"Current" states in bismuth

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Nonlinear effects in bismuth are studied experimentally in weak magnetic fields at helium temperatures. It is shown that even in a zero magnetic field single crystals of bismuth irradiated by radio waves of sufficient amplitude may possess a macroscopic magnetic moment that results from rectification of the high frequency current ("current" states).

A number of nonlinear effects are observed at helium temperatures in bismuth exposed to radiowaves of relatively low intensity. These include second-harmonic generation[1,2], the appearance of a "radio-emf" and of rectified closed currents in the presence of a constant magnetic field,[3–6], and a nonlinear size effect[7]. It is not excluded that this group of effects is related to the observations made by Hsu and Thomas[8] and by Kawamura and Nakahara[9]. The former observed hysteresis in the dependence of the amplitude of transverse sound on the magnetic field in electromagnetic excitation of acoustic oscillations, while the latter observed, approximately in the same region of magnetic fields, hysteresis in the position of the cyclotron-resonance line.

In[10,11] there was noted a dependence of the surface impedance of metals on the amplitude of the high-frequency fields in weak magnetic fields. Further investigation has shown that at radiowave amplitudes $H_\sim$ exceeding a certain critical value $H_\sim^k$ there exist for bismuth magnetic-field regions within which the samples can be in several states[12]. The individual states are characterized by different dependences of the electromagnetic energy absorbed in the sample on the magnetic field. (In[12] the term "surface impedance" was used, but the use of this term seems to be not quite correct under conditions of developed nonlinearity and will be avoided henceforth). By changing the external magnetic field H we can cause a transition of the sample from one state to another. Such transitions take place jumpwise.

The existence of several states can be due to rectification of high-frequency current and to the appearance in the skin layer of a direct current whose direction is determined by the prior history of the sample. As already noted, rectification takes place only in the presence of a constant magnetic field. The rectified current itself produces a magnetic field, so that in the general case the magnetic field in the sample is the sum of the external field and of the field produced by the rectified current. At large radiowave amplitudes, it may turn out that rectified current does not vanish when the external magnetic field is turned off, since the small magnetic field produced by the rectified current itself suffices for the rectification. In other words, the presence of the ambiguous relations observed in[12] may mean that in bismuth exposed to large-amplitude radiowaves there are realized "current" states, i.e., states in which a closed direct current flows around the sample even in the absence of an external magnetic field.

The considerations advanced above can be verified experimentally by direct observation of the magnetic field produced by a self-maintaining rectified current. In addition, in our opinion, the program of future research should include a detailed study of the phenomenological aspect of the effect and attempts to determine the microscopic mechanism leading to the nonlinearity. Performance of part of this program is described in this paper.

PROCEDURE

We used for the measurements single crystal bismuth disks of 18 mm diameter and thicknesses 0.24, 0.4, and 0.58 mm. All the samples had the same orientation of the crystallographic axes, the trigonal axis being directed almost along the normal to the plane of the disk (angle between C_3 and $n \approx 3°$).

The sample was placed inside a system of three inductance coils (see Fig. 1), one of which produced the radio-frequency field, and the two others were used to register the two components of the magnetic moment of the sample. The axis of one of the receiving coils coincided with the axis of the high-frequency coil, while the axis of the other was perpendicular to it. The system of inductance coils together with the sample were placed in liquid helium, the temperature of which was maintained at 1.3°K in most experiments.

The signal from the receiving coil passed through a filter that cut off the high frequency, and was fed after amplification to the Y coordinate of a cathode-ray oscilloscope. The magnetic field was varied sinusoidally. A voltage proportional to the magnetic field was fed to the X coordinate of the oscilloscope. In this form, the circuit served to observe and photograph the jumplike changes of the magnetic moment of the sample (see below).

In a number of experiments we registered the derivatives $\partial M/\partial H$ and $\partial M/\partial H_\sim$ as functions of H, where M is the projection of the magnetic moment of the sample on the axis of one of the receiving coils. To this end, the low-frequency amplifier was switched over into the narrow-band regime, and the signal was fed after amplification to a synchronous detector and then to the y coordinate of a two-coordinate automatic x-y recorder. When recording the signal proportional to $\partial M/\partial H$, the magnetic field was modulated, and when $\partial M/\partial H$ was registered we modulated the amplitude of the high-frequency oscillations.

