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GIGANTIC THERMOEMF OF LAYERED THERMOELECTRIC MATERIALS IN A QUANTIZING MAGNETIC FIELD

In this paper, the thermoEMF of layered thermoelectric material in a strong quantizing magnetic field was calculated from unconventional standpoint, namely as a thermodynamic quantity that does not depend on the scattering mechanisms of free charge carriers in material. In this case it is defined by a linear combination of derivatives of the thermodynamic and chemical potentials with respect to temperature. Based on this representation for the case of a quantizing magnetic field perpendicular to layers, a general expression for the thermoEMF of layered material was obtained. Specific calculations show that in materials with very low effective masses of charge carriers in layer plane and very narrow conduction minibands which govern carrier motion in the perpendicular direction, the oscillation amplitude of thermoEMF even in the quasi-classical range of magnetic fields can reach 10 and more microvolts per kelvin, that can already be considered as a gigantic amplitude. Such amplitude is realized, for instance, in bismuth, though traditionally this material is not considered as layered. In the nearly-ultra-quantum fields the thermoEMF according to the results of calculations made in this paper, with a high degree of nonparabolicity can reach 30 and more millivolts per kelvin even at helium temperatures. Traditional concepts according to which the thermoEMF is a kinetic, i.e. dissipative characteristic of material, essentially regulated by the scattering mechanisms of free charge carriers, cannot explain such great values thereof, even with regard to phonon drag effect. Therefore, it must be assumed that the gigantic thermoEMF of layered thermoelectric materials in a quantizing magnetic field is their purely thermodynamic characteristic. Generation and wide application of such materials might have opened up fresh opportunities for creation of thermoelectric power converters controlled by magnetic field.

Key words: thermodynamic potential, superlattice, quantizing magnetic field, gigantic thermo-EMF, miniband.

Introduction

It is conventionally recognized that the thermoEMF of thermoelectric materials (TEM) both in the absence and presence of a magnetic field is essentially regulated by the scattering mechanisms of free charge carriers [1]. It would seem that this opinion is confirmed by many experimental data, in particular, given in [2]. It is proved by the discovery of phonon drag effect owing to which the thermoEMF can increase considerably [3]. Though [2] and [3], as well as many other earlier and more recent papers, deal with the thermoEMF in the absence of a magnetic field, it is considered that completely analogous situation takes place, at least in a quasi-classical area of quantizing magnetic fields. The theory of thermoEMF of layered thermoelectric materials with much open Fermi surfaces for this case, also with regard to phonon drag effect, was developed in [4], though this paper describes only general formulae which are "not brought to a number". At the same time, for instance, in the alloys of antimony with bismuth and arsenic [5], at helium temperatures in the quasi-classical range of

magnetic fields there were discovered gigantic oscillations of thermoEMF with the amplitude of the order of 10 μV/K that cannot be explained by phonon drag effect, but the authors of that paper attribute them to the specificity of the energy dependence of relaxation time of free carriers in a magnetic field, without specifying the details and physics of the influence of this specificity.

In contrast to traditional views, the authors of [6] propose to consider the thermoEMF in a strong quantizing magnetic field not as a kinetic coefficient, but as a purely thermodynamic, i.e. equilibrium dissipation-free quantity. They justify this proposal by the fact that the so-called "thermomagnetic current" which is determined traditionally and through which the thermoEMF in a quantizing magnetic field is expressed, does not satisfy mass-energy equation which relates coefficient of diffusion to electric conductivity [7]. Therefore, following the approach developed in [8] which takes into account the diamagnetism of free carrier gas disregarded by traditional approach they obtained the formula for the thermoEMF as a thermodynamic quantity:

$$\alpha = \frac{1}{n_0 e}\frac{\partial \Omega}{\partial T} + \frac{1}{e}\frac{\partial \zeta}{\partial T}. \qquad (1)$$

In this formula, n_0 is the concentration of free charge carriers, Ω is the thermodynamic potential of the unit volume of free carrier gas determined through large partition function, T is absolute temperature, ζ is the chemical potential of free carrier gas, the rest of notations are commonly accepted.

