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An investigation of the dispersion law for carriers in bismuth doped with acceptor-type impurities

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The Shubnikov–de Haas effect has been investigated in single-crystal bismuth samples doped with Sn and Pb in fields of up to 60 kOe at liquid-helium temperatures. An increase in the cyclotron masses $m_{cT}(\epsilon_{FT})$ of holes at T with increase in the volume of the Fermi surface was found. In the framework of the Kane two-band model, a gap of $\epsilon_{gT} = (200 \pm 40)$ meV was found at T. The energy dependence of the low cyclotron masses of carriers $m_{cL}(\epsilon_{FL})$ at L on lowering the Fermi level (**H** parallel to the binary axis) was found to be linear. The parameters of the Abrikosov dispersion law for carriers at the point L were determined as $\nu_\chi = 0.99 \times 10^8$ cm/s and $\nu_y = 0.74 \times 10^8$ cm/s. It was shown that the gap ϵ_{gL} at L lies in the interval $0 < |\epsilon_{gL}| < 15$ meV. It was also shown that the band overlap in the acceptor-doped bismuth samples investigated did not depend on the impurity concentration. It was found that the parameter $\epsilon_{0v} + \epsilon_{gL}/2 = (46 \pm 2)$ meV.

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1. INTRODUCTION

The energy spectrum of bismuth has been investigated in a large number of experimental and theoretical papers.[1–4] However, right up to the present time models describing the spectrum of carriers at the points L and T in the reduced Brillouin zone, as well as certain important parameters of the spectrum have not been given unique values, owing to the complexity of the band structure of bismuth and the strong anisotropy of its properties. There are considerable discrepancies in the estimates of the degree of deviation of the hole spectrum from parabolic.[5–8] At the same time the values of the gap parameter of the electron spectrum at L found in various papers do not agree not only in magnitude, but even in sign.[4,7–16]

The introduction of acceptor-type impurities (Pb, Sn) into the bismuth lattice allows the Fermi level to be displaced within wide limits. This opens up the possibility in principle of deriving the energy dependence of the basic parameters of the carrier spectrum at the overlapping extrema of the conduction band L_s and the valence band T_{45}^-,[3] which is essential for checking the theoretical models describing the laws of carrier dispersion at the L- and T-extrema.[1–4,17] No less interesting is the problem of finding new extrema in the valence band of Bi on suitably lowering the Fermi level.

At acceptor impurity concentrations below 0.1 at.% the lattice parameters of Bi do not vary markedly,[3] and the effect of the acceptors is apparently only to make the electron and hole concentrations unequal and to change the electron and hole mobilities.

Theoretical estimates indicate that localization of carriers at impurity centers in Bi has an extremely low probability, owing to the strong screening of the impurity potential by free carriers.[19] It should be noted that even when an impurity level is formed, it inevitably falls within the spectrum of allowed states, owing to the overlap of the valence band with the conduction band, which must lead to delocalization. At the present time there is no direct evidence of the existence of local or quasi-local impurity levels in the spectrum of impurity-doped bismuth.

The investigation of the Fermi surface in impurity-doped bismuth by quantum-oscillation effects is made very difficult because the relaxation times τ of the electrons and holes, which reach $\sim 10^{-9}$ s in pure single crystals of bismuth at liquid-helium temperatures,[20,22] drop rapidly with increase in the impurity concentration. At the same time the dominant scattering mechanism at liquid-helium temperatures becomes scattering by ionized impurities,[5,23–26] which leads to a marked dependence of τ on energy.

