

Shubnikov–de Haas effect and its application to investigation of the energy spectrum of metals, semimetals, and semiconductors

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The importance of every scientific work is determined in the long run by the role that it played in the development of one or another field of physics. From this point of view the 1930 papers of L. V. Shubnikov and V. de Haas in which for the first time the oscillating dependence of the resistance on a magnetic field was found and the first quantum effect in a solid, given later the name of Shubnikov–De Haas (Sh-H) effect, was discovered may be completely evaluated only at present.

The Sh-H effect was the first experimentally observed demonstration of diamagnetic (Landau) quantization of the energy of electrons in solids² and, as a consequence of this, of oscillating dependence on a magnetic field of electron density of states on the Fermi level.

In following years it became clear that the Sh-H effect is a universal and powerful method for studies of the energy spectrum of degenerate electronic systems in metals, semimetals, alloys, and doped semiconductors. After the discovery of the effect in Bi the oscillation of magnetoresistance of this substance was studied in detail in the papers of Ref. 3–10. Later the Sh-H effect was found in zinc, magnesium, beryllium, and niobium,^{11–14} in intermetallic compounds, alloys, and degenerate semiconductors. A^{III}B^V and A^{II}B^{VI} semiconductors and also semiconductors with a narrow forbidden energy gap are especially suitable for its observation.

In recent years the Sh-H effect is being intensively studied in indium antimonide,^{15–18} indium arsenide,^{19–24} mercury selenide,^{25–27} bismuth-antimony alloys, alloys of lead and tin halogenides,^{28–33} and in other semiconductors and also in semimetals (Bi, Sb, As, graphite).^{39–42}

The Sh-H effect is easily observed at liquid helium temperatures in perfect monocrystals of practically all substances which have a degenerate electron system and a relatively high mobility of current carriers ($\mu \geq 10^4$ cm²/V·s). Magnetic fields of the order of tens of kilo-oersteds which may be obtained by superconducting solenoids or cooled electromagnets with ferromagnetic cores are usually sufficient for the observation of the effect. The Shubnikov–de Haas oscillations in a bismuth-antimony alloy monocrystal are shown as an example in Fig. 1.

Comparative ease of observation together with high content of information on the electron system parameters led to the fact that the Sh-H effect has become one of the basic methods of studying electron energy spectra and band structure of metals, semimetals and semiconductors and an important method for the study

of spectral changes under the influence of a high pressure, strong uniaxial deformations, electric field, doping, etc.

1. PHYSICAL FOUNDATIONS AND THEORY OF THE SHUBNIKOV-DE HAAS EFFECT

The quantization of the electron energy in solids under the influence of a magnetic field lies at the foundation of the Sh-H effect. As a result of quantization the quasicontinuous electronic spectrum is transformed into a quasidiscrete one (Fig. 2). For electrons with a quadratic dispersion law the energy ε in the magnetic field H depends upon the quantum numbers n and s and the projection of quasimomentum $P_{||} = (pH)/H$ on the direction of the field H :

$$\varepsilon(n, s, p_{||}) = \hbar\omega_c \left(n + \frac{1}{2} \right) + s\hbar \frac{eH}{2m_s c} + \frac{p_{||}^2}{2m_{||}}, \quad (1)$$

where $n = 0, 1, 2, \dots$; $s = \pm 1$; $\omega_c = eH/m_c c$ is the cyclotron frequency; $\mu_B^* = e\hbar/2m_s c$ is effective Bohr magneton; $m_c, m_s, m_{||}$ are the cyclotron, spin and longitudinal effective masses correspondingly. The separation between adjacent Landau levels $\hbar\omega_c$ becomes larger than the thermal spread ($\sim kT$) of the Fermi boundary in sufficiently strong magnetic fields and at low temperatures. As the magnetic field is increased discrete Landau levels, going up in energy, cross the Fermi

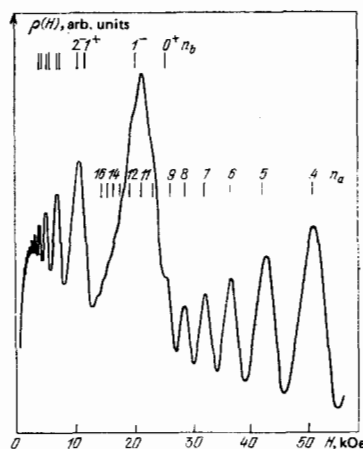


FIG. 1. Shubnikov–de Haas oscillations corresponding to large and small hole sections of the Fermi surface in the Bi_{0.92}Sb_{0.08} sample of *p*-type with the hole concentration $p = 5.3 \times 10^{17}$ cm⁻³ in a magnetic field applied along the binary crystal axis at $T = 1.9$ K. The field marks correspond to the positions of Landau levels calculated according to the model of Ref. 63: n_b —for small sections, n_a —for large sections.

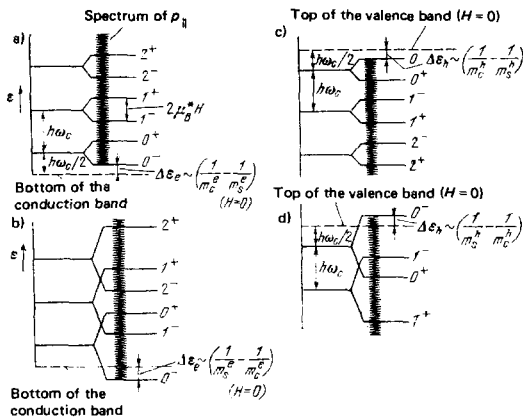


FIG. 2. Diagram of the energy levels of electrons (a,b) and holes (c,d) in a magnetic field.

level. Because the density of states at Landau levels is very high, whenever some Landau level coincides with the Fermi boundary the electron concentration on the Fermi level goes through a maximum producing singularities of all thermodynamic and kinetic coefficients characterizing a particular substance. Periodic changes of the density of states whenever Landau levels coincide with the Fermi boundary as the magnetic field is increased are the cause of the oscillating character of the magnetoresistance.

A year after the discovery of the Sh-H effect the oscillation of the magnetic susceptibility of bismuth in a magnetic field called the de Haas-van Alphen effect was found.⁴³ It was shown that at the same orientation of the magnetic field the same periodicity of the singularities is observed in the de Haas-van Alphen effect as in the Sh-H effect.

Oscillations of the thermoelectric power, Hall effect, thermal heat capacity, thermal conductivity, quasiclassical absorption coefficient of long wavelength sound waves and of other thermodynamic and kinetic characteristics of a metal which now have the general name of quantum oscillation effects were found later. All quantum oscillation effects are observed when the following conditions are satisfied: $\omega_c \tau \gg 1$, $\hbar \omega_c > kT$, $\epsilon_F > \hbar \omega$, where τ is the electron relaxation time and ϵ_F is the Fermi energy.

