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Acoustic-Electric and Acoustic-Plasmonics Properties of Graphene under the Influence of a Surface Acoustic Waves and an External DC

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***Abstract*— The results of the measurement of the induced acoustoelectric (I_{AEC}) current in graphene under the influence of a surface acoustic wave (SAW) fluctuation character of acoustoelectric current at low bias voltage (V_{bias}) are founded. The fluctuation nature of I_{AEC} is manifested in all cases of measurements depending on the action of a SAW and the application of an external electric field near the point of electrical neutrality. Steady electric and fluctuating character acoustoelectric currents near the point of electrical neutrality, which manifests itself at room temperature take place due to the fluctuation of the electric potential of electrons and holes in graphene under the influence of the elastic deformation forces resulting in the generation of SAW on the surface of the piezoelectric crystal. Also results of an investigation of the Raman spectra of graphene under the in-situ SAW influence and an external electric field bias are showed. A significant change in the characteristic of the G- and 2D-peaks, namely splitting of G-peak into two peaks and red-shift 2D- the peak due to the deformation of graphene under the influence of a SAW are ob-served.**

***Keywords*— graphene, surface acoustic wave, acoustoelectric current, plasmons.**

I. INTRODUCTION

The electronic properties of graphene can be controlled not only by electric fields [1], but using the surface acoustic wave [2, 3]. The ability to generate acoustoelectric current in graphene pos-sible under the influence of SAW [2], and to amplification of SAW amplitudes upon application of an external electric bias on graphene on the surface of the piezoelectric crystal [3]. In this pa-per, we investigate the effect of SAW and an external electric field on the occurrence acoustoe-lectric current in graphene and the Raman spectrum of graphene.

II. MATERIALS AND METHODS

In the experiment, as the substrate for SAW devices coated with graphene piezoelectric crystal langasite $La_3Ga_5SiO_{14}$ (LGS) of JSC "Fomos-Materials", RU, www.ratingtechup.ru, is used. To manufacture the SAW devices Y-cut substrate LGS crystal (100) plane parallel to the crystal surface) were used. The electrode structure of interdigital transducer (IDT) - the source of SAW on the surface of the piezoelectric crystal were formed by electron-beam lithography. IDT con-sisted of 50 pairs of electrodes. On the Y-cut crystal LGS substrate IDT were formed to excite SAW with wavelength Λ = 30 microns at the resonant frequency f = 75.33 MHz.

Graphene film is grown by CVD method by original technology [4]. First, the Ni film was de-posited by the self-ion assisted deposition technique on the surface of an oxidized Si(100). The target was high purity Ni (99.9999%). Ion sputtering was performed in 10^{-6} Torr vacuum. The sputtered film thickness was 0.3 μm. The substrate with the sputtered Ni film was then placed in a quartz reactor tube pumped down to a pressure about 10^{-6} Torr and then inserted into a furnace preheated to 950 ^{o}C. When the samples were heated to the reaction temperature, acetylene was admitted into the quartz tube up to a pressure of 0.4 Torr for 5 s and then pumped out and the quartz tube reactor was extracted from the furnace. Transfer of the resulting graphene was done with the aid of polymethylmethacrylate (PMMA) that was spincoated onto the surface of the graphene film to serve as a support. The PMMA/graphene layer was detached from the substrate by wet-etching of the Ni film with a 1% water solution of hydrochloric acid and then manually laid on the piezoelectric LGS substrate between two IDTs. PMMA was then removed from the graphene surface by exposure to acetone in vapor and then liquid form.

Quality graphene were monitored by using Raman spectra on the spectrometer SENTERRA (Bruker).

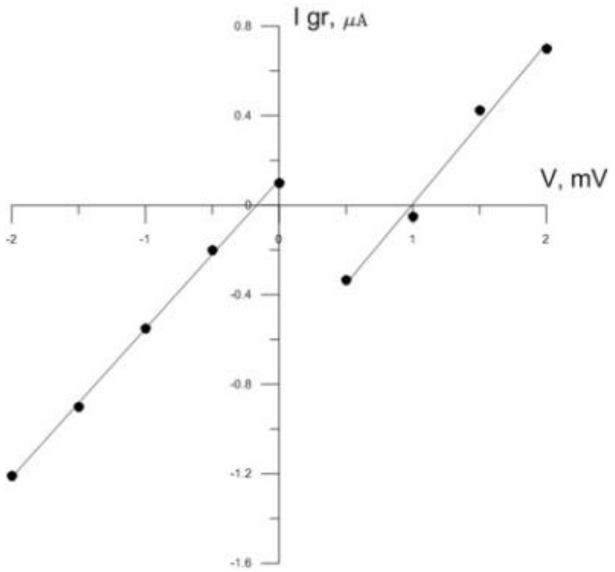
To study the electrical properties of graphene in SAW propagation condition two-Al electrode were formed on the surface of graphene films by electron-beam lithography. The distance be-tween the Al-electrodes is 3 mm.

