"High-temperature" oscillations of the magnetoresistance of bismuth: a possible alternative explanation

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It is shown that the known experimental results on the properties of the "high-temperature" oscillations of the magnetoresistance of bismuth—in particular, the angular dependence of the oscillation periods on the direction of the magnetic field—can be described by the Polyanovskiĭ theory with the use of the condition that the cyclotron frequencies are equal or multiples. [DOI: 10.1063/1.1614238]

The goal of this communication is to explain the unusual "high-temperature" oscillations (HTOs) of the magnetoresistance of bismuth on the basis of the Polyanovskiĭ theory,[1–3] which, with some additional assumptions, can provide an adequate description of the experimental results.

The observation of new quantum oscillations of the magnetoresistance of bismuth, with a period in the inverse magnetic field, ΔH^{-1}, which is 2–3 times shorter than that of the Shubnikov–de Haas oscillations and with an order-of-magnitude smaller amplitude but a slower temperature decay was reported by Bogod and Krasovitskiĭ in 1973.[4] Since then the new oscillations have been studied in detail in crystals of Bi and the alloys $Bi_{1-x}Sb_x$ and also in crystals with donor (Te) and acceptor (Sn) impurities.[5–14] Experiments have been done to study the manifestation of the oscillations in the thermopower[15,16] and the behavior under pressure and uniaxial deformation[17,18] and in strong magnetic fields.[19–21] Based on the data of these experiments it was concluded that the period of the HTOs is not determined directly by the Fermi energy of the electrons E_F^e or holes E_F^h but is somehow related to the value of the band overlap $E_{\rm ov}$. The latter conclusion served as a stimulus for the dubious assertion[22] that the HTOs are "quantum oscillations of the probability of quasielastic intervalley scattering of charge carriers with deep-lying energies," specifically, with states at the bottom of the electron band E_c^e and the top of the hole band E_c^h. However, the deep-lying states are traditionally considered to be completely filled. Although it was shown in a subsequent paper[23] that the deep-lying states in connection with "collisional" broadening of the energy levels can contribute to the conduction, the scope of that work did not encompass a treatment of the detailed picture of the observed oscillation periods in different crystallographic directions and their weak temperature decay.

Polyanovskiĭ[1–3] singled out from the magnetoconductance the term involving intervalley transitions and describing the properties of the HTOs—a small period ΔH^{-1} and weak temperature decay. Meanwhile, Polyanovskiĭ's theory was not accepted by the authors of the experimental papers (see the critique in Ref. 14), since the physical cause of the new oscillations remained unclear. Recently Kirichenko and Kozlov[24] obtained a result similar to Polyanovskiĭ's in an analysis of a completely different object—a layered conductor in which, as in bismuth, there are two types of extremal sections that govern oscillations of the magnetoconductance.

Bismuth is a semimetal, with a weak overlap of the valence and conduction bands, as a result of which electron and hole valleys form. Taking the spectrum of a semimetal in a magnetic field as

$$E_n^e=\left(n+\frac{1}{2}\right)\hbar\Omega^e+\frac{p_z^{e^2}}{2m^e}$$

and

$$E_n^h=E_{\rm ov}-\left(n+\frac{1}{2}\right)\hbar\Omega^h-\frac{p_z^{h^2}}{2m^h}$$

($E_{\rm ov}$ is the overlap energy of the bands), Polyanovskiĭ[1–3] obtained for the conductivity in a magnetic field two terms describing interband transitions, σ^{ee} and σ^{hh}, which determine the Shubnikov–de Haas oscillations[25,26] for the electron and hole valleys, and a term σ^{eh} describing the interband transitions. The last term contains the product of the densities of states in different valleys. This product of two oscillatory characteristics gives rise not only to terms describing the Shubnikov–de Haas oscillations in each of the valleys but also to a "cross" term with combination parameters, arising as a result of the interference of the densities of states in the different valleys. This term in the conductivity can serve as an explanation for the "high-temperature" oscillations of the magnetoresistance of bismuth. It has the form[27]

