GALVANOMAGNETIC PROPERTIES OF MOLYBDENUM CRYSTALS IN STRONG EFFECTIVE FIELDS

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The anisotropy of the galvanomagnetic properties (Hall effect, which is even with respect to a magnetic field of transverse voltage and magnetoresistance) of a high purity single crystal is investigated at helium and hydrogen temperatures in a transverse magnetic field up to 24 kOe. The results are discussed within the framework of the theory of galvanomagnetic phenomena.[1,2]

THE theory of galvanomagnetic phenomena developed in the papers of I. M. Lifshitz et al.[1,2] relates the galvanomagnetic properties of metals, within the limits of large effective fields ($lr \gg 1$; l—mean free path, r—radius of curvature of the electron trajectory in the magnetic field), with the concrete topological features of the Fermi surface for two possible cases: 1) the number of holes is equal to the number of electrons, and 2) the two numbers are not equal.

At the present time the anisotropy of the magnetoresistance and its field dependences have been studied sufficiently thoroughly for a number of metals, including molybdenum.[3,4] However, a rather limited number of investigations have been devoted to the even transverse voltage produced in a plane perpendicular to the current and not reversing sign when the magnetic field direction is reversed, or to the Hall effect; this is connected in part with experimental difficulties in measuring these quantities. The transverse even voltage was observed in the investigation of the galvanomagnetic properties of only four metals: tin,[5] copper,[6] gallium,[7] and tungsten.[8] In [6], the appearance of the transverse even voltage is related to the occurrence of a layer of open trajectories, while in [5] and [8] it is related to the existence of elongated carrier trajectories.

It is of interest to investigate the galvanomagnetic properties of a molybdenum crystal in strong effective fields, particularly to investigate the anisotropy of the transverse even voltage, the dependence of its magnitude on the magnetic field, the anisotropy of the Hall field, and the temperature dependence of the Hall "constant." No such measurements have been made so far for single crystals of molybdenum.

On the basis of investigations of the Fermi surface of molybdenum it can be assumed that this metal has apparently the same number of holes as electrons, and has a closed Fermi surface consisting of electron and hole sheets (see, for example, [1,10]). This was confirmed also by the measurements of the magnetoresistance reported in this paper. The theory of galvanomagnetic phenomena[1,2] predicts for such a metal, in the limit of large effective fields, the appearance of a transverse even voltage and a quadratic growth of the magnetoresistance and transverse even voltage with increasing magnetic field, as well as a linear growth of the Hall field.

In the general anisotropic case, the Hall-field vector $\mathbf{E}_h$ and the even transverse voltage vector $\mathbf{E}_q$ may not be perpendicular to the magnetic field H.[11] As noted in [7], for crystals oriented such that the current is exactly parallel to the crystallographic axis of sufficiently high symmetry (for example, for a cubic crystal, $\mathbf{j} \parallel \langle 100 \rangle$) and the magnetic field lies in a plane perpendicular to the current, the crystal symmetry requires that the transverse even voltage vanish. This was confirmed experimentally.[5]

Thus, a transverse even voltage should be observed in an arbitrarily oriented (relative to the current) sufficiently pure molybdenum crystal placed in a magnetic field.

SAMPLES, MEASUREMENT PROCEDURE

The measurements of the anisotropy and of the field dependences of the magnetoresistances were made on two single-crystal samples of molybdenum obtained by crucible-less zone melting where the zone was heated by electron bombardment[1).][12] One of them was in the form of a cylinder of 3 mm diameter and 40 mm length, with the sample axis making an angle of 35° with the [001] direction, 10° with [011], and 37° with [111]; the second was in the form of a bar with square cross section, 1.8 mm thick and 25 mm long, and a crystallographic orientation such that its axis made an angle 34° with [001], 16° with [011], and 27° with [111]. The distance between the potential contacts was 13 or 9 mm. The samples were cut by an electric-spark method and had a resistance ratio R(300° K)/R(4.2° K) of approximately 3600 and 5200 respectively.

