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ELECTRIC CONDUCTIVITY OF FUNCTIONAL, INCLUDING THERMOELECTRIC, MATERIALS DESCRIBED BY THE FIVAZ MODEL, IN QUASI-CLASSICAL RANGE OF MAGNETIC FIELDS

Development, optimization and use of these or other functional, including thermoelectric, materials for creation of specific devices, elements and systems supposes their adequately precise description on the basis of certain model assumptions of their band spectrum and the mechanisms of charge carrier scattering in them. In some cases the character of band spectrum of these materials directly determines their range of application. An efficient tool of experimental verification, and not infrequently of formation of the above model assumptions, is for instance, investigation of the Shubnikov-de Haas oscillations with different orientations of a magnetic field. However, for a conventional quasi-classical theory taking into account only relaxation time oscillations, in the framework of which, as a rule, processing and interpretation of the experimental results takes place, the specific character of nonparabolicity of charge carrier band spectrum and finite extension of the Fermi surface along the direction of a magnetic field are inessential. This can result in the uncertainty of determination of, for instance, charge carrier concentration, as well as in the controversy of data on the band spectrum of material obtained by different methods. Therefore, the paper introduces an improved theory of the Shubnikov-de Haas oscillations in functional, including thermoelectric, layered materials described by the Fivaz model. The calculations are performed in the framework of Ohm's law applicability for the case when the electric and quantizing magnetic fields are parallel to each other and perpendicular to the layers. In so doing, three factors are taken into account, namely the oscillating dependence of relaxation time on a magnetic field in the range of application of quasi-classical approximation, the nonparabolicity of charge carriers band spectrum described by the Fivaz model and finite extension of the Fermi surface along the direction of a magnetic field.

Key words: Shubnikov-de Haas effect, Fivaz model, relaxation time, density of states, quasi-classical approximation.

Introduction

The Shubnikov-de Haas effect is an efficient tool of studying the band spectrum and mechanisms of charge carrier scattering in functional, including thermoelectric, materials. Investigation of thermoelectric materials in strong magnetic fields, using this effect as well, is addressed in a number of works [1-3]. Study on the Shubnikov-de Haas effect with different orientations of magnetic field permits in the framework of the Lifshits-Kosevich theory [4] to restore the shape of the Fermi surface, as well as to determine the concentration and relaxation times of charge carriers. However, very often in real crystals there are deviations from the Lifshits-Kosevich theory, owing to which, for instance, charge carrier concentrations, found from the Shubnikov-de Haas effect and the Hall effect can differ considerably. The reasons for this difference, in particular, can be nonparabolicity of charge carriers band spectrum and finite extension of the Fermi surface along the direction of a magnetic field that are ignored by conventional Lifshits-Kosevich theory. As a consequence, for instance, at zero concentration of charge carriers, this theory, at least formally, yields physically incorrect result, namely there are oscillations in the absence of the Fermi surface. Theory of electric conductivity of crystals described by the Fivaz model [5], free from these restrictions, is constructed in [6], where it is shown that the nonparabolicity effects can be also pronounced in the case of closed Fermi surfaces. However, in this work, an essential model assumption is made that relaxation time of a longitudinal quasi-pulse is only a function of this quasi-pulse and does not oscillate with a variation of a magnetic field. At the same time, from the results of works [7, 8] it follows that in the case when a fair amount of the Landau levels fit in a narrow miniband describing electron motion perpendicular to layers, a decisive contribution to electric conductivity oscillations is made exactly by the magnetic field dependence of relaxation time. This time already depends on the energy of electron as a whole, rather than on a longitudinal quasi-pulse alone. Therefore, the purpose of this paper is develop a consistent theory of the Shubnikov-de Haas oscillations with regard to all the three factors, namely relaxation time oscillations, band spectrum nonparabolicity in the framework of the Fivaz model and finite extension of the Fermi surface along the direction of a magnetic field.