The high-frequency coil could be connected into one of the arms of a twin-T bridge. This made it possible to register simultaneously the derivative $\partial W/\partial H$ of the power absorbed in the sample with respect to the magnetic field, and the derivative $\partial M/\partial H$, as functions of H.

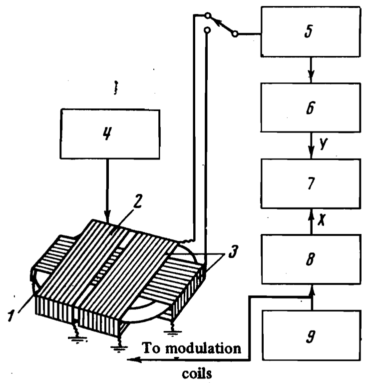

FIG. 1. Block diagram of setup. 1—Sample, 2—high-frequency coil, 3—receiver coil, 4—high-frequency generator, 5—high-frequency filter, 6—low-frequency amplifier, 7—cathode-ray oscilloscope, 8—phase shifter, 9—audio-frequency generator.

In addition, the twin-T bridge was used to record the lines of the radio-frequency size effect for the purpose of determining the orientation of the crystallographic axes of the sample relative to the direction of the high-frequency current and the constant magnetic field.

Certain measurements were performed using only one bridge. The low-frequency receiving coils were removed in this case. The point is that the presence of receiving coils greatly complicates the measurement of the high-frequency electromagnetic field. The high-frequency and the receiving coils form in this case coupled circuits whose resonant frequencies furthermore do not differ too much. On the other hand, if the receiving coils are removed, then the amplitude of the electromagnetic field can be determined from the voltage U across the inductance coil, namely $H_\sim = 0.4\pi nU/\omega L$, where L is the inductance of the coil with the sample, n is the number of turns per unit length, and ω is the frequency. It should be noted, however, that owing to the error in the measurement of U and L, the accuracy of the absolute determination of $H_\sim$ did not exceed 20%.

The constant magnetic field vector was parallel to the plane of the sample in all the experiments. The earth's magnetic field was cancelled out with accuracy 0.5%.

The temperature was determined from the saturated helium vapor pressure. Naturally, we measured in this case the temperature of the helium bath and not that of the sample. The sample had a heat rise which, according to our estimates, did not exceed 0.2—0.3°K when working in superfluid helium.

EXPERIMENT

1. It was necessary to verify that the jumplike changes in the absorbed power in the sample indeed corresponded to changes of the radio-frequency current or, equivalently, to changes of the magnetic moment of the sample. To this end, we recorded simultaneously the plots of $\partial W/\partial H$ and $\partial M/\partial H$ against H. Jumps at $H_\sim > H_\sim^k$ were observed on both curves at the same values of the magnetic field.

In the general case, the jumplike changes of the magnetic moment could be observed by registering the signal from any of the receiving coils. However, at certain orientations of the crystallographic axes of the sample relative to the direction of the high-frequency current, magnetic-moment jumps were observed only when the signal was picked off one of the receiving coils. If, for example, the sample was so placed that the high-frequency current flowed along C_1 or in a direction close to it, then the jumps were observed only when the signal was received from that coil whose axis coincided with the axis of the high-frequency coil. The results reported below pertain just to this case.

2. In all the investigated samples, the presence of several states was observed in two regions of the magnetic field (see Fig. 2). The dimensions of these regions (marked by the letters A and B in Fig. 2), do not depend on the electromagnetic-wave frequency and are determined, at fixed directions of the magnetic field and temperature, exclusively by the value of $H_\sim$. This statement is illustrated in Fig. 3. At given directions of the magnetic field and temperature, we measured the frequency of the electromagnetic wave. We used for the observation a bridge circuit with the receiving low-frequency coils removed. The voltage on the high-frequency coil was chosen each time such that the dimension of region A remained unchanged. The obtained linear dependence confirms the statement made above, since $U \sim L\omega H_\sim$, and the frequency dependence of the inductance is weak because of the small filling factor. A similar check was carried out also for the second region.

