Cyclotron resonance on nonextremal orbits

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We have investigated a new phenomenon, namely, cyclotron resonance (CR) on nonextremal orbits, which occurs on the cyclotron-spectrum spectrum boundary produced artificially by using the sample thickness to cut off the orbits of the electrons belonging to Fermi-surface sections larger than a certain limiting value. This phenomenon uncovers a possibility of investigating the dynamic properties of nonextremal electron groups. The experiments were performed on bismuth at ~9 GHz and ~0.35°K. The effective mass of the electron in a field parallel to the major axis of the "ellipsoid" increases by (8±1)% when the diameter of the "ellipsoid" cross section decreases from 1 to 0.75 times the central diameter. We measured the Doppler shift of the CR line on the extremal orbits with the magnetic field inclined to the sample surface.

We have previously described[1] a new phenomenon, namely cyclotron resonance on electrons belonging to nonextremal sections of the Fermi surface and separated by cutting off the electron trajectories at the thickness of the sample[2].

Cyclotron resonance (CR) is observed as a resonant singularity of the surface resistance of the sample as a function of the magnetic field H. In the case of a convex Fermi surface (FS) and a nonquadratic energy spectrum, such singularities occur at the ends of the smooth spectrum of the cyclotron frequencies Ω = $\Omega(p_H)$ corresponding to the extremal sections of the FS, i.e., to its central section at the limiting point, determined by the direction of the field H.

By cutting off the trajectories whose diameters are larger than than the thickness D of a plane-parallel sample, one produces an artificial boundary Ω_b of the spectrum of the electrons moving along closed trajectories without colliding with the surface of the sample. If the probability of electron scattering in the volume of the sample is much smaller than the probability of its diffuse scattering upon collision with the surface, then CR is observed on such an artificial spectrum boundary $\Omega(p_H)$. The condition for realization of CR consists in this case of simultaneously satisfying, for a certain intermediate section of the FS, two equations, the first of which determines the choice of the boundary section with respect to its diameter

$$2p_{xb} = \frac{e}{c}HD, \tag{1}$$

while the second ensures the resonance condition

$$\frac{\omega}{n} = \Omega_{nb} = \frac{eH}{m_b^* c}; \tag{2}$$

here $m_b^* = m^*(p_{xb})$, ω is the frequency of the measuring microwave field, n is the order of the CR, and $p_x \perp p_H \parallel H$.

An increase of H shifts the limiting section of the convex Fermi surface from its central section toward the reference point, and at $n \gg 1$ the simultaneous satisfaction of Eqs. (1) and (2) takes place on many intermediate sections. However, since the density of states on the intermediate sections is smaller than on the extremal ones, the CR amplitude on the intermediate section is accordingly smaller.

Thus, the cyclotron resonance on the intermediate section of the Fermi surface makes it possible to measure the effective masses of the nonextremal electron groups. This phenomenon affords the only real

opportunity to investigate the dynamic properties of electrons belonging to the intermediate sections. In principle, quantum CR could serve the same purpose[3], but its observation, as shown by estimates, calls for samples having electron relaxation times larger by 2—3 orders of magnitude than in modern samples of record high quality. All other methods of investigating the properties of definite carrier groups yield information only on the extremal sections of the Fermi surface (with the exception, of course, of collective phenomena, such as electric conductivity, which characterize properties of the entire aggregate of carriers in the investigated sample).

EXPERIMENT

The experiments were performed with bismuth single crystals grown from the melt in a dismountable optically polished quartz mold[4]. The samples were disks of 17.8 mm diameter and thickness D = 190 ± 1, 121 ± 2 and 40 ± 3 μ. The average sample thickness was determined by weighing and by measuring the areas of the bases with a BMI-1 microscope. Nonparallelism of the sample surfaces, which was monitored optically, did not exceed the indicated accuracy limits. In the sample 190 μ thick, the threefold C_3 axis was perpendicular to the flat surface of the disk. For the two other samples, the C_3 axis was inclined 42° ± 2° to the surface. The orientation of the samples was determined from the anisotropy of the effective mass of the electrons[5] and was monitored by x-ray diffraction.

