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HIGH-FREQUENCY ACOUSTIC PHENOMENON IN QUASI-TWO-DIMENSIONAL ORGANIC CONDUCTORS

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The propagation of a monochromatic high-frequency longitudinal acoustic wave along the normal to the layer (z-axis) in a bulk quasi-two-dimensional conductor (Q2D), in the absence of an external magnetic field under assumption of arbitrary scattering of the charge carriers from the boundary surface, is studied theoretically. The effect of the Q2D electron energy spectrum features on the attenuation of an ordinary acoustic wave (OAW) and of an anomalous acoustic wave (AAW) is discussed. It is shown that for certain values of the characteristic parameters, the OAW in Q2D is strongly attenuated, and the asymptotic form of the acoustic field in the conductor is determined by the AAW. In the case of a Q2D conductor, the anomalous acoustic field is transmitted by electrons belonging to the belt points of the Fermi surface.

Keywords: High-frequency acoustic wave; quasi-two-dimensional conductor; scattering of conduction electrons; ordinary acoustic wave; anomalous acoustic wave.

1. Introduction

Interest in studying the physical properties of conductors with reduced dimensionality and organic origin has been heightened in connection with the search for new superconducting materials with high critical parameters. A considerable number of the superconductors synthesized in recent years have layered structures and a very distinct anisotropy of the electrical conductivity in the normal (not superconducting) state. Many organic layered conductors have metal-type electrical conductivity even across the layers, and the well-developed concept of quasi-particles carrying a charge in metals can be applied for describing their electronic properties (see Refs. 1–3 for more details). In layered conductors with a quasi-two-dimensional

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(Q2D) electron energy spectrum, the charge carrier energy

$$\varepsilon(\mathbf{p}) = \sum_{n=0}^{\infty} \varepsilon_n(p_x, p_y) \cos\left(\frac{anp_z}{\hbar}\right), \tag{1}$$

depends weakly on the quasi-momentum projection $p_z = \mathbf{p} \cdot \mathbf{n}$ due to the sharp anisotropy of the velocity distribution of the charge carriers, $\mathbf{v} = \partial\varepsilon(\mathbf{p})/\partial\mathbf{p}$ on the Fermi surface $\varepsilon(\mathbf{p}) = \varepsilon_{\mathrm{F}}$. The Fermi surface is a slightly corrugated cylinder with, possibly, small closed cavities belonging to anomalously small groups of charge carriers. Here a is the layer separation and $\hbar$ is Planck's constant. The coefficients ε_n in Eq. (1) rapidly decrease with increasing number n, so that the maximum value of the function $(\varepsilon_{\mathrm{F}} - \varepsilon_0(p_x, p_y)) = \eta\varepsilon_{\mathrm{F}}$ on the Fermi surface $\varepsilon(\mathbf{p}) = \varepsilon_{\mathrm{F}}$ is much smaller than the Fermi energy ε_{F}, i.e., $\eta \ll 1$. η is the quasi-two-dimensionality parameter of the electron energy spectrum.

High-frequency acoustic phenomena in organic layered conductors in the presence of a magnetic field are well-studied, both theoretically and experimentally. These studies show a number of characteristic features which appear distinct from the corresponding properties of 3D conductors (See Ref. 4 and the references therein). Theoretical studies that have been devoted to the investigation of propagation of a high-frequency acoustic wave in metals show that the anomalous acoustic wave (AAW) can also be observed in the absence of a magnetic field.[5,6] This opens new possibilities of studying the electronic properties of the conductors. In this respect, up to now, few theoretical papers appeared concerning high-frequency acoustic phenomena in bulk Q2D conductors,[7,8] as well as in a thin Q2D conductive film[9] in the absence of an external magnetic field. It is well-known that in Q2D conductors, as in the case of metals, there exist an ordinary acoustic wave (OAW) whose velocity of propagation is close to the sound velocity s, and a non-exponentially attenuated anomalous acoustic wave (AAW) determined by electrons whose velocity component in the direction of propagation of the wave reaches the highest value, $v_z^{\max} \approx \eta v_0$. The OAW is transmitted into the conductor by the electrons which are in phase with the acoustic wave, i.e., electrons whose velocities are almost normal to the wave vector. The excitation of the AAW is due to the "dragging" of the acoustic field directly by the conduction electrons. In the case when a longitudinal acoustic wave propagates in the conductor, these are the electrons whose velocities are along the wave vector and their interaction with the electric field accompanying the wave is most effective.