Since the investigated effect does not depend on the frequency, all the subsequent measurements were made in a narrow frequency interval, 0.8—1.0 MHz. It is not convenient to increase the frequency, for this entails a rapid increase of the power released in the sample, and to lower the frequency it would be necessary to decrease the bandwidth of the high-frequency filter.

3. One of the regions in which several states exist includes the point H = 0. An increase of the amplitude of the electromagnetic field led to a broadening of this region, and the initial section of the A(H) plot was nearly linear (Fig. 4). We have verified this circumstance many times and used it subsequently to determine the value of $H_\sim$ when registering the magnetic moment.

As already noted, owing to the connection between the receiving and high-frequency coils, it was impossible to determine by calculation the coefficient k in the relation $H_\sim = kU$. To determine this coefficient we used the following procedure: A bridge circuit, with the receiving coils removed, was used to register the $A(H_\sim)$ dependence. The receiving coils were then placed over the sample, and the A(U) dependence was measured. By comparing the value of $H_\sim^k$ obtained from the first measurement with U^k from the second, or else by comparing the slopes of the plotted straight lines, we obtained the sought coefficient. The results of these two procedures agreed within 5%.

The dependence of the dimension A on the direction of the external magnetic field is shown in Fig. 5. The angle interval in which the points are located is less than π because the two regions in which several states exist become superimposed at small angles between the high-frequency current and the magnetic field. As seen from the figure, the dimension A increases as the vector **H** approaches the direction of the high-frequency currents. Then, first, the slope of the $A(H_\sim)$ line increases, and second, $H_\sim^k$ decreases (the dependence of $H_\sim^k$ on the direction of the magnetic field is given in [12]).

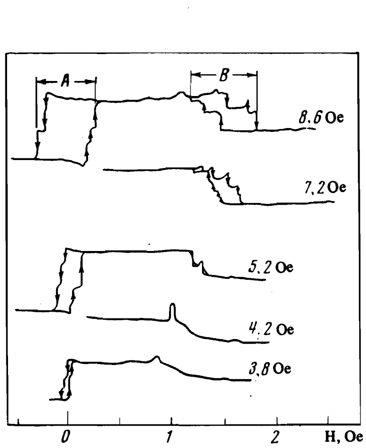

FIG. 2

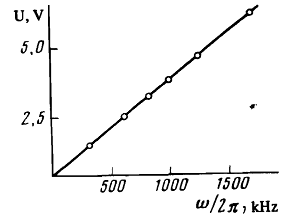

FIG. 3

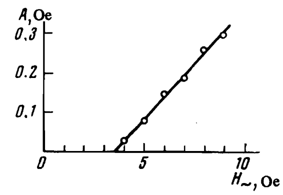

FIG. 4

FIG. 2. Plots of $\partial M/\partial H_\sim$ against H at different values of the amplitude of the high-frequency field for a sample 0.4 mm thick. The signal is picked off the coil whose axis is parallel to the axis of the high-frequency coil; $\angle jH = 15°$, $\angle jC_1 = -4°$. The number alongside each curve designates the amplitude of the high-frequency field at which the recording was made.

FIG. 3. Frequency dependence of the high-frequency coil voltage corresponding to a fixed dimension A. Sample thickness 0.4 mm, $\angle jC_1 = 20°$, $\angle jH = 95°$.

FIG. 4. Plot of $A(H_\sim)$ for a sample 0.4 mm thick; $\angle jC_1 = 2°$, $\angle jH = 50°$.

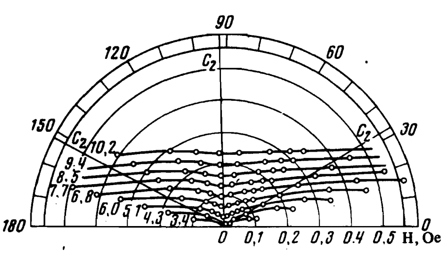

FIG. 5. Dimension A as a function of the direction of the external magnetic field (polar coordinates), d = 0.4 mm, $\angle jC_1 = 2°$. The value of $H_\sim$ in oersteds is marked alongside the curve.

The second region in which several states exist occurs at larger values of $H_\sim$ than the region that contains the origin (see Fig. 2). Even before its appearance, a singularity appears on the plot of $\partial M/\partial H$ against H. An increase of the alternating-field amplitude causes an increase in the value of this singularity and an increase in its distance from the origin.