Generation and registration SAW signal and acoustoelectric current and EMF on graphene per-formed using a high-frequency generator APR-2140 and KEITHLEY 2400 Source Meter according to the electrical circuit given in [3].

Raman spectra of graphene were taken at wavelengths of 488 nm laser excitation, 532 nm and 735 nm, while supplying an external voltage to graphene and SAW.

III. ACOUSTOELECTRONIC PROPERTIES OF GRAPHENE DEPENDING ON AMPLIFICATION CURRENT I_{SAW} FOR SAW POWER AND THE MAGNITUDE AND SIGN OF THE EXTERNAL ELECTRIC FIELD V_{BIAS}

Fig. 1 shows the current-voltage characteristics Igr and I_{AEC} currents through graphene depending on the applied bias voltage Vbias of an external electric field on graphene in the absence of SAW (Fig. 1a, I_{gr} *f* (V_{bias}) and under the influence of SAW (Figure 1b - 1e, I_{AEC} *f* (V_{bias}). I_{gr} - current through the graphene in the absence of SAW, I_{AEC} - induced acousto-electric current through the graphene at different amplification currents I_{saw} for SAW power. Within the statistical error of measurement of current it can be seen a linear relationship I_{gr} *f* (V_{bias}), indicating an ohmic contact on the graphene film. However, for small values V_{bias} (near $V_{bias} \approx 0$) is not possible to measure the value I_{gr} because of large fluctuations in its value due to large measurement errors. It should be noted that in all cases of the measurements of Igr f (V_{bias}) and I_{AEC} *f* (V_{bias}) dependences it do not pass through the point of electrical neutrality, in the absence of V_{bias} (V_{bias} = 0), which indicates that the fluctuation I_{AEC} not only due to large measurement errors. This area shows a decoupling I_{gr} *f* (V_{bias}), Fig.1a and I_{AEC} *f* (V_{bias}), Fig. 1b - 1e.



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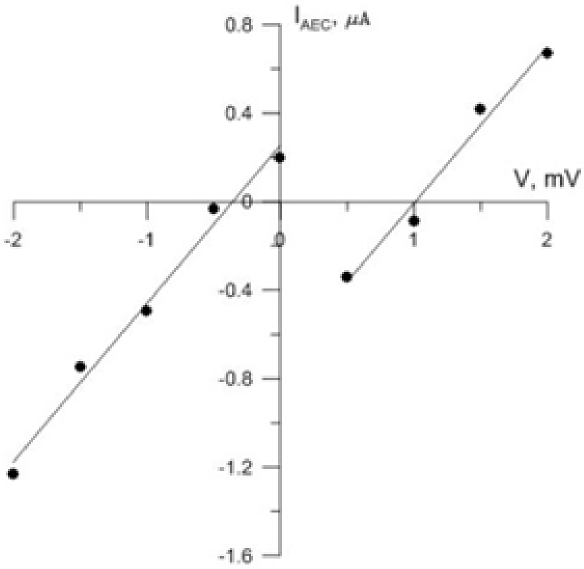

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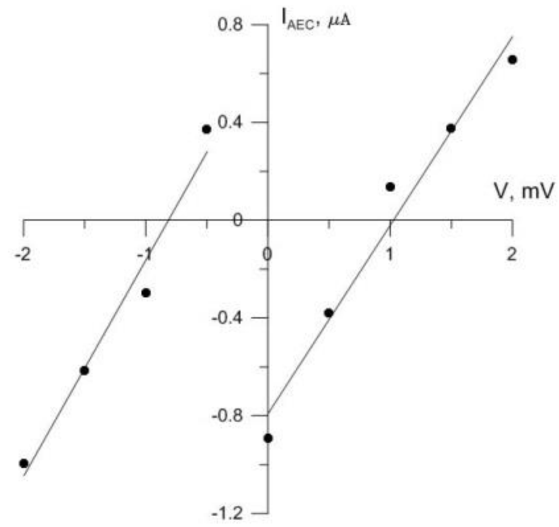

