Phases of the magnetic-breakdown oscillations of the resistance and of the thermoelectric power of beryllium

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The relative shifts of the zeros of the magnetic-breakdown oscillations of the resistance and of the thermoelectric power are measured for [1010] and [1210] beryllium single crystals rotated about the longitudinal axis through an angle $\alpha \leq 2.2^{\circ}$ from the initial position H||[0001]. As $\alpha \rightarrow 0$, an additional shift of the zeros of the oscillations is observed. This shift cannot be ascribed to either an anomalous change of the oscillation frequency or the influence of the Shoenberg effect. The two-dimensional character of the magnetic-breakdown trajectories at H||[0001] and the phase-coherent character of the magnetic breakdown are invoked to explain the observed effect.

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It is known that magnetic breakdown (MB) in beryllium¹ leads to the onset of resistance oscillations and thermopower oscillations³ with giant amplitudes. The dependence of the oscillation amplitude of the resistance on the magnetic field could be well described within the framework of the theory of coherent magnetic breakdown (CMB).⁴ An important confirmation of the phase-coherent character of MB in beryllium was obtained in Ref. 5, where CMB anisotropy of the kinetic coefficients was observed. However, the method used in that reference to register the CMB anisotropy, by means of the phase difference between the ρ_{xx} and ρ_{yy} oscillations for one S-shaped sample, could lead to errors due to the nonequivalent changes for ρ_{xx} and ρ_{yy} in the geometry of the MB trajectories when the magnetic field deviates from the [0001] axis. It seemed of interest to measure the phases of the MB oscillations of the resistance and of the thermopower of beryllium by a direct method which excludes the influence of purely geometric effects.

EXPERIMENT

We measured the phase of the MB resistance and thermopower oscillations of two beryllium samples cut by the electric-spark method from one (0001) plate in the directions [1010] and [1210]. The sample dimensions were $0.5 \times 1 \times 6$ mm, and the resistance ratio was $\rho_{300 \text{ K}} / \rho_{4.2 \text{ K}} \approx 250$. Current and potential leads of copper wire were spark-welded. The (0001) faces of the samples were glued to a polished pyroceram plate in such a way that their axes were parallel. On one side, a common heater was secured to the samples from the top. The heater was made of nichrome wire and had a resistance of approximately 20 Ω . It produced a temperature gradient in the measurements of the thermopower oscillations. The liquid helium was prevented from reaching the heater by a cover glued together of dense paper impregnated in BF-2 resin. The plate with the samples was secured to the rotating platform of the apparatus (Fig. 1), which made it possible to establish any desired orientation of the magnetic field with accuracy not worse than 0.1°.

The magnetic field was produced by a superconducting solenoid operating in the undamped-current regime.⁶ The solenoid winding was made of a KÉTV-2NT cable of 0.7 mm



FIG. 1. Diagram of setup used to rotate the samples in a magnetic field: 1—samples, 2—tank-circuit coil of NMR generator, 3—adjustment screws, 4—springs, 5—support, 6—rotating platform.

diameter, and the superconducting switch was made of a composition bus-bar with cross section 3.5×2 mm. The rate of decrease of the magnetic field was ≤ 1 Oe/h, the homogeneity over a one-cm length was not worse than 10^{-5} . To sweep the field up to 1 kOe we used an additional coil of superconducting wire NT-50 of 0.33 mm diameter placed inside the main solenoid.

The magnetic field was determined by an NMR method. The circuit coil of the generator was located at a distance 5 mm from the samples (Fig. 1). The coil form was made of organic glass, and inside the form was placed aluminum powder imbedded in paraffin. The generator used KP301B field-effect transistors and was placed alongside the coil, so that it could operate at frequencies up to 120 MHz. The generator had nine fixed frequencies in the range from 40 to 120 MHz, which were set by connecting fixed capacitors into the tank circuit. The frequency was tuned continuously by varying the voltage on a varicap whose function was performed by light-emitting diode HP5082-4480. The resonance was recorded by the usual modulation procedure; the modulating magnetic field was produced by alternating current of frequency 167 Hz flowing through the coil that served to sweep the magnetic field. In fields up to 35 kOe, the field strength was determined from the position of the NMR line for the ¹H nuclei present in the material of the tank-circuit coil form; in fields above 35 kOe, we measured the NMR of 27 Al nuclei.