The single crystal, lying on the quartz substrate on which it was grown, was placed in a strip resonator connected in the feedback loop of a traveling-wave-tube oscillator[6]. The self-oscillator operated near the generation threshold. We measured the modulation of the amplitude of the generated microwave signal produced when the magnetic field H applied to the sample was modulated at a frequency 12.5 Hz. A block diagram of the measuring installation is shown in Fig. 1. At small changes of the surface impedance of the sample Z = R + iX, the registered low-frequency signal was proportional to $\partial R/\partial H$. The signal was recorded with an x-y plotter as a function of the magnetic field.

The resonator was a segment of a strip line of resonant length, capacitively loaded on both ends. It was made up of the surface of the sample and of a copper U-shaped strip[7], the distance from which to the sample surface could vary in the range 0.2—1.5 mm. The
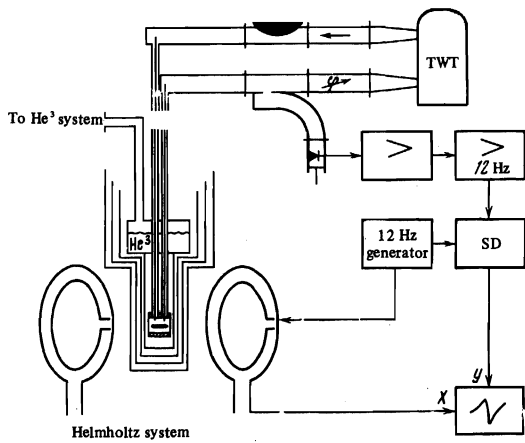
FIG. 1. Block diagram of measuring setup (TWT—traveling wave tube, SD—synchronous detector).
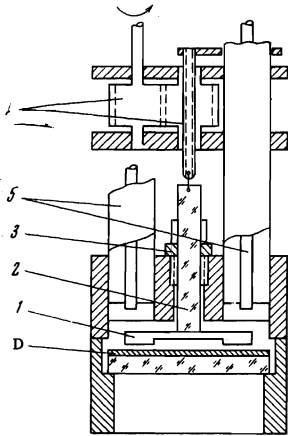

FIG. 2. Tunable strip resonator: D sample disk lying on a quartz substrate; 1—resonant strip; 2—quartz rod; 3—spring retaining sleeve for the rod; 4—tuning mechanism consisting of gears and a screw linked to the rod by a flexible loop; 5—coaxial transmission lines.

FIG. 3. Plot of CR for a bismuth single crystal 190 μ thick; H ∥ C_2, C_3 parallel to the normal, f = 9.16 GHz. The upper curve is plotted with a gain decreased by a factor of 10. The arrows and the numbers under the curve indicate the positions and the orders of the resonance; the lower arrows give the calculated positions of the CR lines on the central section of the Fermi surface without cutoff; H_{co} is the cutoff field. The oscillations in the field <2 Oe are resonances of the magnetic surface levels.

ensuing change in the capacitive loading tuned the oscillator frequency in the range 8.6—9.9 GHz. Figure 2 shows the device that made it possible to displace the strip during the course of a helium experiment. The device consists of a gear and screw that moves downward and upward without rotating, and accordingly presses in or draws out a quartz rod. The strip is glued with epoxy resin "P" to the quartz rod, which slides with little friction in a spring retaining sleeve. A loop of silk wire, linking the quartz rod with the screw, decouples the resonator mechanically from the cover of the instrument, thereby decreasing the level of the noise produced by vibration.

The magnetic field was produced by a system of Helmholtz coils. The earth field was cancelled out with accuracy ~0.01 Oe. A sample temperature 0.35°K was produced during the time of the experiment by pumping off He^3 vapor.

EXPERIMENTAL RESULTS

The main results were obtained in investigations of the sample 190 μ thick, which had the highest quality, namely, the parameter $\omega\tau$ determined in accordance with[8] from the width of the CR line amounted for this sample to 85—90, whereas for the thinner samples the respective values were 35—40 and 15—20.