In the present paper, in order to compare the nature of the attenuation of the OAW and AAW in a Q2D conductor, in the case of arbitrary scattering of the charge carriers from the boundary surface, the amplitudes of the OAW and AAW are theoretically analyzed in the function of certain characteristic parameters defined as the ratios of the charge carriers velocity and sound velocity, the frequency of the incident wave and collision frequency for the charge carriers, and the distances from the boundary surface and charge carriers mean free path.

2. Theoretical Problem

Consider a monochromatic longitudinal acoustic wave propagating along the low conductivity axis (z-axis) of a Q2D bulk conductor ($z \geq 0$). The displacement $U(0,t) = U_0 \exp(-i\omega t)$ of the surface ($z = 0$) is given. We assume that the temperature is fairly low, so that the charge carriers scatter mainly on impurities. In this case, we can confine the discussion to the relaxation-time approximation and analyze the distinctive features introduced due to the Q2D nature of the electron energy spectrum.

The complete system of equations describing the propagation of acoustic waves in conducting media consists of the Boltzmann kinetic equation for the charge carrier distribution function, $f(\mathbf{r},\mathbf{p},t) = f_0(\varepsilon) - \chi(\mathbf{r},\mathbf{p})\partial f_0/\partial\varepsilon$ ($\chi(\mathbf{r},\mathbf{p})$ is the nonequilibrium additive term to the equilibrium distribution function), the Maxwell equations and the equations of elasticity.[10]

The Boltzmann kinetic equation for charge carriers, written in the relaxation-time approximation, takes the form

$$\frac{\partial\chi}{\partial z} - i\frac{\omega^*}{\upsilon_z}\chi = eE_z - i\frac{\omega}{\upsilon_z}\Lambda_{zz}\frac{\partial U}{\partial z}\,, \tag{2}$$

where $\omega^* = \omega + i\nu$, $\boldsymbol{\upsilon} = \{\upsilon_x, \upsilon_y, \upsilon_z\}$ is the velocity of the charge carriers, ν is the collision frequency and Λ_{zz} is the deformation potential tensor component. For simplest energy-momentum relation for the charge carriers (the first two terms in (1)):

$$\varepsilon(\mathbf{p}) = \frac{p_x^2 + p_y^2}{2m} + \eta\upsilon_0\frac{\hbar}{a}\cos\left(\frac{ap_z}{\hbar}\right), \quad \upsilon_0^2 = \frac{2\varepsilon_{\mathrm{F}}}{m}\,, \tag{3}$$

the reduced deformation potential Λ_{zz} depends only on p_z:

$$\Lambda_{zz} = \eta L\,\cos\left(\frac{ap_z}{\hbar}\right). \tag{4}$$

An acoustic wave in a conductor always generates a varying electromagnetic field accompanying it. The longitudinal electric field $E_z(z)$ in conductors with a high charge-carrier density can be derived from the electrical neutrality condition $\langle\chi\rangle = 0$, which is equivalent to the continuity condition for the current, i.e.,

$$j_z(z) = e\langle\upsilon_z\chi\rangle = 0\,. \tag{5}$$

The acoustic field $U(z)$ in the conductor can be described by Eqs. (2), (5) and the equation of the theory of elasticity:

$$\frac{\partial^2 U}{\partial z^2} + \left(\frac{\omega}{s}\right)^2 U = \frac{1}{\rho s^2}\left\langle\Lambda_{zz}\frac{\partial\chi}{\partial z}\right\rangle, \tag{6}$$

where ρ is the density of the medium and s is the acoustic wave velocity.