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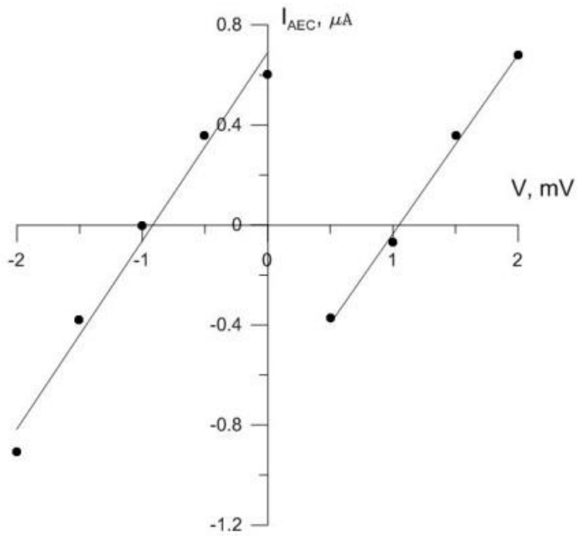

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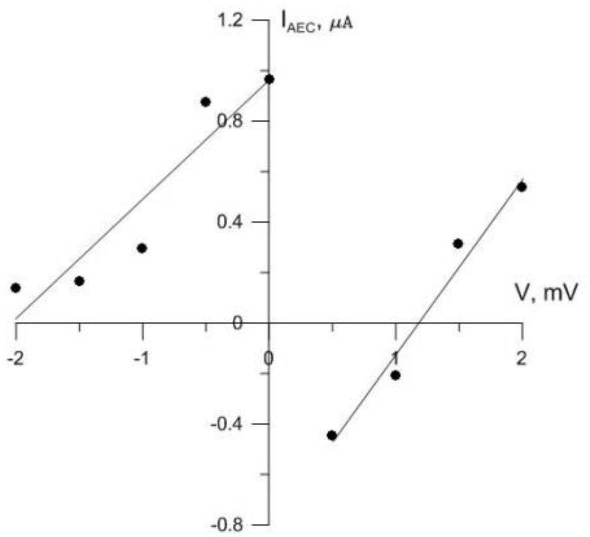


e

Fig. 1. *Dependencies of I_{AEC} f (V_{bias}) under the influence of SAW with a frequency f = 54,2 MHz is shown at various I_{saw} : a) I_{SAW} = 0 μA; b) I_{SAW} = 25 μA; c) I_{SAW} = 50 μA; d) I_{SAW} = 75 μA; e) I_{SAW} = 100 μA. Power SAW source W = 1.07 Wt.*

A more detailed analysis shows that in the low-voltage $V_{bias} \approx 0$, near the point of electrical neutrality [1, 7], there is an interesting feature of the changes in the magnitude and sign of I_{AEC} charge carriers in graphene under the influence of SAW, the magnitude and direction of the applied external electric field potential displacement, V_{bias}. Near field $V_{bias} \approx 0$, both in the positive bias voltage ($V_{bias} > 0$) and negative ($V_{bias} < 0$) there is a change of sign of the current through the graphene I_{gr} and decoupling I_{gr} *f* (V_{bias}), Fig. 1a.

When the SAW turned on the magnitude of the decoupling of I_{AEC} *f* (V_{bias}) dependence near V_{bias} ~ 0 increases (Fig. 1b). With increasing Isaw that gap even more greater (Fig. 1b - 1e). With increasing I_{saw} change of I_{AEC} the sign is observed for large values of negative-bias voltage ($V_{bias} < 0$). For $V_{bias} > 0$ change of current I_{gr} sign for SAW of various power takes place almost at the same value of $V_{bias} \approx 1{,}0$ mV (Fig. 1). The dependence V_{bias} on the Isaw of becomes quadratic when there is a change in the sign I_{AEC}.