Derivation and analysis of a formula for longitudinal electric conductivity

In the derivation of a formula for longitudinal conductivity we will follow the procedure described in [8], but modify this procedure with regard to the Fermi surface closedness and its finite extension along the direction of a magnetic field. The energy spectrum of charge carriers in thermoelectric material described by the Fivaz model in a quantizing magnetic field perpendicular to layers is determined as:

$$\varepsilon\left(n,k_z\right)=\mu^* B\left(2n+1\right)+\Delta\left(1-\cos ak_z\right). \qquad (1)$$

In this formula, n, k_z are the Landau level number and quasi-pulse component in a direction perpendicular to layers, respectively, $\mu^* = \mu_B m_0 / m^*$, μ_B, m_0, m^* are the Bohr magneton, free electron mass and electron effective mass in layer plane, respectively, Δ, a are the half-width of mini band in a direction perpendicular to layers and the distance between translation equivalent layers, respectively, B is magnetic field induction.

The relaxation time of a longitudinal quasi-pulse, as with a conventional approach, will be assumed to be inversely proportional to electron states density in a magnetic field. Using the Poisson formula and taking into account the expansion of the Landau levels due to collisions depending on magnetic field B and electron energy ε the relaxation time for the case of energy spectrum (1) can be written as:

$$\tau\left(\varepsilon,B\right)=\frac{\pi\tau_0}{\kappa_\varepsilon}\left\{1-2\exp\left(-\pi kT_{D0}\chi_\varepsilon/\mu^* B\right)\left[\cos\left(\pi\frac{\varepsilon-\Delta}{\mu^* B}\right)\left[C_0 J_0\left(\pi\Delta/\mu^* B\right)+\right.\right.\right.$$
$$\left.\left.\left.+2\sum_{r=1}^{\infty}\left(-1\right)^r C_{2r}J_{2r}\left(\pi\Delta/\mu^* B\right)\right]-2\sin\left(\pi\frac{\varepsilon-\Delta}{\mu^* B}\right)\sum_{r=0}^{\infty}\left(-1\right)^r C_{2r+1}J_{2r+1}\left(\pi\Delta/\mu^* B\right)\right]\right\}^{-1}. \qquad (2)$$

In this formula, τ_0, T_{D0} are, respectively, relaxation time and the Dingle temperature for an open Fermi surface in the absence of a magnetic field, $\kappa_\varepsilon = \arccos(1 - \varepsilon / \Delta)$ for closed constant-energy surface, and $\kappa_\varepsilon = \pi$ for open constant-energy surfaces, $C_0 = \kappa_\varepsilon$, $C_m = \sin m\,\kappa_\varepsilon/m$, $J_n(x)$ are the Bessel functions of the first kind of actual argument. Therefore, for open surfaces (2) goes over to the formula of [8]. Besides, formula (2), unlike the formula of [8], explicitly takes into consideration the fact that in the absence of the Fermi surface the Shubnikov-de Haas oscillations are absent.

To calculate the electrical conductivity of thermoelectric material, we will need a sum of moduli of electron longitudinal velocities in the occupied Landau subbands. According to [8], with regard to (1) it reduces to calculation of integral of the kind:

$$I = \int_0^\infty \cos\left[\pi\left(\frac{\varepsilon}{\mu^* B} - x\right)\right]\sqrt{2\left(\frac{\varepsilon - \mu^* Bx}{\Delta}\right) - \left(\frac{\varepsilon - \mu^* Bx}{\Delta}\right)^2}\,dx. \quad (3)$$

