INVESTIGATION OF THE FREQUENCY MODULATION OF QUANTUM OSCILLATIONS OF THE MAGNETIC SUSCEPTIBILITY OF BISMUTH

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A detailed study has been made of the periodic oscillations of the magnetic susceptibility of bismuth, associated with the large cross sections of the electron and hole constant-energy surfaces, at temperatures about 0.1° K. It has been discovered that the variation in the hole oscillation frequency is in antiphase with the variation in the electron oscillation frequency. The effect is considerably weaker for the electron high-frequency oscillations than for the hole oscillations. The periods of the frequency modulation correspond to periods of the fundamental low-frequency oscillations, observed for a given magnetic field orientation. Extrapolation to the strong-field region shows, for the last period of low-frequency oscillations, an increase of the electron oscillation frequency and a decrease in the hole frequency. These features of the frequency modulation are explained using a model proposed in [2].

A periodic dependence of the frequency of oscillations on the magnitude of the reciprocal of the magnetic field has been discovered in a study of the de Haas-van Alphen effect in bismuth at very low temperatures.[1] The oscillating dependence of the period on the reciprocal field for large cross sections of the hole constant-energy surface has also been reported by others.[2,3] It was of interest to investigate this effect in more detail for the hole and electron constant-energy surfaces simultaneously, which is important in the explanation of the nature of the effect and the correct interpretation of the results in the determination of the constant-energy-surface parameters from the oscillation data.

RESULTS OF MEASUREMENTS

Investigation of the phenomenon of frequency modulation of the quantum oscillations of the magnetic susceptibility (the dependence of the frequency of the high-frequency oscillations on the magnetic field intensity) was carried out for three principal orientations of bismuth single crystals, with respect to the axis of suspension of a torsion balance at temperatures near 0.1°K, by a method described in [1]. The same samples were used as in [1].

For oscillations associated with the hole part of the Fermi surface, the periodic variation of the frequency appeared very clearly for the magnetic field orientations along planes passing through the trigonal axis and the bisectrix (orientation I) or the trigonal and binary axes (orientation II). For these orientations, the frequency modulation of the high-frequency oscillations was observed only for the directions of the magnetic field such that, in addition to the large cross sections of the Fermi surface associated with the high-frequency oscillations, there were small cross sections of the electron ellipsoids responsible for the low-frequency oscillations. Figures 1 and 2a show, by way of example, the dependence of the magnetic susceptibility anisotropy for the angles ψ equal to 82 and 77° for orientation I, and $\psi = 80°$ for orientation II (ψ is the angle between the magnetic field direction and the trigonal axis). It is evident from these figures that the frequency of the high-frequency oscillations varies periodically with the variation in the magnetic field and the magnitude of the effect increases with increase in the field. The period of the variation in the high-frequency oscillations is shifted in phase, relative to the period of the corresponding low-frequency oscillations, by approximately 45° (Figs. 1 and 2). For orientation II, the effect is observed for a magnetic field directed at $\psi > 50°$ with respect to the trigonal axis. The period of the frequency modulation for this orientation corresponds to the period of the low-frequency oscillations associated with the identical cross sections of two electron ellipsoids. The angular depend-

