Unusual behavior of magnetic moment of $PbMo_6S_8$ single crystal

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Room-temperature measurements reveal a nonlinear field dependence of the magnetic moment and an anisotropy of this moment in fields on the order of several kiloersted in a $PbMo_6S_8$ single crystal, with an approximately cubic structure.

Alekseevskiĭ *et al.* have reported observing a significant ($\sim 20\%$) anisotropy in the magnetic susceptibility χ of a $PbMo_6S_8$ single crystal. This is an extremely surprising result, since although $PbMo_6S_8$ does have a rhombohedral structure, the rhombohedral angle α is nearly a right angle ($\alpha = 89.3°$), and the measurements were carried out in a weak magnetic field H = 5 kOe. We know that there should be no anisotropy of χ in crystals of cubic symmetry in the limit of weak magnetic fields. The observation of an anisotropy of χ in $PbMo_6S_8$ thus might mean that the applied magnetic field was "strong," at least for certain fragments of the Fermi surface of $PbMo_6S_8$, and that the anisotropy of χ would fade away as the field was weakened.

To test this possibility, we measured the field dependence of the magnetic moment of a $PbMo_6S_8$ single crystal for the two orientations $\mathbf{H} \| C_2$ and $\mathbf{H} \| C_3$ at temperatures of 100 K and 290 K. The measurements were computer-controlled by the Faraday method. The data of 10–12 measurements were built up in order to reduce the noise level to 10^{-3} dyn. The mass of the sample was 0.18 g.

Figure 1 shows the results of the measurements at $T = 290$ K. We see that the magnetic moment M is linear in the field up to 800 Oe and does not depend on the orientation of the field. This is as it should be for a cubic crystal in the limit $H \to 0$. The anisotropy of M becomes significant at $H_0 \approx 1$ kOe. In fields $1 < H < 2$ kOe, $M(H)$ is nonlinear, but it becomes linear again as the field is increased further. If we extrapolate the curves of $M(H)$ from $H > 2$ kOe, we find that they intercept the ordinate at $M_0 \approx 4 \times 10^{-4}$ G·cm³/g.

In the measurements at $T = 100$ K, the inflection point on the $M(H)$ curves and the onset of the anisotropy of the magnetic moment shift down the field scale (to 400–500 Oe), while M_0 decreases to 1×10^{-4} G·cm³/g.

In interpreting the results we should take into account the unusual kinetic characteristics of single crystals of ternary chalcogenides of molybdenum, $M_xMo_6S_8$. The temperature dependence of the resistance of $AgMo_6S_8$ and $Cu_{1.8}Mo_6S_8$, for example, has long regions with[2] $\rho(T) \sim T^2$, and the magnetoresistance of samples with $M =$ Pb, Sn, Ag, and Cu is linear in the field in the region of classically weak ($\omega_c \tau < 1$) fields.[3] This behavior, which is not at all typical of ordinary metals, has been attributed to the presence of large, flat faces and sharp edges on the Fermi surface of the ternary

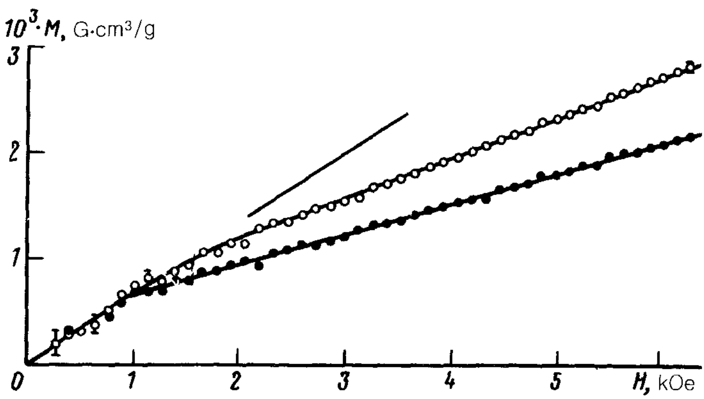

FIG. 1. Field dependence of the magnetic moment of a $PbMo_6S_8$ single crystal at $T = 290$ K. ○—$\mathbf{H} \| C_2$; ●—$\mathbf{H} \| C_3$.

chalcogenides of molybdenum.[2,3] The linear magnetoresistance, for example, would then arise as a result of an averaging of the effect of the field on the electrons at the faces and edges. The imposition of a magnetic field would cause electrons to diffuse along the Fermi surface. As long as an electron remains on a given face, its velocity will remain constant (any field will be "weak"), but when it goes onto a neighboring face, the velocity will change abruptly by a large amount (the same field will become "strong"). A comparison of the results of a model-based calculation valid for all magnetic fields with experimental results on the magnetoresistance and its anisotropy led to the conclusion that the Fermi surface of $PbMo_6S_8$ is approximately a cube.