The Hall effect and the even transverse voltage were measured in the second sample. The potential contacts were made of a thin platinum wire and were spark-welded. The arrangement of the contacts is shown in Fig. 1. Contacts 1–4 should lie in the same plane, which is perpendicular to the direction of the current. Since the magnetoresistance effect is many times larger than the transverse even voltage effect, a slight deviation from the correct contact arrangement leads to a large error in the determination of the even voltage. In the determination of E_q we eliminated the contribution due to the magnetoresistance to the transverse voltage, which is even with respect to the magnetic field, and which is produced across contacts 1–2 and 3–4.

In analogy with the procedure used in [5, 11], we determined the Hall-field vector $\mathbf{E}_h$ from its projections $E_h^{1,2}$ and $E_h^{3,4}$, where

$$E_h^{1,2} = \frac{1}{d}\frac{V_{1,2}(H) - V_{1,2}(-H)}{2},$$

$$E_h^{3,4} = \frac{1}{d}\frac{V_{3,4}(H) - V_{3,4}(-H)}{2};$$

V(H) and V(−H) are the voltages on the corresponding contacts for two opposite directions of the magnetic field, and d is the transverse dimension of the sample. The vector of the even transverse voltage field $\mathbf{E}_q$ was constructed from its projections

$$E_q^{1,2} = \frac{1}{d}\frac{V_{1,2}(H) + V_{1,2}(-H)}{2},$$

$$E_q^{3,4} = \frac{1}{d}\frac{V_{3,4}(H) + V_{3,4}(-H)}{2}.$$

The resultant thermal emf was eliminated by commutation of the current.

In measuring the temperature dependence of the Hall effect, the temperature was varied by pumping on vapor above the liquid helium and hydrogen, and was determined from the vapor pressure. Above 20.4° K, the required temperature was set with the aid of a cryostat and measured with a copper-constantan thermocouple. Measurements of the voltage cross contacts 1–2, 3–4, and 5–6 were with a potentiometer. The sensitivity of the apparatus was 1.5×10^{-8} V/mm, and where necessary a photoelectric amplifier type F-117/2 was used, which raised the sensitivity to 1×10^{-9} V/mm. The crystallographic orientation of the samples was determined by x-ray diffraction.

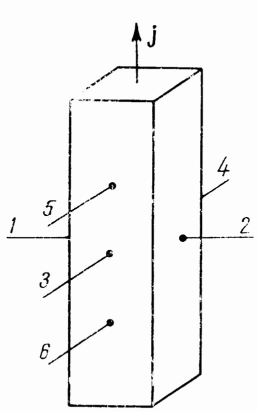

FIG. 1. Arrangement of the potential contacts on the sample. Contacts 1–4 serve to measure the Hall effect and the transverse even voltage, while 5 and 6 are to measure the magnetoresistance.

MEASUREMENT RESULTS, DISCUSSION

We measured the Hall effect, the even transverse voltage, and the magnetoresistance of the molybdenum single crystal in a transverse magnetic field up to 25 kOe, at temperatures 1.8, 4.2, 13.9, 16.6, and 20.4° K, corresponding to an effective field of the order of 10^8–5×10^7 Oe. Figure 2 shows the rotation diagrams of $\Delta R/R$ and of the projections of the field of the even transverse voltage on the direction of the magnetic field and the normal to it (E_{qz} and E_{qy}) relative to the density of the measuring current, for a sample with a ratio R(300° K)/R(4.2° K) ≅ 5200. It is seen from this figure that the vector E_q is not perpendicular to the magnetic field, but makes a certain angle smaller than 90°, and varies with the direction of the magnetic field.

The rotation diagrams for the Hall-field projections E_{hy} and E_{hz} are shown in Fig. 3. The deviation of the vector $\mathbf{E}_h$ from the normal to the magnetic field reaches 20°. Similar deviations were observed in tin[5, 11] and in indium.[11] The angular dependences of E_{qy}, E_{qz}, E_{hy}, and E_{hz}, measured at the other indicated temperatures, are analogous to those given above.

[1)]We take the opportunity to thank L. S. Starostina for the samples and A. A. Kralina and T. V. Ushkova for determining the crystallographic orientation.