Decrease in relaxation time with increase in impurity concentration by several orders does not impose severe restrictions on the investigation of galvanomagnetic and thermomagnetic effects in Bi, but this involves its own inherent difficulties. These are primarily associated with the ambiguity in the interpretation of the experimental data, as a result of the obvious arbitrariness in the choice of a model for the band structure. Additional

complications are caused by the anisotropic nature of the relaxation time,[20] the strongly non-parabolic nature of the dispersion law for carriers at the L-extrema,[5,27] and also by the necessity to take account of intervalley scattering at high temperatures.[22]

There is still another problem of a fundamental nature. In the majority of papers devoted to the investigation of the electro-physical properties of Bi doped with impurities, the experimental data are discussed on the basis of a "rigid"-band model, according to which the band structure of Bi does not vary at impurity concentrations below 0.1 at.%. Until very recently there was no reliable evidence justifying the rigid-band model. In a number of papers the opinion was expressed that even small impurity concentrations alter the parameters of the dispersion law of the carriers in Bi.[28]

In the present work the Shubnikov–de Haas effect in single-crystal bismuth samples doped with Pb and Sn has been investigated. A distinctly non-parabolic nature was found for the law of hole dispersion at T. The magnitude of the gap ε_{gT} at T determined on the basis of the Kane approximation was (200 ± 40 meV). For cross sections of the Fermi surface at L near to the minimum ones (**H** parallel to the binary axis), a linear dependence of the cyclotron mass on energy was found and the value of the parameter $\varepsilon_{ov}+\varepsilon_{gL}/2$ was found to be (46 ± 2) meV (ε_{ov} is the band overlap, and ε_{gL} is the direct gap at L). It was shown that the absolute value of the gap ε_{gL} at L lies in the range $0 \leq |\varepsilon_{gL}| < 15$ meV, and that the amount of band overlap ε_0 does not depend on the impurity hole concentration when the Fermi level in Bi is lowered by tens of meV, which is direct experimental confirmation of the validity of the rigid-band model.

2. MEASURING TECHNIQUES. SAMPLES

In the present work single crystals of Bi doped with Sn and Pb in concentrations of up to 0.04 and 0.1% respectively were investigated. The samples in the form of rectangular parallelepipeds with typical dimension ~ (0.8×0.8×3.0) mm were cut from single-crystal ingots by the electric-spark method. All the samples were cut along the binary axis (C_2), the lateral faces coinciding with the trigonal (C_3) and bisector directions.

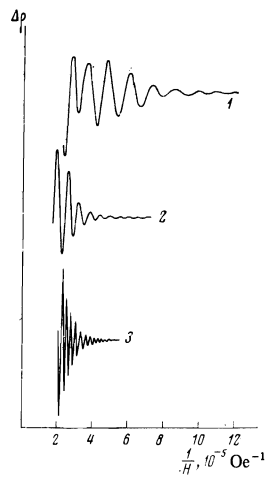

FIG. 1. Shubnikov oscillations in the transverse magnetoresistance $\Delta\rho$ from holes at T in Bi samples doped with acceptor impurities for H ∥ C_3 and T = 4.2 K (monotonic movement of $\Delta\rho$ suppressed): 1—$P_T = 4.2 \cdot 10^{18}$ cm^{-3}; 2—$P_T = 1.27 \cdot 10^{18}$ cm^{-3}; 3—$P_T = 5.5 \cdot 10^{18}$ cm^{-3}.

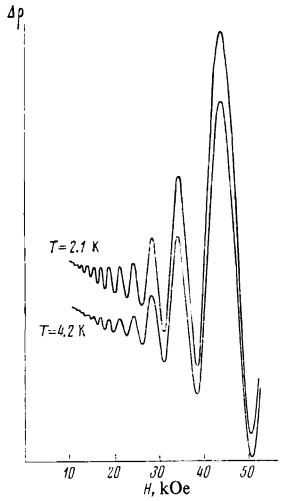

FIG. 2. Shubnikov oscillations in the transverse magnetoresistance $\Delta\rho$ from holes at T in $Bi_{1-x}Pb_x$ samples with $P_T = 1.27 \cdot 10^{18}$ cm^{-3} for H ∥ C_3 and temperatures of T = 4.2 K and T = 2.1 K (monotonic movement partially suppressed).