Then the authors of [6], probably not without reason, restricted themselves to some general manipulations of this formula from which it followed that, in fact, it is equivalent to the formula obtained in [8] which expresses the thermoEMF in a quantizing magnetic field through the entropy of the unit volume of free carrier gas, but in a more vivid form. However, the present author in monograph [9] set forth, in particular, the results of his in-depth treatment of free carrier diamagnetism in layered semiconductors. Therefore, it is not difficult for him to calculate the thermoEMF of layered thermoelectric materials in a quantizing magnetic field by formula (1) and to compare the results to those following from traditional approaches, as well as to some (rather limited!) experimental data. This is the purpose of the present paper.

General formula for the thermoEMF of layered thermoelectric material in a quantizing magnetic field

In this section, on the basis of relation (1), we shall derive a general formula for the thermoEMF of layered thermoelectric material with interlayer distance *a*, placed in a quantizing magnetic field perpendicular to layers. In so doing, we assume that motion of free carriers in plane layer is described by a parabolic dispersion law with the effective mass *m**, and in the perpendicular direction – by arbitrary dispersion law $W(ak_z)$, where k_z is corresponding component of quasi-pulse. Then charge carrier spectrum in a quantizing magnetic field with induction B perpendicular to layers is given by:

$$\varepsilon_{n,k_z} = \mu^* B(2n+1) + W(ak_z). \qquad (2)$$

In this formula, $\mu^* = \mu_B m_0 / m^*$, n is the Landau level number, the rest of notations are commonly accepted. For the layered material with such energy spectrum of free carriers from formula (1) we get the following formula for thermoEMF α:

$$\alpha = \frac{k}{e}\left(\frac{F_1}{F_2}+\frac{F_3}{F_4}\right). \tag{3}$$

The dimensionless functions F_1, F_2, F_3, F_4 which enter into formula (3) are given by:

$$F_1 = -\frac{\pi t \kappa_\gamma}{3}+\frac{b^2}{t}\sum_{l=1}^{\infty}(-1)^l\left[\frac{t}{b\,\mathrm{sh}\left(blt^{-1}\right)}+\frac{\mathrm{ch}\left(blt^{-1}\right)}{\mathrm{sh}^2\left(blt^{-1}\right)}\right]\left\{\int\limits_{w(x)\le\gamma}\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx-\int\limits_{w(x)\ge\gamma}\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]\right\}+$$
$$+\frac{b}{t}\sum_{l=1}^{\infty}\frac{(-1)^l}{\mathrm{sh}\left(blt^{-1}\right)}\left\{\int\limits_{w(x)\ge\gamma}\left(\gamma-w(x)\right)\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]-\int\limits_{w(x)\le\gamma}\left(w(x)-\gamma\right)\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx\right\}+ \tag{4}$$
$$+2b\sum_{l=1}^{\infty}\frac{(-1)^l}{l}\left[\frac{1}{\mathrm{sh}\left(\pi^2 ltb^{-1}\right)}-\frac{\mathrm{ch}\left(\pi^2 ltb^{-1}\right)}{\mathrm{sh}^2\left(\pi^2 ltb^{-1}\right)}\right]\int\limits_{w(x)\le\gamma}\cos\left[\pi lb^{-1}\left(\gamma-w(x)\right)\right]dx,$$

$$F_2 = \int\limits_{w(x)\le\gamma_0}\left(\gamma_0-w(x)\right)dx\,, \tag{5}$$

$$F_3 = 2\pi\sum_{l=1}^{\infty}\frac{(-1)^{l-1}}{\mathrm{sh}\left(\pi^2 ltb^{-1}\right)}\int\limits_{w(x)\le\gamma}\sin\left[\pi lb^{-1}\left(\gamma-w(x)\right)\right]dx+2\pi^3 tb^{-1}\sum_{l=1}^{\infty}\frac{(-1)^l\,\mathrm{ch}\left(\pi^2 ltb^{-1}\right)}{\mathrm{sh}^2\left(\pi^2 ltb^{-1}\right)}\int\limits_{w(x)\le\gamma}\sin\left[\pi lb^{-1}\left(\gamma-w(x)\right)\right]dx+$$
$$-b^2t^{-2}\sum_{l=1}^{\infty}\frac{(-1)^l\,l\,\mathrm{ch}\left(blt^{-1}\right)}{\mathrm{sh}^2\left(blt^{-1}\right)}\left\{\int\limits_{w(x)\le\gamma}\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx+\int\limits_{w(x)\ge\gamma}\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]dx\right\}+bt^{-2}\sum_{l=1}^{\infty}\frac{(-1)^l\,l}{\mathrm{sh}\left(blt^{-1}\right)}\times \tag{6}$$
$$\times\left\{\int\limits_{w(x)\le\gamma}\left(w(x)-\gamma\right)\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx+\int\limits_{w(x)\ge\gamma}\left(\gamma-w(x)\right)\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]dx\right\}$$

