Kinetic properties of ruthenium under magnetic-breakdown conditions. Galvanomagnetic effects

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The magnetoresistance and the Hall effect of the presently purest obtainable ruthenium crystals, with $\rho 273.2$ K/$\rho 4.2$ K up to 3000, were investigated at a temperature 4.2 K and in magnetic fields up to 100 kOe under magnetic-breakdown conditions. It is established that three types of magnetic-breakdown trajectories are formed in ruthenium, namely, open and closed trajectories and a space net. The influence of the trajectories produced after breakdown on the anisotropy and on the field dependences of the magnetoresistance and of the Hall effect are investigated. The formation of magnetic-breakdown trajectories in all three cases is accompanied by quantum oscillations of the magnetoresistance.

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INTRODUCTION

The Fermi surfaces of most metals, both transition and nontransition, and of their various compounds having metallic conductivity (intermetallides, oxides, carbides, etc.), have by now been investigated by experimental methods and by theoretical calculations of the electron energy spectra. It was established as a result of these investigations that many metals have electron-spectrum structures and Fermi surfaces that make possible magnetic breakdown, which leads in most cases to a radical change in the configuration of the electron trajectories. Under magnetic breakdown conditions it becomes therefore possible to control effectively the character of the carrier trajectory in the metal by such external factors as pressure, magnetic fields, ultrasound, or temperature.

In turn, a change in the type of the conduction-electron trajectories can cause a strong change in the electronic properties of a metal, primarily its kinetic properties. Moreover, in a recent paper[1] it is shown theoretically that effects nonlinear in the electric field can appear, in principle, in the kinetic properties of metals under magnetic breakdown conditions. These effects are connected with the "heating" of the electrons on narrow layers of open magnetic-breakdown trajectories.

Ruthenium turned out to be one of the most interesting objects, inasmuch as magnetic breakdown in this metal, depending on the orientation of the magnetic field relative to the crystal, can lead to three different configurations of magnetic-breakdown orbits: open (type A), closed (type B) and space-net (type C). It is therefore of interest, first, to determine the experimental picture of the change of the kinetic properties of ruthenium in all three magnetic-breakdown situations. Second, to establish whether magnetic breakdown oscillations of the resistance are possible in the cases when the magnetic breakdown transforms the open "pre-breakdown" orbits into closed ones (type B) and when it leads to a space net of magnetic breakdown orbits (type C). The point is that in the available papers on magnetic breakdown the observed oscillations of the magnetoresistance are usually connected with open magnetic breakdown trajectories, and doubts have been expressed concerning the possibility of observing resistance oscillations if the magnetic breakdown results in the formation of only closed trajectories. Third, to ascertain whether ruthenium is a promising object for the search for the nonlinear effects predicted in Ref. 1.

We have therefore measured a set of galvanomagnetic and thermoelectric properties of the purest presently available single crystals of ruthenium under magnetic breakdown conditions. This paper is devoted to a study of the influence of magnetic breakdown on the magnetoresistance and on the Hall effect; the results on measurements of the thermoelectric power and the Nernst effect are given in Ref. 2.

1. TOPOLOGICAL FEATURES OF THE FERMI SURFACE OF RUTHENIUM. POSSIBLE MAGNETIC BREAKDOWN TRAJECTORIES

Ruthenium is a transition $4d$ metal with atomic number 44 and hexagonal close-packed lattice. Its electronic structure was investigated by various methods. In Ref. 2, the de Haas–van Alphen (dHvA) method was used to reconstruct the principal sections of the Fermi surface. In Refs. 4–6 were investigated the galvanomagnetic properties of ruthenium, the topology of its Fermi surface, and the degree of compensation. The results of these papers agreed with the data of Ref. 3 and show that ruthenium is a compensated metal. Measurements of the galvanomagnetic properties,[6-8] of the dHvA effects,[3] as well as of the thermoelectric power[2,9-11] offer evidence that magnetic breakdown takes place in ruthenium.

Calculation of the electronic structure of ruthenium for the atomic configuration for $4d^7 5s^1$, with account taken of relativistic corrections, was carried out in Ref. 12. The results of this calculation are in good agreement with all the presently available experimental data.

In accordance with a model constructed from the results of the experimental studies,[3,6] the Fermi surface of ruthenium consists of six sheets.[1)] There are three closed hole sheets, two centered at the points L (the lens $L7h$) and Γ (the sheet $\Gamma 10h$), and the third located between L and M (the sheet $LM7h$). There are also two

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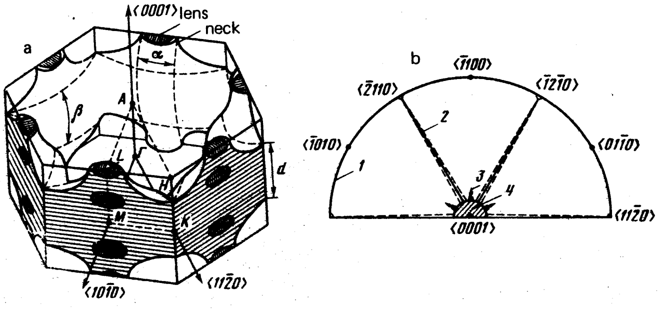

FIG. 1. Multiply connected hole sheets $KM8h$ of the Fermi surface of ruthenium (a) and the stereographic projection (b) of the special directions of the magnetic field at which open trajectories occur (constructed in accordance with the Fermi surface model without allowance for the magnetic breakdown). Line 1 corresponds to the direction of opening along ⟨0001⟩, 2—along ⟨10$\bar{1}$0⟩, 3—along ⟨1$\bar{2}$10⟩, 4—two-dimensional region of open trajectories parallel to the basal plane; at the points ⟨0001⟩ and ⟨1$\bar{2}$10⟩ the thickness of the trajectory layer with open direction parallel to the basal plane vanishes.