The accuracy in orienting the faces with respect to the crystallographic axes was ±1%. Before mounting, the samples were etched in nitric acid and then washed in distilled water and alcohol. The current contacts in the form of thin strips of thin tinned copper foil were soldered to the end faces of the samples with a miniature soldering iron. The potential contacts (copper wire, 50 μm dia.) were welded on with an electric spark welding machine. The distance between the potential contacts did not exceed 1 mm.

The investigation of the Shubnikov–de Haas effect in Bi samples doped with Pb and Sn was carried out in magnetic fields of up to 60 kOe at liquid-helium temperatures on a setup which permitted automatic recording of the $\rho(H)$ and $\partial\rho(H)/\partial H$ curves. In recording the $\rho(H)$ curves, the monotonic trend was suppressed with an analog computing arrangement which generated a signal ~ $\alpha H \pm \beta H^2$, permitting a considerable increase in the accuracy of determining the periods of the Shubnikov oscillations $\Delta(1/H) = eh/cS_{extr}$ and the cyclotron masses $m_c(\varepsilon_F)$.

3. MEASUREMENT RESULTS

Shubnikov oscillations both of light carriers at L and of holes at T were observed in all the investigated samples. One task of the present work was to record the oscillations from small sections of the Fermi surface at L and T with the Fermi level displaced within wide limits. The maximum concentration P_T of the holes at T in the samples investigated was 5.5×10^{18} cm^{-3}.

The transverse magnetoresistance oscillations (**H** ∥ C_3) from a small section of the hole elipsoid at T (Fig. 1) were recorded in fields of up to 60 kOe at liquid-helium temperatures. The cyclotron masses $m_{cT}(\varepsilon_{FT})$ for **H** ∥ C_3 were calculated from the temperature dependence of the amplitude of the Shubnikov oscillations (Fig. 2). It was found that the low cyclotron mass $m_{cT}(\varepsilon_{FT})$ of holes markedly increases on increasing the section of the Fermi surface, which indicates a non-parabolic dispersion law for holes at T.

In some samples the oscillations corresponding to a small section of the Fermi hole surface at T clearly

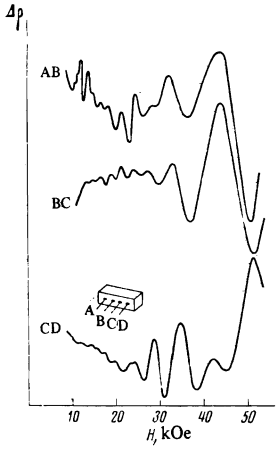

FIG. 3. Shubnikov oscillations in the transverse magnetoresistance $\Delta\rho$ from holes at T in a non-homogeneous $Bi_{1-x}Pb_x$ sample ($x = 1.10^{-3}$) for $H \parallel C_3$ and $T = 4.2$ K measured between neighboring pairs of potential contacts (contact designations shown beside curves).

exhibited a pulsating character. To elucidate the cause of this pulsation, four potential contacts were attached to one of the samples, and the Shubnikov oscillations were recorded consecutively between neighboring pairs of contacts (Fig. 3). As is evident from Fig. 3, the character of the pulsations varies on going from one pair of contacts to another. The most probable cause of pulsations of this type is inhomogeneity in the distribution of impurities in the sample. In the reduction of the experimental data, the Shubnikov oscillations from holes at T with a pulsating character were excluded from the analysis.

Longitudinal magnetoresistance oscillators ($\mathbf{H} \parallel \mathbf{I} \parallel C_2$) from small sections of the Fermi surface at L were recorded in fields up to 12 kOe. The choice of the binary direction for recording the Shubnikov oscillations from light carriers at L is connected with the fact that for $\mathbf{H} \parallel \mathbf{I}$ the oscillations from two sections of equal size near to the minimum have a monochromatic character. This guarantees high accuracy in determining the extremum sections S_L and the cyclotron masses $m_{cL}(\varepsilon_{FL})$.