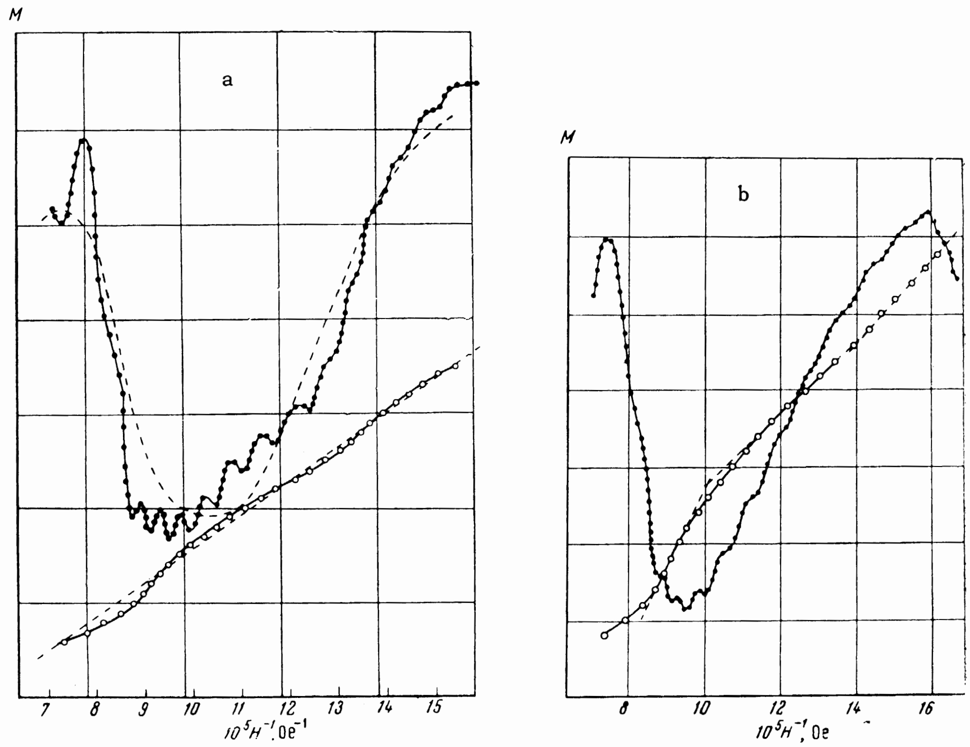

FIG. 1. Dependence of the magnetic moment M on the value of the reciprocal field for bismuth with the magnetic field oriented in a plane passing through the trigonal and bisectrix axes: a) for $\psi = 82°$; b) for $\psi = 77°$. The dependence of the serial numbers of the maxima and minima on 1/H is given in the lower part of the figure.

ence of the variation of the high-frequency oscillation period corresponds to the angular dependence of the cross sections responsible for the low-frequency oscillations.

Analogous variation of the period of the high-frequency oscillations for an electron constant-energy surface is observed for a magnetic field lying in the basal plane. For this orientation, the effect appears only for sufficiently large cross sections of the electron surface, corresponding to the magnetic field directions making $\Psi < 10°$ with the binary axis. In this range of angles, the low-frequency oscillations are associated with two slightly differing cross sections of two electron surfaces. The oscillations associated with the hole constant-energy surface do not appear because this surface does not have a marked anisotropy for this orientation. The periodic variation of the frequency for the electron surface is illustrated in Fig. 2b. Figure 3 shows schematically the dependence of the variation of the oscillation frequency, corresponding to large cross sections of

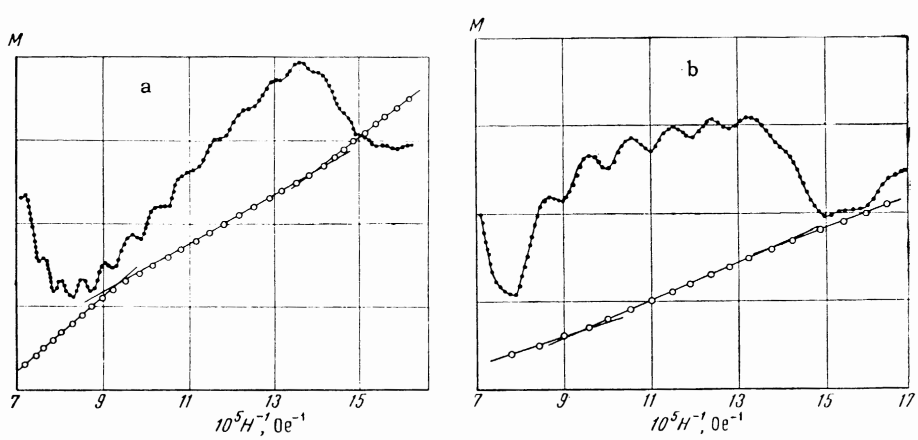

FIG. 2. Dependence M(1/H) for two magnetic field orientations: a) in a plane passing through the trigonal and binary axes, $\psi = 80°$ (ψ is the angle between the trigonal axis and the magnetic field direction); b) in the basal plane, $\Psi = 5°$ (Ψ is the angle between the field direction and the binary axis).