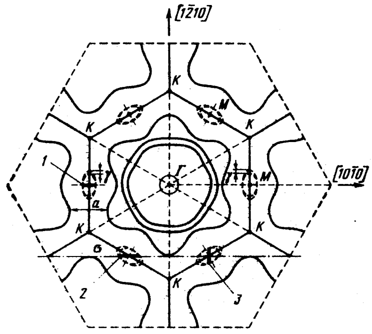

FIG. 3. Projection of multiply connected hole sheet $KM8h$ and of lenses $L7h$ on the plane $M\Gamma K$. The lens sections 1, 2, and 3 correspond to dHvA frequencies $3.2\cdot 10^6$, $4.0\cdot 10^6$ and 3.3×10^6 Oe.

closed electron sheets with center at Γ (the sheets Γ9e and Γ10e). In addition to the closed sheets, there is a multiply connected hole sheet $KM8h$, shown in Fig. 1a and constituting an assembly of "sleeves" connected with one another in the vicinities of the K points and connected by "necks" in the ML directions. Open trajectories are possible on this sheet in the absence of magnetic breakdown (Fig. 1b). The distinguishing feature of the Fermi surface of ruthenium, however, is that the hole lenses $L7h$, located inside the necks of the sheet $KM8h$, are tangent to the latter in the AL direction as a result of the ALH plane, the necks and the lenses are separated by a small energy gap due to spin-orbit splitting. It is precisely this construction of the multiply connected hole sheet and the anisotropy of the spin-orbit gap which admit of several magnetic breakdown types with different configurations of the pre- and post-breakdown trajectories.

A. Open magnetic breakdown trajectories

The intersection of the multiply connected hole sheet $KM8h$ and of the lens $L7h$ by the plane $LMML$ that contains the open magnetic breakdown trajectories is shown in Fig. 2 ($\varphi=0$). The open σ trajectory is shown by a solid line. This magnetic-breakdown situation can be realized at H ∥ ⟨1$\bar{2}$10⟩. Figure 3 shows the projection of the sheet $KM8h$ and of the lenses $L7h$ on the plane $M\Gamma K$. It is seen that, because of the geometric dimensions of the multiply connected hole sheets (the width of the sleeve is a), the layer of the open magnetic-breakdown σ trajectories (dash-dot line) should have a relatively small thickness in the ΓK direction. An investigation of the influence of magnetic breakdown with formation of a narrow layer of trajectories on the critical properties is of interest in connection with the theoretical predictions made in Ref. 1.

B. Closed magnetic-breakdown trajectories

Figures 2 and 4 ($\varphi=0$) show the intersections of the multiply connected sheet $KM8h$ with the planes perpendicular to the directions ⟨1$\bar{2}$10⟩ and ⟨10$\bar{1}$0⟩. In these sections are located open α trajectories that pass over the multiply connected hole sheet in the ⟨0001⟩ direction. However, just as in the preceding case, the presence of a small gap between the hole lens and the neck produces conditions for magnetic breakdown, as a result of which closed post-breakdown trajectories can be formed (shown dashed). Indeed, investigations of the galvanomagnetic properties[6] have shown that magnetic breakdown of this type begins to manifest itself already in magnetic fields as low as 20 kOe.

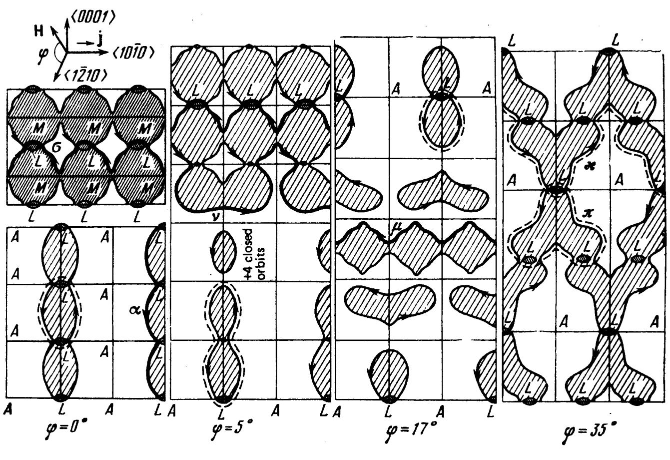

FIG. 2. Characteristic types of sections of the Fermi surface and possible magnetic-breakdown trajectories at H ⊥ ⟨10$\bar{1}$0⟩.

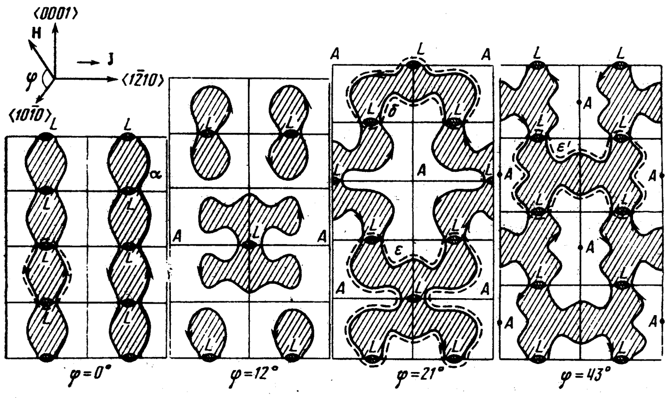

FIG. 4. Typical sections of the Fermi surface and possible magnetic breakdown trajectories for H ⊥ ⟨1$\bar{2}$10⟩.

Because of the anisotropy of the spin-orbit gap (see Figs. 2–4), the magnetic breakdown probability at H ∥ ⟨10$\bar{1}$0⟩ differs from that at H ∥ ⟨1$\bar{2}$10⟩. Naturally, this anisotropy should manifest itself in the kinetic characteristics of ruthenium measured in strong magnetic fields. However, the most interesting question in magnetic breakdown of this type is whether magnetic breakdown oscillation effects can be observed in the galvanomagnetic properties.

C. Magnetic breakdown "space-net" of trajectories

An analysis of the sections of the Fermi surface of ruthenium shows that in certain magnetic-field directions (angle φ) magnetic breakdown between the neck and the lens is possible, with formation of trajectories of the space-net type. Thus, in Fig. 2 ($\varphi = 35°$) and Fig. 4 ($\varphi = 21°$ and $43°$) are shown the sections of a multiply connected hole sheet with the trajectories of this type. The dashed lines show the possible magnetic-breakdown trajectories. Under space-net conditions, the electrons and holes can move randomly both along open trajectories (for example the $\varkappa$ trajectories in Fig. 2) and unclosed ones (the π trajectories on Fig. 2 and the δ, ε, and ε' trajectories on Fig. 4). In strong magnetic fields, however, when the probability of the magnetic breakdown is close to unity, the carriers move mainly in closed trajectories of type π, δ, and ε'. No magnetic breakdown with formation of a magnetic-breakdown space was observed so far in ruthenium. Therefore an investigation of such a magnetic-breakdown situation is of interest. In particular, the possibility of oscillatory phenomena in this magnetic-breakdown situation is a pressing question.