The fact that the observed dependence of the sign change of I_{AEC} depending on the direction of the external electric field E_{ext} applied on the graphene shows that the I_{AEC} depends on the direction of the E_{ext} with respect to the direction of SAW propagation. When theirs directions coincide I_{AEC} is enhanced, which is manifested in the increase in the amplitude of SAW [3]. Otherwise I_{AEC} amplification does not occur or is damped. As the results of measurement of I_{AEC} depending on the V_{bias} and I_{saw} in the case of circuitry switching electric field which is used in the experiment allows observing I_{AEC} amplification by applying a V_{bias} < 0. Magnitude of I_{AEC}, arising under the influence of SAW, increases when the $V_{bias} < 0$ applied and decreases with the $V_{bias} > 0$. At large V_{bias} graphene have common characteristic of the traditional ohmic contact (Fig. 1) and fluctuation potential did not observed [7]. I_{AEC} depends on the direction of the applied electric field [5], and is increased when matching SAW propagation direction and an external electric field, and decreases in the opposite directions.

I_{AEC} decoupling at the low near V_{bias} ~ 0 and the change a sign of the charge carriers by peculiar features of the electronic properties of graphene near the point of electrical neutrality under the influence of a SAW are explained.

According to [1] in graphene there are always a chaotic fluctuation potential due to a nanowave and thermal fluctuations two-dimensional graphene. Near the electrical neutrality point graphene have equal electrons and holes concentration [1] that give the zero carrier concentration, a sharp increase in the resistance of the sample is observed.

Action of SAW causes an elastic deformation of the surface layer of the piezoelectric crystal cell which induces its electric polarization. This causes an increase of the existing fluctuations of charges in the graphene, which is manifested in the form of a widening of the gap of dependence I_{AEC} *f* (V_{bias}) with increasing power of SAW and changing the sign of the charge carriers at the low voltage [5].

The parabolic dependence of I_{AEC} on the I_{SAW} and an V_{bias} at the coincidence of their directions can be explained as generation of I_{AEC} in graphene under the influence of SAW depends on the electron-phonon scattering in graphene, and in piezoelectric substrate. As a result, the electron-phonon scattering, which is large in graphene [1], part of the energy, is lost to this SAW. Energy relaxation of acoustic phonons of graphene depends on the temperature of the crystal lattice [6]. The relaxation energies $\mathcal{E}_k$ of extrinsic piezoelectric surface acoustical phonons (PA) of the substrate and intrinsic deformation acoustical (DA) phonons of graphene can be identified through the inverse linear and quadratic energy dependence of the energy relaxation time,

$$\tau_{PA}(\mathcal{E}_k) \sim \mathcal{E}_k^{-1} \qquad (1)$$

and

$$\tau_{DA}(\mathcal{E}_k) \sim \mathcal{E}_k^{-2} \qquad (2),$$

respectively [6].

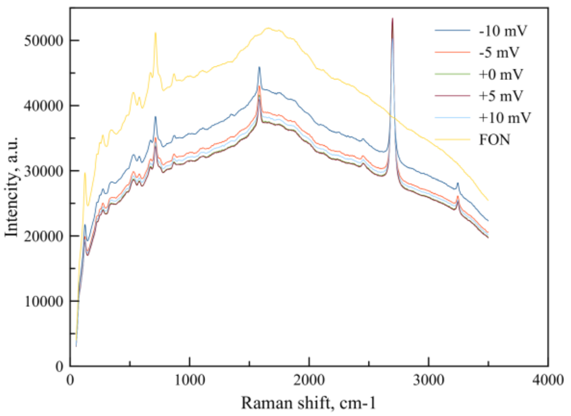
At the low-energy or heat relaxation of acoustic phonons external surface of the piezoelectric substrate is dominant compared to the relaxation of internal deformation acoustic phonons of graphene and qualitatively changes the character of the generation of charge carriers in graphene.

Necessary to consider that any external influence on two- or more- layers graphene has effects not only on existing carriers, but also creates additional carriers [1].

Thus electron-phonon relaxation process in the graphene may be controlled by an external piezoelectric coupling and an external electric field.

IV. FEATURES OF THE RAMAN SPECTRA OF GRAPHENE UNDER THE INFLUENCE OF SURFACE ACOUSTIC WAVE AND AN EXTERNAL ELECTRIC FIELD

Results of the study of the Raman spectra of graphene under the influence “in situ” of SAW and an external electric field bias V_{bias} on the graph showed a significant change in the Raman spectra of graphene and piezoelectric substrate that is associated with the phonon-plasmon interaction in graphene and phonon-electron interaction in piezoelectric crystal, respectively Fig. 2(a - f) and fig.3(a - d).