The general theory of quantum oscillation effects was developed by I. M. Lifshits with co-workers.⁴⁴⁻⁴⁶

It follows from the quantization conditions of Bohr-Sommerfeld that only orbits with discrete values of the area S in the quasimomentum space:

$$S_n = \frac{2\pi e \hbar}{c} H (n + \gamma), \quad (2)$$

where $n = 0, 1, 3, \dots$ and $\gamma \sim 1$, are allowed for an electron in a magnetic field. For a given quantum number n the area of the electron orbit has the same value (2) independently of the longitudinal quasimomentum $p_{||} = (pH)/H$. This means that allowed states for electrons in the quasimomentum space are situated on the discrete tubes cross sections of which by any plane $p_{||} = \text{const}$ determine the trajectory of electrons in this plane (a Landau tube). The shape of the cross section of Landau

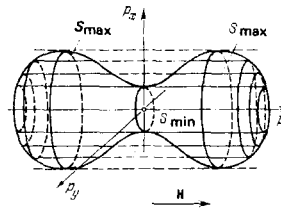


FIG. 3. Landau tubes for the Fermi surface of the dumbbell type in a magnetic field parallel to the principal axis. The minimum and maximum sections of the surface by the planes perpendicular to the magnetic field are shown.

tubes is determined by the shape of the cross section of the corresponding isoenergetic surface (Fig. 3). The singularities of the density of states on the Fermi level in a magnetic field arise whenever a Landau tube takes up a position in which it is touching the Fermi surface. In this case the cross section of the Landau tube by the plane $p_{||} = \text{const}$ is coincident with one of the extremal sections S_{extr} of the Fermi surface (see Fig. 3). Substituting for S_n in the equation (2) by S_{extr} does not depend on the magnetic field (which is correct in the quasiclassical range of fields $\hbar \omega_c \ll \epsilon_F$ to the second order in the small parameter $\sqrt{\hbar \omega_c / \epsilon_F^0}$, where ϵ_F^0 is the Fermi energy at $H = 0$ ⁴⁴), it is possible to find the periodicity of singularities of the density of states. They occur with a constant period Δ in the reciprocal magnetic field equal to

$$\frac{1}{H_{n+1}} - \frac{1}{H_n} = \Delta \left(\frac{1}{H} \right) = \frac{2\pi e \hbar}{c S_{\text{extr}}}, \quad (3)$$

which makes possible a direct determination of the area of the extremal section of the Fermi surface. Equation (3) is called the Lifshits-Onsager equation.

It was shown by Lifshits,⁴⁷ that in the case of a convex Fermi surface the data on the dependence of S_{extr} on the angles which determine the orientation of a magnetic field in relation to crystallographic axes are sufficient to reconstruct the surface uniquely. The problem loses its uniqueness in the case of a complex nonconvex Fermi surface and also in the case of a Fermi surface consisting of several isoenergetic surfaces. In this case, in order to reconstruct the surface, it is necessary to use model representations (construction of the Fermi surface by Harrison's method in the free electron approximation,⁴⁸ calculation by the pseudopotential method, symmetry considerations, data of other experiments, etc.) together with the data from the measurement of oscillations.

The Shubnikov-de Haas effect was used for the determination of Fermi surfaces of many substances: practically, of all metals (including transition metals), of semimetals, and also for the determination of isoenergetic surfaces of many semiconductors. Comparative ease of observation of the Sh-H effect made its use possible in the cases when measurements of other oscillations were complicated or impossible. This relates first of all to the measurements at high pressures, strong uniaxial deformations, strong electric fields, etc.

We note that the Sh-H effect is a kinetic effect and in

contrast to oscillations of thermodynamic parameters in a magnetic field it is connected not only with singularities of the density of states on the Fermi level but also with singularities in electron scattering which have the same periodicity. Let us consider this in greater detail.

The first theory of the magnetoresistance of metals taking into account the quantization of electron energy in a magnetic field was developed by Titeika.⁵¹ He used the following expression for the current density for the calculation of the electrical conductivity:

$$j_x = -e \sum_{n, n'} \sum_{p_{\parallel}, p'_{\parallel}} [W_{\alpha, \alpha'} f_{\alpha} (1 - f_{\alpha'}) - W_{\alpha', \alpha} f_{\alpha'} (1 - f_{\alpha})], \quad (4)$$

where $W_{\alpha, \alpha'}$ is the electron transition probability from the state α to the state α' due to the scattering potential, $f_{\alpha} = f(\epsilon_{\alpha}^{\mathbf{E}})$ is the electron distribution function in the α -state which depends upon the electric field $\mathbf{E} \parallel x$.

Dayvdov and Pomeranchuk⁵² applied the method of Titeika to the particular substance, bismuth, and studied oscillations of magnetoresistance. It was shown in that work that in spite of the fact that the electron effective mass was anisotropic the probability of a transition from one stationary state into another state on scattering by a short range potential was isotropic and depended only on the electron energy.

Later the method of Titeika was applied by Akhiezer and Rumer⁵⁴ to the study of oscillations of resistance of metals in a quantizing magnetic field and by Klinger⁵⁵ and Voronyuk⁵⁶ to calculations of galvanomagnetic phenomena in semiconductors. A rigorous justification of the Titeika method was given by Adams and Holstein⁵⁷ and by Kubo, Hasegawa, and Hashitsume⁵⁸ in a somewhat different way. Adams and Holstein developed a consistent quantum theory of the Sh-H effect using the quantum equation for the movement of the density matrix.⁵⁷ This was the development of the kinetic approach to the study of resistance oscillations in a magnetic field. Simultaneously and independently another approach based on diffusion mechanisms⁵⁹ was developed for the study of quantum oscillations of resistance in the course of elaboration of the theory of the Sh-H effect which is still the object of theoretical studies.

At present only an approximate theory of the Sh-H effect is available. It is developed in the most correct manner for the oscillations of the transverse magnetoresistance. As calculations show, for a strongly degenerate electron gas the transverse conductivity σ_{xx} for all electron scattering mechanisms may be represented as

$$\sigma_{xx} = \sum_{n, n'} \frac{G_{n, n'}(\epsilon_F)}{\sqrt{\epsilon_F - \hbar\omega_c \left(n + \frac{1}{2}\right)} \sqrt{\epsilon_F - \hbar\omega_c \left(n' + \frac{1}{2}\right)}}, \quad (5)$$

where $G_{n, n'}$ is a smooth function of the indices n, n' and the argument ϵ_F , which depends upon the scattering mechanism. The spin is not taken into account in Eq. (5). It follows from Eq. (5) that the conductivity increases sharply at $\epsilon_n = \hbar\omega_c \left(n + \frac{1}{2}\right) - \epsilon_F$. Analysis shows that σ_{xx} has two types of singularities. The scattering of electrons between different Landau sublevels causes the singularities in conductivity of the type $(\epsilon_F - \epsilon_n)^{-1/2}$.

The contribution to conductivity of the quantum level ϵ_n situated near the Fermi level is even more important because in this case the density of both the final and initial states is high. This contribution gives a nonintegrable singularity of the type $(\epsilon_F - \epsilon_n)^{-1}$ which causes logarithmic divergence in the conductivity. Adams and Holstein⁵⁷ calculated both oscillating parts of the transverse conductivity for a quadratic dispersion law:

$$\sigma_{xx} = \sigma_1 + \sigma_2, \quad (6)$$

where σ_1 contains a singularity of the type $(\epsilon_F - \epsilon_n)^{-1/2}$ and σ_2 —a singularity of the type $(\epsilon_2 - \epsilon_n)^{-1}$. The divergence in the conductivity disappears when the finite relaxation time τ of electrons is taken into account.