2. SAMPLES. EXPERIMENTAL PROCEDURE

Measurements of the anisotropy and of the field dependences of the magnetoresistance and of the Hall effect were made at $T = 4.2$ K and in magnetic fields up to 100 kOe on samples measuring $0.5 \times 0.5 \times 12$ mm. The samples were cut along the principal crystal axes by the electric-spark method from a single crystal with $\rho_{273.2K}/\rho_{4.2K} \sim 3000$, grown by the method described in Ref. 13. The error in the orientation of the samples was not more than 2° and was monitored by x-ray diffraction. The potential contacts for the measurement were secured to the sample by the method proposed in Ref. 14. The voltage sensitivity of the measurement setup was 10^{-8} V. The dependences of the magnetoresistance and of the Hall emf on the magnitude and direction of the magnetic field were recorded with a PDP-4 x-y recorder with preamplification of the signal. The scanning rate of the magnetic field produced by the superconducting solenoid was 4 kOe/min. In measurements of the anisotropy of the indicated properties, the sample was rotated in a transverse magnetic field by a friction device at a speed 0.05 rpm.

To separate the oscillating part of the magnetoresistance

$$\Delta\rho = \rho(H) - \rho(H=0) = \Delta\rho_{mon} + \Delta\rho_{osc}$$

(where $\Delta\rho_{mon}$ is the monotonic component and $\Delta\rho_{osc}$ is the oscillating component) we used a method of cancelling out the monotonic part, using the resistance of a tungsten single crystal measuring $1 \times 1 \times 15$ mm. The choice of tungsten was dictated by the fact that,

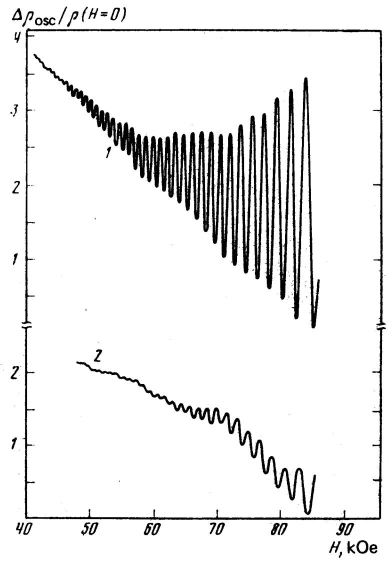

FIG. 5. Typical plots of oscillating contribution to the magnetoresistance for a sample with j ∥ ⟨1$\bar{2}$10⟩ at $T = 4.2$ K: 1—H ∥ ⟨10$\bar{1}$0⟩, 2—direction of H inclined 12° to the ⟨10$\bar{1}$0⟩ axis.

first, its magnetoresistance at all directions of the magnetic field relative to the crystal has a quadratic field dependence close to the field dependence of ruthenium at most orientations of the magnetic field. Second, no oscillations of the magnetoresistance are observed in fields up to 100 kOe in tungsten crystals with a ratio $\rho_{273.2K}/\rho_{4.2K} \sim 10^4$. It was precisely this compensation method which made it possible to record only the oscillating contribution to the magnetoresistance at maximum sensitivity of the installation, and to study the anisotropy of the amplitudes and frequencies of the magnetic-breakdown oscillations. A typical plot of the oscillating part of the magnetoresistance of ruthenium is shown in Fig. 5.

3. MEASUREMENT RESULTS

3.1. Current orientation j ∥ ⟨0001⟩

a) *Anisotropy and field dependences of the magnetoresistance.* Figure 6 shows the anisotropy $\Delta\rho/\rho = f(\varphi)$ and the field dependences of the transverse magnetoresistance $\Delta\rho/\rho$ of a sample with electric-current orientation along the hexagonal axis. As shown by an analysis of the sections of the Fermi surface of ruthenium (Figs. 2–4), the geometry of the experiments with this current orientation is the most convenient for the study of magnetic breakdown of type A, for in this case the direction of the layer of the open magnetic breakdown trajectories is perpendicular to the electric current. In fact, it is seen from Fig. 6a that at H ∥ ⟨1$\bar{2}$10⟩ there appear deep narrow minima of the magnetoresistance, and their depth increases with increasing magnetic field, while the width at half the depth of the minimum $[(\Delta\rho/\rho)_{\max} - (\Delta\rho/\rho)_{\min}]/2$ remains constant at approximately 3°. This fact indicates the presence in the basal plane of open trajectories perpendicular to the current.

This is confirmed also by the form of the field dependences of the magnetoresistance (Fig. 6b), which first increases quadratically with the field when H is directed along the minima and then reveals a tendency to saturation. In fact, according to the model of the Fermi surface of ruthenium, when H is exactly oriented along the ⟨1$\bar{2}$10⟩ axis the usual open trajectories cannot appear in the basal plane because of the necks on the multiply connected hole sheet *KM*8*h* (see Fig. 2). Therefore, in sufficiently weak fields $H < 20$ kOe (even though in this case $\omega\tau > 1$, where ω is the cyclotron frequency and τ is the characteristic relaxation time), all the orbits are closed and the resistance increases quadratically with increasing magnetic field. In strong magnetic fields magnetic breakdown transforms the closed orbits into open ones, and it is this which leads to saturation of the magnetoresistance.