Increasing the concentration of acceptor impurity in Bi led to a marked fall in the frequency of the Shubnikov oscillations from the electron Fermi surface at L (Fig. 4) and to a decrease in the electron cyclotron mass $m_{cL}(\varepsilon_{FL})$. In a sample of Bi doped with tin having a hole

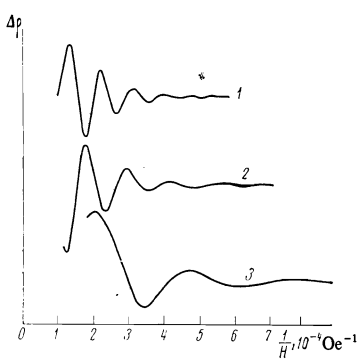

FIG. 4. Shubnikov oscillation in the longitudinal magnetoresistance $\Delta\rho$ from light electrons at L in samples of Bi doped with acceptor impurities for $\mathbf{H} \parallel \mathbf{I}$ and $T = 2.1$ K (monotonic movement of $\Delta\rho$ suppressed): 1—$P_T = 3.51 \cdot 10^{17}$ cm^{-3}; 2—$P_T = 6.86 \cdot 10^{17}$ cm^{-3}; 3—$P_T = 1.27 \cdot 10^{18}$ cm^{-3}.

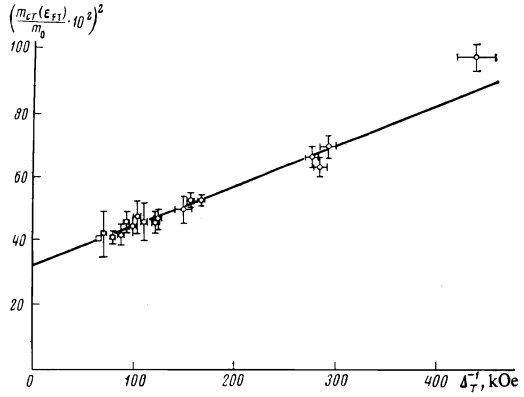

FIG. 5. Dependence of the square of the low cyclotron mass $m_{cT}(\varepsilon_{FT})$ of holes at T on the reciprocal of the period of the Shubnikov oscillations $\Delta_T^{-1} \sim S_{extr}$ for $\mathbf{H} \parallel C_3$ in Bi samples doped with acceptor impurities: □—pure Bi[35]; ○—data from the present work.

concentration of $P_T = 5.5 \times 10^{18}$ cm^{-3} at the point T oscillations from light holes at L were found.[37] It should be noted that the first indirect evidence for the existence of hole extrema at L were derived by investigating the angular dependence patterns of the transverse magnetoresistances and also the anisotropy and field dependences of the Hall coefficient.[29-33]

4. DISCUSSION OF RESULTS

The dependence of the low cyclotron mass $m_{cT}(\varepsilon_{cT})$ on the reciprocal of the Shubnikov oscillations $\Delta_T^{-1} \sim S_{extr}(\mathbf{H} \parallel C_3)$ for the hole ellipsoid at T is shown in Fig. 5. The increase in $m_{cT}(\varepsilon_{FT})$ with increase in Δ_T^{-1} is a consequence of the non-parabolic nature of the hole spectrum at T. In view of the lack of reliable data on the band structure at T in Bi, it is rational to use the two-band Kane model as a first approximation to describe the non-parabolic hole spectrum at T. According to this model

$$[m_{cT}(\varepsilon_{FT})]^2 = [m_{cT}(0)]^2 + 2e\hbar m_{cT}(0)/\pi c\varepsilon_{gT}\Delta_T, \qquad (1)$$

where $m_{cT}(0) \sim \varepsilon_{gT}$ the cyclotron mass at the top of the valence band at T, and ε_{gT} is the gap at T. The continuous line in Fig. 5 has been drawn from formula (1) with the conditions that $m_{cT}(0) = 5.7 \times 10^{-2} m_0$, $\varepsilon_{gT} = 200$ meV. The accuracy in determining ε_{gT} was ± 40 meV. Attempts have also been made previously[5-7] to estimate the gap ε_{gT} experimentally, but the data obtained in the present work are based on the results of direct measurement and may therefore be considered the most reliable.