The expressions for σ_1 and σ_2 for a finite temperature taking broadening of Landau levels into account are:

$$\begin{aligned} \frac{\sigma_1}{\sigma_0} &= \frac{5x}{\sqrt{2}} \sqrt{\frac{\hbar\omega_c}{\epsilon_F}} \sum_{r=1}^{\infty} \frac{(-1)^r \sqrt{r}}{\text{sh}(rx)} e^{-2\pi r/\omega_c \tau} \cos\left(\frac{2\pi r \epsilon_F}{\hbar\omega_c} - \frac{\pi}{4}\right), \\ \frac{\sigma_2}{\sigma_0} &= \frac{3\pi x}{8} \frac{\hbar\omega_c}{\epsilon_F} \sum_{r=1}^{\infty} \frac{(-1)^r r}{\text{sh}(rx)} e^{-2\pi r/\omega_c \tau} \cos\left(\frac{2\pi r \epsilon_F}{\hbar\omega_c} - \frac{\pi}{2}\right), \end{aligned} \quad (7)$$

where $x = 2\pi^2 kT/\hbar\omega_c$, σ_0 is the conductivity in the quasi-classical limit which has for one type of carriers with the concentration n the asymptotic behavior in strong fields $\sigma_0 = e^2 n/m\tau\omega_c^2$, where m is the conductivity effective mass.

If the spin splitting of Landau levels is taken into account, then an additional factor $\cos(\pi r g/2)$, where g is the effective g -factor, determining the ratio of the energy of spin splitting $\Delta\epsilon_s$ to the magnitude of the cyclotron quantum: $g = \Delta\epsilon_s/\hbar\omega_c$, appears in equations (7).

Thus, the Sh-H oscillations make it possible to determine on the basis of Eqs. (7) the magnitude of the cyclotron mass m_c , the relaxation time τ and the spin splitting factor g ⁴⁸ as well as the oscillation period related to one of extremal sections of the Fermi surface for a given orientation of the magnetic field. For this purpose the amplitude of oscillations at the same field at different temperatures are compared to the amplitudes at different fields but at the same temperature. The g -factor, as a rule, is determined from the ratio of amplitudes of the first and second harmonics of oscillations related to the same extremal section of the Fermi surface. It is not difficult to see that amplitudes of the first and all odd harmonics of oscillations are equal to zero if the g -factor is equal to $(2k+1)$, where $k=0, 1, 2, \dots$. The fundamental harmonic has a doubled frequency and the oscillations curve exhibits an apparent increase by a factor two of the extremal section of the Fermi surface. This phenomenon is called the spin damping of oscillations. It is often observed in metals and semiconductors with a large effective g -factor and with a strongly anisotropic Fermi surface (for example, in Bi, Sb, As, their alloys, in graphite, etc.).

The relaxation time τ in Eqs. (7) may be related to some effective temperature $T_D = \hbar/\pi k\tau$ which was introduced for the first time by Dingle⁶⁰ for the characterization of the Sh-H oscillations. We note that the relaxation time τ determining the broadening of Landau levels can be substantially different from the relaxation

time of current carriers τ_0 at $H=0$. In fact, the magnitudes of τ and τ_0 correspond to completely different conditions under which the current carriers are scattered. Thus, the relaxation time τ_0 is the average for all electrons on the Fermi surface which describes isotropic scattering of electrons with the Fermi momentum at $H=0$. On the other hand, τ of Eqs. (7) describes the scattering by a Landau level of electrons which have a very small magnitude of the longitudinal momentum component $p_{||}$.

In particular, as was shown by Brown,⁶¹ the relaxation times τ_0 and τ in the case when scattering by ionized impurities dominates are related by the approximate relation:

$$\tau_0 \approx \left(\frac{\varepsilon_F}{\hbar}\right)^{1/2} \tau^{3/2}, \quad (8)$$

from which it follows that τ and τ_0 can substantially differ even by an order of magnitude.

The oscillating parts of the conductivity σ_1 and σ_2 differ between themselves by the phase shift $\pi/4$ and by the factor $(\hbar\omega_c/\varepsilon_F)^{1/2}$. If the ratio $\varepsilon_F/\hbar\omega_c$ is not too large so that a small number of Landau levels participates in transport processes the term σ_2 must dominate. The term σ_1 is substantial at small oscillation amplitudes when a large number of quantum levels participates in the conduction process. Predominance of σ_1 or σ_2 can be experimentally discerned first of all by the phase of oscillations.

Therefore, it is possible to conclude that the oscillations of the transverse magnetoresistance allow one to determine with high precision the extremal sections of the Fermi surface. The spin splitting factor, the Dingle temperature and the cyclotron masses of current carriers can be determined with lower precision.⁴⁹ It is necessary to keep in mind during a determination of an extremal section of the Fermi surface using oscillation effects that the Lifshits-Onsager equation (3) is valid only in the case when the extremal section does not depend upon the magnetic field and this is equivalent to a lack of dependence of the Fermi surface on the magnetic field. Indeed, the oscillations of the Fermi energy in a magnetic field are of the second order in the small parameter $(\hbar\omega_c/\varepsilon_F)^{1/2}$. On the approach to the ultra-quantum region of fields this parameter approaches the value of one and the change in the Fermi energy becomes important. Thus, at small values of quantum numbers n of the oscillations it is necessary to consider the movement of the Fermi level in a magnetic field during the determination of the quasiclassical extremal section S_{extr} of the Fermi surface. This effect is most important when there are present several groups of current carriers with different cyclotron masses because the shift of the Fermi level in this case is determined by the effects of flow of current carriers in a magnetic field between extrema in the spectrum. It is possible to take the movement of the Fermi level into account rigorously only by using a particular model of the electron energy spectrum (see, for example, Ref. 62).

In contrast to magnetoresistance the Hall coefficient R_H does not depend on scattering in the first order of

the parameter $1/\omega_c\tau$ and, therefore, does not experience oscillations. Oscillations of the Hall effect appear only in the second order of scattering. Thus, their amplitudes must be considerably smaller than the amplitudes of magnetoresistance oscillations.

Stradling and Artcliffe⁶³ while studying oscillations of the Hall effect in InSb found that the amplitudes of oscillations of the nondiagonal component ρ_{xy} had only 1% of the amplitude of oscillations of the diagonal component ρ_{xx} and were shifted in phase by $\pi/4$. A similar relation between amplitudes and phases of oscillations of ρ_{xy} and ρ_{xx} was found in n-GaSb and other doped semiconductors with a simple isoenergetic surface of electrons or holes.^{64,65}

Up to now we have considered oscillations of the transverse magnetoresistance for $\mathbf{j} \perp \mathbf{H}$. Oscillations of the longitudinal magnetoresistance ($\mathbf{j} \parallel \mathbf{H}$) have special features.

