhanced sensitivity (91 kHz/%) and linear response in the range 20–98%RH. For further improvements of the sensor performance an acoustic wave with stronger piezoelectric properties propagating in GO-based structure should be found.

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