To exclude the influence of the magnetic fluxes frozen into the superconducting winding of the solenoid on the inhomogeneity of the field, we determined in each NMR measurement the distribution of the field along the solenoid axis, and the samples were placed in the region of maximum field homogeneity. The initial sample setting with $H \parallel [0001]$ was effected by using the extrema of the magnetoresistance (minimum in the case of rotation and local maximum for inclination). Even though both samples were cut from a single welloriented plate, their [001] axes turned out to deviate by an angle $\approx 1^{\circ}$. The phases of the resistance MB oscillations and thermopower oscillations were therefore measured for each sample separately. We determined the positions of the oscillations at fixed angles sample-rotation α around the longitudinal axis in a range $|\alpha| \leq 2.2^{\circ}$. The MB oscillations were recorded as functions of the current I flowing through the field-sweep coil; the additional field produced by this coil was determined from a calibration curve plotted by NMR. It was found that the additional field H has a nonlinear dependence on the current I:

$$H = AI + BI^2, \tag{1}$$

and the coefficients A and B turned out to be monotonically increasing functions of the field H_0 produced by the main solenoid. A typical calibration is shown in Fig. 2, in coordinates that are convenient for the separation of the linear and quadratic terms. The nonlinearity of the sweep-coil calibration is apparently due to the fact that it is located inside a relatively large superconducting solenoid.

The NMR signal was registered for each plot of the MB oscillations. This provided an absolute tie-in with the magnetic field. To verify that the measured phases of the MB oscillations are independent of the presence of the modulating magnetic field, frequency modulation of the oscillations of the NMR generator was used in some experiments. The absolute accuracy of the determination of the magnetic field



FIG. 2. Calibration of the field-sweep coil at $H_0 = 53081.9$ Oe. The line was drawn by least squares. The maximum deviation of the experimental points from the line does not exceed 0.2 Oe. The coefficients of expression (1) are in this case A = 41.199 Oe/A and B = 0.0809 Oe/A².

was 0.5-1 Oe, and the relative accuracy was not worse than 0.2 Oe. The magnetic field of the current flowing through the samples in the course of the resistance measurements did not exceed 1 Oe.

Each plot of the MB oscillations contained, depending on the strength of the main-solenoid field, from 2 to 12 oscillations. The voltages proportional to the measured signal, to the current through the field-sweep coil, and the voltage from the output of the synchronous detector of the registration circuit of the NMR resonance were fed to digital voltmeters and then punched on a tape for further computer reduction. In the course of the reduction we determined the absolute value of the magnetic field, identified the maxima and minima of the MB oscillations, drew the straight line corresponding to the monotonic part of the measured resistance or thermopower signal, and determined the coordinates of the points of intersection of the measured oscillatory dependence and this straight line. The values of the magnetic field at these points-the zeros of the oscillations-were used in the subsequent calculations of the phases and frequencies of the MB oscillations. All the measurements were performed at T = 4.2 K.

RESULTS

Figure 3 shows plots of the MB oscillations of the resistance and of the thermopower, plotted for a sample with orientation [1010] at H||[0001]. The resistance oscillations were observed against the background of a large monotonic component whose presence could lead to errors in the determination of the phases and frequencies of the oscillations. If we express the dependence of the resistance on the field in the form

$$\rho(H) = \rho_{\rm mon}(H) + \rho_{\rm amp} \sin(2\pi F/H + \varphi), \qquad (2)$$

the observed zeros of the oscillations can be shifted relative to their real positions by an amount

$$\Delta H = \frac{H^3}{2\pi F \rho_{\rm amp}} \frac{d\rho_{\rm mon}}{dH}.$$
(3)

For the resistance plot shown in Fig. 3, $\Delta H \approx 8$ Oe. In the plots of the thermopower, the monotonic component



FIG. 3. MB oscillations of the resistance (1) and of the thermopower (2), plotted for a sample with orientation $[10\overline{1}0]$ at H $\|[0001]$. The scale for the thermopower is arbitrary. The dashed lines show the monotonic components of the signals.

was much lower than the oscillatory component, and the correction of type (3) was within the magnetic-field measurement error. In this connection, the main results of the present paper were obtained from measurements of the MB oscillations of the thermoelectric power.