Thus it follows from the classical theory that for $\mathbf{j} \parallel \mathbf{H}$ the magnetic field does not influence the magnetoresistance at all in a metal with an isotropic dispersion of current carriers. This is connected with the fact that the movement of current carriers along a magnetic field is not distorted. Taking the quantization of the electron motion in a magnetic field into account gives a nonzero longitudinal magnetoresistance even in the case of a simple dispersion law and isotropic scattering^{66,67}. A change of the longitudinal magnetoresistance is caused in a quantizing magnetic field by the dependence on H of the probability of scattering of current carriers. We shall not consider the complicated problem of the calculation of the monotonic part of the longitudinal magnetic field which is solved at present only with some simplifying assumptions.^{68,69} From the point of view of the theory of the Sh-H effect only the oscillating part of the longitudinal magnetoresistance is of interest. The most important conclusion in this case is that with the same orientation of the magnetic field with respect to the crystallographic axes the period of the Sh-H oscillations does not depend upon the mutual orientation of the current \mathbf{j} and the field \mathbf{H} . However, the amplitude of the oscillations in ρ_{xx} in many cases is not described by Eqs. (7) for σ_1 and σ_2 given above. First of all, this refers to the temperature dependence of the oscillation amplitude which, according to Eqs. (7), for $k(T + T_D) > \hbar\omega_c$ is proportional to the Dingle factor $\exp[-k(T + T_D)/\hbar\omega_c]$. Thus, an anomalous dependence of the oscillation amplitude on T at which the amplitude decreases with decreasing temperature may arise in the longitudinal magnetoresistance oscillations. Such a dependence was observed, for example, in pure bismuth in the temperature range (12–1.3)K with the magnetic field oriented in the basis plane of the crystal.⁷⁰ The anomaly is caused by the fact that the largest contribution to the oscillating part of $\bar{\rho}_{xx}$ comes from transitions between the different Landau subbands which in strong magnetic fields must be accompanied by large changes of the longitudinal momentum $p_{||}$ (Fig. 4). Participation of phonons with large quasimomenta $\hbar q$ in order of magnitude equal to the Fermi surface dimensions is necessary to satisfy the momentum conservation law in the

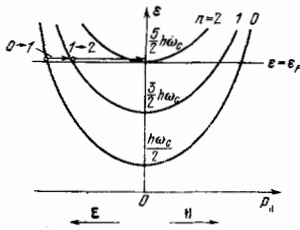


FIG. 4. Electron transitions between Landau subbands with different quantum numbers n which are characteristic for the longitudinal conductivity in a magnetic field with $\mathbf{j} \parallel \mathbf{H}$.

transitions in question. Freezing out of such phonons at low temperatures decreases the efficiency of scattering of electrons by phonons which is responsible for the longitudinal magnetoresistance oscillations and causes a decrease of the amplitude of oscillations with decreasing temperature. Calculations show that the oscillating part of the longitudinal magnetoresistance $\tilde{\rho}_{xx}$ in the case of electron scattering by phonons with the wave vector q_0 and frequency ω_{q_0} is described by the following expression:

$$\tilde{\rho}_{xx} = \text{const} \cdot \sqrt{H} \frac{x}{\text{sh } x} (\omega_{q_0}^2 N_{q_0} + \alpha), \quad (9)$$

where

$$N_{q_0} = (e^{h\omega_{q_0}/kT} - 1)^{-1}, \quad x = \frac{2\pi^2 kT}{h\omega_c},$$

α is a constant which takes into account the influence of scattering within the limits of one Landau subband and the influence of residual scattering by impurities.

Concluding this section, we note an interesting phenomenon related to the peculiarities of the Sh-H oscillations which are manifested in the appearance of additional frequencies equal to combinations of the frequencies corresponding to the sections of the Fermi surface. This phenomenon is observed in semimetals doped with different impurities and is a specific feature of the Sh-H effect because it is not observed in oscillations of the magnetic moment M (de Haas-van Alphen effect) in the same single crystal. The appearance of combination frequencies in the Sh-H oscillations was observed in Bi

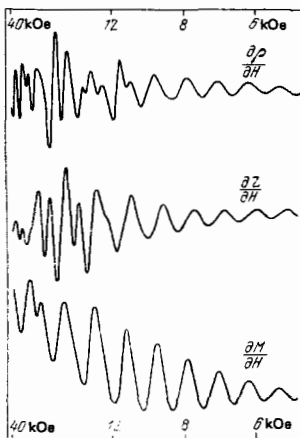


FIG. 5. Oscillations of the derivative of the magnetoresistance $\partial\rho/\partial H$, surface impedance $\partial Z/\partial H$ and magnetic moment $\partial M/\partial H$ in a magnetic field in the Bi-Te alloy.

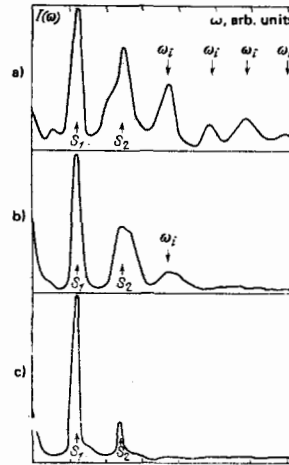


FIG. 6. Spectral composition $I(\omega)$ of the oscillation curves shown in Fig. 5: (a) $\partial\rho/\partial H$, (b) $\partial Z/\partial H$, (c) $\partial M/\partial H$.

and Bi_{1-x}Sb_x alloys doped with the donor impurity Te.⁷¹ As an illustration, recordings of oscillations of $\partial\rho/\partial H$, $\partial Z/\partial H$ and $\partial M/\partial H$ of a single crystal Bi-Te alloy sample in a field H parallel to the bisector axis are shown in Fig. 5 (Z is the surface impedance). For this orientation of H the oscillations contain two frequencies ω_1 and ω_2 corresponding to the two small sections S_1 and S_2 of the electron Fermi surface. It can be seen that the curves $\partial\rho/\partial H$ and $\partial Z/\partial H$ are qualitatively different from the oscillations of $\partial M/\partial H$. The spectral density curves $I(\omega)$ obtained by Fourier analysis of the oscillation curves of Fig. 5 are given in Fig. 6. The two peaks at the frequencies ω_1 and ω_2 determining the sections S_1 and S_2 are observed on the $I(\omega)$ curve corresponding to the oscillations of $\partial M/\partial H$. The harmonic composition of the oscillations of $\partial\rho/\partial H$ and $\partial Z/\partial H$ is much more complicated, and corresponding spectral curves contain besides the peaks at the frequencies ω_1 and ω_2 peaks at the frequencies described by the formula $\omega_i = n\omega_1 + m\omega_2$, where m and n are integers.

The results of Fourier analysis of the oscillations of $\partial\rho/\partial H$ are given in Table I.