$$F_4 = \kappa_\gamma+2\pi^2 tb^{-1}\sum_{l=1}^{\infty}\frac{(-1)^l\,l}{\mathrm{sh}\left(\pi^2 ltb^{-1}\right)}\int\limits_{w(x)\le\gamma}\cos\left[\pi lb^{-1}\left(\gamma-w(x)\right)\right]dx+\sum_{l=1}^{\infty}\frac{(-1)^l\,blt^{-1}}{\mathrm{sh}\left(blt^{-1}\right)}\left\{\int\limits_{w(x)\ge\gamma}\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]dx-\right. \tag{7}$$
$$\left.-\int\limits_{w(x)\le\gamma}\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx\right\}.$$

The dimensionless parameters (complexes) and the dimensionless function for layered material described by the Fivaz model which enter into these functions have the following values:

$$t = kT/\Delta,\ b = \mu^* B/\Delta,\ \gamma = \zeta/\Delta,\ w(x) = 1-\cos x,\ \kappa_\gamma = \arccos(1-\gamma)\,. \tag{8}$$

In such a case, Δ is the half-width of a narrow miniband which determines the motion of free carriers in the direction perpendicular to layers, ζ is the temperature- and magnetic field-dependent chemical potential of free carriers subsystem. The latter is determined from the following equation:

$$\int\limits_{w(x)\le\gamma_0}\left(\gamma_0-w(x)\right)dx=\int\limits_{w(x)\le\gamma}\left(\gamma-w(x)\right)dx+2\pi t\sum_{l=1}^{\infty}\frac{(-1)^l}{\mathrm{sh}\left(\pi^2 ltb^{-1}\right)}\int\limits_{w(x)\le\gamma}\sin\left[\pi lb^{-1}\left(\gamma-w(x)\right)\right]dx+$$
$$+b\sum_{l=1}^{\infty}\frac{(-1)^{l-1}}{\mathrm{sh}\left(blt^{-1}\right)}\left\{\int\limits_{w(x)\le\gamma}\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx+\int\limits_{w(x)\ge\gamma}\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]dx\right\}. \tag{9}$$

Here, in the case of nondegenerate gas $\kappa_\gamma = 0$, and at $\gamma = 2$ $\kappa_\gamma = \pi$.

In its turn, parameter γ_0 is related to free carrier concentration by the following relation:

$$n_0 a h^2 / 4\pi m^* \Delta = (\gamma_0 - 1)\arccos(1-\gamma_0) + \sqrt{2\gamma_0 - \gamma_0^2}\,. \tag{10}$$

Formulae (3) – (7) with regard to (8) – (10) completely determine the thermoEMF of layered thermoelectric material as a purely thermodynamic quantity. It should be noted that, on the one hand, the authors of [6] who proposed formula (1) did not indicate the exact values of magnetic field induction starting from which this formula, hence all the relations obtained on its basis in this paper, become valid. On the other hand, electron gas diamagnetism also exists in weak magnetic fields, including in the zero magnetic field limit. Therefore, it is worthwhile to consider and compare to known theoretical results and experiment the corollaries of formula (1), hence, of formulae (3) – (7) in the zero magnetic field limit. In this limit the relation (5) is not changed, and the relations (4), (6) and (7) acquire the form:

$$F_1(0) = -\frac{\pi t \kappa_\gamma}{3} + t\sum_{l=1}^{\infty}(-1)^l\left(\frac{1}{l}+\frac{1}{l^2}\right)\left\{\int\limits_{w(x)\le\gamma}\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx - \int\limits_{w(x)\ge\gamma}\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]\right\} + \\ + \sum_{l=1}^{\infty}\frac{(-1)^l}{l}\left\{\int\limits_{w(x)\ge\gamma}\left(\gamma-w(x)\right)\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right] - \int\limits_{w(x)\le\gamma}\left(w(x)-\gamma\right)\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx\right\}, \tag{11}$$