It is seen from Fig. 3 that the MB oscillations of the thermoelectric power s are shifted relative to the MB of the oscillations of the resistivity ρ by one-quarter of the period. If the values of the field at the points a and b of Fig. 3 are designated by H_s and H_ρ , then the phase difference between the oscillations of the thermopower and the resistance is

$$\varphi_s - \varphi_\rho = 2\pi F (1/H_\rho - 1/H_s).$$
 (4)

It follows from our measurements that $\varphi_s - \varphi_{\rho} = -90^{\circ} \pm 2^{\circ}$ regardless of the orientation of the sample relative to the magnetic field.

We investigated the relative change of the MB oscillation phase upon rotation of the sample around the longitudinal axis. To this end we chose one of the zeros of the oscillations (this was usually a point of type *a* in Fig. 3), and plotted the dependence of its position on the angle α between the hexagonal axis of the sample and the magnetic field. Such a rotation leads to a change in the area of the extremal section of the Fermi-surface sheet responsible for the oscillations (the "cigar" in the case of beryllium); this yields the change of the position of the oscillation $\delta H / H = \delta F / F$. In the case of small rotations

$$\delta F/F = 1/\cos\alpha - 1 \approx \frac{1}{2}\alpha^2,\tag{5}$$

i.e., the relation

$$\delta H / H = \alpha^2 / 2 \tag{6}$$

should be satisfied.

The experimental plots of $\delta H/H$ against α^2 , obtained for the samples [1010] and [1210], are shown in Fig. 4. The relative change of phase $\delta H/H$ can generally be reckoned from any arbitrary point. The experimental plots of H/Hagainst α^2 shown in Fig. 4 were drawn in such a way as to satisfy the relation (6) at large α . The validity of this relation at $\alpha > 3^\circ$ follows from the results of direct measurements of $F(\alpha)$ (Ref. 7).



FIG. 5. Dependence of the additional phase shift of the MB oscillations of beryllium on the magnetic field.

It is seen from Fig. 4 that relation (6) is satisfied only at sufficiently large inclination angles of the magnetic field away from the hexagonal axis. When the magnetic field approaches the hexagonal axis, systematic deviations from (6) set in, and are larger the stronger the magnetic field. The range of angles in which the additional displacement of the oscillations is observed differs somewhat for samples [1010] and [1210], but the values of the displacements at H||[0001] are equal. The latter means satisfaction of the condition $\rho_{xx} = \rho_{yy}$, which is a consequence of the symmetry.

The dependences of $\delta H/H$ on α^2 near the hexagonal axis are approximated in Fig. 4 by straight lines. Results of more detailed measurements of $\delta H/H$ vs α^2 offer evidence that these dependences have a certain structure that manifests itself more clearly when the magnetic field becomes stronger (dash-dot curve in Fig. 4b).

The relative displacements of the $\delta H/H$ oscillations measured at **H** $\|[0001]$ can be recalculated into a phase shift

$$p = -2\pi \frac{F}{H} \frac{\delta H}{H}.$$
(7)

The dependence of the phase shift on the magnetic field intensity is shown in Fig. 5.

q



FIG. 4. Angular dependences of the relative shift of the phase of the MB oscillations of beryllium: a—sample [1010], b—[1210]. The measured values of the thermopower are: $\Delta - H_0 = 23.54$; $\bullet - 35.25$; $\odot - 52.56$ kOe. The measurement data of the resistance are: $5 \bigtriangleup - H_0 = 63.21$ kOe.

The expected change of the oscillation frequency at $\alpha^2 = 10^{-3}$ is according to (5) $\delta F/F = 5 \times 10^{-4}$. To determine $\delta F/F$ with accuracy of the order of 10^{-4} from plots of several oscillations, the accuracy of the measurement of the period should be of the same order. In a field of 50 kOe, this requirement corresponds to a relative accuracy of 0.05 Oe. Although the experimental accuracy attained by us did not give grounds for hoping to determine the angular dependence of the oscillation frequency, we determined the average value of F by using the measured thermopower oscillations. The obtained value $F = 9.42 \pm 0.02$ MHz is in good agreement with the results of others.^{4,7}

DISCUSSION

MB oscillations of the thermopower and of the resistance of beryllium are determined by the extremal sections of the electronic cigars, and the phase shift, measured in the present study, between them is $-\pi/2$. A similar phase shift was observed by us for ruthenium,⁸ in which the MB oscillations of the thermopower and of the resistance are determined by the extremal sections of the whole ellipsoids. These results contradict the phenomenological analysis of Ref. 9. According to that reference, the thermopower oscillations should be shifted relative to the resistance oscillations by $+\pi/2$, and the sign of the phase shift should be determined by the type of the extremal sections responsible for the oscillations (" + " for the electron and " - " for the hole sections). Our data seem to indicate that the phase shift between the MB oscillations of the thermopower and the resistance is equal to $-\pi/2$ independently of the type of the extremal sections responsible for the oscillations.