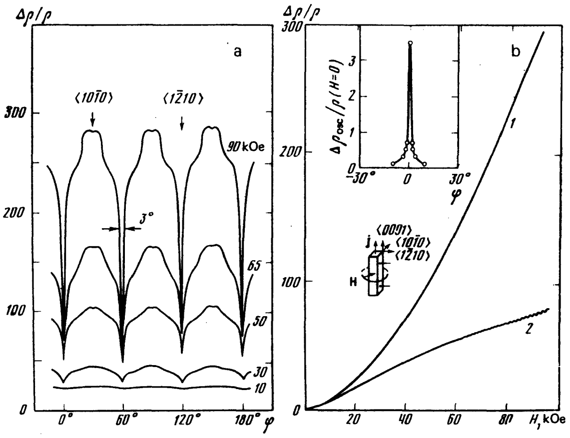

FIG. 6. Anisotropy (a) and field dependences (b) of transverse magnetoresistance of sample with j ∥ ⟨0001⟩ at $T = 4.2°$ K. In case b: 1—H ∥ ⟨10$\bar{1}$0⟩, 2—H ∥ ⟨1$\bar{2}$10⟩. Inset—anisotropy of the amplitude of the oscillations in a field 85 kOe, $\varphi = 0$ corresponds to H ∥ ⟨1$\bar{2}$10⟩.

b) *Oscillations of the magnetoresistance.* In accordance with Ref. 15, the appearance of open magnetic-breakdown trajectories in ruthenium is indicated also by oscillations of the magnetoresistance at the minima of the rotation diagrams, i.e., at H ∥ ⟨1$\bar{2}$10⟩. The frequency of the observed oscillations is $(4.10 \pm 0.05) \cdot 10^6$ Oe, and agrees well with the frequency of the dHvA oscillations (4.0×10^6 Oe, Ref. 3), corresponding to the area of the extremal intersection of the *L*7*h* lens with the *LMML* plane at H ∥ ⟨1$\bar{2}$10⟩ (Fig. 3, section 2). The agreement between the frequencies of the oscillations shows that the layer of the magnetic-breakdown trajectories passes through the lens and lies precisely in the *LMML* plane. If such magnetic breakdown trajectories were not produced, then the ordinary Shubnikov oscillations would correspond to section 1 of Fig. 3 and have a frequency 3.3×10^6 kOe, something not observed in the experiment.

The inset in Fig. 6b shows the anisotropy of the amplitudes of the osciallations of the magnetoresistance. Attention is called to the sharp decrease of the oscillation amplitude when the magnetic field direction deviates from the ⟨1$\bar{2}$10⟩ axis: deviation by an angle ~1.5° causes the amplitude to decrease by a factor of 2, and deviation by 10° suppresses the oscillations. This fact is evidence that the produced magnetic-breakdown layer of open σ trajectories is quite narrow (see Fig. 3).

c) *Hall effect.* At H ∥ ⟨1$\bar{2}$10⟩, anomalies are observed also in the Hall effect. Figure 7 shows the dependences of the Hall emf E_h and of the derivative of the magnetoresistance $d(\Delta\rho/\rho)/dH$ on the magnetic field. It is seen that the change of the slope of the field dependence of the Hall emf occurs at the same value of the magnetic field as the change from quadratic dependence to saturation

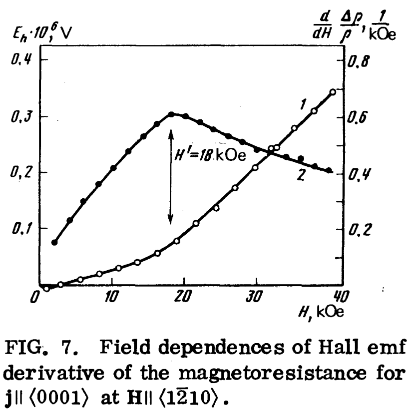

FIG. 7. Field dependences of Hall emf E_h (curve 1) and of the derivative of the magnetoresistance for a sample with j ∥ ⟨0001⟩ at H ∥ ⟨1$\bar{2}$10⟩.

in the magnetoresistance.

Figure 8 shows the anisotropy and the field dependence of the Hall coefficient R. It is seen, first, that at $\mathbf{H}\parallel\langle 1\bar{2}10\rangle$ in fields $H \geqslant 18$ kOe the absolute value of R increases sharply. For other orientations of the magnetic field, at which the formation of open magnetic breakdown trajectories is impossible, no such effect is observed. Second, in magnetic fields weaker than H' (but at $\omega\tau > 1$), the anisotropy of the Hall coefficient is small and amounts to only 10%. On the contrary, at $H > H'$, a sharp anisotropy of the Hall coefficient appears. The value of R increases by three times (compared with R in the "pre-breakdown" situation) in precisely those directions of H relative to the crystal at which realization of open magnetic breakdown trajectories is possible.

The appearance of minima on the rotation diagram at $\mathbf{H}\parallel\langle 1\bar{2}10\rangle$ can also be attributed with ordinary open trajectories produced on the multiply connected hole sheet in accordance with the stereographic projection (Fig. 1b), when $\mathbf{H}$ does not lie strictly in the (0001) plane. However, the entire aggregate of the experimental facts for the sample with $\mathbf{j}\parallel\langle 0001\rangle$, and in particular the value of the frequency and the sharp anisotropy of the amplitude of the oscillations, and also the anomalies in the field dependences of the Hall emf and of the magnetoresistance, can apparently not be explained without taking the magnetic breakdown into account.

3.2. The current orientations $\mathbf{j}\parallel\langle 10\bar{1}0\rangle$ and $\mathbf{j}\parallel\langle 1\bar{2}10\rangle$

On samples with the electric field directed along the axes $\langle 10\bar{1}0\rangle$ and $\langle 1\bar{2}10\rangle$ it is possible to observe experimentally the magnetic breakdown situations of type B and C.

a) *Anisotropy and field dependences of the magnetoresistance.* Samples with these orientations reveal a strong anisotropy of the magnetoresistance (Fig. 9). At a magnetic field direction $\mathbf{H}\parallel\langle 0001\rangle$, the plots of $\Delta\rho/\rho = f(\varphi)$ of both samples show deep and broad minima, and the field dependence of $\Delta\rho/\rho$ in these minima tends to saturation. This behavior of the magnetoresistance was attributed in Ref. 6 to geometrical decompensation due to the presence, on the $KM8h$ hole sheet, of a thick β layer of orbits of electronic character (Fig. 1). Measurements in strong magnetic fields confirm this conclusion.