For the samples with the C_3 axis close to the normal, the second region is observed only in the case when the magnetic field lies inside an angle interval 60° between those binary axes which make the smallest angles with the directions of the high-frequency currents (Fig. 6). The position of this 60° interval relative to the direction of the high-frequency currents can be varied by rotating the sample inside the system of the measuring coils. We have observed a continuous change of this interval, from the value −34°—+26° corresponding to Fig. 6, up to the value −2°—+58°. With further rotation of the sample (the rotation was through 4°), one of the binary axes intersected the direction of the high-frequency currents, and to observe the second region the magnetic field had to be oriented in the range from +2° to −58°.

4. The increase of the temperature led to a narrowing of the regions having several states. The change of the width A was due in this case to the increase of $H^k_\sim$ and to the decrease of the slope of the $A(H_\sim)$ line. The change in the dimension A depended on the orientation of the constant magnetic field. When the temperature was changed from 1.3 to 4.2°K, for example, the critical amplitude of the electromagnetic field increased by approximately three times at small angles between H and the high-frequency current, and only by two times when this angle was close to $\pi/2$. The situation gives qualitatively the appearance that raising the temperature decreases the anisotropy of the dependence of A on the magnetic field direction.

5. Jumplike changes of the magnetic moment of the sample could be observed on the oscilloscope screen. Photographs taken from the screen are shown in Figs. 7a and 7b. In the simplest case (Fig. 7a) there are only

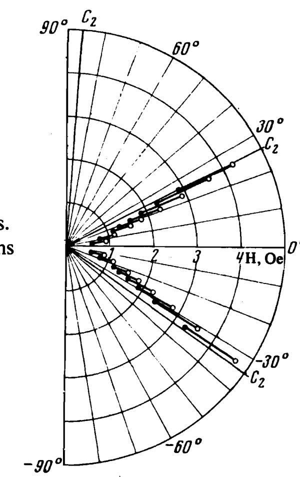

FIG. 6. Position of the second region with several states as a function of the direction of the magnetic field. The angles are reckoned from the direction of the high-frequency currents. The light circles designate the positions of the last jump of the magnetic moment with increasing field, while the black circles correspond to the last jump of the magnetic moment with decreasing field. Sample thickness 0.4 mm, $H_\sim$ = 7.2 Oe.

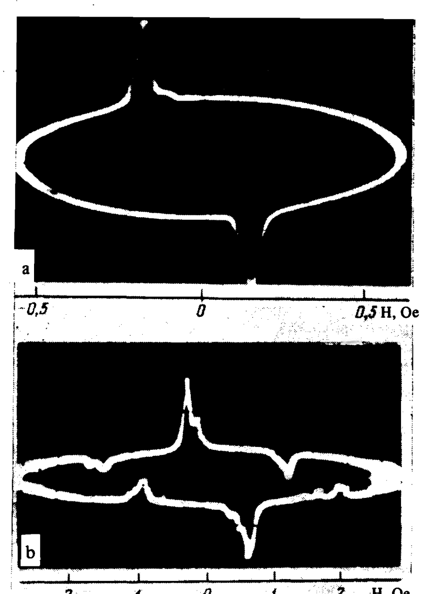

FIG. 7. Jumplike changes of the magnetic moment of the sample. The signal comes from the coil whose axis coincides with the axis of the high-frequency coil. The beam moves counterclockwise over the oscilloscope screen when viewed from the observer's position: d = 0.4 mm, $\angle jH = -4°$. a) $\angle jH = 95°$, $H_\sim$ 22 Oe, Ω = 76 Hz; b) $\angle jH = -24°$, $H_\sim$ = 15 Oe, Ω = 37 Hz, both regions are seen.

two brightly expressed pips corresponding to the flipping of the magnetic moment. In the more complicated situation, as well as when the power absorbed in the sample or the dependence of $\partial M/\partial H$ on H is registered, two regions are observed (Fig. 7b), in each of which there are several jumplike changes of the magnetic moment. We note that in the region that contains the origin, the magnetic moment of the sample increases if the magnetic field increases, and decreases with decreasing magnetic field, while in the second region the signs of $\partial M/\partial t$ and $\partial H/\partial t$ are opposite.