The angular brackets in the expressions above indicate standard integration over the Fermi surface

$$\langle\cdots\rangle = -\frac{2}{(2\pi\hbar)^3}\int(\cdots)\frac{\partial f_0}{\partial\varepsilon}d^3p\,. \tag{7}$$

The behavior of the acoustic field in the conductor generally depends on the type of scattering of the charge carriers on the boundaries. In this respect, the kinetic equation (2) must be supplemented with the corresponding boundary condition

$$\chi_-(\mathbf{p}, z=0) = P\chi_+(\mathbf{p}, z=0)\,, \tag{8}$$

where χ_- and χ_+ are the nonequlibrium terms due to the conduction electrons incident on and reflected from the surface $z = 0$. The properties of the sample surface are taken into account through the specular reflection parameter P. It is defined in such a way that $P = 1$ for purely specular reflection and $P = 0$ for purely diffuse reflection.[11] Hence, values of P in the range $0 < P < 1$ might be interpreted as probabilities for specular reflection of the conduction electrons.

3. Results and Discussion

After Fourier transforming Eqs. (2), (5) and (6) with respect to z, and using the appropriate boundary conditions (8), one obtains the following algebraic solution for the acoustic field in the Q2D conductor

$$U^k = \frac{1}{-(\omega/s)^2 + k^2(1+G_k)} \left\{ -\frac{ik}{\pi} U(0) \left(1 + G_k + \frac{P-1}{P+1} G_k \right) + \int_{-\infty}^{\infty} K(\omega,k,r) r U^r dr \right\}, \tag{9}$$

where $U(0)$ is the displacement at $z = 0$. The functions G_k and $K(\omega,k,r)$ are

$$G_k = \frac{1}{\rho s^2}\frac{\omega^*}{\omega}\left[\left\langle \frac{\omega^2\Lambda_{zz}^2}{v_z^2}\frac{1}{-(\omega/s)^2+k^2}\right\rangle - \left\langle \frac{\omega k}{\omega^*}\frac{\Lambda_{zz}}{-(\omega/s)^2+k^2}\right\rangle^2 \left\langle \frac{1}{-(\omega/s)^2+k^2}\right\rangle^{-1}\right] \tag{10}$$

$$K(\omega,k,r) = \frac{P-1}{P+1}\frac{i\omega^{*2}\omega k}{\pi\rho s^2}\left[\left\langle \frac{\Lambda_{zz}^2 v_z^2}{|v_z|[-k^2v_z^2+\omega^{*2}][-r^2v_z^2+\omega^{*2}]}\right\rangle\right]. \tag{11}$$

Using the expressions for the dispersion relation Eq. (3) and deformation potential Eq. (4) and integrating over the Fermi surface by Eq. (7), one obtains the following expressions for G_k and $K(\omega,k,r)$

$$G_k = \frac{mL^2}{2\pi^2\hbar^2\rho v_0^2 a}\frac{\omega\omega^*}{k^2s^2}\left[1 - \sqrt{1 - \left(\frac{k\eta v_0}{\omega^*}\right)^2}\right], \tag{12}$$

$$K(\omega,k,r) = -\frac{i}{\pi}\frac{P-1}{P+1}\frac{mL^2\omega\eta}{\pi^3\hbar^2\rho s^2 v_0 a}\left[A - B\right], \tag{13}$$

where the notations are

$$A = \mathrm{ArcSinh}\left[\left|\frac{k\eta v_0}{\omega^*}\right|\right]\sqrt{\left(\frac{\omega^*}{k\eta v_0}\right)^2 - 1}\,,$$

$$B = \mathrm{ArcSinh}\left[\left|\frac{r\eta v_0}{\omega^*}\right|\right]\sqrt{\left(\frac{\omega^*}{r\eta v_0}\right)^2 - 1}\,.$$