However, the upper integration limit in this integral is not correct. By analogy with formula (2), the upper integration limit should be equal to $\varepsilon / \mu^* B$. This statement is true inasmuch as, according to the Poisson summation formula, the integration variable x comes from essentially positive value $2n + 1$ in formula (1). The above limitation also explicitly takes into account the absence of the Shubnikov-de Haas oscillations in the absence of the Fermi surface [6]. Therefore, in conformity with procedure [8] with regard to (2), as well as the correct upper limit in (3), the formula for full electric conductivity of thermoelectric material at low temperatures with neglecting the field and temperature dependence of chemical potential acquires the following form:

$$\sigma = \sigma_0 + \sigma_1 + \sigma_2 + \sigma_3. \quad (4)$$

In so doing:

$$\sigma_0 = \frac{8\pi^3 e^2 m^* a \tau_0 \Delta^2}{h^4}\left(1 - \frac{\sin 2\kappa_\zeta}{2\kappa_\zeta}\right), \quad (5)$$

$$\begin{aligned}\sigma_1 = 2\sigma_0 R_T R_D \Bigg\{ &\cos\left(\pi\frac{\zeta - \Delta}{\mu^* B}\right)\left[C_0 J_0\left(\frac{\pi\Delta}{\mu^* B}\right) + 2\sum_{r=1}^\infty (-1)^r C_{2r} J_{2r}\left(\frac{\pi\Delta}{\mu^* B}\right)\right] - \\ &-2\sin\left(\pi\frac{\zeta - \Delta}{\mu^* B}\right)\sum_{r=0}^\infty (-1)^r C_{2r+1} J_{2r+1}\left(\frac{\pi\Delta}{\mu^* B}\right)\Bigg\}, \end{aligned} \quad (6)$$

$$\begin{aligned}\sigma_2 = -\frac{8\pi^3 e^2 m^* a \tau_0 \Delta^2}{h^4} R_T R_D \Bigg\{ &\cos\left(\pi\frac{\zeta - \Delta}{\mu^* B}\right)\Bigg[\left(\tilde{C}_0 - \tilde{C}_2\right) J_0\left(\frac{\pi\Delta}{\mu^* B}\right) + \\ &+\sum_{r=1}^\infty (-1)^r \left(2\tilde{C}_{2r} - \tilde{C}_{2r+2} - \tilde{C}_{2r-2}\right) J_{2r}\left(\frac{\pi\Delta}{\mu^* B}\right)\Bigg] + \\ &+\sin\left(\pi\frac{\zeta - \Delta}{\mu^* B}\right)\sum_{r=0}^\infty (-1)^r \left(2\tilde{C}_{2r+1} - \tilde{C}_{2r+3} - \tilde{C}_{|2r-1|}\right) J_{2r+1}\left(\frac{\pi\Delta}{\mu^* B}\right)\Bigg\}, \end{aligned} \quad (7)$$

$$\sigma_3 = -\frac{8\pi^3 e^2 m^* a\tau_0\Delta^2}{h^4} R_D^2 \left\{ \left[C_0 J_0\left(\frac{\pi\Delta}{\mu^* B}\right) + 2\sum_{r=1}^{\infty}(-1)^r C_{2r} J_{2r}\left(\frac{\pi\Delta}{\mu^* B}\right) \right] \times \right.$$
$$\times \left[\left(\tilde{C}_0 - \tilde{C}_2\right) J_0\left(\frac{\pi\Delta}{\mu^* B}\right) + \sum_{r=1}^{\infty}(-1)^r \left(2\tilde{C}_{2r} - \tilde{C}_{2r+2} - \tilde{C}_{2r-2}\right) J_{2r}\left(\frac{\pi\Delta}{\mu^* B}\right) \right] + \tag{8}$$
$$\left. +2\left[\sum_{r=0}^{\infty}(-1)^r C_{2r+1} J_{2r+1}\left(\frac{\pi\Delta}{\mu^* B}\right) \right] \left[\sum_{r=0}^{\infty}(-1)^r \left(2\tilde{C}_{2r+1} - \tilde{C}_{2r+3} - \tilde{C}_{|2r-1|}\right) J_{2r+1}\left(\frac{\pi\Delta}{\mu^* B}\right) \right] \right\},$$