In order to explain the appearance of combination frequencies in the Sh-H effect it is necessary to take into account the fact that according to Eq. (6) the oscillating part of the transverse conductivity σ_{xx} contains two different contributions σ_1 and σ_2 . It was shown by Lifshits⁴⁵ that at large quantum numbers the dominating term σ_1 (at $n \gg 1$, $\sigma_1 \gg \sigma_2$) is expressed linearly in terms of the oscillating part of the derivative of the magnetic momentum, $\sigma_1 \sim \partial M/\partial H$. This relationship is valid for

TABLE I.

| Observed frequencies ω_i , rel units | Relative height of the maximum $I(\omega_i)$ | Combination of frequencies $n\omega_1 + m\omega_2$ | Observed frequencies ω_i , rel units | Relative height of the maximum $I(\omega_i)$ | Combination of frequencies $n\omega_1 + m\omega_2$ |
|---|--|--|---|--|--|
| 11 | 1.00 | $\omega_1 = 11$ (section S_1) | 38 | 0.46 | $\omega_1 + \omega_2 = 38$ |
| 22 | 0.62 | $2\omega_1 = 22$ | 49 | 0.25 | $2\omega_1 + \omega_2 = 49$ |
| 27 | 0.92 | $\omega_2 = 27$ (section S_2) | 60 | 0.30 | $3\omega_1 + \omega_2 = 60$ |
| 32 | 0.25 | $3\omega_1 = 33$ | 72 | 0.14 | $4\omega_1 + \omega_2 = 71$ |

any shape of the Fermi surface and for any mechanism of scattering of current carriers.⁴⁴

The absence of combination frequencies in the de Haas-van Alphen effect allows one to relate their appearance to the term σ_2 an analogue of which is absent in the oscillating component of $\partial M/\partial H$. The contribution of σ_2 to the Sh-H oscillations increases with increasing magnetic field or with a decrease of quantum numbers n and may become very large near the ultra-quantum field limit, the term σ_2 diverging logarithmically if the linewidth Γ of the Landau level approaches zero. Adams and Holstein have shown⁵⁷ that the oscillation amplitudes of σ_1 and σ_2 are connected with the monotonic $\nu_0(\epsilon)$ and oscillating $\nu(\epsilon)$ components of the electron density of states by the following relations:

$$\begin{aligned} \sigma_1 &\sim \nu_0(\epsilon) \nu(\epsilon) \sqrt{\tau}, \\ \sigma_2 &\sim \nu^2(\epsilon) \tau. \end{aligned} \quad (10)$$

The following mechanism of increasing the amplitude of the oscillating part of the density of states $\nu(\epsilon)$ is possible in doped single crystals. At low temperatures scattering by ionized impurities is the main mechanism of relaxation in such materials determining the linewidth of the Landau level $\Gamma \sim \hbar/\tau$. Efficiency of this scattering is determined by the screening radius of impurity centers r_D which for a degenerate electron system is equal to

$$r_D = \left[\frac{4\pi e^2}{\kappa} (\nu_0(\epsilon_F) + \nu(\epsilon_F)) \right]^{-1/2}, \quad (11)$$

where κ is the dielectric permittivity and $\nu_0(\epsilon_F) + \nu(\epsilon_F)$ is the total density of states at the Fermi level.

According to Eq. (11), the oscillations of the density of states cause the oscillations of the screening radius r_D which in turn leads to oscillations of Γ and τ . As a result, the expansion of the term σ_2 [see Eq. (10)] in terms of the harmonics of basic frequencies ω_1 and ω_2 will contain combination frequencies $n\omega_1 + m\omega_2$. This effect is less important for the term σ_1 because, according to (10), $\sigma_1 \sim \nu_0(\epsilon) \sqrt{\sigma_2}$.

The appearance of combination frequencies in the Sh-H oscillations indicates that this effect has a strong nonlinearity due to which a single crystal in a magnetic field may operate as a frequency mixer. The presence of combination frequencies allows us to obtain additional information on the screening radius and on the character of the Friedel oscillations of electron density around charged centers.

2. SHUBNIKOV-de HAAS OSCILLATIONS UNDER CONDITIONS OF MAGNETIC BREAKDOWN IN METALS

The cause of oscillations of the magnetoresistance of a metal related to the nonmonotonic character of change of the electron density of states at the Fermi level in a magnetic field was considered earlier.

The other cause of oscillations of magnetoresistance is magnetic breakdown, i.e., the possibility of a transition of a conduction electron from one quasiclassical orbit to another in its motion in a magnetic field. An interesting phenomenon which has been given the name

of gigantic magnetic breakdown oscillations of resistance occurs as a result of interband magnetic breakdown.

The detailed analysis of interband magnetic breakdown was made by Blount⁷². The breakdown probability is the main characteristic of the breakdown

$$w = e^{-H_0/H}, \quad (12)$$

where $H_0 = (m_c/e\hbar)\Delta^2/\epsilon_F$, Δ is the energy gap between bands which separates quasiclassical orbits in the breakdown region. A very interesting situation arises in the fields $H \sim H_0$. The point is that the asymptotic behavior of the magnetoresistance $\rho(H)$ is very sensitive to the shape of the trajectory of electrons in a magnetic field. The transition from closed trajectories to open ones changes $\rho(H)$ by several orders of magnitude. The particle-wave dualism of the magnetic breakdown dynamics of conduction electrons is manifested in fields $H \sim H_0$. Two approaches to the interpretation of the magnetic breakdown are possible in this case. In one of them—the stochastic approach—electrons are considered as classical particles making stochastic jumps between orbits with the probability w . In the other approach the electron is considered as a wave for which the breakdown region is the semitransparent tunnel barrier through which the wave passes with the amplitude equal to \sqrt{w} . The second type of breakdown is called coherent. The occurrence of either one of these two cases in a metal is determined by the number and character of defects and impurities in the samples under investigation. The coherent magnetic breakdown is observed in pure perfect single crystals of metals in strong magnetic fields.

The results obtained on beryllium⁷³⁻⁷⁵ may be given as an example of coherent breakdown. In this metal the breakdown occurs between the small orbit of the “cigar” (“needle”) and the large orbit of the “crown” (“monster”) (Fig. 7). The breakdown is accompanied by gigantic oscillations of $\rho(H)$ which are periodic in the reciprocal of the magnetic field (Fig. 8). Their period is connected with the extremal area of the small orbit by the Lifshits-Onsager equation (3).

The narrow layers of trajectories responsible for the oscillations are determined not only by extremal sections of the “cigar” but also by narrow partitions in the “crown” to which the noncentral sections of the “cigar” correspond. Because of this, modulation of the oscillation amplitude (beats) with the difference frequency oc-

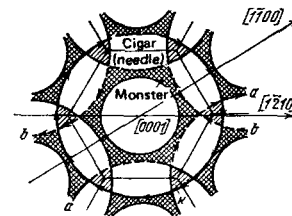


FIG. 7. Trajectory of electrons in Be (continuous thick line) in the case of magnetic breakdown between the two parts of the Fermi surface—the “crown” (“monster”) and the “cigar” (“needle”) at $H \parallel [0001]$.