$$F_3(0) = -\sum_{l=1}^{\infty}\frac{(-1)^l}{l}\left\{\int\limits_{w(x)\le\gamma}\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx + \int\limits_{w(x)\ge\gamma}\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]dx\right\} + t^{-1}\sum_{.l=1}^{\infty}(-1)^l \times \\ \times\left\{\int\limits_{w(x)\le\gamma}\left(w(x)-\gamma\right)\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx + \int\limits_{w(x)\ge\gamma}\left(\gamma-w(x)\right)\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]dx\right\}, \tag{12}$$

$$F_4(0) = \kappa_\gamma + \sum_{l=1}^{\infty}(-1)^l\left\{\int\limits_{w(x)\ge\gamma}\exp\left[lt^{-1}\left(\gamma-w(x)\right)\right]dx - \int\limits_{w(x)\le\gamma}\exp\left[lt^{-1}\left(w(x)-\gamma\right)\right]dx\right\}. \tag{13}$$

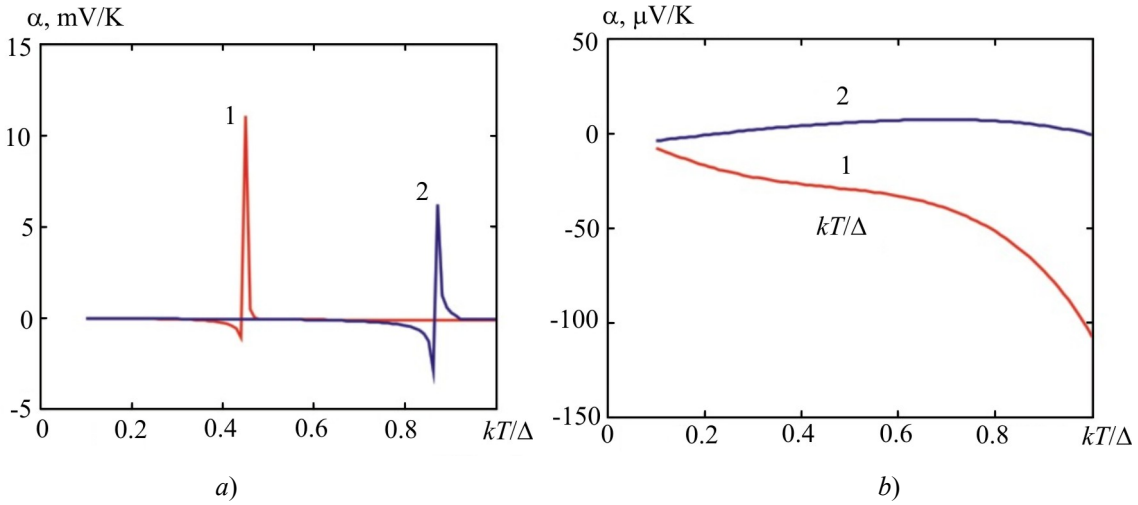
The equation which determines the chemical potential takes on the form:

$$\int\limits_{w(x)\le\gamma_0}\left(\gamma_0 - w(x)\right)dx = t\int_0^{\pi}\ln\left\{1+\exp\left[\frac{\gamma-w(x)}{t}\right]\right\}dx\,. \tag{13}$$

ThermoEMF of layered thermoelectric material in a weak magnetic field

The results of calculation of thermoEMF of layered thermoelectric material in a weak magnetic field are given in Fig. 1.