In formulae (5) – (8), ζ is chemical potential of charge carrier gas in material, $C_0 = \kappa_\zeta = \arccos(1 - \zeta/\Delta)$ for closed Fermi surfaces, and $C_0 = \kappa_\zeta = \pi$ for open Fermi surfaces, $C_m = \sin mC_0/m$, $\tilde{C}_m = C_m/C_0$. Reducing multipliers R_T and R_D are given below:

$$R_T = \frac{\pi^2 kT/\mu^* B}{\operatorname{sh}\left(\pi^2 kT/\mu^* B\right)}, \tag{9}$$

$$R_D = \exp\left(-\pi k T_{D0}\kappa_\zeta/\mu^* B\right). \tag{10}$$

The former in a known way takes into account temperature “blurring” of oscillations, and the latter is caused by expansion of the Landau levels due to electron scattering on impurity ions. Exactly this scattering is dominant in the range of existence of the Shubnikov-de Haas oscillations. The nonparabolicity of conduction band of thermoelectric material in formulae (6) – (8) is manifested in the presence of the Bessel functions, and finite extension of the Fermi surface along the direction of the field affects the Shubnikov-de Haas oscillations through modulating coefficients C_m and $\tilde{C}_m$ that become, respectively, zero and unity at $\zeta = 0$, i.e. with disappearance of the Fermi surface. Thus, if the Fermi surface disappears, the Shubnikov-de Haas oscillations disappear “in principle”, rather than “in probability” and “on the average”. Here lies one of the basic distinctions of the proposed approach from the conventional one.

Components of longitudinal electric conductivity entering into formula (4) can be interpreted as follows. Component σ_0 is electric conductivity of thermoelectric material at low temperatures in the absence of a magnetic field. Component σ_1 determines the main contribution to the Shubnikov-de Haas oscillations due to relaxation time oscillations in the range of quasi-classical approximation applicability. Component σ_2 shows itself even in the absence of relaxation time oscillations. It is caused by quantization of phase space in a magnetic field and can become dominant with increasing induction of a magnetic field. In the range of application of quasi-classical approximation, component σ_2 results only in phase shift and certain renormalization of amplitude of the main oscillations determined by component σ_1. Component σ_3 describes the so-called “slow” oscillations [8] whose frequency does not depend on the sections of the Fermi surface by planes perpendicular to a magnetic field. Owing to this, “slow” oscillations are not subject to temperature “blurring”, but decay only due to scattering processes. However, these oscillations disappear alongside with common “quick” oscillations with disappearance of the Fermi surface.

Analysis shows that in the effective mass approximation remain only components σ_0, σ_1, σ_2 and they, respectively, are:

$$\sigma_0 = \frac{32\pi^3 e^2 m^* a\tau_0\Delta\zeta}{3h^4}, \tag{11}$$

$$\sigma_1 = 2\sigma_0 R_T R_D \sqrt{\frac{\mu^* B}{\Delta}} \left[\cos\left(\frac{\pi\zeta}{\mu^* B}\right) C\left(\sqrt{\frac{\zeta}{\mu^* B}}\right) + \sin\left(\frac{\pi\zeta}{\mu^* B}\right) S\left(\sqrt{\frac{\zeta}{\mu^* B}}\right) \right], \tag{12}$$

$$\sigma_2 = -\frac{32\pi^2 e^2 m^* a \tau_0 \Delta^2}{h^4} R_T R_D \left(\frac{\mu^* B}{\Delta}\right)^{3/2} \left[\sin\left(\frac{\pi\zeta}{\mu^* B}\right) C\left(\sqrt{\frac{\zeta}{\mu^* B}}\right) - \cos\left(\frac{\pi\zeta}{\mu^* B}\right) S\left(\sqrt{\frac{\zeta}{\mu^* B}}\right) \right]. \tag{13}$$

In formulae (12) and (13), $C(x)$ and $S(x)$ are the Fresnel cosine- and sine-integrals, respectively. Inasmuch as $C(0) = S(0) = 0$, in this case we also take into account the fact of disappearance of oscillations with the disappearance of the Fermi surface. Transition to conventional quasi-classical approximation in these formulae is matched by condition $\zeta / \mu^* B >> 1$. Then $C(x) = S(x) \approx 0.5$ and we get:

$$\sigma_1 = \sigma_0 R_T R_D \sqrt{\frac{2\mu^* B}{\Delta}} \cos\left(\frac{\pi\zeta}{\mu^* B} - \frac{\pi}{4}\right). \tag{14}$$