This idea can apparently also be used to explain the unusual behavior of the magnetic moment of $PbMo_6S_8$. The results in Fig. 1 may be interpreted as the sum of a Pauli paramagnetism which is isotropic and linear in the field (on the one hand) and (on the other) an orbital diamagnetism which is anisotropic and nonlinear at $H < 2$ kOe. As usual, the paramagnetic moment is determined by the density of states at the Fermi level, and the corresponding susceptibility is $\chi_p = \mu_B^2 N(\epsilon_F)$.

To calculate the diamagnetic orbital moment, as in the case of the magnetoresistance, we need an expression valid at all fields. Unfortunately, there is no such expression at the moment, so we are forced to restrict the discussion to some qualitative considerations. An orbital diamagnetism arises as an electron moves along curved trajectories, so it becomes noticeable when an electron manages to move from one flat face of the Fermi surface to a neighboring one. Hence the field ($H_0 \approx 1$ kOe) at which the anisotropy appears in M at $T = 290$ K can be used to estimate the radius of curvature of the edges of the Fermi surface, k_r: $r_H = c\hbar k_r / eH_0 \approx 1$, where r_H is the radius of the cyclotron orbit, and l is the mean free path. Assuming[4] $l \approx 10$–20 Å, we find $k_r \approx (1.5$–$3) \times 10^3$ cm^{-1}. This result is smaller by a factor of $\sim 5 \times 10^4$ than the size of the Brillouin zone, $2\pi/a \approx 10^8$ cm^{-1}. Lowering the temperature to 100 K leads to an increase in l by a factor ~ 2 [according to the results of measurements of $\rho(T)$], and an anisotropy of the magnetic moment arises in weaker fields.

We know that in the limit of a weak magnetic field, i.e., under the condition $r_H \gg 1$, the Landau–Peierls orbital diamagnetism χ_L is isotropic at any point on the Fermi surface. At $H > H_0$ this condition is violated for the edges of the Fermi surface, and an anisotropic component associated with reference points of the Fermi surface arises.[5] To estimate this component, we can apparently make use of the fact that in the case of a spherical Fermi surface it is the same as $\chi_L = -\frac{1}{3}(m/m^*)^2 \chi_p$, where m^* is the effective cyclotron mass. Assuming that a similar m^* dependence holds for a polyhedral Fermi surface, and finding m^* by averaging over k_z,

$$m^* = (k_z^{max})^{-1} \int_0^{k_z^{max}} [(\hbar/2\pi)\oint dk/v_\perp]\, dk_z ,$$

we find $\chi_L(C_3)/\chi_L(C_2) = 1.5$ for a cubic Fermi surface.

From the experimental data we can easily distinguish the orbital component of χ, which is equal to the difference between the slope of the $M(H)$ curves at $H > 2$ kOe and that in the limit $H \to 0$ (i.e., χ_p):

$$\chi_p(H \to 0) = 6.6 \times 10^{-7}\ \mathrm{cm^3/g}$$

$$\chi_L(C_2) = -2.68 \times 10^{-7}\ \mathrm{cm^3/g}$$

$$\chi_L(C_3) = -3.72 \times 10^{-7}\ \mathrm{cm^3/g}.$$

The ratio $\chi_L(C_3)/\chi_L(C_2) = 1.4$ agrees well with that (1.5) which has been suggested for a cubic Fermi surface. A conversion of the Pauli susceptibility χ_p to a density of states leads to a value of $N(\epsilon_F)$ which is 20% higher than the specific-heat value.[6]

In summary, the unusual behavior of the magnetic moment of $PbMo_6S_8$ can be explained by assuming that the Fermi surface of this compound has a nearly cubic shape, with large flat faces and sharp edges. This shape of the Fermi surface describes a metal which has conducting "channels" in three mutually perpendicular directions. The extent of the mutual effects of these channels determines the radii of curvature of the edges of the Fermi surface. The small radius of curvature found in the present study is evidence of essentially noninteracting channels. Such channels could apparently form as a result of the filling of d states of molybdenum, specifically, $e_g\,(x^2 - y^2)$ orbitals.[7]

The anisotropy observed in the magnetic moment is the first experimental observation of the diamagnetic contribution of reference points of the Fermi surface which was predicted 35 years ago.[5]

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