It can be seen from Fig. 9 that when the magnetic field is inclined away from the $\langle 0001\rangle$ axis by more than 30° there are observed, first, a sharp increase of the magnetoresistance and, second, anisotropy of the dependences of $\Delta\rho/\rho$ on φ in a range of angles φ from 30° to 90°. In this angle interval, the magnetoresistance varies like $\Delta\rho/\rho \propto H^n$, where n takes on values from 1.7 to 2.0. The unlimited growth of the magnetoresistance with increasing field, particularly at $\mathbf{H}\parallel\langle 10\bar{1}0\rangle$ and at $\mathbf{H}\parallel\langle 1\bar{2}10\rangle$, points to the absence of open α trajectories (Fig. 1a) and cannot be explained on the basis of a stereographic projection (Fig. 1b) constructed without allowance for the magnetic breakdown. This behavior of the magnetoresistance is the result of magnetic breakdown of type B, which transforms the open trajectories of the α layer into closed ones (Figs. 2 and 4, $\varphi = 0$). It is precisely this interpretation which corresponds to the results of the measurement of the dHvA effect: in Ref. 3 were observed oscillations due to closed sections of the multiply connected hole sheet passing through the LML line. The presence of magnetic breakdown of this type even in weak fields as low as 20 kOe was observed in Ref. 6. We note that the asymptotic behavior of the field dependence of $\Delta\rho/\rho$ in this magnetic-breakdown case agrees well with the theoretical analysis carried out in Refs. 16 and 17.

It is seen from Figs. 2 and 4 that magnetic breakdown of type C formation of a space net of trajectories is possible in a sample with $\mathbf{j}\parallel\langle 10\bar{1}0\rangle$ at $\varphi \sim 35°$ and in a sample with $\mathbf{j}\parallel\langle 1\bar{2}10\rangle$ at angles close to 21° and 43°. Repeated recording of the plots of $\Delta\rho/\rho$ against the field in the region of these angles reveals an oscillatory contribution which is superimposed on the monotonic field dependence of the magnetoresistance. To investigate the nature of these oscillations, a method of cancelling the monotonic component was used. This made it possible to separate for samples with orientation of the current $\mathbf{j}$ along $\langle 10\bar{1}0\rangle$ and $\langle 1\bar{2}10\rangle$ the oscillating contribution to the magnetoresistance

$$\Delta\rho_{osc}/\rho(H=0) = [\rho(H) - \rho_{mon}(H)]/\rho(H=0),$$

to measure the magnetic frequencies F and the ampli-

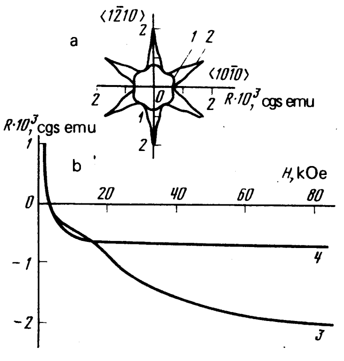

FIG. 8. Anisotropy (a) and field dependences (b) of the Hall coefficient at $\mathbf{j}\parallel\langle 0001\rangle$: 1—$H = 15$ kOe, 2—$H = 85$ kOe, 3—$\mathbf{H}\parallel\langle 1\bar{2}10\rangle$, 4—$\mathbf{H}\parallel\langle 10\bar{1}0\rangle$.

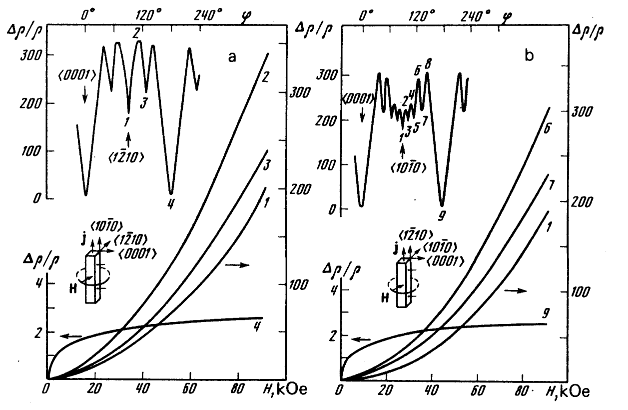

FIG. 9. Anisotropy at $H = 90$ kOe and field dependences of the transverse magnetoresistance for the samples: a) $\mathbf{j}\parallel\langle 10\bar{1}0\rangle$, b) $\mathbf{j}\parallel\langle 1\bar{2}10\rangle$.

tudes of the oscillations, and to study their anisotropy.

b) *Oscillations of the magnetoresistance.* From the frequencies F of the oscillations—see Fig. 10—it is seen that for the sample with the current orientation $\mathbf{j} \parallel \langle 10\bar{1}0\rangle$ at $\mathbf{H} \parallel \langle 1\bar{2}10\rangle$ the oscillating contribution is formed by trajectories that lie in the plane *ALM* (Fig. 3, Sec. 1), while for the sample with $\mathbf{j} \parallel \langle 1\bar{2}10\rangle$ at $\mathbf{H} \parallel \langle 10\bar{1}0\rangle$ the magnetic breakdown trajectories lie in the plane *LMML*, which is perpendicular to the $\langle 10\bar{1}0\rangle$ axis (Fig. 3, section 3). The frequency anisotropy of the oscillations observed for these samples at other directions of the magnetic field is also determined by the corresponding sections of the hole lens *L7h* and agrees well with the dHvA frequencies.[3]

The anisotropy of the amplitudes of the oscillations $\Delta\rho_{osc}/\rho\ (H=0) = f(\varphi)$ of both samples (Figs. 10a and 10b) is quite strong and is characterized by strongly pronounced maxima. At the minima of the dependence of $\Delta\rho_{osc}/\rho\ (H=0)$ on φ in the angle interval $0 < \varphi < 60°$ the oscillations do not vanish and can be observed in experiment. In the interval $60° < \varphi < 90°$, no oscillating contribution was observed at the sensitivity of the installation.

As can be seen from the figures, the plots of $\Delta\rho_{osc}/\rho\ (H=0) = f(\varphi)$ for samples with different orientations differ substantially. Thus, for the sample with $\mathbf{j} \parallel \langle 10\bar{1}0\rangle$ (Fig. 10a) there are two maxima of the oscillation amplitudes, and the interesting fact in this case is that the position of the outer maximum $\varphi = 34°$ corresponds to that magnetic-field direction at which the magnetic breakdown trajectory of type C should be realized (Fig. 2, $\varphi = 35°$). At $\mathbf{H} \parallel \langle 1\bar{2}10\rangle$ magnetic breakdwon of type B is realized, as is evidenced by the rather large amplitude of the oscillations. However, as seen from Fig. 10a, when **H** is inclined away from the $\langle 1\bar{2}10\rangle$ axis the amplitude increases and $\varphi \sim 5°$ a maximum is observed. This experimental fact becomes understandable upon examination of Fig. 2, $\varphi = 5°$, from which it follows that when **H** is inclined away from the $\langle 1\bar{2}10\rangle$ axis the magnetic breakdown likewise produces closed trajectories, but elongated ones that pass through several Brillouin zones. It is seen that this is precisely the reason for the increase of the oscillation amplitude.

