Nonlinear size effect in bismuth

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It is shown that when a semimetal is placed in a constant magnetic field parallel to its surface and is exposed to radio waves, a system of dc spikes is produced in addition to the spikes of the high-frequency current in the interior of the sample. The appearance of the dc spikes is due to the change in the length of the electron path in the skin layer under the influence of the magnetic field of the wave itself. In samples of limited dimensions, the presence of a system of dc spikes, the distance between which depends on the magnitude of the constant magnetic field, should lead to the appearance of a nonlinear size effect. This effect is experimentally investigated in bismuth.

Several authors[1-3] have shown recently that, at least in semimetals, various nonlinear phenomena can be observed even at relatively small electromagnetic-wave powers. In particular, direct current is produced in a skin layer under the influence of electromagnetic radiation.

It is known that spikes of high-frequency current can be produced in the interior of a metal[4] in the presence of a static magnetic field under the conditions of the anomalous skin effect, and are due to the individual motion of the electrons of the extremal sections of the Fermi surface. One can expect the presence of direct current concentrated at the surface of the sample to lead analogously to the onset of direct-current spikes in the interior of the sample.

Let us examine the qualitative aspect of the phenomenon. We confine ourselves to the case of low frequencies $\omega\tau \ll 1$, $\delta/r \ll 1$ (ω is the frequency of the electromagnetic wave, τ is the electron relaxation time, δ is the depth of the skin layer, and r is the Larmor radius). In this case we can assume that the electron move in a quasistatic but spatially strongly inhomogeneous electromagnetic field. Since the phase shift between the alternating electric field E and the magnetic field H_1 differs from $\pi/2$, superposition of the magnetic field of the wave itself on the static magnetic field H_0 causes the two half-cycles of the alternating current to become unequal, owing to the change in the path length of the effective electrons in the skin layer. As a result, an electric current proportional to H_1E and having a dc component appears in the skin layer.

It was easy to estimate the density j_2 of the direct current if the field H_0 is parallel to the surface of the metal. The high-frequency current j_1 is proportional to $\sigma_0 E\delta r^{-1}$, where σ_0 is the static conductivity. The influence of the magnetic field is estimated from the relation $j_2 \sim H_1 dj_1/dH_0$. Using the estimate $H_1 \sim cE/\omega\delta$, we obtain $j_0 \sim \sigma_0 E(\omega\tau)^{-1} eEl\epsilon_F^{-1}$ where l is the electron mean free path and ϵ_F is the Fermi energy.

In a magnetic field parallel to the surface of the metal, the electrons of the extremal section of the Fermi surface, after passing through the skin layer, are again focused at a depth 2r as they move along the trajectories and produce at this depth a direct-current spike. At the same distance from the surface there is a spike of fields H_1 and E, which leads to the appearance of direct current at the depths 2r and 4r. Thus, a system of direct-current spikes is produced. It is clear from the foregoing arguments that a similar system of spikes is produced also for the current $\tilde{j}_2$ at the frequency 2ω.

In a sample of limited dimensions, the presence of a system of dc spikes, the distance between which is determined by the value of the external magnetic field H_0, should lead to the appearance of a nonlinear size effect, similar to the radio-frequency size effect[4]. The present article is devoted to this phenomenon.

1. THEORY

We shall show that a more rigorous allowance for the change in the electron path length in the skin layer to the proper magnetic field of the wave leads to the appearance in the skin layer of a direct current j_2 and to the appearance in the interior of the sample of dc spikes. Assume that an electromagnetic wave is incident on the surface of a metal occupying the half-space $z > 0$. The electric field $\mathbf{E}$ of the wave is parallel to the x axis. A static magnetic field $\mathbf{H}_0$ is parallel to the y axis. We assume that the quasistationarity condition $\omega\tau \gg 1$ and the strong-magnetic-field condition $\Omega\tau \gg 1$ are satisfied ($\Omega = eH_0/mc$ is the cyclotron frequency). As the model of the Fermi surface of the metal we use a cylindrical surface $p_x^2 + p_z^2 = p^2$ (a similar model can be used in the case of bismuth, the electronic part of the Fermi surface of which consists of three strongly elongated ellipsoids). The length of the cylinder is assumed to be p_H, so that the electron density is $n = 2\pi p_H p^2 h^{-3}$.