Formula (14) corresponds to conventional Lifshits-Kosevich theory for the Fermi surface with the only extreme, namely maximal section by plane $k_z = 0$, perpendicular to a magnetic field. Exactly this formula, or its modifications for the cases of more compound Fermi surfaces with many extreme sections by planes perpendicular to a magnetic field, is generally used for processing experimental data on the Shubnikov-de Haas effect in functional, including thermoelectric, materials, in the case when the electrical and magnetic fields are parallel to each other. Identically, the formula for component σ_2 acquires the form:

$$\sigma_2 = -\frac{16\sqrt{2}\pi^2 e^2 m^* a \tau_0 \Delta^2}{h^4} R_T R_D \left(\frac{\mu^* B}{\Delta}\right)^{3/2} \sin\left(\frac{\pi\zeta}{\mu^* B} - \frac{\pi}{4}\right). \tag{15}$$

From formulae (14) and (15) it is seen that in the case of crystals described by the Fivaz model the condition of applicability of conventional approach whereby components σ_2 and σ_3 are neglected has the form of $\mu^* B / \Delta << 1$. In this case, for instance, component σ_2 is about $\Delta / \mu^* B$ times lower than σ_1. However, in strongly anisotropic crystals the ratio $\mu^* B / \Delta$ is not too low or even high as compared to unity, owing to which component σ_2 is not only negligibly small, but can become dominant. In this case, theory developed in [6] is correct.

Using standard trigonometric formulae, the total oscillating part of electric conductivity for a closed Fermi surface under the conditions of applicability of quasi-classical approximation can be written as:

$$\sigma_{os} = \sigma_0 R_T R_D \sqrt{1 + a^2} \sqrt{\frac{2\mu^* B}{\Delta}} \cos\left(\frac{\pi\zeta}{\mu^* B} - \frac{\pi}{4} + \phi\right). \tag{16}$$

Parameters a and ϕ describe, respectively, renormalization of the amplitude of oscillations and their phase shift. These parameters are determined as follows:

$$a = 3\mu^* B / 2\pi\zeta, \phi = \mathrm{arctg}\, a. \tag{17}$$

Hence, under conditions of applicability of quasi-classical approximation, when $\mu^* B / \Delta << 1$, renormalization of amplitude and phase shift of the Shubnikov-de Haas oscillations as compared to conventional Lifshits-Kosevich theory are small, provided that for a concrete closed Fermi surface the ratio ζ/Δ is not too small as compared to unity.

It formally follows from formula (16) that nonparabolicity described by the Fivaz model does not show itself anyhow in the Shubnikov-de Haas oscillations, if the Fermi surfaces are closed. However, in reality it is not the case. The point is that with the same concentration of charge carriers and closed Fermi surface the Fermi energy of charge carriers in thermoelectric material described by the Fivaz model is somewhat lower than in the effective mass approximation. This difference is due to the fact that with the same energy $\varepsilon < 2\Delta$, i.e. under condition of closed constant-energy surfaces, the density of states in the Fivaz model is higher than in the effective mass approximation. Therefore, even in quasi-classical approximation, as it follows from formulae (16) and (17), the said nonparabolicity should be manifested in the reduction of oscillation frequency, their phase delay and some variation of their relative contribution. The nonparabolicity is even more prominent if instead of quasi-classical formula (16) one uses shrewd formulae (5) – (8) in the framework of the Fivaz model and (11) – (13) in the effective mass approximation. It is evident from Fig.1 which shows that the nonparabolicity described by the Fivaz model results in phase delay of oscillations and rather perceptible reduction of their amplitude. This reduction, just as phase delay, is attributable to the fact that density of states in the Fivaz model is higher, hence, oscillation frequency and relaxation time is lower than in the effective mass approximation.