$$\sigma^{eh}=\frac{3}{8}\sigma_{\rm cl}\frac{\hbar(\Omega^e+\Omega^h)}{E_{\rm ov}}\sum_{k,l=1}^{\infty}\frac{(-1)^{k+l}}{\sqrt{kl}}\times\left\{A\left[\frac{2\pi^2k_BT}{\hbar}\left(\frac{k}{\Omega^e}-\frac{1}{\Omega^h}\right)\right]\sin\frac{2\pi}{\hbar}\times\left(k\frac{E_F^e}{\Omega^e}+l\frac{E_{\rm ov}-E_F^e}{\Omega^h}\right)+A\left[\frac{2\pi^2k_BT}{\hbar}\left(\frac{k}{\Omega^e}+\frac{l}{\Omega^h}\right)\right]\times\cos\frac{2\pi}{\hbar}\left(k\frac{E_F^e}{\Omega^e}-l\frac{E_{\rm ov}-E_F^e}{\Omega^h}\right)\right\}. \quad (1)$$

Here Ω^e and Ω^h are the cyclotron frequencies of the electrons and holes, $A(x)=x/\sinh(x)\approx x\exp(-x)$ for $x\gg 1$.

Taking into consideration that

$$\frac{2\pi E_F^e}{\hbar\Omega^e}=\frac{cS^e}{\hbar eH},\quad \frac{2\pi E_F^h}{\hbar\Omega^h}=\frac{cS^h}{\hbar eH},\quad E_{\text{ov}}-E_F^e=E_F^h,$$

we obtain Polyanovskiĭ's result:

$$\sigma^{eh}=\frac{3}{8}\sigma_{\text{cl}}\frac{\hbar(\Omega^e+\Omega^h)}{E_{\text{ov}}}\sum_{k,l=1}^{\infty}\frac{(-1)^{k+l}}{\sqrt{kl}}\times\left\{A\left(\frac{2\pi^2k_BT}{\hbar\Omega^-}\right)\sin\left(\frac{cS^+}{\hbar eH}\right)+A\left(\frac{2\pi^2k_BT}{\hbar\Omega^+}\right)\cos\left(\frac{cS^-}{\hbar eH}\right)\right\}. \tag{2}$$

Equation (2) contains the combination areas

$$S^{\pm}=kS^e\pm lS^h \tag{3}$$

and the combination inverse cyclotron frequencies

$$\frac{1}{\Omega^{\pm}}=\frac{k}{\Omega^e}\pm\frac{1}{\Omega^h}. \tag{4}$$

According to Eq. (2) there are two series of oscillations, with periods

$$\Delta H^{-1}=\frac{2\pi\hbar e}{cS^{\pm}}, \tag{5}$$

the temperature decay of which is governed by the factor

$$\exp\left(-\frac{2\pi^2k_BT}{\hbar\Omega^{\mp}}\right). \tag{6}$$

We are mainly interested in the oscillations determined by the combination area S^+, for which the period ΔH^{-1} is smaller than for the Shubnikov–de Haas oscillations and the temperature decay determined by the combination frequency Ω^- is slower than for the Shubnikov–de Haas oscillations. These oscillations can explain the HTOs. The second type of oscillations—long-period, with faster temperature decay, have not been detected in experiment.

The results obtained explain well the whole complex of experimental observations. For example, adding a donor impurity (Te) to bismuth increases the electron concentration and decreases the hole concentration. Additionally, E_F^e increases and E_F^h decreases, but the period of the HTOs remains unchanged,[13] since the sum of the areas (3) remains practically unchanged. An analogous result is obtained when an acceptor impurity (Sn) is added to bismuth. When the isovalent impurity antimony is added to bismuth, both energies E_F^e and E_F^h decrease, and accordingly both areas S^e and S^h decrease, leading to an increase in the oscillation period ΔH^{-1} (Ref. 12). Similarly, one can explain the results of a study[18] on the influence of uniaxial deformation on the periods of the HTOs and also the fact that the ultraquantum limit at high magnetic field is reached simultaneously for both the Shubnikov–de Haas and "high-temperature" oscillations.[19] (Note that this would not be the case if the HTOs were related to some energy other than the Fermi energy, as in the model of Ref. 22).