To find the response of the electron system to an external perturbation, it is necessary to solve the Boltzmann kinetic equation. In the τ-approximation we have

$$v_z\frac{\partial f}{\partial z}+\Omega\frac{\partial f}{\partial\varphi}+\tau^{-1}f=g, \tag{1}$$

where $f df_0/d\epsilon$ is the nonequilibrium part of the distribution function, $\varphi = \Omega t$ is the dimensionless time of motion along the orbit, and g describes the external perturbation.

When solving the kinetic equation we shall use the methods developed in the theory of anomalous penetration of the electromagnetic field into a metal[4]. Neglecting the field E_z and the boundary conditions for the distribution function at z = 0, and continuing the field E in even fashion to the region $z < 0$, we employ the Fourier transformation:

$$A_k=\int_{-\infty}^{\infty}dzA(z)e^{-ikz},\quad A(z)=(2\pi)^{-1}\int_{-\infty}^{\infty}dkA_ke^{ikz}. \tag{2}$$

We can then easily obtain the electric current j_k

$= e\langle v f_k \rangle$. The angle brackets correspond to integration over the Fermi surface. In our case $v_z = v \cos\varphi$, $v_x = -v \sin\varphi$ and

$$\langle B\rangle = 2mp_H h^{-3}\int_0^{2\pi} B\,d\varphi. \tag{3}$$

To find the linear response it is necessary to put $g = g_1 = e\mathbf{E}\cdot\mathbf{v}$. The solution of the kinetic equation (1) with this right-hand side is well known. Under the conditions of the anomalous skin effect ($kr \gg 1$), the Fourier component of electric conductivity is

$$\sigma_k = \frac{2\sigma_0}{\pi}\frac{1-\sin 2kr}{kr}, \quad \sigma_0 = \frac{ne^2\tau}{m}, \quad k>0. \tag{4}$$

The Fourier component of the electric field is obtained from Maxwell's equations

$$E_k = -\frac{i\omega}{c}\frac{2H_1(0)}{k^2 - 4\pi i\omega c^{-2}\sigma_k}, \tag{5}$$

where $H_1(0)$ is the amplitude of the alternating magnetic field on the surface of the metal. As shown in [4], electric field $E(z)$ can penetrate under these conditions into the interior of the sample, forming spikes at values of z that are multiples of $2r = D$.

In the calculation of the quadratic response there are two possibilities, viz., one can take into account in the kinetic equation the terms quadratic in the electric field E, and one can take into account the influence of the alternating magnetic field H_1 on the electron trajectory. It is well known that at $\omega\tau \ll 1$ the influence of H_1 is the more significant. Since we shall henceforth be interested only in quantities that are constant in time, g_2 should be taken in the form

$$g_2 = -\frac{e}{4mc}\operatorname{Re}\left(H_1\frac{\partial f_1^*}{\partial\varphi}\right), \tag{6}$$

where the asterisk denotes the complex conjugate.

Substituting (6) in (1), we can obtain the value of f_2 and, using (3), calculate the density of the direct current $j_{k2} = \langle v_x f_2\rangle$. Under the condition $kr \gg 1$ we obtain

$$j_{k2} = \frac{e^3 v p_H \tau}{\pi\omega h^3}\int_{-\infty}^{\infty} dq K(q)\operatorname{Im}(E_{k-q}E_q^*), \qquad K(q) = \operatorname{sgn} q - \sin(2qr) \tag{7}$$

Here $\operatorname{sgn} x = x/|x|$, and E_k is given by (5). From (7), taking the inverse Fourier transform, we obtain the distribution of the direct current over the sample thickness:

$$j_2(z) = \frac{ne^3 l}{2\pi\omega p^2}\operatorname{Im}\{2E(z)\tilde{E}^*(z) + E(z)[\tilde{E}^*(z-D) - \tilde{E}^*(z+D)]\}, \qquad \tilde{E}(z) = \pi^{-1}\int_0^{\infty} dq\sin(qz)E_q. \tag{8}$$

The value of j_2 agrees with the estimate given in the Introduction.

The behavior of the current j_2 at small z will be determined by estimating the integrals contained in (8):

$$j_2 \sim \zeta\ln\zeta, \quad \zeta = \frac{z}{2r}\left(\frac{2r}{\delta_0}\right)^{2/3} \ll 1,$$

where $\delta_0^2 = c^2/2\pi\sigma_0\omega$. The distribution of the direct current over the thickness of the samples at arbitrary z can be obtained by using the function $E(z)$ given in [5]. This distribution is shown schematically in Fig. 1.