The main result of the present paper is the determination of the angular dependence of the shift of the MB oscillations of the resistance and of the thermopower of beryllium (Figs. 4 and 5). If the angle α between the magnetic field direction and the [0001] axis exceeds $\alpha_0 = 0.5 - 1.5^\circ$ (depending on the field strength), the shift of the MB oscillations as a function of the angle α is well described by their relation (6). It follows therefore that the MB oscillation frequency F at $\alpha > \alpha_0$ is determined by the extremal sections of the cigar, in agreement with the results of direct measurements of $F(\alpha)$ cited in Ref. 7. An interesting singularity can arise at $\alpha \rightarrow 0$, where an additional shift of the MB oscillations appears and contradicts (6). Since this additional shift depends on the magnetic field intensity, it cannot be attributed to geometric singularities of the shape of the Fermi surfaces of beryllium. Nor is it possible to attribute the shift to an anomalous change of the frequency of the MB oscillations, caused by some other factors, say the magnetic breakdown on a non-extremal orbit. In the latter case, the transition to the normal dependence (6), meaning a return to the extremal orbits, should be accompanied by a jump of the phase of the MB oscillations, something not observed in experiment (Fig. 4).

One other possible explanation of the additional shift of the MB oscillations might be the Shoenberg effect, which consists in the fact that if the de Haas-van Alphen effect is strong the internal field in the sample differs from the applied field.¹⁰ It is known¹¹ that the Shoenberg effect in beryllium manifests itself in the appearance of low-frequency modulation of the MB oscillations. It must be noted, however, that measurements of the additional shift φ were made by us at magnetic field values corresponding both to maxima and to minima of the low-frequency modulation, and all the data obtained fit well the common smooth dependence shown in Fig. 5. This result demonstrates that the Shoenberg effect is not the cause of the onset of the additional shift of the MB oscillations.

Thus, the observed additional shift of the MB oscillations cannot be attributed to either anomalous changes of their frequency or to the influence of the Shoenberg effect.

The decisive factors in the explanation of the observed effect are the two-dimensional character of the MB trajectories at $H \parallel [0001]$, and the phase-coherent character of the MB in beryllium. The two-dimensionality of the magnetic breakdown net under CMB conditions leads to an increase of its sensitivity to variations of the phase of the wave function of the electron, compared with the one-dimensional case of a linear string of orbits coupled by magnetic breakdown. In the one-dimensional case the role of the phase indicator is played only by small closed sections of the Fermi surface, and the changes of the phase of the wave function of the electron when moving along the orbit-the linkages between them-drop out of the final expressions for the oscillations of the kinetic coefficients. This occurs because the interference of the wave functions is possible only on closed orbits. In the two-dimensional case there is a network of intersecting orbits and, in addition to the interference of the wave functions interference on the linkage orbits occurs on the small closed sections of the Fermi surface. It is this which leads to the high sensitivity of the kinetic coefficients to small changes of the phase of the wave function. Such a change occurs, e.g., in a magnetic-breakdown transition of carriers from one sheet of the Fermi surface to another. According to Ref. 12, the jump of the phase of the wave function in such a transition depends on the magnetic field intensity, to which it apparently is possible to attribute the observed dependence of the phase shift of the MB oscillations on the field (Fig. 5). In principle, other causes can also lead to small changes of the phase of the electron wave function.

There is at present no rigorous theory of CMB in the two-dimensional case. There is only an estimate of the range of directions of the magnetic field in which the influence of the two-dimensionality cannot be neglected. According to Ref. 13, it amounts to $\alpha_0 \sim \kappa^{1/2}$, where $\kappa = e\hbar H / cp_0^2$ is the quasiclassicism parameter, and p_0 is the characteristic momentum. For beryllium, this estimate at H = 50 kOe yields $\alpha_0 \approx 10^{-2}$ rad, in reasonable agreement with the experimental data of Fig. 4.

Thus, the experimental results of the present paper can be regarded as a manifestation of the two-dimensional character of the phase-coherent magnetic breakdown in beryllium.

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