From the figure it is seen that, at least at relatively low values of γ_0, the thermoEMF of layered thermoelectric material first increases with a rise in temperature to sufficiently high values of several mV/K, then drastically changes its polarity, reaching 7 – 11 mV/K, following which it starts decaying. Such a situation is due to a purely thermodynamic nature of the thermoEMF in the case under study. From formula (1) it follows that the thermoEMF is governed by two competing processes.



a) *b*)

Fig. 1. Temperature dependences of the thermoEMF of layered thermoelectric material in a weak magnetic field. In Fig. 1 a curves 1, 2 correspond to the values of γ_0 *equal to 0.5 and 1, and in Fig. 1 b – 1.5 and 2.*

On the one hand, with a rise in temperature, there is increase in the thermodynamic potential, which contributes to increase in the value of thermoEMF. On the other hand, with a rise in temperature, the chemical potential (at least in impurity material) decreases, which contributes to decrease in the value of thermoEMF. This is precisely why the temperature dependence of thermoEMF is such as above. For the foregoing reasons, a similar behaviour of thermoEMF must also take place at higher values of γ_0, i.e. free carrier concentrations. However, the higher these concentrations, the slower, all other factors being equal, the thermodynamic and chemical potentials vary with temperature. Consequently, the value of thermoEMF, all other factors being equal, with increase in free carrier concentration must decay, and its maximum must shift towards higher temperatures. Exactly for this reason in the analyzed temperature range at concentrations corresponding to values of γ_0 equal to 1.5 and 2 the maximum values of thermoEMF do not become apparent. Note that with traditional consideration, the thermoEMF of impurity material, with a rise in temperature, all the time while we are in the impurity region, is generally increased, because in terms of traditional assumptions a reduction in the chemical potential should contribute to thermoEMF increase.

Nevertheless, let us make a more detailed comparison between the obtained results and the experimental data. Let us consider, for instance, materials based on bismuth telluride. According to [1] and our estimates given, e.g. in [10], for these materials it can be assumed that $\Delta = 1.2$ eV, $m^* = m_0$, $n_0 = 3 \cdot 10^{19}\,\text{cm}^{-3}$, $a = 3$ nm. We obtain that at 300 K for *n*-type material the Seebeck coefficient must have been equal to – 1.95 μV/K. However, the true value of thermoEMF of this material under said conditions is two orders of magnitude higher. Therefore, we come to conclusion that in the zero magnetic field and at normal temperatures the above mechanism, at least for traditional thermoelectric materials, does not work. Although from the plot in Fig. 1 *a* it follows that in the zero magnetic field this mechanism might have been "activated" at higher temperatures, in traditional thermoelectric materials it will not be the case, since for them the respective "activation temperature" is higher than melting temperature.

Moreover, an essential fault of using this approach in the absence of a magnetic field is impossibility to explain the thermoEMF anisotropy which is found in many layered materials [11], as long as "purely thermodynamic" thermoEMF according to general formula (1) must be a scalar, rather than a tensor quantity.

However, a different situation can be realized in materials with narrow allowed energy bands. If, for instance, $\Delta = 0.06$ eV, the above effect might have become apparent already at temperature 278 K or at higher carrier concentration at temperature 556 K. Instead, this does not take place either because the dissipation-free thermoEMF in the weak field limit exists only formally, without being such in reality, or because carrier concentration in these materials is too high to achieve the temperature which is needed for "activation" of the effect.

Gigantic thermoEMF of layered thermoelectric material in a quantizing magnetic field

To determine the thermoEMF in a quantizing magnetic field at low temperatures, we leave in formulae (4), (6) and (7), as well as in equation (9) only the terms which do not explicitly depend on the magnetic field and oscillating functions of the magnetic field. The field dependences of thermoEMF for this case in quasi-classical magnetic fields are given in Fig. 2, and in a wider range of magnetic fields, including ultra-quantum ones, in Fig. 3.

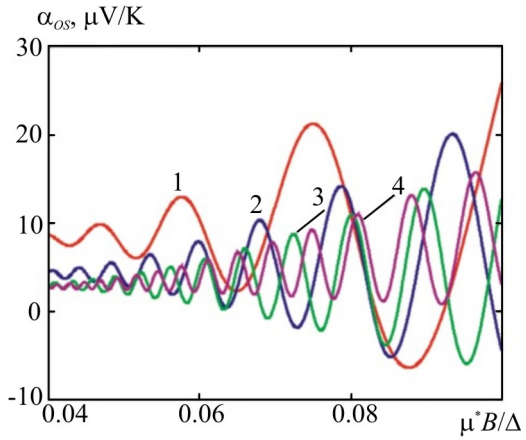


Fig. 2. Field dependences of the thermoEMF at $kT/\Delta = 0.03$ in a quasi-classical range of magnetic fields. Curves 1 – 4 are for the values of γ_0 from 0.5 to 2 with increment 0.5.