The corresponding dispersion function

$$D_k = (1 + G_k)\left(\frac{ks}{\omega}\right)^2 - 1\,, \tag{14}$$

besides the zeros $k = \pm k_0$ with

$$k_0 = \frac{\omega}{s}(1 - c - (c\alpha)^2 - ic\delta + c\sqrt{(1 + c\alpha^2 + \delta)^2 - \alpha^2})\,, \tag{15}$$

also has a branch point $k_1 = \pm\omega^*/(\eta v_0)$. The branch point is due to a belt on the Fermi surface with the maximum value of the electron velocity $v_z^{\max}$. Here,

$$c = \frac{mL^2}{\pi^3\hbar^2\rho a v_0^2} \sim \frac{m}{M}\,, \quad \alpha = \frac{\eta v_0}{s}\,, \quad \delta = \frac{\nu}{\omega}\,,$$

where m and M are the electron and ion mass, respectively.

Using the inverse Fourier transformation on Eq. (9), the acoustic field in the conductor can be represented as a sum of an OAW (U^{OAW}) whose velocity of propagation is close to the sound velocity s, and an AAW (U^{AAW}) determined by electrons with velocity $v_z^{\max} \approx \eta v_0$

$$U(z) = U^{\mathrm{OAW}}(z) + U^{\mathrm{AAW}}(z)\,. \tag{16}$$

The amplitude of the OAW can be determined if the residues at the roots of the dispersion equation (14) are calculated

$$U^{\mathrm{OAW}}(z) = U(0)\frac{f^2 + 2c[1 + i\delta - \sqrt{(1 + i\delta)^2 - \alpha^2 f^2}]}{f^2 + cf^2\alpha^2[(1 + i\delta)^2 - \alpha^2 f^2]^{-1/2}}\exp\left[if\alpha\frac{b}{\delta}\right], \tag{17}$$

where $b = z/\eta l$, and $f = (s/\omega)k_0$.

Formulas derived above for the zeros of the dispersion function as well as the amplitude of the OAW, Eqs. (15) and (17) are exact ones. For high frequencies $\omega \gg \nu$ ($\delta \ll 1$), as assumed ($\omega \approx 10^{10}\ \mathrm{s}^{-1} - 10^{13}\ \mathrm{s}^{-1}$), the adiabatic parameter $c = 10^{-4}$ is much smaller than δ, i.e., in Q2D conductor the inequality $c^2 \ll \delta$ is well-satisfied. In this connection, the amplitude of the AAW as well as expressions given below concerning the asymptotic form of the imaginary part of k_0, i.e., the coefficient of attenuation will be calculated in first approximation in the parameter c.

The amplitude of the AAW is determined by the contour integral along the "edges" of the cut in the complex plane from the branch point $k_1 = \pm\omega^*/(\eta v_0)$ of the dispersion function, Eq. (14), to infinity. The following result has been obtained

$$U^{\mathrm{AAW}}(z) = -icU(0)\sqrt{\frac{1}{2\pi}\frac{P-1}{P+1}\frac{\alpha^2}{t}} \times \left(1 - \frac{\alpha(1+i\delta)}{2x\sqrt{L}(1+c\alpha^2 x^{-1/2}/4)}\right)\left(i\frac{\delta}{b}\right)^{3/2} \exp\left[i\frac{b}{\delta}(1+i\delta)\right], \quad (18)$$

where the following notations are used

$$t = (1+i\delta)^2 - \alpha^2\,, \quad x = (1+i\delta)^2 - \alpha^2 L\,,$$

$$L = 1 - \frac{c}{2}\left(1 - \sqrt{[(1+i\delta)^2 + c\alpha^2/4]^2 - \alpha^2}\right).$$