On the curve for the sample with $\mathbf{j} \parallel \langle 1\bar{2}10\rangle$ (Fig. 10b) there are three amplitude maxima, and just as for the sample with $\mathbf{j} \parallel \langle 10\bar{1}0\rangle$, the two outer maxima, $\varphi = 20°$ and $\varphi = 42°$, correspond to magnetic breakdown situations of type C (Fig. 3, $\varphi = 21°$ and $\varphi = 43°$), while the maximum at $\varphi \sim 5°$ is of the same origin as the similar maximum of the sample with $\mathbf{j} \parallel \langle 10\bar{1}0\rangle$.

The minima of the amplitudes of the oscillations correspond to magnetic field directions such that the magnetic breakdown does not lead to substantial changes in the configuration of the trajectories (see, e.g., Fig. 4, $\varphi = 12°$).

c) *Hall effect.* Measurements of the anisotropy and of the field dependences of the Hall coefficient on samples with $\mathbf{j} \parallel \langle 10\bar{1}0\rangle$ and $\mathbf{j} \parallel \langle 1\bar{2}10\rangle$ have shown that, just as for the sample with $\mathbf{j} \parallel \langle 0001\rangle$, R reverses sign and becomes negative on going over to strong effective magnetic fields. In accordance with Ref. 6, this is observed at all directions of **j** and **H** relative to the crystal.

It follows from the measurements that when the magnetic field is directed along the $\langle 1\bar{2}10\rangle$ axis the Hall coefficient depends on the direction of the electric current **j**. Thus, for the samples with $\mathbf{j} \parallel \langle 0001\rangle$ and $\mathbf{j} \parallel \langle 10\bar{1}0\rangle$ at $H = 90$ kOe the values of R are respectively -1.99×10^{-3} and -1.34×10^{-3} cgs emu. In case (c) of Sec. 3.1, the increase of R at $\mathbf{H} \parallel \langle 1\bar{2}10\rangle$ (curve 3 on Fig. 8) was due to formation of a magnetic-breakdown σ layer of open trajectories. However, in the measurements at $\mathbf{H} \parallel \langle 10\bar{1}0\rangle$ on the sample with $\mathbf{j} \parallel \langle 1\bar{2}10\rangle$, as well as on the sample $\mathbf{j} \parallel \langle 0001\rangle$ (curve 4 of Fig. 8), the $R(H)$ field dependences saturate even in weak magnetic fields, and have no anomalies with further increase of H. This indicates that magnetic breakdown of type B has practically no effect on the Hall coefficient, i.e., it is not accompanied by magnetic-breakdown decompensation. In contrast to weak magnetic fields,[6] the anisotropy of the Hall coefficient in ruthenium in strong magnetic fields changes substantially only at $\mathbf{H} \parallel \langle 1\bar{2}10\rangle$, i.e., under conditions when magnetic breakdown of type A takes place.

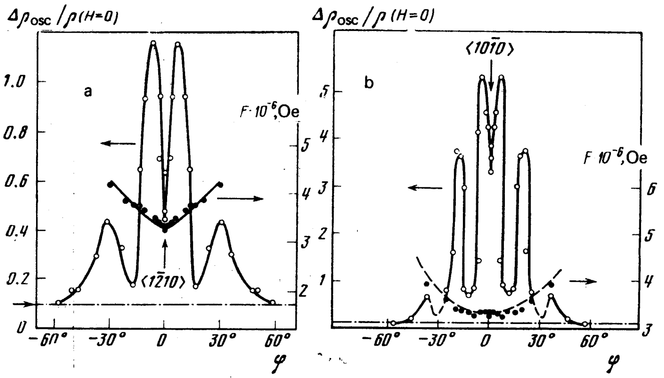

FIG. 10. Anisotropy of the amplitudes ($H = 85$ kOe) and the frequencies F of the oscillations of the magnetoresistance for the samples with $\mathbf{j} \parallel \langle 10\bar{1}0\rangle$ (a) and $\mathbf{j} \parallel \langle 1\bar{2}10\rangle$ (b); in the latter case the dashed lines show the dHvA frequencies from Ref. 3. The dash-dot lines show the noise level.

We note also that our present measurements in strong magnetic fields have confirmed the validity of the conclusions[6] that at $\mathbf{H} \parallel \langle 0001\rangle$ on the multiply connected hole sheet *KM8h* there is realized a thick β layer of closed orbits of electronic character (Fig. 1a). Measurement of the Hall coefficient made it possible to determine the thickness d_β of this layer, namely[18,19]

$$d_\beta = 4\pi^3/SRe = 0.78 \pm 0.08\ \text{Å}^{-1},$$

where S is the area of the intersection of the Brillouin zone with the plane normal to **H** and is equal to 6.24 Å^{-2}, e is the electron charge, and $R = 1.59 \cdot 10^{-3}$ cgs emu. The obtained value of d_β agrees well with the dimension of the multiply connected hole sheet in the direction *HKH* (0.8 Å^{-1}), obtained from measurements of the dHvA effect.[3]

4. DISCUSSION

A. Open magnetic-breakdown trajectories

The results of the measurements of the galvanomagnetic properties of the ruthenium sample with current orientation along the ⟨0001⟩ axis have shown that in strong magnetic fields at H || ⟨1$\bar{2}$10⟩ a narrow minimum appears on the rotation diagram of the magnetoresistance, and the field dependence of $\Delta\rho/\rho$ tends to saturate and is accompanied by oscillations. At this direction of the magnetic field, corresponding anomalies are observed also in the Hall effect. These singularities exist only at H || ⟨1$\bar{2}$10⟩ and point, in our opinion, to the formation of open magnetic-breakdown σ trajectories in the basal plane.

It is suggested in Ref. 9 that these singularities are not connected with the onset of a layer of open magnetic breakdown trajectories, but are the result of the magnetic-breakdown "decompensation" that appears when α-layer trajectories that are open along the hexagonal axis (Figs. 1, 2, 4) are transformed into closed ones (magnetic-breakdown situation B). However, the magnetic-breakdown decompensation hypothesis does not explain the entire aggregate of the results of the measurements of the galvanomagnetic properties, for the following reasons.