Figure 3 shows a typical plot of $\partial R/\partial H$ against the magnetic field. We see that the character of the curve changes near the CR cutoff field H_{co} on the extremal section: a sharp decrease takes place in the CR ampli-
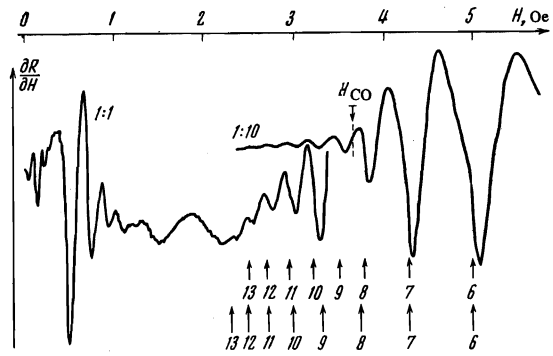
tude, but the resonant peaks are observed in field intervals from H_{co} = 3.67 ± 0.05 to ~2 Oe for the 190 μ sample, in the interval from H_{co} = 6.58 ± 0.07 to ~4 Oe for the 121 μ sample, and in the interval H_{co} = 17.4 ± 0.2 to 8 Oe for the 40 μ sample. The presented values of H_{co} were calculated with allowance for the orientations of the sample relative to the momentum of the electrons of the central section of the Fermi surface p_{xc} = 5.58 × 10^{-22} g-cm/sec, measured in[9]. In the indicated field intervals, no periodicity of the oscillations is observed as a function of the reciprocal field—a periodicity that is rigorously maintained for the CR on the extremal central orbit at H > H_{co}. As shown by experiment, when the frequency is varied in the range 8.7—9.8 GHz, the resonant field of each n-th resonance changes at H < H_{co} in accordance with the conditions (1) and (2).

To verify that the resonances observed at H < H_{co} were due to electrons belonging to nonextremal sections of the Fermi surface, we performed measurements with the magnetic field inclined at a small angle ϑ to the sample surface. If the resonant electrons belong to nonextremal and consequently noncentral sections of the Fermi surface, they have a velocity component v_H along the direction of the field H and as a result of the Doppler effect their resonant frequency should shift by $kv_H \sin\vartheta$ (k is the wave vector of the microwave field in the metal and is parallel to the normal to the metal surface). As a result, the resonant field H_n should shift by an amount ΔH_n in accordance with the formula

$$\frac{\Delta H_n}{H_n} = \frac{kv_H \sin\vartheta}{\omega}.$$

Figure 4 shows plots of CR at different inclination angles ϑ, and the obtained dependence of $\Delta H_n/H_n$ on the inclination angle ϑ is shown in Fig. 5. When the plot was constructed it was assumed that in the field H < H_{co} the CR Doppler shift is equal to

$$\frac{\Delta H_n}{H_n} = \left(\frac{\Delta H_n}{H_n}\right)_e - \left(\frac{\Delta H_n}{H_n}\right)_a,$$

where $(\Delta H_n/H_n)_e$ is the direct result of the experiment, and the correction $(\Delta H_n/H_n)_a$ is due to the anisotropy of the effective mass of the central-section electrons, determined from the same experiments by observing the CR at H > H_{co}, for which there is no Doppler shift. This correction reaches ~3% at ϑ = 10°.
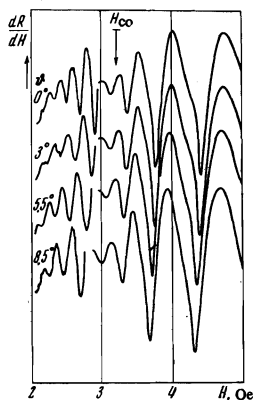
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FIG. 4. CR plot at different inclination angles ϑ of the magnetic field H to the surface of a sample 190 μ thick; H $\parallel$ C_1 at $\vartheta = 0$.
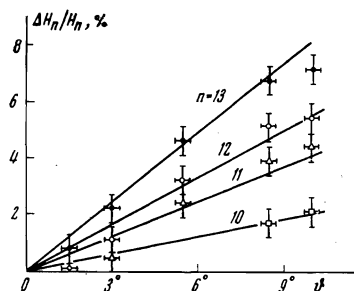

FIG. 5.   FIG. 6.