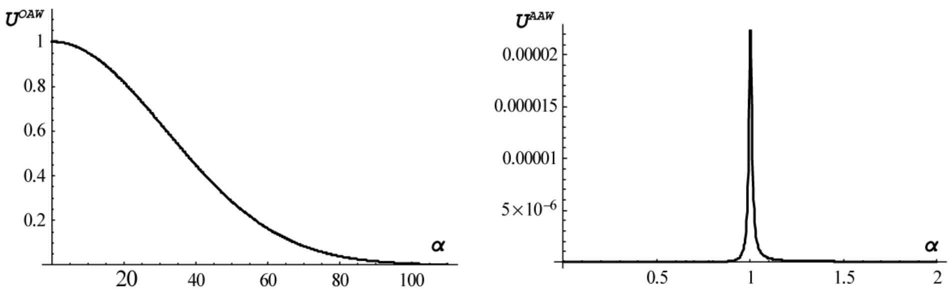
The asymptotic form of the acoustic field in a Q2D conductor depends essentially on the parameters $\alpha = \eta v_0/s$, and $b = z/\eta l$. In the following, the attenuation of the OAW and AAW in a bulk Q2D conductor is analyzed in function of these parameters as well as the dependence of the amplitude of the AAW on the specular reflection parameter P.

The dependence of the amplitude of the OAW on the parameter α is shown in Fig. 1(a). When $\alpha \ll 1$ ($\eta v_0 \ll s$), the attenuation of the OAW is due to the electron scattering. In this case, the coefficient of attenuation does not depend on the value of α as well as on the frequency of the incident acoustic wave ω

$$\Gamma = \mathrm{Im}\, k_0 = c\frac{\nu}{s}\,. \quad (19)$$

As ηv_0 approaches s, the coefficient of attenuation approaches zero, and at $\alpha = 1(\eta v_0 = s)$ the amplitude of the OAW reaches its highest value because the electrons which participate in the attenuation have velocities which are almost normal to the wave vector. These are the electrons which are in phase with the wave.

The coefficient of attenuation increases and the amplitude of the OAW decreases radically, with increasing α. For $\alpha \gg 1$ ($\eta v_0 \gg s$), the OAW undergoes collisionless



(a) (b)

Fig. 1. The dependence of the amplitude of: (a) OAW, (b) AAW versus parameter $\alpha = \eta v_0/s$ for $\delta = 10^{-2}$, $c = 10^{-4}$ and $b = 0.05$.

($\nu/\omega \to 0$) attenuation. The coefficient of attenuation is linear in the frequency

$$\Gamma = \operatorname{Im} k_0 = c\alpha \frac{\omega}{s} \,. \tag{20}$$

In this case, the OAW is strongly attenuated and the acoustic field in the Q2D conductor is completely determined by the AAW.

The dependence of the amplitude of the AAW on the parameter α shows that it has its maximum value also at $\alpha = 1$ [Fig. 1(b)]. In the case of the AAW, the maximum amplitude is due to the participation of the electrons whose velocities are along the wave vector and their interaction with the electric field accompanying the wave is most effective.

Figures 2(a) and 2(b) show the corresponding dependence of the amplitudes of both waves on the parameter $b = z/\eta l$. In a bulk Q2D dimensional conductor, at small distances from the boundary surface, the amplitude of the AAW is insignificant compared to the OAW (as in the case of an isotropic conductor). At $\alpha \gg 1$, and $\delta \gg 1$, the OAW is attenuated at a distance of order of $\eta l \delta$, and the AAW at a distance of order of mean free path of the conduction electrons ηl. It means that the asymptotic form of the acoustic field at $b \gg \delta$, i.e., at distance larger than that reached by electrons in a period of oscillation, is determined by the quasi-wave given by Eq. (18). Specifically, for $\alpha = 100$, the amplitude of the OAW is of the same order as the amplitude of the AAW at $b = 0.196$, and for values larger than this, it decreases radically. For larger values of α, the OAW is attenuated at even smaller distances from the surface, i.e., at smaller b.

