Anisotropy of the properties of single crystals of ternary chalcogenides of molybdenum

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The critical magnetic fields and the anisotropy of the magnetoresistance of the single crystals $Pb_2Mo_6S_{7.5}$, $Sn_2Mo_6S_{7.5}$, and $Cu_{1.9}Mo_6S_8$ have been measured. Analysis of the results suggests a particular shape of the Fermi surface.

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The most important properties of superconducting ternary chalcogenides of molybdenum—the exceedingly high critical magnetic fields and the high critical current densities, which have been observed in samples of ternary chalcogenides of molybdenum with lead—still await a consistent, convincing explanation.[1] Extremely important relative data can be found by studying the properties of ternary chalcogenides of molybdenum in the form of single cystals. Such a study can yield information about the topology of the Fermi surface of these compounds. In this letter we are reporting measurements of the critical magnetic fields H_{c2} and of the magnetoresistance $\rho(H)$ of ternary chalcogenides of molybdenum with lead, tin, and copper as the third component.

The single crystals were grown from the melt in a chamber filled with pure argon at a pressure of 170–180 atm. During the growth, the sample temperature was raised to 1900–2000 °C and held there for 1 to 3 h; then the temperature was reduced at a rate from 1 to 3 deg/min. The phase composition of the resulting single crystals was determined with an x-ray diffraction camera. In addition to the principal lines corresponding to the Chevrel phase, the diffraction patterns reveal faint lines corresponding to the free third component for the compounds with lead and tin, and they reveal lines of MoS_2 for the compound with copper. The basic superconducting characteristics (T_c and dH_c/dT) of these crystals agree well with data in the literature. The width of the transition to the superconducting state is 0.5–1 K; above the transition, the resistance increases linearly with the temperature.[1] The ratios of the resistance at $T = 300$ K to the resistance extrapolated to 0 K are 60, 10, and 15 for $Sn_2Mo_6S_{7.5}$, $Pb_2Mo_6S_{7.5}$, and $Cu_{1.9}Mo_6S_8$, respectively. Samples used for measurements of the resistance, with a cross section from 0.5×0.5 to 1×1 mm and a length of 5–10 mm, were cut by an electric-arc method from bars oriented by the method inverse x-ray diffraction. In this letter we are reporting measurements carried out for nine samples of various compositions in various orientations.

A superconducting solenoid produced a magnetic field up to 150 kOe. The current in the sample flowed perpendicular to the magnetic field in all cases. To measure the anisotropy of H_{c2} and $\rho(H)$ over the temperature range 5–15 K, we used the apparatus of Ref. 2, supplemented with a copper block with a heater and thermom-

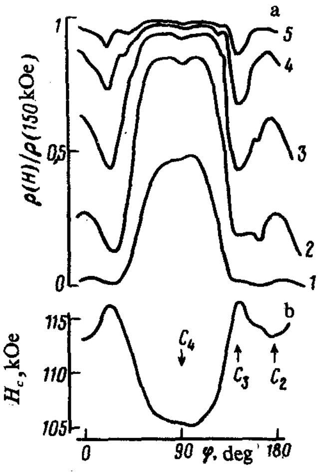

FIG. 1. a—Angular dependence of the resistance of a $Cu_{1.9}Mo_6S_8$ sample oriented along the C_2 direction near the S–N transition at $T = 4.2$ K. 1) $H = 105$; 2) 110; 3) 115; 4) 120; 5) 125 Oe. b—Anisotropy of the critical field.

eters. The temperature was regulated at the specified level within 0.02 K. The measurements were carried out at $J \approx 1$–10 mA to avoid an effect of the measurement current on the results for H_{c2}.

The critical magnetic fields mesured for the $Cu_{1.9}Mo_6S_8$ samples depended on the direction of the field with respect to the crystallographic axes. The magnitude of this anisotropy reached 8 kOe at $T = 4.2$ K. Since the width of the transition curve is 10–20 kOe, depending on the temperature, and is always greater than the anisotropy of H_{c2}, we found it convenient to determine the anisotropy of H_{c2} from the curves of $\rho(\phi)$ recorded near the S—N transition, i.e., the transition to the normal state. Figure 1b shows some recordings of this type, for a sample with axis along the C_2 direction; Fig. 1b shows the corresponding $H_{c2}(\phi)$ dependence. As H_{c2} we adopted that magnetic field at which the resistance of the sample is equal to half the resistance in the normal state. We see from these results that $H_{c2}(\phi)$ has maxima at $\mathbf{H} \| C_3$. A similar dependence was found in a sample in the C_2 orientation, prepared by a chemical-transport method.[1)]

The angular dependence of the resistance in the magnetic fields corresponding to the vicinity of the transition to the normal state was also recorded for $Pb_2Mo_6S_{7.5}$ and $Sn_2Mo_6S_{7.5}$ samples in various orientations. It turned out that the signal from these samples depends only slightly on the direction of the magnetic field and that the possible anisotropy of the critical magnetic field is less than 3%.

We studied the magnetoresistance in the normal state at both $T > T_c$ and $T < T_c$ (in the latter case, with $H > H_{c2}$). The magnetoresistance of the $Sn_2Mo_6S_{7.5}$ and $Cu_{1.9}Mo_6S_8$ samples in fields above 30 kOe varied linearly with the field. The magnetoresistance of the $Pb_2Mo_6S_{7.5}$ was slight in fields up to 150 kOe and varied in proportion to H^2. This quadratic dependence of the magnetoresistance corresponds to the region of weak effective magnetic fields, apparently a result of the low ratio of the resistance of the $Pb_2Mo_6S_{7.5}$ samples.