At values of z that are multiples of $D = 2r$, antisymmetrical bursts of direct current are produced. The amplitude of the spikes decreases like $N^{-2/3}$ at large numbers N, and the signs of the spikes oscillate. No static electric field was produced, but the current j_2 gives rise to a static magnetic field $\mathbf{H}_2$, the value of which, apart from a numerical factor of order of unity, is $H_1^2(0)/(2\pi)^3 H_0$, and whose direction coincides with that of $\mathbf{H}_0$.

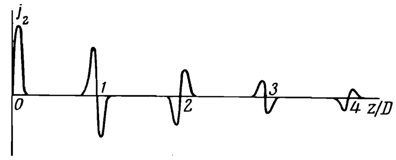

FIG. 1. Dependence of the density of the current j_2 on the distance to the surface of the metal.

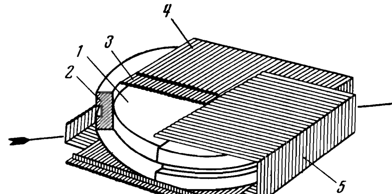

FIG. 2. Arrangement of measuring coils: 1—bismuth sample, 2—rotating substrate, 3—high-frequency inductance coil, 4 and 5—low-frequency coils.

The same method can be used to obtain the Fourier component of the current $\tilde{j}_{k2}$ at the frequency 2ω:

$$\tilde{j}_{k2} = \frac{\sigma_0 e}{2\pi^2\omega p}\int_{-\infty}^{\infty} E_{k-q}E_q[K(q) - K(k)]\,dq. \tag{9}$$

It follows therefore that effects analogous to those described above should exist also at double the frequency.

2. EXPERIMENT

The experiments were performed on single-crystal bismuth in the form of a disk of 18 mm diameter and thickness d = 0.58 mm. The normal to the plane of the disk made an angle of 3° with the trigonal axis. At helium temperatures, the electron mean free path in the metal was sufficient to register the lines of the radio-frequency size effect.

The sample was placed on a rotating polystyrene substrate inside a system of three inductance coils as shown in Fig. 2. To increase the homogeneity of the fields, the number of turns of the coils was chosen such that the transverse dimension of the winding exceeded the sample diameter. The system of inductance coils together with the sample were placed in a helium bath whose temperature was maintained at 1.3°K.

One of the coils (coil 3 in Fig. 2) was used to produce an alternating magnetic field at frequencies 0.5–2.5 MHz and amplitude $H_1(0)$ up to 5 Oe. At these magnetic-field amplitudes, the heat rise of the sample relative to the helium bath did not exceed 0.2°K, according to our estimates. The two other coils formed a bridge circuit of the Bloch type, operating at frequencies ω_M from 100 to 1000 Hz. At frequencies below 100 Hz, the alternating magnetic field was practically homogeneous over the sample thickness. The amplitude of low-frequency alternating field E_M was of the order of 0.1 Oe.

The signal from the receiving coil was fed to a filter that suppressed the high frequency, and then, after narrow-band amplification and synchronous detection, to the y coordinate of an automatic potentiometer. The x-coordinate of the potentiometer received a voltage proportional to the static magnetic field H_0. The magnetic field H_0 was parallel to the plane of the sample. The earth's field was cancelled out accurate to 1%.

Since the axes of the low-frequency coils were mutually perpendicular, there was no signal from the receiving coil in the absence of a high-frequency electromagnetic field. Turning on the high-frequency field led, according to (8), to the appearance of direct current in the skin layer and to the appearance of direct-current spikes in the interior of the sample. As a result, the sample acquired a macroscopic magnetic moment, the magnitude and direction of which were determined by the sum of the fields $\mathbf{H}_0 + \mathbf{H}_M \cos \omega_M t$. In the general case the direction of the static magnetic field did not coincide with the axis of any of the low-frequency coils, and both the magnitude and the direction of the magnetic field were modulated. The modulation of the field caused a change in the magnitude and direction of the macroscopic magnetic moment of the sample, which in turn induced an emf in the receiving coil. As a result, the signal obtained in the experiment was a mixture of the derivatives of the projections of the macroscopic magnetic moment $\mathbf{M}$ of the sample on the receiving-coil axis with respect to the magnetic field and to the azimuthal angle.

A typical experimental curve is shown in Fig. 3. For comparison, the same figure shows a plot of the radium-frequency size effect obtained at $H_1 < 0.1$ Oe. As seen from the figure, a nonmonotonic behavior of the macroscopic magnetic moment of the sample is observed at approximately those values of the magnetic field at which the lines of the radio-frequency size effect occur. The left edge of the lines on the $M(H_0)$ curve is shifted by an amount Δ relative to the left edge of the lines of the radio-frequency size effect towards weaker magnetic fields, and the shift increases with increasing intensity of the high-frequency magnetic field. The angular dependence of the position of the lines on the $M(H_0)$ curve at a fixed amplitude H_1 is close to the angular dependence of the lines of the radio-frequency size effect.