First, if the magnetic-breakdown decompensation actually takes place at H || ⟨1$\bar{2}$10⟩, then it should occur also at H || ⟨10$\bar{1}$0⟩, inasmuch as at the latter orientation, as shown by measurements of the magnetoresistance of a sample with j || ⟨1$\bar{2}$10⟩ (Fig. 9b), magnetic breakdown also transforms open trajectories of the α layer into closed ones. But in this direction of the magnetic field no anomalies similar to those which occur for the sample with j || ⟨0001⟩ at H || ⟨1$\bar{2}$10⟩ are observed in either the magnetoresistance, (Fig. 6 and Fig. 9b) or in the Hall effect (Fig. 8), for the sample with either j || ⟨0001⟩ or j || ⟨1$\bar{2}$10⟩.

Second, in contrast to the open trajectories, the decompensation effects due to closed trajectories should not be sensitive to the current orientation. However, at one and the same direction of the magnetic field H || ⟨1$\bar{2}$10⟩, in measurements made on the sample with j || ⟨0001⟩ (Fig. 6b), saturation of the magnetoresistance is observed, while for the sample with j || ⟨10$\bar{1}$0⟩ the magnetoresistance increases quadratically (Fig. 9a). The assumption of a magnetic-breakdown layer of open σ trajectories in the basal plane explains these experimental facts and agrees well with the current diagram $\rho(H) = AH^2\cos^2\alpha$ (where α is the angle between the directions of the electric current and the direction of the opening in **k** space), inasmuch as $\alpha = \pi/2$ for j || ⟨0001⟩ and $\alpha = 0$ for j || ⟨10$\bar{1}$0⟩. At H || ⟨10$\bar{1}$0⟩ no open magnetic breakdown trajectory can be realized (see Fig. 3). The effects indicated above are likewise absent at this direction of the magnetic field.

Third, the magnetic-breakdown decompensation hypothesis contradicts the results of measurements of dHvA effect[3]: if such a decompensation were to occur at H || ⟨1$\bar{2}$10⟩, then the oscillations of the magnetoresistance in this direction of H should, in accordance with Ref. 3, have a frequency 3.2×10^6 Oe (Fig. 4, lens section 1). However, the frequencies observed in experiment at H || ⟨1$\bar{2}$10⟩ amount to $(4.10 \pm 0.05) \cdot 10^6$ Oe for the magnetoresistance, $(4.05 \pm 0.05) \cdot 10^6$ Oe for the thermoelectric power,[2,9,10] and $(4.05 \pm 0.05) \cdot 10^6$ Oe for the Nerst emf.[10] These values agree well with the dHvA frequency 4.0×10^6 Oe, which corresponds to the area of the extremal intersection of the lens with the *LMML* plane (Sec. 2 on Fig. 3).

These experimental facts show that the magnetic breakdown in ruthenium at H || ⟨1$\bar{2}$10⟩ leads to open σ trajectories located in the *LMML* plane and perpendicular to the ⟨0001⟩ direction. The anisotropy of the oscillation amplitude (Fig. 6b) shows that the layer of the open trajectories is quite narrow.

The narrowness of the magnetic breakdown layer is evidenced also by an estimate of its relative width $\delta k_z / k_F$. Since the half-widths of the minimum on the $\Delta\rho/\rho = f(\varphi)$ curve (Fig. 6a) and of the maximum on the dependence of $\Delta\rho_{osc}/\rho$ $(H=0)$ on φ (Fig. 6b) are not sensitive to the magnetic field or to the purity of the sample (analogous measurements were made by us on a sample with $\rho_{273.2K}/\rho_{4.2K} = 1200$), the angular dimension of the singularity does not depend on the mean free path of the electrons and is determined only by the topology of the Fermi surface. Then $\tan\Delta\varphi = \delta k_z / LL$, where $\Delta\varphi \approx 1.5°$ is the half-width of the burst of the oscillation amplitude (Fig. 6b), δk_z is the width of the layer of the open magnetic breakdown trajectories, and $LL = \Gamma M = 0.68\Gamma K$ is the distance between the lenses and the ⟨10$\bar{1}$0⟩ direction of the *LMML* plane. According to this estimate, δk_z amounts to $0.014\Gamma K$, i.e., $\delta k_z / k_F \sim 2 \cdot 10^{-2}$.

B. Closed magnetic breakdown trajectories

As was already shown (Fig. 2, $\varphi = 0$ and Fig. 4, $\varphi = 0$) magnetic breakdown in ruthenium can cause the α trajectories that are open along ⟨0001⟩ to become closed. In our experiments the influence of such a magnetic breakdown on the magnetoresistance was observed on samples with orientations j || ⟨1$\bar{2}$10⟩ and j || ⟨10$\bar{1}$0⟩. The results of the measurements [case (b) of Sec. 3.2] have shown that in strong magnetic fields at the directions H || ⟨10$\bar{1}$0⟩ and H || ⟨1$\bar{2}$10⟩ there is no thick α layer of trajectories open along ⟨0001⟩, and the galvanomagnetic properties have a behavior typical of closed trajectories corresponding to magnetic breakdown of type B.

A distinguishing feature of this magnetic breakdown situation in ruthenium is, first, that the energy gap between the neck and the lens in anisotropic: it is maximal in *LH* direction of the Brillouin zone and is equal to zero ("conical" tangency point[3]) along *LA* (Figs. 2 and 3). Consequently, on the noncentral sections parallel to the planes *ALM* (H || ⟨1$\bar{2}$10⟩ and *AHK* (H || ⟨10$\bar{1}$0⟩, the gap increases with increasing k_z measured from the point *L*. By virtue of this, the width of the layer of the "post-breakdown" closed trajectories increases with increasing magnetic field. Second, not all the trajectories of the α layer can be transformed into closed ones as a result of magnetic breakdown. In the "post-breakdown" region of magnetic fields there should remain a very narrow γ layer of trajectories open along ⟨0001⟩

(Figs. 1 and 2). The possible existence of such a layer is evidenced by the sharp minima of the magnetoresistance at H || ⟨10$\bar{1}$0⟩ and H || ⟨1$\bar{2}$10⟩, designated by the number 1 on Fig. 9, which do not vanish even in strong fields ~100 kOe.