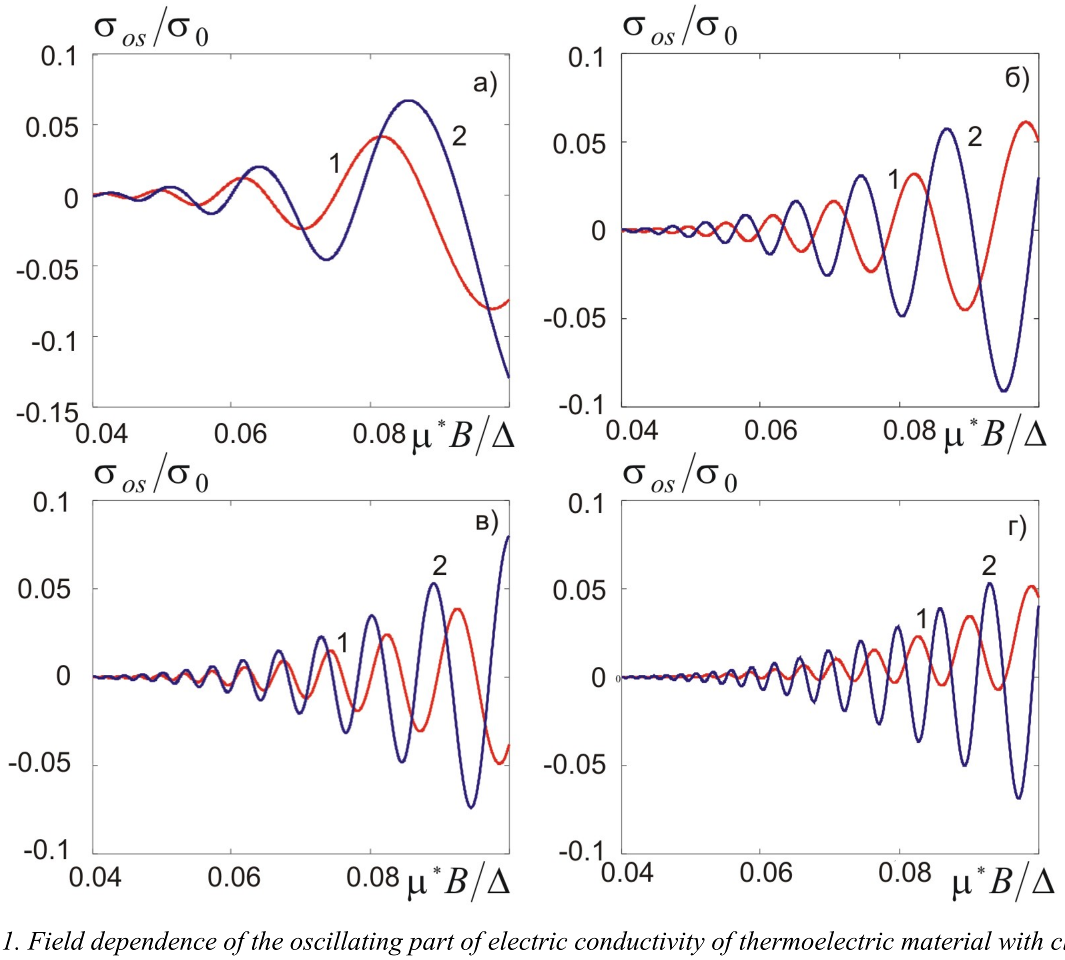

Fig. 1. Field dependence of the oscillating part of electric conductivity of thermoelectric material with closed Fermi surface in conformity with shrewd formulae with ζ/Δ value equal to: a) – 0.5;b) – 1;c) – 1.5; d) – 2. In each figure curves 1 (red) are plotted in the framework of the Fivaz model, curves 2 (blue) – in effective mass approximation.

For comparison, Fig. 2 gives the field dependence of the oscillating part of longitudinal electric conductivity of thermoelectric material in the framework of the Lifshits-Kosevich theory.