The width of the pips observed on the screen is determined by the time during which the magnetic moment changes. The order of magnitude of this time is 10^{-3} sec. (The receiving system passed pulses of width $\gtrsim 3 \times 10^{-5}$ sec.).

Using photographs from the oscilloscope screen, we can easily estimate the value of the magnetic field $\mathscr{H}$ produced by rectification of the high-frequency current. To this end it is necessary to compare the signal due to the sinusoidal variation of the external magnetic field $H = H_a \cos \Omega t$ with the signal corresponding to the flipping of the magnetic moment of the sample. An elementary calculation in which it is assumed that the field $\mathscr{H}$ is homogeneous inside the sample, yields for $\mathscr{H}$ the expression

$$\mathscr{H} = p \frac{D}{d} \frac{H_a S}{ab} \cos \alpha,$$

where a and b are the semiaxes of the ellipse observed on the oscilloscope screen and measured in length units, S is the area between the pip and the ellipse, α is the angle between the axis of the receiving coil and the direction of H, D is the height of the receiving coil, and d is the sample thickness. The coefficient p, which is of the order of unity, reflects the fact that part of the magnetic flux produced by the rectified current is closed inside the receiving coil. In our case, the sample thickness was 0.4 mm, the receiving coil had a rectangular cross section and consisted of two sections of 6 mm width each, separated by an interval of 3 mm. The height of the coil was 1.4 mm, and the sample was placed at the center of the coil, so that the gaps between the sample and the coil were ~ 0.5 mm. With these dimensions, the coefficient p ranged from 1.5 to 3.

Using the parameters corresponding to Fig. 7a, we obtain $\mathscr{H} \approx 0.7 - 1.4$ Oe. For Fig. 7b in the central region $\mathscr{H} \approx 0.7 - 1.4$ Oe, and in the region that does not cover the origin we have $\mathscr{H} \approx 0.2 - 0.4$ Oe.

Examination of the picture on the oscilloscope screen has revealed that at amplitudes $H_\sim$ greatly exceeding $H_\sim^k$ there is one more characteristic value of the high-frequency field. If the wave amplitude exceeds this value, then in the field region whose dimension approximately coincides with A there are observed time-periodic variations of the magnetic moment of the sample. The period of these variations is ~ 10^{-3} sec. We plan to publish the results of a detailed investigation of this effect later.

6. On the whole, effect was well reproducible from sample to sample, although the values of $H_\sim^k$, the slopes of the lines $A(H_\sim)$, etc., differed slightly. It appears that these differences are due to small difference in the sample quality and are not determined by the differences between their thicknesses. A weak deformation of the sample with thickness 0.58 mm, such that the lines of the radiofrequency size effect have not yet vanished, has led to a vanishing of the region with several states in strong fields, but did not destroy the region that encloses the origin.

The central region on all the samples had as a rule a certain asymmetry. This asymmetry is noticeable in Fig. 2, namely, jumplike changes of the magnetic moment are observed at different values of the magnetic field, depending on the side from which zero is approached. Among the possible causes for the appearance of asymmetry are the insufficient compensation of the earth's magnetic field, and the inhomogeneity of the high-frequency field along the sample.

7. It was of interest to trace the behavior of the magnetic moment of the sample at electromagnetic-field amplitudes lower than critical. As seen from Fig. 8, which was plotted point by point, by numerical integration of the $\partial M/\partial H_\sim = f(H_\sim)$ curves, an abrupt change of the macroscopic magnetic moment is observed in weak magnetic fields, and an increase of the amplitude of the high-frequency field leads to an increase of its absolute magnitude. The curves corresponding to different $H_\sim$ are not similar to each other. With increasing $H_\sim$, the largest increase takes place in the initial section of the M(H) curve, and the magnetic-field interval in which an abrupt change of the magnetic moment takes place becomes narrower.

In the sample 0.4 mm thick, the increase of the absolute value of the magnetic field occurs up to $H \approx 0.7$ Oe, followed by a decrease, and the M(H) curve crosses the abscissa axis in a magnetic field $H \approx 3.5$ Oe. A similar curve was obtained also with the sample 0.6 mm thick. For this sample, the maximum was reached at $H \approx 0.4$ Oe, and the abscissa axis was crossed at $H \approx 2.4$ Oe.