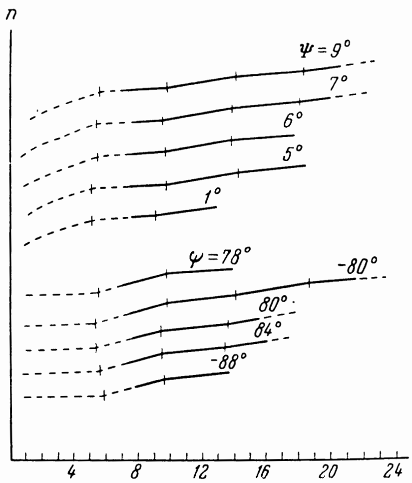

FIG. 3. Schematic representation of the variation in the frequency of oscillations corresponding to large cross sections of the hole (lower part of the figure) and electron (upper part of the figure) surfaces, plotted as a function of 1/H. The upper curves correspond to the direction of the magnetic field making an angle Ψ with the binary axis; the lower curves correspond to the direction of the magnetic field making an angle ψ with the trigonal axis. The extrapolations to the strong-field region are shown dashed.

the hole and electron surfaces, on the reciprocal field.

DISCUSSION OF RESULTS

The features of the frequency modulation discussed above are in good agreement with the model of electron transitions between closed constant energy surfaces when the magnetic field is varied.[2,4] These transitions are associated with the anisotropy of the constant-energy surfaces and with the various distances between the Landau levels for certain orientations of the magnetic field with respect to the crystallographic axes. The transitions begin to be important when the value of μH (μ is the effective Bohr magneton) for one of the surfaces becomes comparable with the Fermi energy E for the same surface. If for other constant-energy surfaces the distance between the Landau levels is much less than the Fermi energy, then, when the magnetic field is varied, electrons may leak periodically between the surfaces at the frequency at which levels pass through the Fermi level in the surfaces with high values of μH. The main feature of this effect for bismuth should be the opposite variation of the frequency of the electron and hole oscillations, which is in good agreement with experimental data. It is evident from Figs. 2 and 3 that, for the same cross sections associated with the low-frequency oscillations, an increase in the hole oscillation frequency is always accompanied by a reduction in the electron oscillation frequency, and conversely. The extrapolation of the curves in Fig. 3 to the strong-field region gives for the last period (the last Landau level above the Fermi level) an increase in the frequency of oscillations associated with the electron ellipsoid, and a reduction in the frequency for holes. The extrapolation was carried out in most cases on the basis of the experimentally determined frequency modulation periods and, in some cases, on the basis of the periods of the corresponding low-frequency oscillations.

Since the change in the electron and hole density should be proportional to the density of states, then, obviously, for a group with a lower density of states, the effect will be weaker. According to [1], the density of states at the Fermi boundary of an electron surface is approximately one fourth as small as the density of states at a hole surface. Therefore, the magnitude of the frequency modulation effect for electrons should be considerably smaller than for holes, which is in good agreement with the experimental data (cf. Figs. 1 and 2).

In those cases when the periods of the low-frequency oscillations are associated with three relatively small and different cross sections of the electron surfaces, the effect may be irregular and complex, which is also in agreement with experiment. The relatively simple cases are illustrated in Fig. 1. The $\psi = 82°$ orientation of the magnetic field in Fig. 1 corresponds to the cross section S_1, close to the principal minimum cross section, of one electron ellipsoid and the two identical cross sections $S_2 \approx 2S_1$ of two other electron ellipsoids. The observed frequency modulation period corresponds to the cross sections S_2. Figure 1b shows the period corresponding to the cross section S_1. In some cases, two frequency modulation periods are observed simultaneously, these periods being associated with different cross sections of electron ellipsoids and the period corresponding to the cross section S_2 appearing more strongly in the strong-field region.