For constructing the plots the following crystal band parameters were taken: $\Delta = 0.01$ eV, $m^* = 0.01\ m_0$. These parameters correspond to rather strong crystal anisotropy, since if we take $a = 3$ nm, it turns out that the respective effective mass ratio in directions perpendicular and parallel to the layers is 85, though this is far from being a limit for layered functional materials, including thermoelectric. Moreover, it is assumed that $T = 3$ K and $T_D = 1.5$ K. With such parameters, magnetic fields indicated in the plots correspond to the range from 0.04 to 0.1 T. It was also considered that under condition of constant charge carrier concentration and absolute zero temperature the chemical potential ζ of electron gas in the Fivaz model is related to chemical potential ζ_{em} in the effective mass approximation by the following ratio:

$$(\zeta/\Delta - 1)\arccos(1 - \zeta/\Delta) + \sqrt{2\zeta/\Delta - (\zeta/\Delta)^2} = (2\zeta_{em}/\Delta)^{3/2}/3. \quad (18)$$

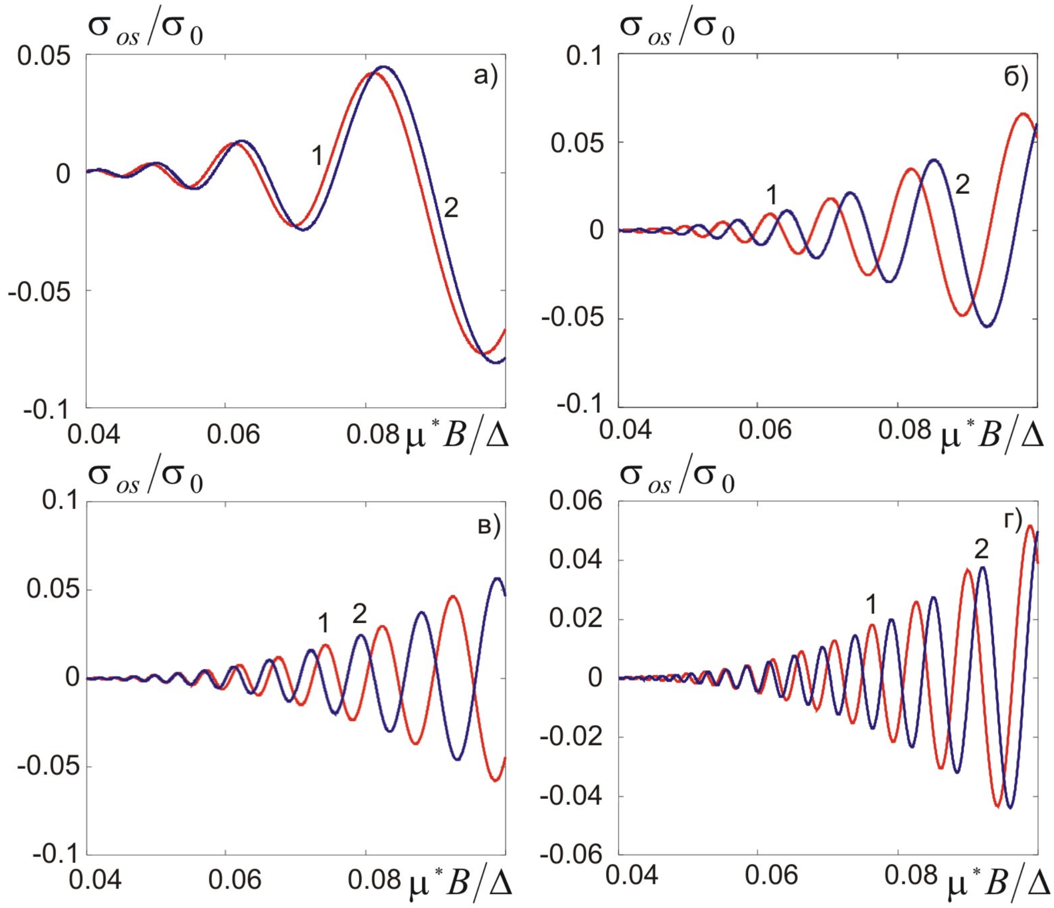

Fig. 2. Field dependence of the oscillating part of electric conductivity of thermoelectric material with closed Fermi surface in the framework of the Lifshits-Kosevich theory with ζ/Δ value equal to: a) – 0.5;b) – 1;c) – 1.5;d) – 2. In each figure curves 1 (red) are plotted in the framework of the Fivaz model, curves 2 (blue) – in the effective mass approximation.

From comparison of Figures 1 and 2 it is seen that with each value of the ratio ζ/Δ curves 1 and 2 constructed by precise formulae differ much wider than the same curves plotted in the framework of the Lifshits-Kosevich theory. This takes place because the Lifshits-Kosevich theory takes into account only an indirect effect of nonparabolicity described by the Fivaz model on the longitudinal electric conductivity of thermoelectric material. This effect is produced through the value of the Fermi energy, i.e. through the frequency of Shubnikov-de Haas oscillations and through the Dingle factor, as long as it depends on the density of states on the Fermi level, and, other things equal, it reduces the amplitude of oscillations the greater, the greater is this density. Whereas shrewd formulae take into account not only an indirect, but also a direct effect of nonparabolicity on the oscillations of conductivity through the Bessel functions. Moreover, shrewd formulae explicitly take into account finite extension of the Fermi surface along the direction of a magnetic field. With the stipulated problem parameters the relative contribution of the oscillating part of electric conductivity reaches 5 – 7 %. This is much higher than in typical metals where this contribution rarely exceeds 0.1 %. The latter fact simplifies considerably the process of investigation of band structure of such functional, including thermoelectric, materials.