FIG. 5. Relative CR Doppler shift at H $<$ H_{co} against the angle of inclination of the magnetic field. The numbers on the right mark the orders n of the resonance. The straight lines were drawn approximately through the experimental points.

FIG. 6. Dependence of the Fermi-velocity component v_H along the magnetic-field direction, referred to the wave velocity $v = \omega\delta_i$, on the Fermi-momentum component p_H. The values of p_H/p_{H_0} were determined, in view of the known shape of the Fermi surface of bismuth [9], from the diameter of the electron trajectory in its cutoff field. p_{H_0} is the momentum at the limiting point. The straight line is calculated; the value $\delta_i = 2 \times 10^{-4}$ cm was assumed; the Fermi velocity was calculated using an ellipsoidal model of the Fermi surface.

Figure 6 shows a plot of the experimentally determined value of $v_H/v = v_H/\omega\delta i$ against the electron Fermi-momentum component p_H. The component p_H of the resonant electrons corresponding to each order of the resonance at H $<$ H_{co} was determined from the diameter of the electron trajectory in its cutoff field, the Fermi-surface shape being assumed known[9].

DISCUSSION

First we note some experimental results confirming the interpretation proposed above for the phenomenon. Measurements of the resonant values of the field H_n at different frequencies ω of the microwave measuring field have shown that H_n varies in accordance with the CR conditions reported above. The Doppler shift $\Delta H_n/H_n$ of the observed resonances is proportional to the small angle ϑ of the inclination of the field H to the surface of the sample (Fig. 5). $\Delta H_n/H_n$ increases with increasing order of the resonance; this means that resonances of higher order occur for electrons having a higher velocity component v_H, i.e., on Fermi-surface sections farther from the central section.

It should be noted that owing to the inversion symmetry of the Fermi surface, when the field is inclined to the metal surface, there are present equivalent elec-
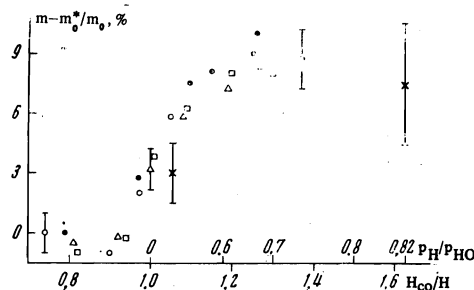

FIG. 7. Dependence of the effective mass m* of the electrons at H $\parallel$ C_2 on the value of H_{co}/H. The mass of the electrons of the central section is $m_0^* = 0.0092m_e$. Light symbols—experiments with sample of thickness D = 190 μ at frequencies f equal to: $\circ$—8.99 GHz, $\square$—9.50 GHz, $\triangle$—9.75 GHz; $\bullet$—D = 121 μ, f = 8.69 GHz; $\times$—D = 40 μ, f = 9.65 GHz. The values of p_H/p_{H_0} were determined as indicated in the caption of Fig. 6.

trons that drift along the field H both into the interior of the metal and in the opposite direction. One should therefore observe a splitting of the resonant peaks. However, the resonance due to the electrons moving towards the surface usually has a smaller amplitude[10], since these electrons interact more weakly with the electromagnetic wave moving into the interior of the metal than the electrons moving together with the wave. The fact that experiment revealed only a shift of the peaks towards weaker fields with simultaneous decrease of the amplitude by a factor ~ 1.5—2 (Fig. 4) at a maximum inclination angle $\vartheta \sim 10°$ is apparently due to the insufficient resolution of the resonances. A shift of the resonant peaks without splitting of the resonances, in magnetic field inclined to the surface of the sample has been observed in many studies, for example in[11].