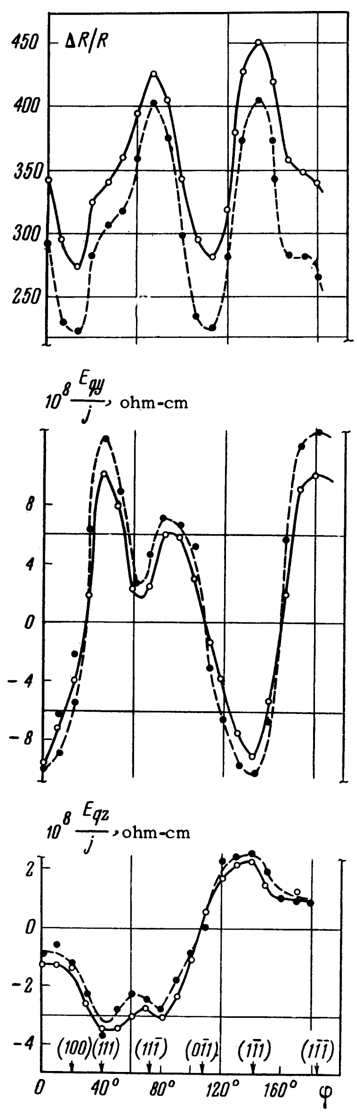

FIG. 2. Rotation diagrams, measured at helium and hydrogen temperatures, of the magnetoresistance in an identical effective field H[R (273.2°K)/R (t) = 5 – 10^7 Oe], and the projection of the magnetic field of the transverse even voltage E_{qy}/j and E_{qz}/j for identical value of H[R(273.2°K)/I(T)]$^{1/2}$ = 1 × 10^6 Oe; ○–T = 4.2°K, ●–T = 20.4°K.

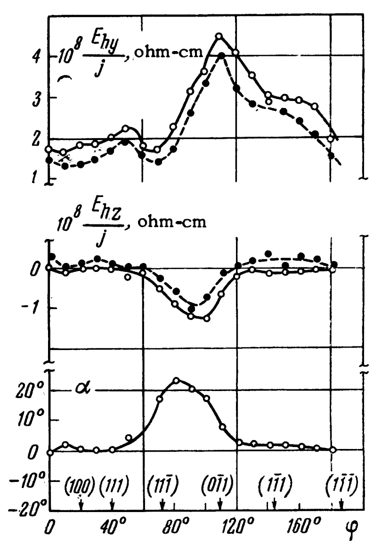

FIG. 3. Rotation diagram of the projections of the Hall field E_{hy}/j and E_{hz}/j at T = 4.2°K (○) and T = 20.4°K (●), and dependence of the deviation of the Hall field α from the normal to the magnetic field on the orientation of the magnetic field H = 15 kOe.

Plots of ΔR/R and of E_q and E_k against the magnetic field are shown in Fig. 4 for two different orientations of the field relative to the crystallographic axes of the sample. As seen from this figure, ΔR/R and E_q vary quadratically with changing magnetic field, and E_h varies linearly, as predicted by the theory[1,2] for compensated metals with a closed Fermi surface. At other directions of the magnetic field, the character of these plots remains the same.

1. Thus, as expected, a transverse-voltage electric field which is even with respect to the magnetic field is produced by current in a molybdenum single crystal placed in a magnetic field, and is of the same order of magnitude as the Hall field.

In [5,8], the occurrence of an even voltage is related to the existence of elongated closed carrier orbits. From this point of view, the presence of an even voltage in the molybdenum crystal can be attributed to the appreciable anisotropy of its Fermi surface. However, a discussion of the connection between the anisotropy of the even trans-