The lines from different electron ellipsoids were similar in shape but could differ in sign (see Fig. 3b). Reversal of the direction of the constant magnetic field did not change the sign of the lines. The sign was determined by the orientation in the skin layer of the velocity of the electrons of the given ellipsoid.

The direction of the high-frequency currents and the direction perpendicular to it break up the plane of the sample into four quadrants. The lines from the ellipsoids in which the extremal-section electron velocities in the skin layers were located respectively within the limits of the first and third or second and fourth quadrants had opposite signs. By simultaneous rotation of the sample and of the static magnetic field relative to the system of the measuring coils, it was possible to reverse the sign of the line without changing its position relative to the magnetic field. The corresponding line was not observed if the velocity of the external-section electrons of any of the ellipsoids was parallel or perpendicular to the high-frequency current in the skin layer.

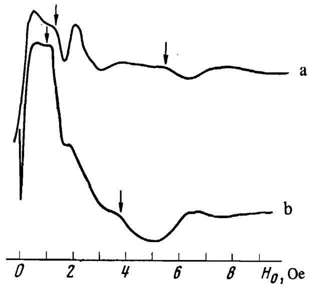

FIG. 3

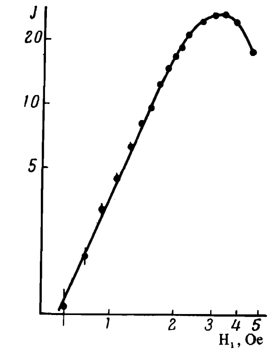

FIG. 4

FIG. 3. Radio-frequency size-effect line (curve a), $H_1 = 0.1$ Oe, and nonlinear size effect line (curve b). The angle between C_2 and H_0 is 13°, $H_1 \parallel C_1$, $H_1 = 2.3$ Oe. The arrows mark the starts of the lines. The lines from the two different ellipsoids on curve b have opposite signs.

FIG. 4. Dependence of the amplitude of the nonlinear size effect lines (in arbitrary units) on H_1, the angle between C_1 and H_0 is 5°, and $H_0 = 2$ Oe.

An increase in the intensity of the alternating magnetic field led to an increase in the amplitude of the observed lines, $J \sim H_1^2$, up to $H_1 \sim 2$ Oe (see Fig. 4). A simultaneous increase took place in the line width and, as already noted, the left edge of the lines shifted towards weaker magnetic fields. An increase of the frequency at fixed H_1 caused narrowing of the lines. An increase of the temperature of the helium bath led to a sharp increase in the amplitude of the nonlinear size effect, so that the lines were not observed at 4.2°K.

At certain orientations of the magnetic field relative to the crystallographic axes of the sample, lines in the doubled field were observed on the $M(H_0)$ curves, i.e., lines whose positions, reckoned from the left edge, conformed with the condition $2D = d$. These lines were most intense at directions at H_0 close to the binary axis C_2. The lines in the doubled field had the same shape as the lines in the single field (for which $D = d$), but were of opposite sign.

In weak fields $H_0 < 1$ Oe, an abrupt change is observed in the macroscopic magnetic moment of the sample when the magnetic field is increased (Fig. 3b). The corresponding line, the amplitude of which exceeds at certain H_0 the amplitude of the size-effect line by an order of magnitude, is closely connected with the appearance of "current states" in bismuth[3]. In the present paper, as a rule, we chose fields H_1 such that there were no hysteresis phenomena. We propose to report a detailed investigation of the macroscopic magnetic moment of bismuth in a future article.

3. DISCUSSION

The appearance of a direct-current spike in an oppositely located skin layer is not the only cause that leads to a nonlinear size effect. In that magnetic field in which the diameter of the electron orbit becomes comparable with the sample thickness, the size effect on the current j_2 receives a contribution proportional to $E(0)E(1)$ ($E(N)$ is the amplitude of the high-frequency field in the N-th spike).from the change in the number of returns of the electrons to the skin layer, which is connected with the scattering of the electrons from the opposite side of the plate. The magnitude of this contribution is determined by the coefficient of specularity of the electron scattering from the surface of the sample, and cannot be obtained in the approximation used by us. However, the cutoff of the electron orbits cannot make a contribution to the nonlinear size effect observed in the doubled field.