At distances such as $\delta \ll b \ll 1$, and when $\alpha \gg 1$, the asymptotic behavior of the AAW is governed by the power-law attenuation of form $z^{-3/2}$ instead of exponential damping.

Comparing the nature of attenuation of both waves in Q2D conductor, one can conclude that the value

$$\alpha_c = \delta/c \,, \quad (\alpha_c \gg 1)$$

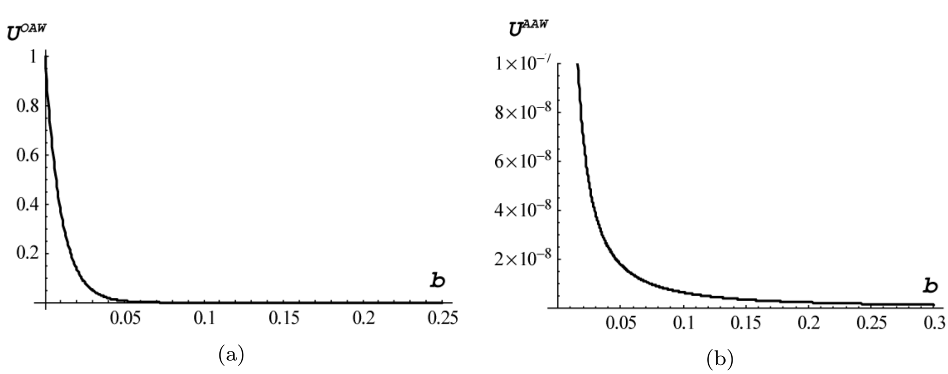


Fig. 2. The dependence of the amplitude of: (a) OAW, (b) AAW on the parameter $b = z/\eta l$ for $\delta = 10^{-2}$, $c = 10^{-4}$ and $\alpha = 100$.

can be considered as a "critical" value for the parameter α below which the acoustic field is determined by the OAW but above which the AAW is dominant.

Let us consider the influence of the scattering of conduction electrons from the boundary surface on the amplitude of the AAW. The graphics for the dependences given here were obtained for $P \approx 1$ which means that we have chosen almost specular reflection of the conduction electrons from the boundary surface. If a purely specular reflection is considered, then the corresponding boundary conditions with $P = 1$ should be taken into account. In that case the following result is obtained for the amplitude of the AAW

$$U_s^{\mathrm{AAW}}(z) = -icU(0)\frac{\sqrt{2/\pi}}{2}\frac{\alpha^4}{t^2}\left(\frac{i\delta}{b}\right)^{3/2}\exp\left[i\frac{b}{\delta}(1+i\delta)\right]. \qquad (21)$$

If P is decreased, which is the probability that the specular reflection is decreased, the amplitude of the AAW given by Eq. (18) is increasing but is still much smaller compared to the amplitude of the AAW given by Eq. (21). The attenuation length of the AAW is slightly increased, and the OAW is strongly attenuated at much smaller distances from the surface. Specifically, for $P = 0.5$, $\alpha = 100$ and $\delta = 0.01$, the OAW is strongly attenuated at $b = 0.5$, and for the same set of parameters, the AAW is attenuated even at $b = 1000$ but its amplitude is much smaller than $U_s^{\mathrm{AAW}}(z)$. The asymptotical behavior of the AAW due to the decreasing of P remains unchanged.

The analyses concerning the attenuation of the OAW and AAW for different values of the characteristic parameters show that the electrons which are involved in the transmittance of the acoustic energy in the Q2D conductor feel the surface of the sample only weakly. Those are so called "glancing" electrons belonging to the belt point of the corrugated cylinder open in the direction of propagation of the acoustic wave. Their velocity vector is almost parallel to the boundary surface, and the corresponding projection onto the normal to layers is $v_z^{\max} = \eta v_0$. This is the velocity of propagation of the AAW.