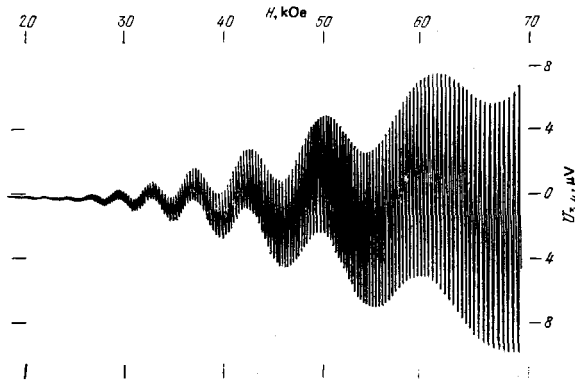


FIG. 8. Magnetic breakdown oscillations of $d\rho/dH$ in Be in the case of breakdown between the "crown" and the "cigar."

curs over quite a broad range of fields. It is possible to evaluate from experimental data the magnitude of Δ which is equal to ~ 0.08 eV for beryllium. The value of H_0 for beryllium is ~ 110 kOe.

Experiments made on transition metals of the 5th group have shown that the breakdown field H_0 increases on passing from vanadium to niobium. The magnetic breakdown in these metals occurs through the gap which is caused by the spin-orbit interaction. Hence, the study of magnetic breakdown oscillations of the resistance is a good method for investigating the spin-orbit interaction in transition metals. Comparison of experimental data with calculations allows one to evaluate the validity of a chosen theoretical model. Let us note also that gigantic magnetic breakdown oscillations are used at present for precise determination of the magnitude and gradient of a magnetic field^{74,75}.

3. APPLICATIONS OF THE SHUBNIKOV-de HAAS EFFECT

a) Study of rearrangement of the energy spectrum of metals under extremal conditions

We shall illustrate the use of the Sh-H effect for studying the rearrangement of the electron spectrum of metals on the example of investigating Lifshits topological phase transitions⁷⁶ of the 2.5 order. These transitions are observed in metals, semimetals and doped semiconductors when the external parameters are altered. The 2.5 order transition is a jumplike change of the topology of the Fermi surface at some critical value of an external parameter responsible for the electron spectrum change. Topological phase transitions can be observed most simply in substances subjected to a high pressure or a strong uniaxial elastic deformations. Topological Lifshits transitions accompanying isotropic compression were observed for the first time in the studies of bismuth and its alloys,⁷⁷ and also in the studies of cadmium,⁷⁸ tellurium and its alloys.⁷⁹ Uniaxial stresses of the lattice changing the crystal symmetry substantially increase the probability of observation of the 2.5 order transitions. The topological phase transitions of bismuth-antimony alloys doped by donor and acceptor type impurities⁸⁰⁻⁸² are most striking and diverse.

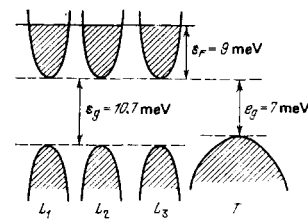


FIG. 9. Position of electron and hole extreme close to the Fermi level in the $\text{Bi}_{0.924}\text{Sb}_{0.076}$ alloy of n -type.

We illustrate these transitions on the example of the Bi-Sb semiconductor alloy of n -type. The Fermi surface of such an alloy consists of three electron quasiellipsoids situated in the L -points of the Brillouin zone (centers of the pseudo-hexagonal faces). The energy spectrum diagram is shown in Fig. 9.

The change of sections and volumes of electron surfaces under uniaxial stress was determined by means of recording the Sh-H effect at different orientations of the magnetic field.

The current carrier concentration in a doped semiconductor alloy is completely determined by the concentration of doping impurities and does not change if there is no overlapping of separate extrema of the conduction and valence bands in the process of the energy spectrum change. The change of the volumes of separate ellipsoids at constant total concentration of current carriers can occur evidently only at the expense of other ellipsoids due to the redistribution of current carriers between extrema. It is convenient to observe experimentally the change of the extremal section of those isoenergetic surfaces which grow when electron transitions occur. Their growth, evidently, must stop at the moment when complete depopulation occurs of those extrema in which a decrease of the current carrier concentration takes place.

The following types of topological transitions occur in the semiconducting Bi-Sb alloy of n -type in the case of a uniaxial compression deformation along the binary (C_2) and the bisector (C_1) axes⁸¹:

1) In the case of a compression along C_1 or an extension along C_2 the volume of one of the quasiellipsoids increases and that of the other two decreases. A transition of the type 3EL \rightarrow 1EL at which electrons from three extrema flow to one extremum takes place at the critical value of the deformation. The dependence of the sections of increasing and decreasing isoenergetic surfaces is shown in Fig. 10, a.

2) In the case of a compression along C_2 or an extension along C_1 the volume of two quasiellipsoids increases and that of the third ellipsoid decreases. A transition of the type 3EL \rightarrow 2EL as a result of which all electrons are transferred to two equivalent extrema takes place at the critical value of the deformation (Fig. 10, b).

The topological transition of the type 3EL + 1 Hole \rightarrow 2EL + Hole which causes the disappearance of one electron quasiellipsoid⁸² is observed in the case of uni-

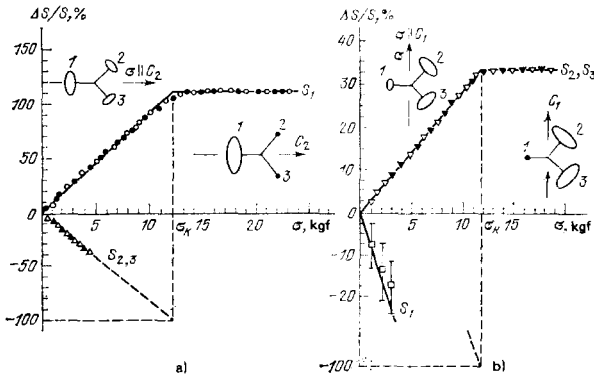


FIG. 10. Dependence of the extremal sections of the Fermi surface on the load in the case of deformation along the axes C_2 (a) and C_1 (b) in the $\text{Bi}_{0.924}\text{Sb}_{0.076}$ alloy of n -type.

axial deformations in pure bismuth in which in the original state there are equal numbers of electrons and holes filling three electron and one hole energy surfaces. Electron transitions of the type semiconductor-metal with formation of isoenergetic surfaces $2EL + 1$ Hole or $1EL + 1$ Hole, which cause the appearance of electrons and holes in equal numbers and the beginning of the Sh-H oscillations with frequencies corresponding to the sections of electron and hole isoenergetic surfaces (Fig. 11), take place in the undoped semiconducting Bi-Sb alloy in the case of a uniaxial deformation along the C_2 and C_1 axes.

b) New possibility of studying the density of states in doped semiconductors

Recently an interesting possibility was found experimentally of obtaining the energy dependence of the density of states in the impurity band of a strongly doped semiconductor by investigating the Sh-H effect under pressure. A study was made of gallium antimonide of the n -type with two types of extrema in the conduction band situated at the Γ - and L -points of the Brillouin zone and separated by a small energy gap ($\epsilon_{\Gamma L} \sim 100$ meV). The conduction band structure of GaSb is shown in Fig. 12. Only the lower Γ -extremum which is characterized by a sufficiently small density of states and by the small value of the effective mass at the bottom of the band, $m_{0\Gamma} \approx 0.04 m_0$ (m_0 is the mass of a free elec-

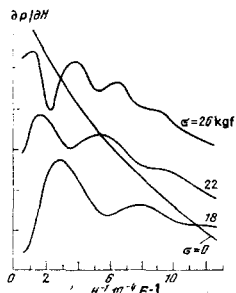


FIG. 11. Appearance of the Shubnikov-de Haas oscillation in the case of $\mathbf{H} \parallel C_1$ in the semiconducting alloy $\text{Bi}_{0.925}\text{Sb}_{0.075}$ for a uniaxial deformation $\sigma \parallel C_2$ as a result of the semiconductor-metal transition. Loads σ (in kgf) shown near the corresponding curves.