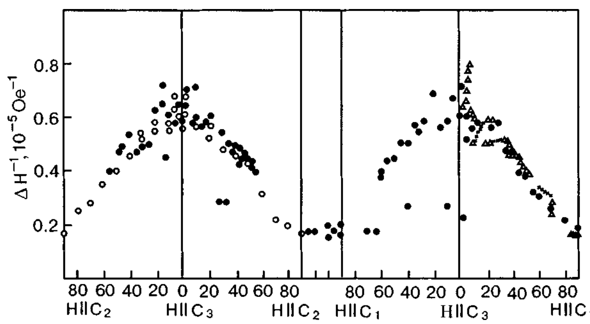

FIG. 1. Angular dependence of the periods ΔH^{-1} of the "high-temperature" oscillations in bismuth, obtained in measurements of the diagonal and off-diagonal components of the magnetoresistance tensor (●—data of Ref. 7; △—mean period;[9] ×—period obtained from the angular shift of the extrema[9]) and from thermopower measurements (○—Ref. 16).

The most important result of the experimental study of HTOs is the dependence of the oscillation periods ΔH^{-1} on the orientation of the magnetic field with respect to the crystallographic directions[7,9,16] (Fig. 1). Let us discuss these findings.

As was noted in Refs. 1–3, the HTOs are due to intervalley transitions of electrons which occur at resonant values of the magnetic field, fields at which Landau levels in the different valleys are simultaneously found near the Fermi energy. However, the frequency with which such events occur as the magnetic field is varied is determined exclusively by the frequency with which Landau levels pass through the Fermi energy in the band with the higher cyclotron frequency (lower cyclotron mass), and so this does not give rise to a new period ΔH^{-1} of the magnetoresistance oscillations. The new period is due to the appearance of a combination of cyclotron processes in the two valleys. Polyanovskiĭ's theory, of a formal mathematical construction, does not allow one to understand the physical nature of the simultaneous oscillatory terms. As we have said, the crossing terms appear as a result of the interference of the oscillatory dependences of the densities of states in the different valleys. The reason why the amplitudes of these oscillations are not small (undetectable) is apparently of a quantum nature, like that which was pointed out by Adams and Holstein[26] in an analysis of the Shubnikov–de Haas oscillations in the single-band case, having to do with the influence of electric field on the collision integral.

It is important to note that for the intervalley conversion transitions, the density of states has features (maxima) in both the initial and final states. In addition, expression (2) for the "high-temperature" oscillations, unlike the case of the Shubnikov–de Haas oscillations,[26] does not contain the value of the chemical potential, and the energies E_F^e and E_F^h are related through a constant, nonoscillatory quantity—the overlap energy E_{ov}. For this reason the amplitude of the oscillations is not very sensitive to temperature smearing of the Fermi boundary, i.e., the temperature decay of the oscillation amplitude is weak.[27]

Experimental observations show that the amplitude of the HTOs in bismuth increases with temperature in the range 1.5–10 K, passes through a maximum (at 10 K), and then falls off slowly.[9,14] The authors justifiably surmised that high-frequency thermal phonons excited at the higher tem-

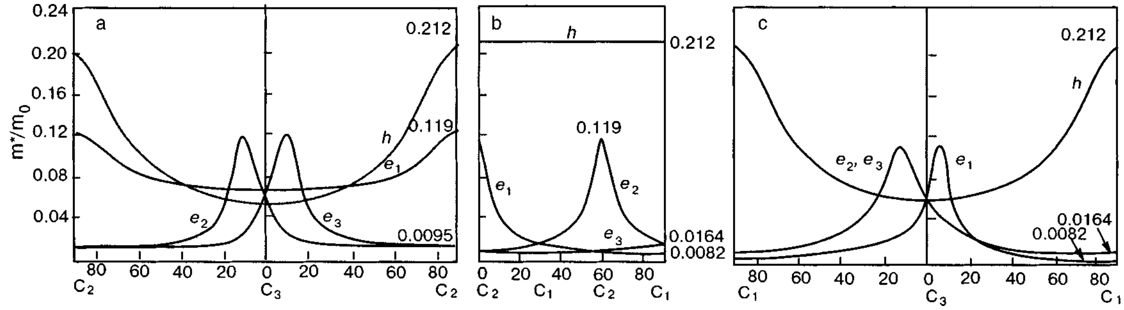

FIG. 2. Angular dependence of the cyclotron mass in bismuth, constructed from the data of Refs. 28–30.

peratures play a role in the formation of the HTOs. This role may reduce to the circumstance that the turning on of electron–phonon scattering leads to the appearance of free electron states below the Fermi energy, which are needed for isoenergetic intervalley transitions.