The detailed investigation of the behavior of the AAW in a Q2D conductor will enable some parameters characteristic for their electronic energy spectrum to be obtained such as the corrugation parameter η, as well as the distribution of the charge carriers velocities on the Fermi surface. It can also be very useful in obtaining information about the dispersion relation and the relaxation properties of the charge carriers which in Q2D conductors generally become more involved than in metals.

Ultrasonic waves constitute excellent tools to probe different electronic states in organic Q2D superconductors, namely, the Mott-insulating, antiferromagnetic, metallic and superconducting states, within a pressure interval of a few hundred bars. Ultrasonic techniques can be used to study quasiparticles and magnetic excitations effects on the elastic properties of superconductors, and attenuation measurements are probably the key experiments that are still to be performed on organic and high T_c superconductors to assess the nature of the superconducting state. Indeed, the smallness of the crystals prohibits the application of standard ultrasonic methods to these materials. The usual pulse echo method cannot, however, be utilized directly since the propagation length (∼0.3 mm) does not allow time separation of transmitted and reflected echoes. Although difficult to adapt to thin organic crystals, pulsed ultrasonic velocity experiments have already been carried out on a few compounds of the κ-(BEDT-TTF)$_2$X family.[12–14] It consists of measuring the phase shift and the amplitude of the first elastic pulse transmitted through the crystal and a delay line; this is why the technique yields only velocity and attenuation variations. A modified ultrasonic technique is used to investigate the elastic velocity in organic superconductors κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ and κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br in their normal state.[12] It was found that the velocity of longitudinal waves propagating along a direction perpendicular to the layers has a rather low value around 2000 m/sec, in agreement with the Q2D character of the structure. Its value can be varied by means of an external pressure in the experiment. On the other hand, it could alter the amplitude of the AAW wave as well as conditions for its attenuation mentioned in the text above. It was also suggested that magnetic field (directed perpendicularly to the layers plane) measurements up to 9 Tesla can also be done by using the same setup as without magnetic field.[12] Actually, in the presence of the magnetic field directed along the normal to the layers, the absorption of the acoustic waves in the case of Q2D organic conductors should be mainly determined by renormalization of the charge carrier energy in the field of the wave. However, if the magnetic field is tilted at angle θ from the normal to the layers, then angular oscillations of the velocity of the conduction electrons as well as the coefficient of attenuation are expected to be observed which is also in agreement with the Q2D character of these conductors.

The newest experimental data[15] show that ultrasonic velocity and attenuation measurements on the quasi-two-dimensional organic conductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br point clearly toward competition between metallic (superconducting) and insulating (antiferromagnetic) phases at low temperatures deep in the Fermi liquid part of the P–T phase diagram.

It could be useful to mention that acoustic measurements of the Q1D organic conductor (TMTSF)$_2$PF$_6$ have also been performed. It was shown that the behavior of the sound velocity and attenuation observed at the transition to the spin-density-wave phase (SDW) ground state 12 K depends on the particular mode propagating in the crystal.[16]

4. Conclusion

In this paper, we have analyzed the attenuation of the OAW and AAW in a Q2D conductor, in the case of arbitrary scattering of the charge carriers from the boundary surface. The model of the simplest charge-carrier dispersion relation is considered, which in many cases permits a correct comprehension of the nature of the electron transport in layered organic conductors. The behavior of both the OAW and AAW is theoretically analyzed in the function of certain characteristic for the propagation of high-frequency acoustic wave parameters. We found that in a Q2D conductor in the absence of an external magnetic field for larger values of the parameter α and at large distances from the boundary surface, the OAW is strongly attenuated and the acoustic field is determined by the AAW. Both waves have substantially different velocities of propagation. The anomalous acoustic field in the case of a Q2D conductor is controlled by the conduction electrons belonging to the belt point of the corrugated Fermi surface. The theoretical analysis of high-frequency acoustic phenomena with or without magnetic field seems to be very useful since they are highly informative and can be used successfully for investigating the electronic structure of layered organic conductors in detail.

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