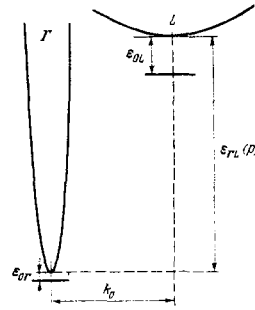


FIG. 12. Structure of the conduction band of gallium antimonide doped by donor impurities. $\epsilon_{0\Gamma}$ and ϵ_{0L} are the energies of splitting of the impurity level from the Γ and L bands.

tron), is filled at the impurity electron concentration $n_{\Gamma} \approx 10^{18} \text{ cm}^{-3}$.

Under the influence of pressure the energy gap $\epsilon_{\Gamma L}$ between Γ - and L -extrema decreases at a rate of 10 meV/kbar, and the bottom of the Γ -conduction band approaches the bottom of the empty L -band.

The Fermi surface of Γ -electrons in GaSb is close to spherical with good accuracy. Therefore, the study of the Sh-H oscillations due to Γ -electrons under pressure is a convenient and precise method of determining their concentration by using the simple relation

$$n_{\Gamma} = \frac{8}{3\pi^2} S_{\text{extr}}^{3/2} \quad (13)$$

where h is the Planck constant, and S_{extr} is the extremal section which is calculated from the oscillation period $\Delta(1/H)$ using the Lifshits-Onsager formula (3). The Sh-H oscillations due to Γ -electrons in the GaSb sample with the original concentration $n_{\Gamma} = 1.35 \cdot 10^{18} \text{ cm}^{-3}$ at several values of pressure⁸³ are given as an example in Fig. 13. The experiment shows that the concentration n_{Γ} is constant in the region of low pressures and starting from some value of pressure $p = p'$, which is the lower the higher the doping level of a semiconductor,

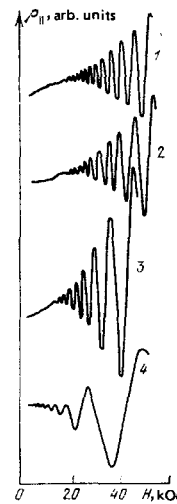


FIG. 13. Sh-H oscillations of the longitudinal magnetoresistance $\rho_{||}$ of the GaSb sample with $n_{\Gamma} = 1.35 \cdot 10^{18} \text{ cm}^{-3}$ (at $p = 0$) for different pressures p equal to (in kbar) 1.0 (1), 3.4 (2), 5.6 (3), and 8.3 (4).

n_{Γ} decreases and becomes equal to zero at $p = p_k \approx 9$ kbar. It follows from the analysis that the decrease of the concentration n_{Γ} under pressure occurs due to Γ - and L -levels approaching each other and Γ -electrons flowing to the impurity level split from the L -band.

The number of transferred electrons $n_i(\epsilon')$ is determined by the position of the Fermi level on the energy scale and by the density of states in the impurity band $g_{iL}(\epsilon')$:

$$n_i(\epsilon') = \int_{-\infty}^{\epsilon'} g_{iL}(\epsilon) d\epsilon, \quad (14)$$

where

$$\epsilon' = \epsilon_{\Gamma}^{\Gamma}(p) - [\epsilon_{\Gamma L}(p) - \epsilon_{iL}] = \epsilon_{\Gamma}^{\Gamma}(p) - \left[\epsilon_{\Gamma L}(0) - \left| \frac{\partial \epsilon_{\Gamma L}}{\partial p} \right| p - \epsilon_{iL} \right] \quad (15)$$

($\epsilon_{\Gamma}^{\Gamma}$ is the Fermi energy in the Γ -band) (see Fig. 12). On the other hand, it follows from the condition of electrical neutrality that

$$n_i(\epsilon') = N_d - N_a - n_{\Gamma}(\epsilon'), \quad (16)$$

where N_d, N_a are permanent donor and acceptor concentrations in the semiconductor. It follows from Eqs. (14)–(16) that the density of states $g_{iL}(\epsilon')$ is

$$g_{iL}(\epsilon') = \frac{\partial n_i(\epsilon')}{\partial \epsilon'} = - \frac{\partial n_{\Gamma}(\epsilon')}{\partial \epsilon'}. \quad (17)$$

The Γ -band with known parameters of the electron spectrum may be used as a kind of a standard for the determination of the Fermi level on the energy scale. Changing the relative positions of Γ - and L -extrema by pressure and controlling n_{Γ} by the Sh-H oscillations it is possible to plot by points the density of states in the L -band. This was done in the paper of Ref. 83. The dependence of g_{iL} on ϵ for the GaSb sample with an original concentration of Γ -electrons equal to $2.8 \cdot 10^{17} \text{ cm}^{-3}$ is given in Fig. 14. It was shown that the density of states at the impurity level may be described with good accuracy by the following Gaussian curve

$$g_{iL}(\epsilon) = \frac{N_d}{\gamma \sqrt{\pi}} e^{-(\epsilon - \epsilon_0)^2 / \gamma^2} \quad (18)$$

and the parameter γ , determining the linewidth of the level, can be found.

The dependence of the density of states in Γ - and L -conduction bands and in the impurity L -band for samples with original concentration of Γ -electrons equal to $2.0 \cdot 10^{17}$ and $1.14 \cdot 10^{18} \text{ cm}^{-3}$ is given in Fig. 15.

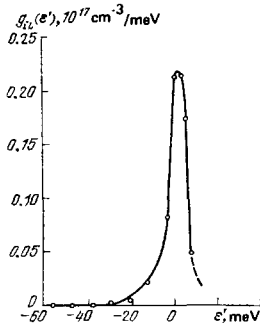


FIG. 14. Density of states in the impurity L -band in the GaSb sample with $n_{\Gamma} = 2.8 \cdot 10^{17} \text{ cm}^{-3}$ at $p = 0$.

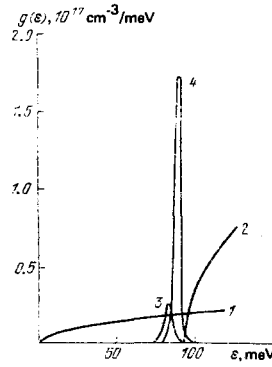


FIG. 15. Density of states in the conduction band of the GaSb sample doped by Te. (1) Γ -band; (2) L -band (for one L -extremum); (3) impurity L -band (sample with $n_{\Gamma} = 2 \cdot 10^{17} \text{ cm}^{-3}$ at $p = 0$); (4) impurity L -band (sample with $n_{\Gamma} = 1.14 \cdot 10^{18} \text{ cm}^{-3}$ at $p = 0$).