Moreover, from Figure 1 it is seen that with increasing the ratio ζ/Δ, i.e. charge carrier concentration, the difference between curves plotted with and without regard to nonparabolicity described by the Fivaz model is increased. The greatest difference between said curves is traced at $\zeta/\Delta = 2$, i.e. for a “transient” Fermi surface. This takes place because the above surface has no tangential planes perpendicular to magnetic field direction, while with any positive value $\zeta/\Delta < 2$ there are two such planes parallel to plane $k_z = 0$ and symmetric with respect to it.

In conclusion, it is necessary to note a series of additional factors determining the validity and the range of application of the results obtained in this paper. First, the results of this paper are valid when the oscillating part of electric conductivity is a small value as compared to the permanent part. Second, in [8] where the oscillating part of electric conductivity was considered for the case of strongly open Fermi surfaces, it is noted that in this case, apart from the phase shift determined by the relations (17), there is an additional shift of electric conductivity oscillation phases of the order kT_D/Δ. Comparison between the calculations by the author of [8] and the experiment shows that this shift is considerable enough. But then it appears that analysis performed by the author of [8] in the field when the contribution of the oscillating part of electric conductivity is small, is not correct, because in the case of $kT_D/\Delta \geq 1$ the length of electron mean free path is small or, at least, insufficiently large as compared to the distance between translation equivalent layers, and, hence, it is necessary to use other approaches, such as described in [7, 9]. However, discussion of this point is beyond the scope of this paper.

Conclusions and recommendations

1. Under conditions of oscillating dependence of relaxation time on a magnetic field, the nonparabolicity of band spectrum of functional, including thermoelectric, materials described by the Fivaz model, is manifested in phase delay of longitudinal electric conductivity oscillations and their amplitude reduction. These peculiarities become apparent with closed Fermi surfaces.
2. Band spectrum nonparabolicity is the more prominent the greater is charge carrier concentration. Peculiarities of nonparabolicity manifestation are mainly due to fundamental differences between the electronic density of states in the Fivaz model and in the effective mass approximation.
3. The results obtained in the paper can be used for the diagnostics of band structure of materials described by the Fivaz model with closed Fermi surfaces.

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