The possibility of electron intervalley conversion transitions is obvious when the cyclotron frequencies on the extremal orbits in the different valleys are equal, since in that case the electron states remain coherent. That was the case considered in the paper by Kirichenko and Kozlov.[24] However, in bismuth it is only when the magnetic field direction is along the trigonal axis C_3 that the cyclotron frequencies of electrons and holes are approximately equal (the cyclotron effective masses are equal to $0.0630m_0$ for electrons and $0.0639m_0$ for holes). When the field deviates from the C_3 axis, and especially for a field direction along the bisector C_1 and binary C_2 axes, the cyclotron effective masses of electrons and holes are considerably different. Meanwhile, the HTOs in bismuth are observed for arbitrary directions of the magnetic field.

We note that the possibility of electron intervalley conversion transitions remains present for multiples of the cyclotron frequencies as well. Indeed, if the cyclotron frequencies for the two valleys are multiples of each other, then, with a frequency equal to the lower of the two cyclotron frequencies, the coherence of the electron states is periodically recovered, i.e., the conditions for a resonant transition of an electron from an orbit in one valley to an orbit in another valley are restored.

We choose the harmonics k and l in formula (2) such that their ratio corresponds to a multiple of the cyclotron frequencies, i.e., we let

$$\frac{k}{l}=\frac{\Omega^e}{\Omega^h}=\frac{m^h}{m^e}=K, \qquad (7)$$

where $K=1,2,3...$ are integers. This assumption was also made in Refs. 1 and 2.

To check stated assumptions we calculated the possible oscillation periods according to Eq. (5) with the use of relation (7). Figure 1 illustrates the experimental data for the periods ΔH^{-1} of the HTOs in bismuth upon variation of the magnetic field direction. The angular dependence of the periods ΔH^{-1} for the bisector plane C_3C_2 was taken from Refs. 7 and 16 and for the binary plane C_3C_1 from Refs. 7 and 9. It is seen that the values of the HTO periods ΔH^{-1} have a considerably larger scatter than for the Shubnikov–de Haas oscillations (see Ref. 7); in particular, the pattern for the bisector plane C_3C_2 does not have mirror symmetry. In Ref. 9, observations of the HTOs were made as the angle of rotation of the magnetic field in the binary plane C_3C_1 was varied with a step of 1–2°, and it was found that in many cases two close periods were observed, and in some directions oscillations were not detected.

The results of calculations of the periods ΔH^{-1} according to formulas (5) and (3) with the use of (7) are extremely sensitive to the initial data for the cyclotron masses m^* and the cross-sectional areas S in bismuth, and for this reason we give in Figs. 2 and 3 the angular dependences of m^* and S adopted in the calculations. The angular dependences for the masses were constructed according to the data of cyclotron resonance studies,[28–30] the data for the areas were generalized according to studies[31–34] of the Shubnikov–de Haas and de Haas–van Alfven effects in bismuth. The values of m^* and S for the principal directions of the magnetic field (along the C_1, C_2, and C_3 axes) correspond to the numbers given in the review by Edelman.[35]

If the periods ΔH^{-1} are calculated according to Eqs. (5) and (3) for the values of the harmonics $k=l=1$, then good agreement with experiment is obtained only for $\mathbf{H}\|\mathbf{C}_3$

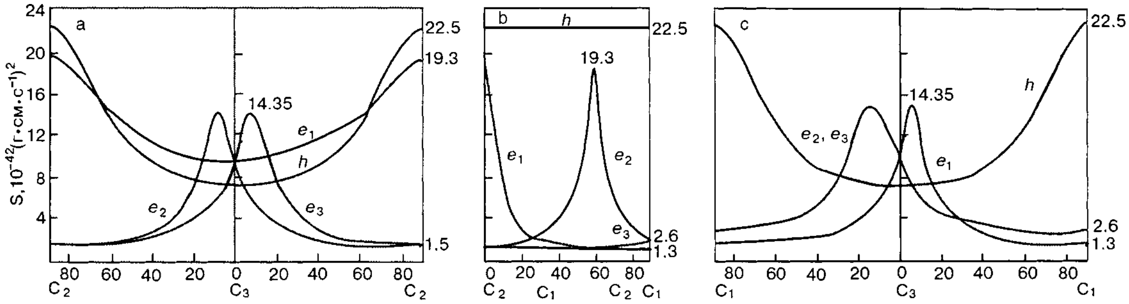

FIG. 3. Angular dependence of the areas of sections through the hole and electron ellipsoids in bismuth, constructed according to the data of Refs. 31–34.