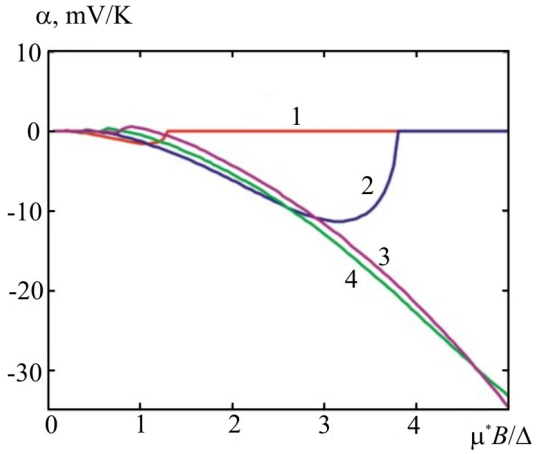


Fig. 3. Field dependences of the thermoEMF at $kT/\Delta = 0.03$ in a wide range of magnetic fields. Curves 1 – 4 are for the values of γ_0 from 0.5 to 2 with increment 0.5.

From the figures it is seen that even in the quasi-classical range of magnetic fields the oscillation amplitude of thermoEMF for the analyzed range of carrier concentrations reaches 6 – 15 μV/K. Such amplitude according to [5] can be considered as gigantic. With increasing carrier

concentration, the oscillation frequency of thermoEMF grows, and the amplitude drops. It is noteworthy that in [5], materials whose thermoEMF oscillations were studied experimentally are not considered as layered. However, our approach here is applicable to them, at least when a dominant role is played by the so-called "small groups" of free charge carriers. From the results presented in [5] it follows that thermoEMF oscillations in the investigated semi-metallic alloys of antimony with arsenic can also be of a purely thermodynamic nature.

In the nearly ultra-quantum fields the thermoEMF first reaches rather high maximum, and then decays due to compression of the Fermi surface in the direction of a magnetic field because of free carrier condensation in the lower Landau subband. However, the stronger the magnetic field, the more difficult it is to compress the Fermi surface, by virtue of which with increasing concentration of free carriers, the thermoEMF maximum is drastically increased and shifted towards the area of stronger magnetic fields. Therefore, at high concentrations in the considered range of magnetic fields this maximum does not become apparent, and thermoEMF even at low temperatures achieves rather high values, namely of the order of 35 mV/K.

Let us now analyze the possibility of reaching the effect of drastic thermoEMF increase in a quantizing magnetic field. In traditional materials based on superlattices of *Al-Ga-As* system for which $\Delta = 0.06\,\text{eV},\ m^* = 0.5\,m_0$ reaching said effect requires magnetic fields with inductions of the order of 2600 *T* which are hardly achievable. However, for drastically anisotropic superlattices, for instance, based on graphene for which $\Delta = 0.01\,\text{eV},\ m^* = 10^{-3} m_0$ it is sufficient to have fields with inductions of the order of 0.86 *T* which are quite common today. Therefore, such materials that are "controlled" by a quantizing magnetic field can be used as cooling materials for very low temperatures.

ThermoEMF of layered thermoelectric material in a quantizing magnetic field at high temperatures

To pass over to the case of high temperatures, we ignore the oscillating terms in formulae (4), (6) and (7) equation (9). Then, as a result of calculations we get the field dependences of thermoEMF shown in Fig. 4.