Figure 2 shows the angular dependence of the resistance of $Sn_2Mo_6S_{7.5}$ sample for the case in which the rotation axis makes an angle of 10–15° with the C_3 axis. These

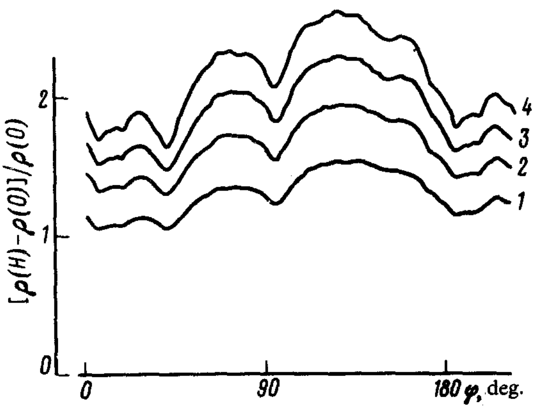

FIG. 2. Magnetoresistance rosettes recorded for a $Sn_2Mo_6S_{7.5}$ sample at $T = 11$ K. 1—$H = 90$; 2—110; 3—130; 4—150 kOe.

curves exhibit minima, whose relative depth increases with increasing magnetic field and with decreasing temperature. The anisotropy of the magnetoresistance of the $Pb_2Mo_6S_{7.5}$ and $Cu_{1.9}Mo_6S_8$ is less pronounced; some illustrative curves of $\rho(\phi)$ for a $Pb_2Mo_6S_{7.5}$ sample are shown in Fig. 3. The minima of this rosette lie along fourfold axes.

Comparing the data on the anisotropy of H_{c2} with the data on the anisotropy of $\rho(H)$, we can attempt to find the approximate shape of the Fermi surface of the ternary chalcogenides of molybdenum. Since H_{c2} depends on the curvature of the Fermi surface, with the maximum values of H_{c2} corresponding to the flattest parts of the Fermi surface, we may conclude from the data obtained for ternary chalcogenides of molybdenum with copper that the flat parts of the Fermi surface run perpendicular to threefold axes. Examining the $\rho(\phi)$ curves, on the other hand, we see that the minima of the magnetoresistance lie along the fourfold axes. In the case of strong effective magnetic fields, i.e., under the condition $\omega_c \tau \gg 1$, the minima on the $\rho(\phi)$ curve for the compensated metal lie in the region of open directions of the Fermi surface. In the present case we have $\omega_c \tau \sim 1$, so that the anisotropy of $\rho(H)$ may be determined to a large extent by the anisotropy of the collision integral. An increase in the field or a decrease in the temperature, either of which leads to an increase in $\omega_c \tau$, causes an increase in the anisotropy of $\rho(H)$. It may therefore be assumed that in the present case the direction of the minima of the magnetoresistance corresponds to open directions of

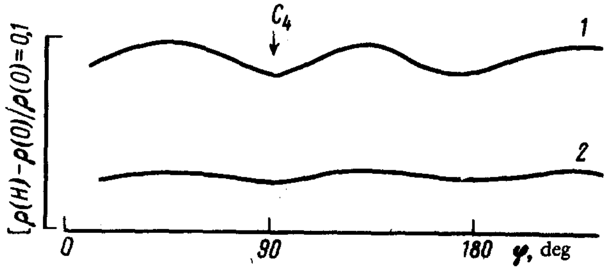

FIG. 3. Magnetoresistance rosettes orientation in a 150-kOe field. 1—T for a $Pb_2Mo_6S_{7.5}$ sample in the C_4 = 11.5; 2—15 K.

the Fermi surface. Under the assumption that the Fermi surface of the ternary chalcogenides of molybdenum with copper, lead, and tin are not greatly different, we can offer the following model for this Fermi surface: an octahedron with vertices which lie on fourfold axes and with faces which are perpendicular to threefold axes. Open directions pass through the vertices.[1)] This shape of the Fermi surface is apparently similar to the open sheet of the Fermi surface of a ternary chalcogenides of molybdenum given in Ref. 4. As mentioned above, the anisotropy of the resistance in the vicinity of the S–N transition for the ternary chalcogenides of molybdenum with lead and tin is small (much smaller than that for the chalcogenide with copper). At present, the reason for this difference is not clear. One possibility is a difference in the properties of the Chevrel phases of the first and second groups.[5] We do not, furthermore, rule out the possibility that optical vibrational modes of tin and lead may be responsible for the difference.

We should point out the anisotropy of H_{c2} in the ternary chalcogenides of molybdenum with tin has been observed previously.[6,7] Decroux *et al.*[7] have carried out detailed measurements of the anisotropy of H_{c2} for ternary chalcogenides of molybdenum with lead and tin. They found anisotropy $\sim 20\%$. An anisotropy of similar magnitude in $Sn_2Mo_6S_{7.5}$ was reported in Ref. 6, where it was suggested that this anisotropy might be related to an anisotropic defect distribution in the crystal. It should be kept in mind, however, that in the study of the anisotropy in Ref. 7 the current through the sample was usually not perpendicular to the external field, and the angle between the field and the current and different values for different orientations of the sample, while in present experiments the current always flowed perpendicular to **H**.

Measurements with higher-quality single crystals will probably lead to a more reliable determination of the shape of the Fermi surface of the ternary chalcogenides of molybdenum.

[1)]We wish to thank Doctor Chevrel for graciously furnishing the sample.

[2)]The flat parts of the Fermi surface may be one of the factors[3] which lead to the linear dependence of the magnetoresistance on the field which we observed for ternary chalcogenides of molybdenum with copper and tin over a broad range of the magnetic field.

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