As seen from (8), the additional direct current in the skin layer, which is proportional to $E(0)E(N)$, appears also when the next spike of the high-frequency field emerges to the surface. In magnetic fields where $r = d/2$ and $r = d/4$, in the case of a Fermi surface in the form of a strongly elongated ellipsoid, the contributions to the nonlinear size effect made by the emergence of the spikes of the high-frequency and direct current to the surface, are of the same order of magnitude and cannot be distinguished in experiment.

With a Fermi surface of this shape, the dimension of the electron orbit is determined by the projection of the magnetic field $\mathbf{H}_0$ on the major axis of the ellipsoid. Therefore the experimentally observed lines can be regarded as superpositions of a certain monotonic variation on the derivative dM_1/dH (where M_1 is that part of the macroscopic magnetic moment of the sample which is connected with the motion of the electrons of one of the ellipsoids, and H is the projection of $\mathbf{H}_0$ on the axis of this ellipsoid).

If the Fermi surface consists of only one ellipsoid, then reasoning analogous to that given in the introduction makes it easy to trace the variation of the sign of the received signal when the orientation of the high-frequency and constant magnetic fields is altered. Let the axis of the ellipsoid be directed along y and let the fields $\mathbf{H}_1$ and $\mathbf{H}_0$ makes angles φ and χ respectively with these directions. Then $j_{1x} \sim \sigma_0 \delta r^{-1} E_1 \cos\varphi$, where $\delta^3 \sim c^2 r/\sigma_0 \omega$ and $r = vmc(eH_0|\cos\chi|)^{-1}$. Differentiating with respect to $H_0 \cos\chi$ and multiplying by $H_1 \cos\varphi$, we obtain for the total current $J_{2x} \sim j_{2x}\delta \sim cH_1^2 \cos^2\varphi/H_0 \times \cos\chi$. Since the experimental yields the derivative of the projection of the moment of the sample on the $\mathbf{E}$ direction and the modulating field H_M is directed along $\mathbf{H}_1$ the received signal is proportional, according to[6], to $H_1^2 H_M \cos^2\varphi \sin(2\varphi)/H_0^2 \cos^2\chi$.

It is seen from this expression that the sign of the registered signal does not depend on the direction of the field H_0, and when the sample is rotated relative to the system of the measuring coils the sign changes when the ellipsoid axis makes angles 0, $\pi/2$, π, and $3\pi/2$ with the direction of $\mathbf{H}_1$. It is precisely this behavior which was observed in the experiment. To obtain a similar result in the case of a Fermi surface consisting of three ellipsoids, it is necessary to assume that at those values of H_0 at which the dimension of the electron orbit of any of the ellipsoids becomes comparable with the sample thickness the main contribution to the high-frequency conductivity is made by the electrons from just this ellipsoid.

The shift of the experimentally observed lines relative to the lines of the radio-frequency size effect (registered at small H_1) towards weaker magnetic fields may be due to the presence of the field $\mathbf{H}_2$. The order of magnitude of Δ coincides with the estimate for the field H_2 obtained from formula (8), although the conditions $H_1 \ll H_0$ and $\Omega\tau \gg 1$ are not strictly satisfied in the experiments. In the case of a Fermi surface in the form of one ellipsoid, the projection of the field $\mathbf{H}_2$ on the ellipsoid axis is proportional to J_{2x}, and in the case of rotation of the field H_0 it reverses sign simultaneously with the change of the sign of the projection of H_0. This means that the lines shift in the same direction regardless of the direction of $\mathbf{H}_0$ relative to the ellipsoid axis.

We have calculated the dependence of Δ on the direction of the field $\mathbf{H}_0$ for a Fermi-surface model in the form of three cylinders that are rotated 120° relative to one another. The results of the calculations are too cumbersome to be presented here. However, even in this case, at an arbitrary direction of $\mathbf{H}_0$ the shift of the lines from any cylinder should be towards weaker magnetic fields.

It appears that the nonlinear size effect can be observed not only in semimetals, but also in metals with large carrier density. The point is that the power released in the sample is proportional to $\delta H_1^2 \sim H_1^2 n^{-1/3}$, so that at the same superheat of the sample relative to the helium bath the field H_1 can be increased by a factor $n^{1/6}$. Since the magnetic moment of the sample is of the order of H_1^2/H_0, and the field H_0 in which the size-effect lines are observed is proportional to $n^{1/3}$, it is possible to obtain in this field a magnetic moment of the same magnitude as in bismuth, without increasing the heat rise of the sample.

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