When investigating the strongly non-parabolic spectrum of carriers at L in Bi doped with acceptor impurities, the Fermi energy of holes ε_{FT} at T was employed in the present work as an independent energy scale. This was calculated according to the two-band model as

$$\varepsilon_F = \varepsilon_{par} - \frac{\varepsilon_g}{2} + \left(\left(\frac{\varepsilon_g}{2}\right)^2 + \varepsilon_{par}^2\right)^{1/2}, \qquad (2)$$

$$\varepsilon_{par} = e\hbar/m_c(\varepsilon_F)c\Delta \qquad (3)$$

using the data given in Fig. 5. The dependence of the low cyclotron mass $m_{cL}(\varepsilon_{FL})$ of electrons and holes at $L(\mathbf{H} \parallel \mathbf{I})$ on the Fermi energy of holes ε_{FT} at T is shown in Fig. 6. As a first approximation this dependence has a linear character.

From the Abrikosov[4] theory it follows that for sections S of the Fermi surface at L near to the minimum one, the following relationship is valid with a fair degree of accuracy:

$$S=2\pi m_{cL}(0)\varepsilon_{FL}(1+\varepsilon_{FL}/\varepsilon_{gL}), \quad (4)$$

$$m_{cL}(\varepsilon_{FL})=m_{cL}(0)(1+2\varepsilon_{FL}/\varepsilon_{gL}), \quad (5)$$

where $m_{cL}(0)$ is the cyclotron mass at the bottom of the band, ε_{gL} is the direct gap at L. It should be noted that relationships (4) and (5), which are valid for the case $p_z \approx 0$ (z is the direction of elongation of the Fermi surface at L) practically do not change in character on going from a normal spectrum to an inverted one, since at $p_z = 0$ the carrier dispersion law at L contains the square of the gap parameter $\gamma - \gamma_0$.[4] It is only necessary to remember that in the case of an inverted spectrum ($\gamma - \gamma_0 < 0$) the Fermi energy is reckoned from the saddle point, while $\varepsilon_{gL} = 2|\gamma - \gamma_0|$.

From expressions (4) and (5) one easily derives

$$m_{cL}(\varepsilon_{FL})/m_0 = 2\alpha(\varepsilon_{FL}+\varepsilon_{gL}/2) = 2\alpha(\varepsilon_{ov}+\varepsilon_{gL}/2-\varepsilon_{FT}), \quad (6)$$

$$[m_{cL}(\varepsilon_{FL})/m_0]^2 = (\alpha\varepsilon_{gL})^2 + 2\alpha eh/\pi c m_0 \Delta_L, \quad (7)$$

$$\Delta_L^{-1} = (2\alpha\pi c m_0/eh)[(\varepsilon_{FL}+\varepsilon_{gL}/2)^2 - (\varepsilon_{gL}/2)^2], \quad (8)$$

where the parameter α determines the gap dependence of the cyclotron mass $m_{cL}(0)/m_0 = \alpha\varepsilon_{gL}$ at the bottom of the band, m_0 is the free electron mass, ε_{ov} is the band overlap, and Δ_L^{-1} is the reciprocal of the period of Shubnikov oscillations from the Fermi surface at L.