Effective masses of electrons of nonextremal Fermi-surface sections. From the measured values of the resonant field one can determine the dependence of the effective mass of the electrons on their momentum component p_H, as shown in Fig. 7. The effective masses m* were calculated from the formula

$$m^* = eH_n n/\omega c.$$

The values of H_n at H $>$ H_{co} were determined by comparing the CR line shape calculated in accordance with Chambers[12] with that plotted experimentally, and at H $<$ H_{co}, when the resonances have practically a sinusoidal shape, H_n were taken to be the field values corresponding to the inflection points marked in Fig. 3 by arrows. The justification for this method of determining H_n follows from experiments on CR in thicker samples in the absence of cutoff. In this case the resonant values of the field $H_n = m^*\omega c/en$ calculated using the effective mass m* calculated from the period ΔH^{-1} of the CR correspond to the inflection points in the plot of that part of the curve where the resonances take the form of sinusoidal oscillations.

The momentum component p_H corresponding to each value of m* for the known shape of the bismuth Fermi surface[9] was determined from the diameter of the electron trajectory in its cutoff field. The points obtained for different samples at different frequencies, which are collected in Fig. 6, fit a single curve within the limits of the experimental errors, thus confirming once more the proposed interpretation of the phenomenon.

It is known that the energy spectrum of the bismuth

electrons differs from quadratic (see, for example,[5,9]), but the $m^*(p_H)$ dependence obtained in the present paper, where p_H is directed along the major axis of the electronic "ellipsoid" of the Fermi surface, yields new quantitative information concerning this fact.

Distribution of microwave field in the surface layer of bismuth. The decrease of the CR amplitude, as seen from Fig. 3, begins at a field somewhat larger than H_{co}. This can be connected with the fact that the fraction of the electrons that take part in the resonance begins to decrease already at $(H - H_{co})/H_{co} \sim \delta_{eff}/D$, where δ_{eff} is the skin-layer depth at which the electro-magnetic field attenuates appreciably. Assuming $H = H_8$—a field of resonance of order $n = 8$—we have in Fig. 3 $(H_8 - H_{co})/H_{co} \sim 0.05$. From this we obtain the estimate $\delta_{eff} = 10^{-3}$ cm, which is approximately ten times larger than the value $\delta \approx 0.8 \times 10^{-4}$ cm for the anomalous skin effect in bismuth, obtained as a result of measuring the high-frequency surface impedance[13].

Information on the distribution of high-frequency fields in bismuth can also be obtained by studying the Doppler shift of the CR on nonextremal orbits. Following[14], we represent an electromagnetic wave propagating along the inner normal z to the metal surface in the form

$$E(z, t) = E_0 \exp i(-z/\delta_i + iz/\delta_r + \omega t),$$

where the inductive depth $\delta_i = 1/k$ characterizes the wave velocity, and the active depth δ_r characterizes its damping. As shown above, for electrons having a velocity component v_H along a magnetic field H making an angle ϑ with the surface of the sample, the resonant field H_n shifts by a relative amount

$$\frac{\Delta H_n}{H_n} = \frac{k v_H \sin \vartheta}{\omega} = \frac{v_H \sin \vartheta}{\omega \delta_i}.$$

If the velocity v_H is known by calculation with the aid of a model, then the foregoing formula enables us to obtain δ_i. To determine δ_i, we use the experimental results shown in Fig. 5. Recognizing that the measurement accuracy is $\Delta H_n/H_n \sim 20\%$, the values of v_H for electrons of nonextremal sections can be obtained with sufficient accuracy by calculation using the ellipsoidal model of the Fermi surface.

The value of δ_i determined from the Doppler shift of the resonances by the described method amounts to $\delta_i = (2.3 \pm 0.5) \times 10^{-4}$ cm (at a frequency $f = 9.14$ GHz). The values of δ_i calculated from the shift of resonances of different orders ($n = 11, 12, 13$) have a scatter $\pm 0.5 \times 10^{-4}$ cm, which agrees with the error in the determination of $\Delta H_n/H_n$.