DISCUSSION

The experimental data reported above indicate, in our opinion, that current states are indeed realized in bismuth, but provide no direct indication of the nonlinearity mechanism that causes these states to appear.

There are several competing mechanisms capable of causing rectification of the high-frequency current. These include modulation of the conductivity by the self-field of the wave and heating of the electron system or of the entire crystal as a whole, which leads to the appearance of a Nernst-Ettingshausen emf. Finally, the presence of a system of surface levels, the energy spectrum of which is altered by the high-frequency-wave field, should also lead to a nonlinearity. One can hope that an explanation of the cause of the nonlinearity will be afforded in the future by a detailed investigation of the behavior of the magnetic moment of the samples exposed to electromagnetic waves with amplitude smaller than $H_\sim^k$.

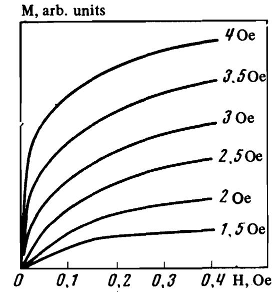

FIG. 8. Dependence of the magnetic moment of the sample on the external magnetic field at $H_\sim < H_\sim^k$, d = 0.4 mm, $\angle jC_1 = -4°$, $\angle jH = 90°$. The value of $H_\sim$ is marked on the curve.

As shown by Gurevich and co-workers[13,14], regardless of the microscopic mechanism that leads to the nonlinearity, a conducting medium in which there is a stationary weakly damped acoustic or thermal flux, or an electromagnetic-energy flux, can become unstable against a transition into a state with nonzero macroscopic magnetic moment. To this end it is necessary that the flux density exceed a certain critical value. A stationary magnetic field is produced in the medium, and changes periodically in a direction transverse to the flux. At the same time, circulating electric currents appear in the interior of the sample.

In our experiments, there was a flux of electromagnetic energy (damped radiowave) and a heat flux connected with heating of the sample by the radiowave were present along the normal to the surface of the sample. An estimate of the critical flux of electromagnetic energy for bismuth, given in[14], coincides in order of magnitude with the flux density corresponding to the amplitude of the high-frequency field, namely $H_\sim^k$. Moreover, Gurevich and Ioffe have predicted a transition of the sample to a vibrational state at even higher values of the sample flux[14,15]. Nonetheless, there is no complete correspondence between the experiments described above and the data in[13-15]. The theoretical papers contain the requirement that the flux be weakly damped, whereas in experiment the skin-layer depth was much smaller than the sample thickness. In addition, the experimentally observed transition to the current state was determined by the amplitude $H_\sim$ of the high-frequency field, and not by the energy flux density.

Regions with several states are observed in magnetic fields where the radius of the Larmor orbit of the bismuth electrons greatly exceeds the depth of the skin layer. As the electron moves in its orbit, it is acted upon by the entire magnetic field, which is the sum of the external field and the field produced by the rectified current. The latter, under ideal conditions, is homogeneous in the interior of the sample and experiences an abrupt change only in the skin layer. It can be assumed that the rectification efficiency and the field of the rectified current are determined by two parameters, by the amplitudes of the high-frequency wave and by the value of the total magnetic field

$$\mathscr{H}=f(H_\sim, H_0)=f(H_\sim, H+\mathscr{H}),$$

where it is assumed for simplicity that H and $\mathscr{H}$ are directed along one straight line.

Using the experimental data, we can attempt to select the function $f(H_0)$. Thus, in the presence of two stable states with transitions between them, corresponding to Fig. 7a, the function f should take the form shown in Fig. 9a. To verify this, let us find graphically the solutions of the equation $\mathscr{H} = f(H_0)$. Let, for example, H = 0. Then $f(H_0) = f(\mathscr{H})$, where $\mathscr{H}$ is regarded as an independent variable, and the solutions are obtained by the intersection of the plot of $y = f(\mathscr{H})$ with the straight line $y = \mathscr{H}$. As seen from Fig. 9b, in this case there are three solutions, two stable and corresponding to nonzero $\mathscr{H}$, and the other unstable.