Comparison of the dependence of $m_{cL}(\varepsilon_{FL})$ on ε_{FT} given in Fig. 6 with formula (6) permits one to conclude that the quantities 2α and $\varepsilon_{ov}+\varepsilon_{gL}/2$ can be determined from Fig. 6 with satisfactory precision. The value $\varepsilon_{ov}+\varepsilon_{gL}/2 = (46 \pm 2)$ meV found in the present work permits determination of the parameter $\varepsilon_{FL}+\varepsilon_{gL}/2$ as (34 ± 2) meV for pure Bi, which agrees well with data from optical measurements.[34] From the value of 2α calculated for the case $\mathbf{H} \parallel z$ using the relationship[4]

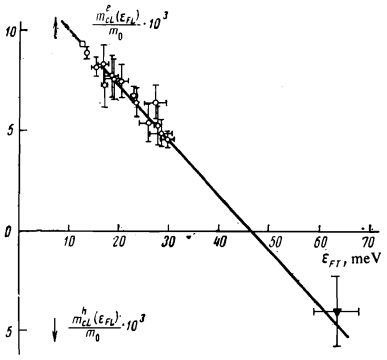

FIG. 6. Dependence of the cyclotron mass of electrons $m^e_{cL}(\varepsilon_{FL})$ and holes $m^h_{cL}(\varepsilon_{FL})$ at $L(\mathbf{H} \parallel \mathbf{I})$ on the Fermi energy ε_{FT} of holes at T: □—pure Bi[35]; ○ and ▼—data from the present work.

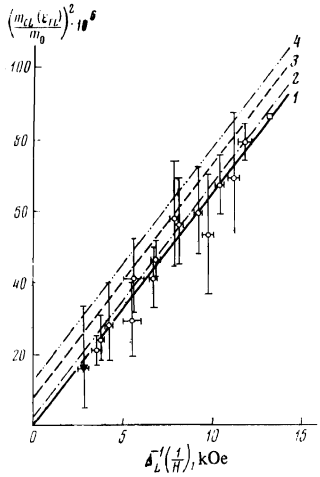

FIG. 7. Dependence of the square of the cyclotron masses $m_{cL}(\varepsilon_{FL})$ of electrons and holes at L on the reciprocal of the period of the Shubnikov oscillations $\Delta_L^{-1}(\mathbf{H} \parallel \mathbf{I})$: □—pure Bi[35]; ○ and ▼—data from the present work for electrons and holes at L respectively. Theoretical curves are drawn from formula (7) for the following cases: 1—$\varepsilon_{gL} = 0$; 2—$\varepsilon_{gL} = 10$ meV; 3—$\varepsilon_{gL} = 20$ MeV; 4—$\varepsilon_{gL} = 26$ meV.

$$m_{cL}(\varepsilon_{FL})|_{H\parallel z} = \frac{1}{v_x v_y}\left(\varepsilon_{FL}+\frac{\varepsilon_{gL}}{2}\right), \quad \frac{v_x}{v_y} = \frac{S_{max}}{S_{mid}},$$

where S_{max} and S_{mid} are the maximum and mean main cross sections of the Fermi surface at L in Bi,[35] the parameters of the Abrikosov dispersion law for carriers at L were calculated to be $v_x = 0.99 \cdot 10^8$ cm/s, and $v_y = 0.74 \cdot 10^8$ cm/s (x-axis parallel to the binary axis).

The dependence of the square of the low cyclotron mass $m_{cL}(\varepsilon_{FL})$ of light carriers at L on the reciprocal of the period of the Shubnikov oscillations $\Delta_L^{-1} \sim S_{extr}$ has a linear character in accordance with the predictions of theory (see (7)) (Fig. 7). The same can be said of the dependence of Δ_L^{-1} on $\varepsilon_{FL}+\varepsilon_{gL}/2$ (Fig. 8). In constructing the latter dependence the values of $\varepsilon_{FL}+\varepsilon_{gL}/2$ were taken from Fig. 6. The continuous curves in Figs. 7 and 8 were drawn from the formulae (7) and (8) respectively for several values of the gap ε_{gL} in the range 0

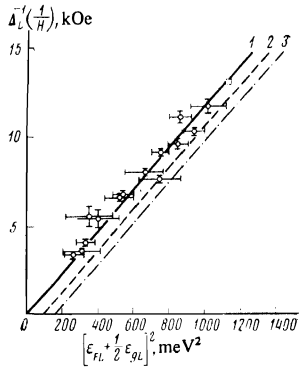

FIG. 8. Dependence of the reciprocal of the period of the Shubnikov oscillations from electrons at $L(\mathbf{H} \parallel \mathbf{I})$ on the square of the parameter $\varepsilon_{FL}+\varepsilon_{gL}/2$ for $T = 2.1$ K: □—pure Bi[35]; ○—data from the present work. Theoretical curves are drawn from formula (8) for the cases where: 1—$\varepsilon_{gL} = 0$; 2—$\varepsilon_{gL} = 20$ meV; 3—$\varepsilon_{gL} = 26$ meV.