When the inclination angle of the field is increased, an angle ϑ_0 is reached at which the electrons move to a skin layer depth $\delta_r = 2\pi n v_H \sin \vartheta_0/\omega$ within a time equal to the cyclotron period. The angle ϑ_0 is characterized by an abrupt change in the amplitude of the observed cyclotron resonances; its measurement yields the value of δ_r. It follows from Fig. 4 that when the angle of inclination of the field changes to $\vartheta \sim 8.5°$, the sharp decrease of the CR amplitude does not take place. If we assume that the angle ϑ_0 has not yet been reached in this case, then we obtain for δ_r the estimate $\delta_r \gtrsim 10^{-3}$ cm.

A penetration of the HF field to a depth δ_{eff} much larger than the value of δ obtained by measuring the surface impedance was observed in an investigation of a bismuth single crystal for transparency to a microwave[15], where a value $\delta_{eff} \sim 20\delta$ was obtained. A similar slow fall-off of an hf field in the interior of a metal was observed in indium in an investigation of the splitting of the CR at the limiting point when the field was inclined[14] and in an investigation of the size effect[16]. A value $\delta_{eff} \approx 8\delta$ was obtained in both papers. As noted in[16], the reason for this difference between δ and δ_{eff} lies in the fact that the depth δ_{eff} depends strongly on the distribution of the hf field in the surface layer of the sample when the surface impedance (the measurement of which yields δ) is constant. It follows from the theory of the anomalous skin effect[17] that the electromagnetic field in the metal attenuates much more slowly than exponentially, and a difference by one order of magnitude between δ and δ_{eff} is justified.

Observation of specular CR. In[7], CR was observed at $H < H_{co}$ for electrons specularly reflected from the sample surface. A distinguishing feature of specular CR is its behavior when the frequency is varied: with decreasing frequency, the resonant field increases, i.e., it changes in a manner opposite to that in ordinary CR. As already mentioned, for the resonances observed in the present study, such anomalous behavior with varying frequency is not observed, so that they cannot be specular cyclotron resonances, all the more since the calculated resonant values of the field should be entirely different for the specular cyclotron resonances.

The differences between the experiments reported in this paper and the experiments on the observation of specular cyclotron resonances[7] was due to the quality of the investigated samples. The samples used in[7] were of worse quality and had a parameter $\omega\tau \sim 10-20$. The absence of specular CR in the given experiments can be attributed to the fact that in high-grade samples collisions with the surface make a larger contribution to the electron scattering than collisions in the volume. As seen from Fig. 3, the relative width of the CR lines is ~ 10 times smaller than that of the oscillations connected with transitions between magnetic surface levels of the electrons reflected from the surface[18]. The samples used in[7] had approximately the same relaxation time for electrons moving in a volume along closed orbits and for electrons that experience specular reflection from the surface. An increase in the volume relaxation time leads to a decrease in the width and to an increase in the amplitude of the CR on closed nonextremal orbits, and the latter not only become observable but also mask the CR with specular electron reflection.

One of the samples investigated in the present paper (120 μ thick) was mechanically damaged, after which its parameter $\omega\tau$, determined from the width of the CR line, decreased by approximately a factor of 2. The number of observed orders of CR at $H < H_{co}$ then decreased from 5 to 2, their amplitude decreased, and an additional minimum was seen on the experimental plot; its position in the field had the anomalous frequency dependence characteristic of specular CR and observed in[7].

Conclusion. We have investigated certain characteristic features of cyclotron resonances on electrons of nonextremal sections of the Fermi surface, and have demonstrated the possibility of using this phenomenon for the study of properties of these groups of electrons. The most significant results of the study are the meas-

urements of the effective masses of nonextremal groups of bismuth electrons, which give a very lucid and exact representation of the deviation of the electron energy spectrum from quadratic.

The new phenomenon was investigated in bismuth, which is a very convenient study object; in principle, however, it can be investigated in other metals in which the effect of cyclotron-resonance cutoff has been observed (for example, Sn[19], In[20]). All that we need for this purpose is single crystals of these metals of extremely high grade, better than those investigated to date. Their preparation, of course, is a difficult task, but the effort would be justified by the fact it makes the only realistic method of investigating nonextremal carrier groups applicable to these metals.

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