If we know the form of the function $f(H_0)$, an analogous construction can be easily carried out for arbitrary H, since the plot of $f(H + \mathscr{H})$ is obtained from the plot of $f(\mathscr{H})$ by a shift along the abscissa axis by an amount H. The thin straight line in Fig. 9b corresponds to that value of H at which a jumplike transition from one state to another occurs.

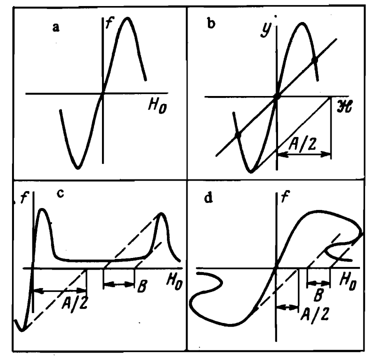

FIG. 9

The function $f(H_0)$ could have been reconstructed from the registered experimental dependence of $\mathscr{H}$ on the external magnetic field, if the absolute value of $\mathscr{H}$ were known with sufficient accuracy. However, the accuracy with which this quantity has been obtained in the present study is insufficient for this purpose.

Favoring the foregoing considerations are two circumstances. First, at small $H_\sim$ the nonlinearity, and with it also the rectified current, should vanish. Therefore at $H_\sim \ll H_\sim^k$ for H, with the exception of possibly a small region near zero, one can write $f(H_0) \approx f(H)$. Qualitatively, the f(H) curve at small $H_\sim$ (Fig. 8) coincides with that shown in Fig. 9a. Second, since the condition for the appearance of several stable states in the vicinity of the point H = 0 is $\partial f/\partial H_0|_{H=0} = 1$ (see Fig. 9b), the derivative $\partial\mathscr{H}/\partial H$ should increase rapidly as $H_\sim$ approaches $H_\sim^k$. Indeed, by differentiation with respect to H we obtain at H = 0

$$\frac{\partial \mathscr{H}}{\partial H}(H_\sim, 0)=\{[f'(H_\sim, 0)]^{-1}-1\}^{-1},$$

from which we see that as $H_\sim \to H_\sim^k$ the derivative $\partial\mathscr{H}/\partial H|_{H=0} \to \infty$. The abrupt increase of this derivative with increasing $H_\sim$ was observed in experiment.

It is easy to imagine a function $f(H_0)$ that makes it possible to obtain a large number of stable states. Let the function f be single-valued. Then, in analogy with the procedure used above, it is easy to verify that additional solutions of the equation $\mathscr{H} = f(H_0)$ appear if at a certain value of the amplitude of the high-frequency field there appear on the $f(H_0)$ curve additional points at which $\partial f/\partial H_0 \geq 1$. Such a point can be located sufficiently close to H = 0. In this case there exists only one region in which there are more than three states of the sample. On the other hand, if the point with $\partial f/\partial H_0 \geq 1$ is far from the origin, then there exist two regions with several states (this case is shown in Fig. 9c). The increase of the magnetic field leads in this case to a jumplike increase of the magnetic moment of the sample in both regions, whereas experiments have revealed a jumplike decrease of the magnetic moment in strong fields with increasing field.

The experimental data can be reconciled by assuming that the function $f(H_0)$ is not single-valued and is of the type shown in Fig. 9d. The criterion for the appearance of additional states with this form of $f(H_0)$ is not condition $\partial f/\partial H_0 \leq 1$ at one of the points of the ambiguity region.

CONCLUSION

The appearance of current states in bismuth is in our opinion the most pronounced of the presently known manifestations of nonlinearity in metals. The presence of an abrupt boundary of the effect, namely a field at which a jumplike transition takes place, raises hopes of using current states for an exact measurement of some combination of microscopic quantities. It is necessary here, of course, to have a theory that expresses the value of the transition field in terms of microscopic parameters, and to construct this theory it is necessary, in turn, to know the nonlinearity mechanism.

It should be noted that the onset of current states, as well as of other nonlinear effects, can apparently be observed not in semimetals only. To observe a developed nonlinearity in an ordinary metal it is necessary to increase the amplitude of the high-frequency field. This increase, however, will not necessarily lead to an increase of the overheating of the sample[7].

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