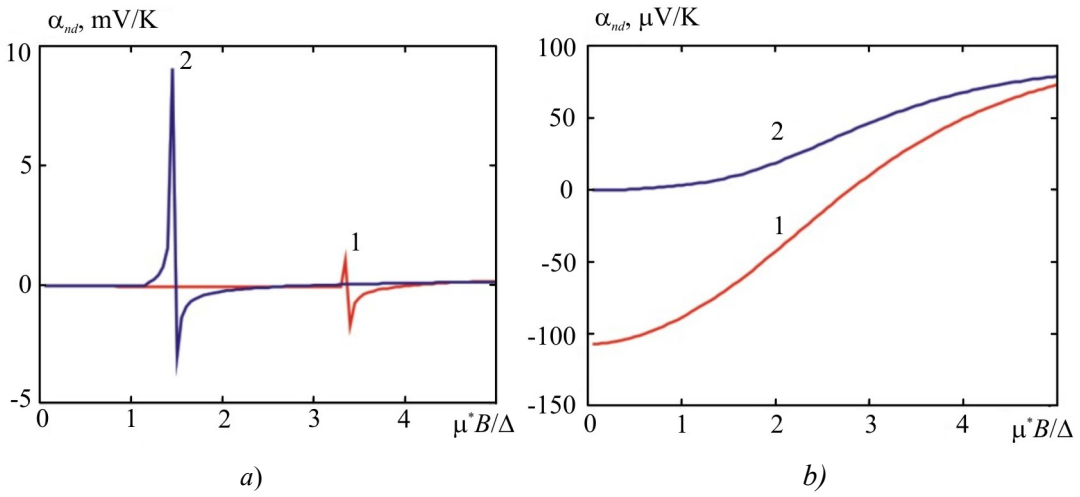


Fig. 4. Field dependences of the thermoEMF of layered thermoelectric material at high temperature ($kT/\Delta = 1$) in a wide range of magnetic fields. In Fig. 4 a curves 1, 2 correspond to the values of γ_0 equal to 0.5 and 1 a in Fig. 4 b to 1.5 and 2.

From Fig. 4 *a* it is seen that there is an optimal carrier concentration whereby the effect of drastic thermoEMF increase in a magnetic field manifests itself in the best way. It is explained by the following physical reasons. On the one hand, magnetic field increase from thermodynamic standpoint is equivalent to material cooling. An as long as the curves in Fig. 4 *a*, *b* are constructed with the highest value of parameter $kT/\Delta = 1$ from the considered range, it is clear that with magnetic field increase the thermoEMF peak must take place at least for those carrier concentrations whereby it occurs in the range of $0 \le kT/\Delta \le 1$ (within the approach used) in the absence of a magnetic field. At different carrier concentrations in the same temperature range this peak must not be observed. On the other hand, at high temperatures the chemical potential of carriers with a rise in temperature monotonously decreases, and with magnetic field increase, on the contrary, monotonously increases. So, both the possibility itself of thermoEMF peak origination in a quantizing magnetic field and its value are governed by the ratio between the velocities of these competing processes. With low carrier concentrations these processes proceed equally fast, and with high concentrations − equally slow. Therefore, at high temperature there is optimal carrier concentration, hence the degree of material nonparabolicity when the difference in velocities of these processes is the greatest, hence the thermoEMF peak is most pronounced. The same reasons account for the reversal of thermoEMF polarity in a quantizing magnetic field. Thus, control of the figure of merit of generator materials with the aid of a quantizing magnetic field is also possible, if the effective masses of carriers in layer plane are sufficiently small. At the same time, miniband widths must be optimal in the sense that the temperature whereby thermoEMF peak takes place must be sufficiently high so that the material could be considered to be "generator" and at the same time the approach used here would be valid. According to Fig. 4 *a*, in the considered range of carrier concentrations the peak thermoEMF values change approximately in the range of (1 ÷ 9) mV/K, hence, they can be also considered to be gigantic.

Conclusions

1. Based on the idea of thermoEMF in a quantizing magnetic field as a purely thermodynamic quantity, it was shown that both at low and high temperatures the value of thermoEMF of layered thermoelectric materials in the presence of this field can reach 1 – 35 mV/K.
2. At low temperatures the value of thermoEMF in a quantizing magnetic field close to ultra-quantum limit with growing concentration of free carriers is increased. Whereas at high concentrations there is optimal carrier concentration whereby the peak of thermoEMF is expressed in the best way.
3. The thermoEMF, hence the figure of merit of layered cooling and generator thermoelectric materials can be efficiently controlled with the aid of a quantizing magnetic field, provided that the effective masses of charge carriers in layer plane are sufficiently low, and carrier concentrations and miniband widths which determine carrier motion in the direction perpendicular to layers have optimal values.

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