Therefore, due to the fact of coexistence of impurity states with band states there exists in principle a possibility of investigating disordered systems by studying the properties of band electrons to which the experimental methods effectively used in the physics of crystalline solids may be applied.

c) Study of long duration nonequilibrium processes in semiconductors

In concluding this section we consider the possibility of applying the Sh-H effect to the study of long duration relaxation processes in semiconductors.

Ternary compounds $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ doped by elements of the third group (In, Ga, Al, Te) are semiconductors in which long duration relaxation processes are manifested most clearly.⁸⁴⁻⁸⁶ In particular, the In dopant in the $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ alloys with a concentration of about one atomic percent forms a large capacity impurity level the position of which in the band spectrum of the semiconductor depends on the tin concentration x , on pressure, and on magnetic field.^{87,88} It is established experimentally that the In impurity level stabilizes the Fermi level in the alloy. In the case when the impurity level is situated in the conduction band or in the valence band the Fermi level in the alloy is coincident with the impurity level and the equilibrium concentration of free carriers is completely determined by the position of the latter.

The most characteristic special feature of $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ with the In dopant is the possibility of generation by a magnetic field or by light of a nonequilibrium metallic state the relaxation time of which is of the order of hours and tens of hours. The relaxation time depends exponentially on the temperature so at $T > 20 \text{ K}$ the time τ decreases to values of fractions of a second.⁸⁴ This property of the $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ alloys with 1 at.% of In makes possible the production of a long-lived nonequilibrium state (for example, a state with a large nonequilibrium electron concentration). Let us consider, as an example, one of the ways of producing a nonequilibrium electron concentration in the conduction band by a magnetic field.

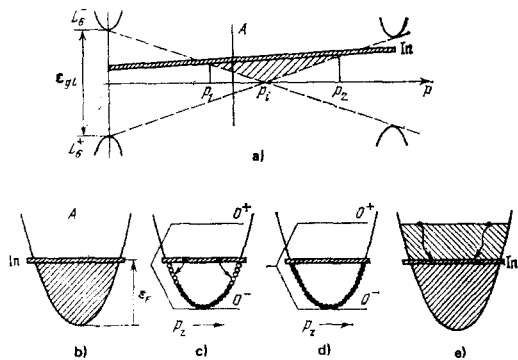


FIG. 16. Energy diagrams illustrating the formation of a non-equilibrium metal in a magnetic field (cf. text).

For this purpose by varying the composition and the pressure the original state of the alloy is chosen in which the impurity level is situated in the conduction band close to its bottom. The energy spectrum of the original state of the alloy at $H=0$ corresponds to the section A in the diagram in Fig. 16, a and is shown schematically in Fig. 16, b. The nonequilibrium electron concentration is produced by the following cycle.

A strong magnetic field corresponding to the ultra-quantum region of fields for the original electron concentration is switched on at a temperature $T \geq 20$ K, and as a result electrons are set on the one-dimensional Landau parabola with quantum numbers $n=0, s=-1$. The density of states in the band grows with the growth of the magnetic field proportionally to the degree of degeneracy of electronic states with respect to the longitudinal momentum $\sim eH/\pi\hbar c$. Due to this and also due to the fact that the Fermi level is fixed to the impurity level, a flow starts of electrons from the impurity level into the band in correspondence with its growing capacity (Fig. 16, c). The flow process occurs rather fast at $T \geq 20$ K and the electron concentration in the band becomes higher than original concentration (Fig. 16, d). Then, the magnetic field is kept constant and the crystal is cooled down to liquid helium temperatures at which the electron flow time from the level into the band or vice versa increases to several hours. The magnetic field is switched off after that.

The nonequilibrium electron concentration equal to the equilibrium concentration in the magnetic field is "frozen" in the conduction band of the $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ alloy as a result of this cycle. The Fermi level takes the nonequilibrium position above the impurity level of In and the slow process of the reverse flow of electrons out of the band into the impurity level begins. Nonequilibrium electrons form the degenerate Fermi gas in the conduction band which does not differ by its properties from the electron system of an ordinary metal. The $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ behaves under these conditions as a single band metal with the Fermi surface slowly shrinking in time.

The Sh-H effect allows us to prove experimentally the existence of such a state with a nonequilibrium electron concentration. For this purpose it is necessary to record the Sh-H effect at times separated by equal time

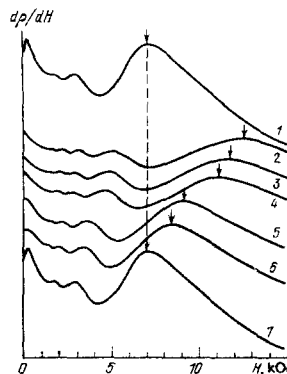


FIG. 17. Recordings of the Shubnikov-de Haas oscillations of $\partial\rho/\partial H$ for the $\text{Pb}_{0.75}\text{Sb}_{0.25} + 0.5$ at. % In alloy at $p = 5.4$ kbar, $\mathbf{H} \parallel \langle 100 \rangle$ which correspond to the equilibrium (1, 7) and non-equilibrium (2-6) states of electrons in the conduction band (cf. text).

intervals (for example, every 20-30 min). Because the recording of one oscillation curve takes 2-3 minutes, the electron concentration in the band during the recording may be considered constant. The curves recorded spaced by sufficiently large time intervals differ in their oscillation periods and show a slow decrease with time of the electron concentration in the band.

The successive Sh-H oscillations curves due to non-equilibrium electrons in $\text{Pb}_{0.75}\text{Sn}_{0.25}\text{Te}$ alloy with 0.5 at.% of In obtained in Ref. 84 are shown in Fig. 17. The curves 3-6 are recorded 5, 10, 30, and 60 minutes respectively after the field was switched off at the temperature of 2 K. The curves 1, 7 correspond to the equilibrium state (1—before the formation of the non-equilibrium state in a magnetic field, 7—after the "destruction" of the nonequilibrium state by heating the crystal up to ~ 20 K).

A similar nonequilibrium state may be produced by excitation of electrons from an impurity level by infrared radiation. The electron concentration in the conduction band at any level of infrared pumping may also be determined by means of recording the Sh-H oscillations.⁸⁵

Some cases interesting from the authors' point of view of using the Sh-H effect for finding and studying new phenomena in contemporary solid state physics were considered above. The examples given serve only as illustrations of wide possibilities for using this effect under conditions when application of other methods turns out to be practically difficult or simply impossible and consequently is not exhaustive. However, they are, from our point of view, sufficiently convincing illustrations of wide and often unexpected possibilities of the Sh-H effect which was discovered 50 years ago. Let us hope that also in future the field of application of the Sh-H effect will be continually expanding and finding ever more new applications.

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