The magnetoresistance oscillations observed at H || ⟨10$\bar{1}$0⟩, j || ⟨1$\bar{2}$10⟩ and at H || ⟨1$\bar{2}$10⟩, j || ⟨10$\bar{1}$0⟩ are of magnetic-breakdown origin, and are not ordinary Shubnikov–de Haas oscillations, which are not accompanied by a change of the configuration of the electron trajectories. This is indicated by the aggregate of the experimental results.

First, the frequencies of the oscillations correspond to the extremal intersections of the lens by the planes *LMML* at H || ⟨10$\bar{1}$0⟩ and *ALM* at H || ⟨1$\bar{2}$10⟩, i.e., precisely those intersections in which MP trajectories of type B should be located (sections 1 and 3 on Fig. 3). Second, we observed no oscillating contributions containing two frequencies that would stem in the case of the Shubnikov–de Haas effect from different extremal sections of the lenses at a given direction of the magnetic field, as is the situation in the case of the dHvA effect.[3] Third, the amplitude of the observed oscillations has a sharply pronounced anisotropy (Fig. 10), which cannot be explained by means of the Shubnikov–de Haas effect.

Thus, the observed oscillations of the magnetoresistance are of magnetic-breakdown origin.

C. Magnetic breakdown "space net"

As already shown in Sec. 3.2, the frequency of the oscillations of the magnetoresistance in the samples with j || ⟨1$\bar{2}$10⟩ and j || ⟨10$\bar{1}$0⟩ (Fig. 10) agrees with the frequencies of the oscillations of the dHvA effect,[3] and the positions of the maxima of the amplitudes of the oscillations correspond to the directions of the magnetic field, at which the magnetic breakdown can produce elongated closed trajectories (B) or a space net (C). However, the amplitude of the oscillations at H || ⟨1$\bar{2}$10⟩ (Fig. 10a) is smaller by one order of magnitude than at H || ⟨10$\bar{1}$0⟩ (Fig. 10b). This fact shows directly that the point of contact between the lens and the multiply connected hole sheet *KM*8*h* is a conical point (Fig. 11). In fact, at H || ⟨1$\bar{2}$10⟩ the areas of the lens sections change strongly in the k_z direction and the oscillating signal averages out, inasmuch as

$$\sigma_{osc} \sim \int \exp(icS_L(k_z)/e\hbar H)\, dk_z,$$

where σ_{osc} is the oscillating contribution to the conductivity, and $S_L(k_z)$ is the lens cross section area. At H || ⟨10$\bar{1}$0⟩ there is a relatively broad layer of extremal sections with equal areas.

This conclusion is confirmed also by the entire picture of the anisotropy of the amplitudes of the oscillations of the magnetoresistance of samples with j || ⟨10$\bar{1}$0⟩ and j || ⟨1$\bar{2}$10⟩. In the former case, at any direction of H, the magnetic-breakdown trajectories always pass through conical points (Fig. 11, H || ⟨1$\bar{2}$10⟩), and in the latter the situation is similar to the case H || ⟨10$\bar{1}$0⟩. This interpretation is confirmed also by results of measurements of the thermoelectric power.[2]

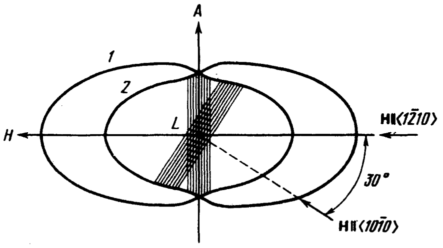

FIG. 11. Section of multiply connected hole sheet *KM*8*h* (1) and of the lens *L*7*h* (2) by the plane *ALH* and in the region of the conical points.

It should be noted that the observed oscillations of the magnetoresistance, in the case when closed trajectories change into magnetic breakdown trajectories of the space-net type, is a rather interesting experimental fact. The point is that in measurements on other metals in similar situations, no oscillations were observed and at the present time there are no theoretical estimates of the amplitude of oscillations of this type.

Thus, the aggregate of the results of the performed measurements cannot be explained without taking magnetic breakdown into account. The presence of magnetic breakdown in ruthenium is evidenced by the following experimental facts:

a) the strong anisotropy of the amplitudes of the oscillations of the magnetoresistance, thermoelectric power,[2,9,10] and Nernst effect,[10]

b) the correspondence of the frequencies of the observed oscillations to precisely those sections of the *L*7*h* lens which take part in the formation of the magnetic breakdown trajectories;

c) the strong deviations from the Kohler rule[6] and the change of the form of the field dependences of the magnetoresistance and of the Hall emf, which take place only at those magnetic field directions at which a radical magnetic breakdown restructuring of the electron trajectories is possible.

The existence of magnetic breakdown in ruthenium is confirmed also by the dHvA frequencies[3] observed in strong magnetic fields and due to closed trajectories of type B.

In conclusion we note the main result obtained in the investigation of the galvanomagnetic properties of ruthenium in strong magnetic fields, where the influence of the magnetic breakdown on these properties is substantial.

1) The behavior of the galvanomagnetic properties is determined mainly by the multiply connected hole sheet *KM*8*h*. Its topological singularities and the structure of the electron spectrum of ruthenium create possibilities for such effects as geometrical decompensation and magnetic breakdown. Three types of electron trajectories are realized in magnetic breakdown: A—closed trajectories are transformed into a layer of open magnetic breakdown trajectories; B—the open trajectories

that are open along ⟨0001⟩ are transformed into closed ones; C—a space net of magnetic-breakdown trajectories is realized.

2) All three types of magnetic breakdown are accompanied by quantum oscillations of the magnetoresistance.

3) Magnetic breakdown in ruthenium leads to formation of a rather narrow layer of open trajectories with relative width $\delta k_z/k_F \sim 2 \cdot 10^{-2}$. These circumstances make ruthenium a promising object for the search for nonlinear effects theoretically predicted in Ref. 1.

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1) In the designation of the sheets, the first letter denotes the symmetry point of the Brillouin zone at which the given sheet is centered (two letters denote the direction in the zone); the number corresponds to the number of the band (zone) to which the given sheet belongs; the letters *e* and *h* designate an electron or hole sheet.

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