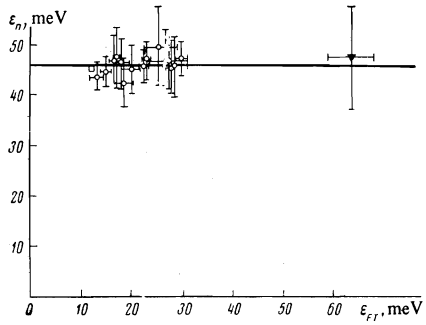

FIG. 9. Dependence of the amount of band overlap ε_{ov} on the Fermi energy of holes at T in the acceptor-doped samples of Bi investigated for the case where $\varepsilon_{gL}=0$; □—pure Bi[35]; ○ and ▼—data from the present work for electrons and holes at L respectively.

$\leqslant \varepsilon_{gL} \leqslant 26$ meV (the magnitude of the parameter 2α was determined from Fig. 6 and was not varied later). As is evident from Figs. 7 and 8, the gap ε_{gL} in Bi does not exceed 15 meV in absolute value (the best agreement of the experimental data with the theoretical straight lines is observed for $\varepsilon_{gL}=0$). In this connection it should be noted that according to the data of Yastrebova[15] and Kostial'[16] the gap parameter $\gamma-\gamma_0$ in Bi does not exceed a few millielectron-volts ($\gamma-\gamma_0<0$) (see also[4]). The value of $\varepsilon_{gL}=(26\pm3)$ meV for the gap at L in Bi obtained by Édel'man[35] therefore appears to be much too high.

Calculation of ε_{FT} and ε_{FL} using formulae (2) and (3) showed that the amount of overlap ε_{ov} of the valence and conduction bands does not change noticeably on lowering the Fermi level by ~ 50 meV using an acceptor impurity (Fig. 9). The dependence shown in Fig. 9 was obtained for the particular case $\varepsilon_{gL}=0$, but variation of ε_{gL} within the reasonable limits of $0\leqslant|\varepsilon_{gL}|<15$ meV did not alter the character of this dependence qualitatively. Thus, the rigid band model for Bi doped with impurities in a concentration of <0.1 at.% has found reliable experimental confirmation (see also[40]).

From the data obtained in the present work it is possible to determine the maximum critical concentration P_T^* of holes at T for which the electron Fermi surface at L disappears. In the calculations the value of the minimum cross-section S_{min} of the hole Fermi surface at T corresponding to $\varepsilon_{FT}=46$ meV was taken, and it was considered that $S_{max}/S_{min}=3.32$.[35]

The investigations of the angular dependence of the section of the hole ellipsoid at T carried out in the present work showed that for $P_T<5\cdot10^{18}$ cm^{-3} the anisotropy of the ellipsoid does not, within the limits of experimental error, change with increase in P_T. For P_T^* a value of $2.6\cdot10^{18}$ cm^{-3} was found; this was appreciably less than the integral hole concentration of $P^*=4\cdot10^{18}$ cm^{-3} at which the electrons at L disappear at liquid-helium temperatures.[5,36,37] The value of P^* was determined in[5,36,37] from the Hall coefficient $R_\infty=1/ecP^*$ obtained in extremely high magnetic fields. In the latter case information on the distribution of holes between the various extrema in the valence band of Bi was naturally lost. The discrepancy between the values of P^* and P_T^* can only be explained by part of the impurity holes $P^*-P_T^*=1.4\cdot10^{18}$ cm^{-3} being consumed in filling additional hole extrema in the valence band of Bi. These extrema may be those at $\Sigma(TW)$[3,24,26,38,39] situated at a depth of ~ 10 meV below the Fermi level in pure Bi.