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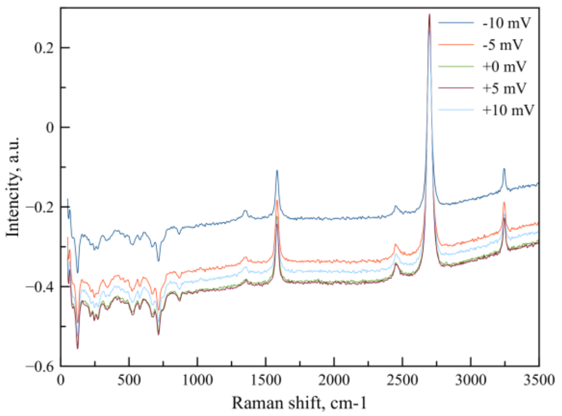

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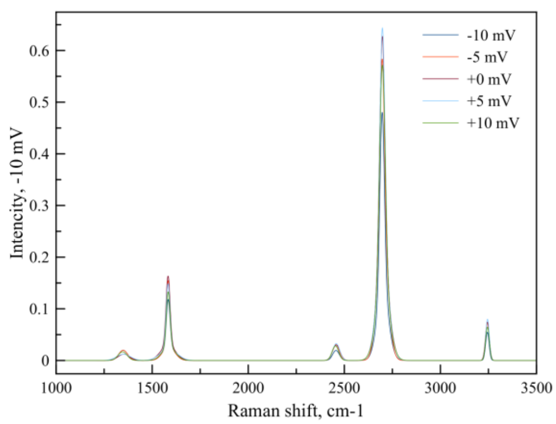

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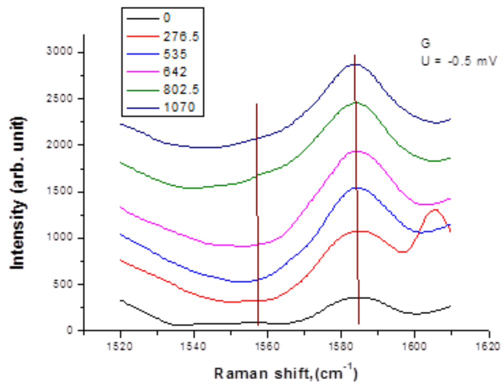
Fig.2. *Raman spectra of LGS crystal - graphene and the substrate (a); Raman spectra of graphene given without background spectra of LGS crystal substrate in the frequency range 0 - 3500 cm^{-1} (b); 1000 - 3500 cm^{-1} (c) at different V_{bias}.*

The analysis of Raman spectra of graphene in the typical D-, G- and 2D- peaks of graphene and satellite the (D + D '') and (2D ') - peaks shows the changes of their intensities depending on the value and sign of the V_{bias} applied to the graphene. Intensivists of G- and 2D- peaks of graphene at negative bias voltage (V_{bias} = - 10 mV; V_{bias} = - 5 mV) higher than without application of an external bias (V_{bias} = - 0 mV), Fig. 2a. Intensity of G- and 2D- peaks for positive V_{bias} = + 5 mV and V_{bias} = +10 mV are reduced relative to V_{bias} = 0 mV, and practically the equal between them for these voltages V_{bias} = + 5 mV and V_{bias} = +10 mV, Fig. 2c. Although there is not a clear and unambiguous change in intensity of the G- and 2D- peaks dependence on the sign and magnitude of the V_{bias}, due to the inhomogeneity of used graphene.

But it is quite clearly that the external electric displacement has an effect on the intensity of the Raman peaks of graphene (Fig. 3a - 3f).