Figure 4 shows the dependence of the high-frequency oscillation period on the reciprocal of the magnetic field, $\Delta(1/H)$, for $\psi = 82°$ (orientation I). Obviously, the frequency modulation amplitude for this group of carriers should depend on the degree of degeneracy of the upper level in the group (with a large value of μH), i.e., it should be inversely proportional to the number of

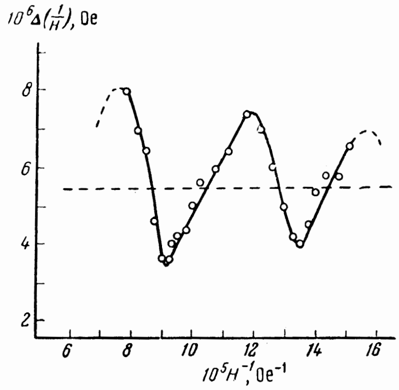

FIG. 4. Dependence of the period of the high-frequency oscillations Δ on the reciprocal field for $\psi = 82°$ and the magnetic field oriented in a plane passing through the trigonal and bisector axes.

levels in this group. Within the experimental error, this representation is in qualitative agreement with the experimental data.

A unique relationship between the oscillation frequency and the extremal cross sections of the Fermi surface holds only in the weak-field region, when the frequency-modulation effect is practically absent. If the range of fields in which the high-frequency oscillations are observed is less than 1.5—2 periods of the low-frequency oscillations, which determine the frequency modulation, the extremal cross section cannot be determined exactly. Depending on the range of fields (Fig. 3), we obtain either two high or too low values of the extremal cross section. Naturally, the data for the large extremal cross sections of the constant-energy surfaces of bismuth, obtained by observing oscillations only in strong magnetic fields, are not exact.

In [1] the extremal cross sections of the constant-energy surfaces were determined from the average value of the frequency of the high-frequency oscillations, obtained by averaging over several low-frequency periods. However, the high-frequency oscillations, corresponding to the maximum principal cross section S_3 of the electron constant-energy surface, were observed only in the range of fields greater than 7000 Oe, and to determine S_3 we used the data obtained in the range $(9-14) \times 10^{-5}$ Oe^{-1}. It is evident from Fig. 3 that in this range the oscillation frequency increases. The true value of S_3, obtained by averaging over several low-frequency periods, should be $S_3 = (19.5 \pm 1) \times 10^{-42}$ $g^2 \cdot cm^2 \cdot sec^{-2}$. The electron density, calculated taking into account this correction, is $n_e = (2.82 \pm 0.1) \times 10^{17}$ cm^{-3}, which agrees much better with the hole density $n_h = (2.76 \pm 0.2) \times 10^{17}$ cm^{-3}.[1]

The effect discussed here may be one of the reasons for the distortion of the form of the low-frequency oscillation curves in the region of strong fields. Electron transitions from groups with large μH to groups with small μH should reduce the frequency of the low-frequency oscillations and, consequently, the corresponding part of the $M(1/H)$ curve becomes flatter. For the reverse transitions, the frequency of the low-frequency oscillations should increase and the corresponding part of the curve should become steeper. This change in the form of the $M(1/H)$ curve of the low-frequency oscillations is in agreement with experimental data. The flat transition from a minimum to a maximum (Fig. 2a) corresponds to a reduction in the frequency of oscillations associated with large cross sections of the hole surface. When the hole oscillation frequency increases, a steeper transition from a maximum to a minimum is observed. For the electron high-frequency oscillations (Fig. 2b), the opposite is observed.

In conclusion, we take this opportunity to record our deep gratitude to A. I. Shal'nikov for his interest in this work.

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