In conclusion we would like to take the opportunity of expressing our sincere gratitude to G. A. Ivanov and D. V. Gitsu for supplying some of the samples of Bi doped with acceptor impurities and for their interest in the work.

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Nonlinear magnetoelectric effect in ferromagnetic semiconductors

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A magnetization proportional to the square of the electric field is observed in ferromagnetic semiconductors in optical-band fields. It is shown that the effect is proportional to the energy of the s-d exchange interaction of the carriers with the magnetic atoms. The nonlinear magnetization at $\omega \gg v_F q$ is proportional to q^4 and n; at $\omega = 0$ it is proportional to q^2 and is independent of n (q and ω are the wave vector and frequency of the nonlinear magnetization, while n is the carrier density). Action of two laser beams of equal frequency on a crystal produced a specified inhomogeneous static distribution of the magnetization M_z, the magnitude and wave vector of which was determined by the values of the angle θ; the value of q can vary in a wide range from 0 to $2q_L$ when the angle θ is varied from 0 to π, where q_L is the wave vector of the light and θ is the angle between the directions of the laser beams. The distribution of the magnetization can be determined from the diffraction of a sounding light beam; according to estimates, to produce a magnetization such that the intensity of the first diffraction maximum is of the order of the intensity of the sounding light, the required lasers are of quite low power. The nonlinear magnetization leads to a nonlinear interaction of the optical-band waves; generation at the difference frequency by means of a nonlinear ferromagnet is considered and, in contrast to the known results of nonlinear optics, the spatial-synchronism regime turns out to be less effective at difference frequencies lower than some definite value.

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1. Landau and Lifshitz have indicated in their monograph[1] that, for certain definite magnetic symmetry classes, a magnetoelectric effect can exist wherein a magnetization (polarization) proportional to the electric (magnetic) field can be produced in a crystal. Dzyaloshinskiĭ[2] has shown subsequently that the antiferromagnetic Cr_2O_3 has a magnetic symmetry that makes this effect possible, as was subsequently observed experimentally[3] in Cr_2O_3.

However, magnetization proportional to the square of the electric field can exist in all magnets. We have named this the nonlinear magnetoelectric effect. At sufficient field amplitudes, naturally, the nonlinear effect can be appreciable. We shall investigate below the nonlinear magnetoelectric effect in ferromagnetic semiconductors in optical-band electric fields. Many ferroelectric semiconductors have forbidden bands on the order of 1 eV (see, e.g., the reviews[4–6]) and are by the same token transparent enough in the optical band for which high-power lasers are available.

The physical meaning of the considered effect can be explained in the following manner: It is known that a transverse electric field E excites electron-density oscillations in second order in the field.[1)] In ferromagnetic semiconductors, the s–d exchange energy AS, where S is the spin of the magnetic atom, exceeds the Fermi energy E_F of the carriers,[2)] up to the highest possible values of the concentration n, so that the carrier spins have all the same direction. The z component s_z of the electron spin density of these polarized carriers is determined by the electron density, so that the resultant Δs_z, which is proportional to E^2, alters in turn the effective magnetic field H_{eff} that acts on the spin of the magnetic atoms as a result of the s–d exchange interaction and is proportional to A. This produces a nonlinear magnetization proportional to the s–d exchange energy and the square of the electric field. By the same token the experimental observation of this effect can be used to investigate exchange interaction (to measure its magnitude and its dependence on the parameters).

2. We shall consider wide-band ferromagnetic semiconductors in which the widths of the conduction band W is large in comparison with AS. The Hamiltonian of the crystal is written in the form

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{sd} + \mathcal{H}_{\text{Coul}}; \tag{1}$$

here