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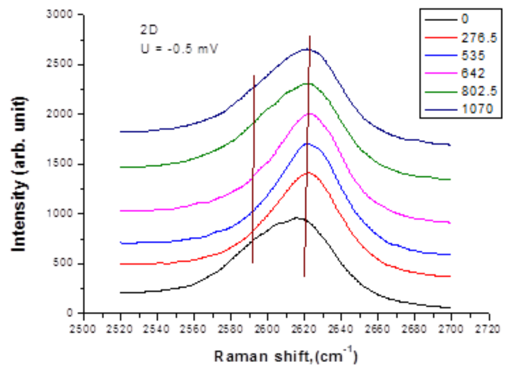

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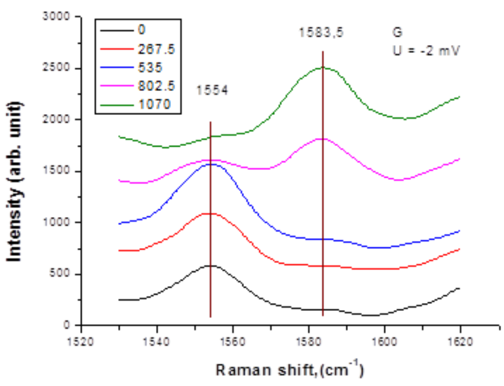

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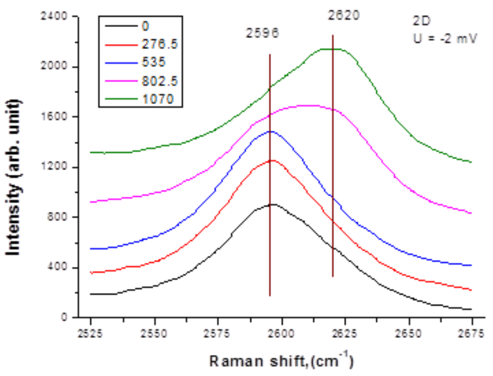

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Fig.3. *Spectra of characteristic G- peaks (a, c) and 2D-peaks (b, d) depending on the graphene SAW power (on the inset shown SAW power in mWt) at different V_{bias}.*

Figure 3 (a - d) shows the changes in the Raman spectra of graphene in the characteristic G- and 2D- peaks depending on the action of SAW and changes its output at different V_{bias}. An effect of SAW changes the character and appearance of the Raman spectra (Fig. 3a – 3d) of graphene. As seen from Fig. 3a and 3b, which show the Raman spectra of graphene at various SAW power and at low V_{bias} = - 0.5 mV (or its absence V_{bias} = 0 mV, which are not shown since identical spectra) with increasing SAW power graphene (I_{saw}) spectrum intensity shift occurs at almost unchanged their absolute intensity relative to the Raman base excitation of piezoelectric substrate. This suggests that an increase of the SAW power themselves the intensity of characteristic G- and 2D-peaks of graphene does not change but the excitation level of piezoelectric substrate changes, in our case LGS- substrate. At a small V_{bias} or absence V_{bias} Raman spectra changes occurs substantially in piezoelectric substrate than graphene. The intensity Raman spectra growth only for base LGS – substrate excitation (Fig.2a). The intensity of the G- and 2D- peaks of graphene does not change, nor change their basic resonance frequency 1584 cm^{-1} (G- peak, Fig. 3a) and 2620 cm^{-1} (2D- peak, Fig. 3b).

But when use a negative bias voltage on the graphene (V_{bias} = - 1,0 mV, - 1,5 mV, - 2,0 mV) and increase of SAW power occur splitting of G- peak into two peaks at the frequencies 1583.5 cm^{-1} and 1554 cm^{-1} (Fig. 3c) and offset 2D- peak frequency of 2620 cm^{-1} to 2596 cm^{-1} (Fig. 3d) is observed. The difference for frequencies splitting of G- peak and 2D-peak is ~ 30 cm^{-1} and ~ (24 - 28) cm^{-1}, respectively. This experimental fact indicates the effect of I_{AEC} stimulated by SAW and E_{ext} in graphene on the intensity and form of G- and 2D- peaks of graphene.

Similar results - the splitting of G-peak into two peaks and red-shift of 2D- the peak observed in the Raman spectra when there are the mechanical deformation of tension and compression of graphene [7]. The authors explain these results by change of shift of the phonon frequencies in the deformed graphene. When deformation of graphene formed deformation defects, which is manifested in red-shift 2D- peak. Splitting of G-peak into two G+ - and G- - peaks occurs because of the shift of the phonon frequency of graphene perpendicular or parallel to the axis of deformation of graphene, respectively [7]. The difference of splitting frequencies under uniaxial deformation of graphene for Δ G+ - peak ~ 10.8 cm^{-1} and ΔG- - peak ~ 31.7 cm^{-1} with respect to G- peak, in the absence of deformation, and the shift of the 2D-peak is 64 cm^{-1}.