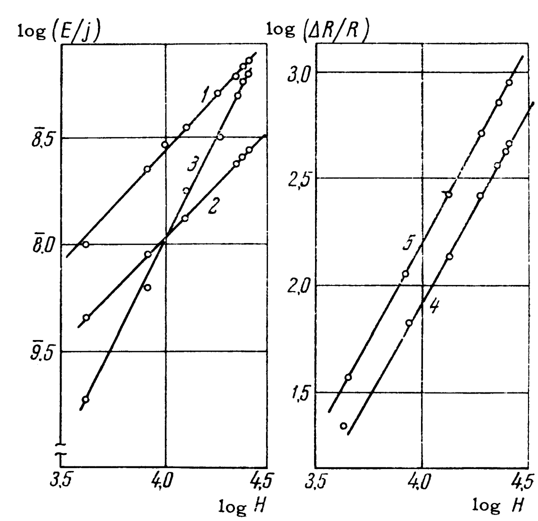

FIG. 4. Absolute value of the Hall field (curve 1, φ = 105° and curve 2, φ = 70°), the even transverse voltage (curve 3, φ = 70°), and the magnetoresistance (curve 4, φ = 105° and curve 5, φ = 70°) against the magnetic field at 20.4°K.

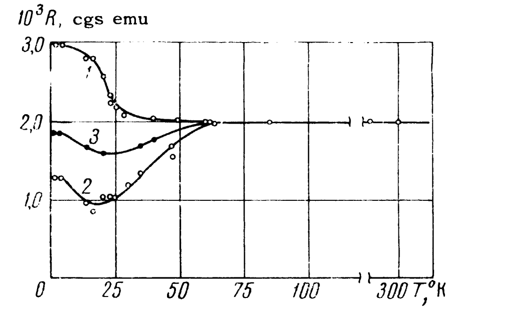

FIG. 5. Temperature dependence of the Hall "constant" of single-crystal molybdenum: 1—magnetic field lies in the $(\bar{0}11)$ plane (φ = 180°), 2—magnetic field in $(1\bar{1}\bar{1})$ plane (φ = 72°), 3—values obtained by averaging the Hall "constant" over all directions of the magnetic field.

verse voltage in molybdenum with the details of the form of its Fermi surface is premature at present, owing to the limited amount of experimental data.

It must be noted that, unlike the Hall field, the transverse even voltage, and the magnetoresistance as well, decreases rapidly with increasing temperature. The values of the even voltage, measured at a definite orientation of the magnetic field in the same effective field, but at different temperatures (4.2 and 20.4° K), are not the same, nor are the values of the magnetoresistance. The influence of the temperature and of the magnetic field will be considered in detail separately.

2. It is shown in [1] that, for arbitrary conduction-electron dispersion, the anisotropy of the Hall effect depends in the complicated manner on the form of the collision integral and on the dispersion law. However, in the case when the holes and electrons are equal in number ($n_e = n_h = n$), the Hall effect will obviously be a maximum for those directions of the magnetic field for which the mobility of the electrons and the holes differ most.

Measurement of the Hall effect show that the effect is largest when the magnetic field coincides with the (011) plane, and comes closest to the [100] axis than in all possible cases (Fig. 3). At $\varphi = 108°$ the field H makes an angle of 10° with the [100] axis.

Quite independently of this experiment, it follows from the Larmor model for the Fermi surface of molybdenum[9,10] that it is precisely at this direction of the magnetic field (for a given sample orientation) that the electron orbits differ most from the Hall orbit in their configuration and dimensions, and this proves to some degree the differences in the mobilities. This assumption confirms the results of experiments aimed at the observation of cyclotron resonance on molybdenum,[13] where a strong increase of the effective carrier masses was observed when the direction of **H** approached the vicinity of axis of the type ⟨100⟩, and also measurements of the anisotropy of the magnetoresistance. Thus, the increase in the Hall effect of molybdenum, when the magnetic field comes sufficiently close to the [100] axis, can be qualitatively related with details in the form of the Fermi surface of this metal, namely to the occurrence of a thick layer of electron orbits which differ strongly from the hole orbits.

The temperature dependence of the Hall "constant" is shown in Fig. 5. The anisotropy of the effect comes into play near 60° K and increases with decreasing temperature. If we average the Hall "constant" over the angle (middle curve), then a rather weak temperature dependence of this average quantity is observed. The Hall "constant" is positive. This means (as can be readily seen from the expression for the Hall "constant" obtained for the case $n_e = n_h$ and the approximation of quadratic anisotropic dispersion (see [1]), that the average hole mobility in the molybdenum is larger than the average mobility of the conduction electrons.

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