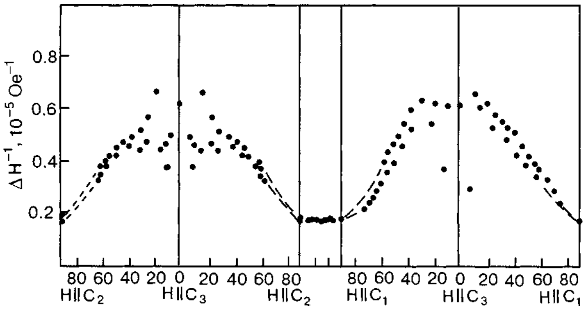

FIG. 4. Angular dependence of the calculated values of the periods ΔH^{-1} of the "high-temperature" oscillations in bismuth. For the trigonal plane C_1C_2, as in Fig. 1, a 30° sector is given; the rest of the pattern is mirror symmetric.

($\Delta H^{-1}=0.63\times 10^{-5}$ Oe^{-1}). For the direction $\mathbf{H}\|\mathbf{C}_2$ the calculated value of ΔH^{-1} (0.253×10^{-5} Oe^{-1}) is almost one and a half times larger than the experimentally observed value (0.18×10^{-5} Oe^{-1}). For the direction $\mathbf{H}\|\mathbf{C}_1$ the difference reaches a factor of 2.5: the calculation gives 0.44×10^{-5} Oe^{-1} while the experimental value is approximately 0.18×10^{-5} Oe^{-1}. Accordingly, the angular dependences of ΔH^{-1} for the bisector (C_3C_2) and especially for the binary (C_3C_1) planes differ appreciably from the picture observed experimentally. Consequently, this version of Polyanovskiĭ's theory does not give numerical agreement with experiment.

A completely different result is obtained when condition (7) is used. Figure 4 shows the results of calculations of the periods ΔH^{-1} with the use of that condition.

The calculations were done for values up to $K=10$, since beyond that the error of the calculations grows strongly. The dashed curves in Fig. 4 show proposed extensions of the calculated curves of ΔH^{-1} to values for magnetic field directions along the C_1 and C_2 axes. It is seen that the calculated picture (Fig. 4) reproduces the experimentally observed picture (Fig. 1) to a satisfactory degree of approximation. (One must consider the error of both the experimental observations and of the initial data for the calculations.)

We note that in the case when high harmonics participate in the formation of the oscillations (for a strong deviation of the field from the C_3 axis), the oscillations take on a less perfect sinusoidal form and their amplitude decreases.[6,9,16,18,19]

On account of the Dingle smearing of the levels, oscillations of an interference nature can be observed not only at exact integer values of the ratio m^h/m^e but also close to these values. For an exact integer value of the ratio $k/l=m^h/m^e$ the inverse combination cyclotron frequency tends toward zero, i.e., the temperature suppression of the oscillations because of smearing of the Fermi distribution vanishes, and only the Dingle smearing remains. The same result was obtained by Kirichenko and Kozlov[24] in considering a layered conductor in which the cyclotron frequencies on two cross sections are equal.

We note that the oscillations shown by the dark dots in Fig. 4 cannot be designated as hole or electron; they are of an interference nature and are combination oscillations.

We note another important result of the calculations: for the planes (C_3C_1) and (C_3C_2) (see Fig. 4) the calculated points form two chains of possible oscillation periods with slightly different values, as has been noted repeatedly by the authors of experimental papers (see Refs. 9 and 13, etc.)

Thus formulas (2)–(5), which were obtained by Polyanovskiĭ for explaining the "high-temperature" oscillations of the magnetoresistance in bismuth, give a completely successful description of the properties of those oscillations and, in particular, describe the angular dependences of the oscillation periods ΔH^{-1} with respect to the magnetic field direction. In our view it remains necessary to solidify the physical grounds for the nature of the combination areas and to explain the temperature dependence of the oscillation amplitude at lower temperatures (<10 K). The above considerations are only of a preliminary character.

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