Under the influence of SAW graphene, located on the surface of the piezoelectric crystal experiencing the same strain that is experiencing the surface of the piezoelectric crystal during propagation of SAW. This was visualized using X-ray diffraction of synchrotron radiation synchrotron BESSY II (Berlin, Germany) by deformation of graphene on a surface of the piezoelectric Y- cut LGS- crystal when exposed to SAW having a wavelength of 30 microns and a resonance frequency f = 75.33 MHz [5].

The external electric field on a double layer and three layer graphene and more layer create an additional charge carriers [1], and the processes of decay and gain the intensity of plasmons and shifts of wavelength plasmons, and to control by it [8].

The dependence of the intensities of G- and 2D-peaks of graphene on the I_{AEC}, induced by the SAW and E_{ext} in graphene is due to plasmons properties of graphene when exposed to infrared light, the exciting Raman shift. Previously is shown [8] that an increase in the intensities of the G- and 2D- peaks in the Raman spectra of graphene with noble-metal nanostructures, containing embedded in it nanoparticles Ag. Nanoparticles Ag, surrounded by a shell SiO_2 dielectric resonators generates plasmons its exposed to infrared light. When illuminated metal nanoparticles (NPs-Me) with size (a_{Me}) which is much smaller than the light absorption layer, the skin layer (l_{sl}) of metal, $a_{Me} \ll l_{sl}$, NPs-Me surface generate plasmons. At multiple reflection light from the dielectric shell with at a frequency of the plasmons excitation, the generation power of radiation increases with resonance, localized plasmons inside the shell. This energy of localized plasmons propagates without loss across graphene over long distances up to (10 – 100) μm up to graphene boundaries, and repeatedly reflected from the boundaries of graphene "puddles" increase the intensity of the generation of plasmons, which manifests itself in increasing the intensity of the characteristic Raman spectrum G- and 2D- peaks of graphene [8].

As a result, deformation of graphene by the effect of the SAW "superlattice" is formed [9], which is fully modulated "superlattice" by the surface of the piezoelectric crystal and SAW parameters [5]. And this "superlattice" allows you to initiate a long-lived graphene plasmons [9] and a tremendous increase in the intensity of the surface plasmon, which is observed in our experiment as increase in the intensity of the characteristic G- and 2D- peaks Raman spectrum of graphene under the influence of a SAW, and an external electric bias. In addition, the nanoscale morphological defects as atomic steps and kinks on epitaxially grown on SiC graphene, can be sources of plasmon radiation [10].

Based on the analogy of our results with the results in [6 - 10], we assume that the observed splitting of the G-peak two G + - and G- - peak and redshift the 2D-peak in the Raman spectra of graphene caused by deformation of graphene under action of SAW and influence of the external electric field.

Thus, by using Raman spectroscopy of graphene under the influence of in-situ SAW and an external electric field bias to graphene substantial change of characteristic of G- and 2D- peaks, the G-peak splitting into two peaks, and 2D-peak red shift due to the deformation of the graphene under the influence of SAW is found.

V. RESULTS AND CONCLUSIONS

1. Results of the IAEC measurements induced in graphene under influence of SAW establish the specific fluctuation of IAEC near the point of electrical neutrality, at low voltages of external bias, Vbias. The fluctuation nature of the IAEC is manifested in all cases of measurements depending on the effect of SAW and Vbias that observing at room temperature in air.

2. The parabolic dependence of the IAEC f(Isaw) explained by mechanism of the relaxation of acoustic phonons of piezocrystal substrate.

3. The results of the study of the Raman spectra of graphene under the influence of “in situ” SAW and Eext on a graphene showed a significant change in the characteristic of the G- and 2D- peaks: G-peak splitting into two G+-, G- -peaks and red-shift the 2D- peak due to the deformation of graphene under the influence of a SAW and Vbias.

4. The change intensity of the G- and 2D- peaks of graphene under the effects SAW and Eext ex-plained by plasmonic properties of graphene, which increased as a result of multiple reflections from the "superlattice", formed by deformation of graphene under the influence of SAW and Eext and at the boundaries of graphene "puddles". The ability to control the magnitude and direction of IAEC induced in graphene by SAW have of practical importance.

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