Effect of electron-electron interaction on the state density in the low-dimensionality compound Mo_2S_3

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(Submitted 1 October 1985)
Pis'ma Zh. Eksp. Teor. Fiz. **42**, No. 9, 377–380 (10 November 1985)

A special experiment has been carried out to determine the anomalous part of the magnetic susceptibility of the low-dimensionality compound Mo_2S_3. This part of the susceptibility is associated with an anomaly in the state density of conduction electrons.

Anomalies arise in the electron state density during repulsion between conduction electrons in a metallic system with disorder.[1] These anomalies lead to an anomalous temperature dependence for thermodynamic properties such as the magnetic susceptibility and the specific heat. Experiments on the anomalous part of the magnetic susceptibility provide information on the magnitude and sign of the constant of the electron-electron interaction, λ_c. The temperature dependence of the anomalous part of the magnetic susceptibility, χ_a, can be used to determine the effective dimensionality of the electron subsystem. This possibility is particularly important when it is difficult to measure the anisotropy of the conductivity directly. There are serious difficulties, however, in experimentally studying the anomalous magnetic susceptibility associated with an anomaly in the state density of conduction electrons. These difficulties stem from the fact that the effect is small, even in comparison with the signal from a small ($\sim 10^{-5}$) paramagnetic impurity. In the present experiments we have been able to not only see the effect in its pure form but also to study its temperature dependence, thanks to two factors: the quasi-one-dimensional nature of the effect, with a high density of quasi-one-dimensional filaments in the Mo_2S_3 compound studied, and the particular design of the experiment.

We studied the temperature dependence of the magnetic susceptibility χ of a polycrystalline Mo_2S_3 powder over the temperature interval 4.2–300 K. The measurements are taken by a Faraday weight method. The temperature dependence of the electrical conductivity σ is measured by four-contact potentiometric method with the same samples, pressed into tablets.

The crystal structure of Mo_2S_3 was studied in Ref. 2. The Mo atoms in Mo_2S_3 form metal chains of two types, with metal-metal bonding between Mo atoms.

According to the data of Ref. 1, an anomaly in the state density of conduction electrons and thus in χ_a is determined by the diffusion coefficient (d) of the conduction electrons:

$$\chi_a = \frac{3\xi(3/2)(g\mu_B)^2}{16\sqrt{2}\pi^{3/2}\sqrt{\hbar DkT}}\left\{\frac{2}{\ln(T_0/T)} + 2\lambda_c\right\}, \qquad (1)$$

where $T_0 = (2\gamma\pi)\omega_D\exp(\lambda_c^{-1})$. The behavior in (1) is observed during the quasi-one-dimensional motion of interacting electrons. According to the data of Ref. 3, at temperatures below 76 K in Mo_2S_3 single crystals there is an exponential retardation of relaxation processes, and the system can easily and reversibly be put in a state with a frozen high-temperature phase by means of rapid cooling. Figure 1 shows curves of the

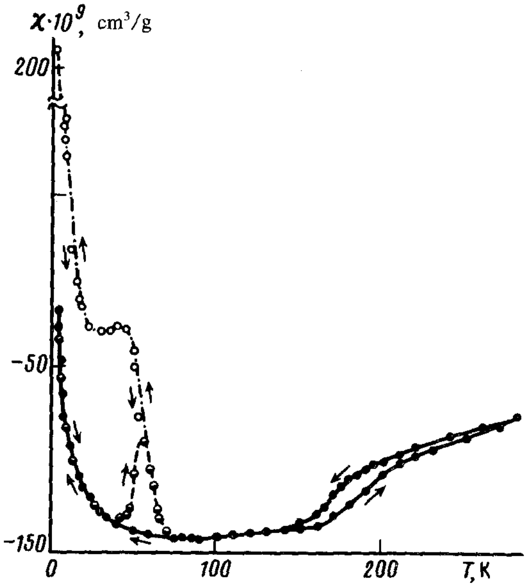

FIG. 1. Temperature dependence of the magnetic susceptibility χ of a polycrystalline Mo_2S_3 powder measured at various cooling and heating rates. ●—The sample is cooled from 80 to 30 K in 1 h (χ_r); ◓—the sample is heated from 30 to 80 K in 1 h; ○—the sample is cooled and heated in the same temperature interval over 14 h (χ_s).

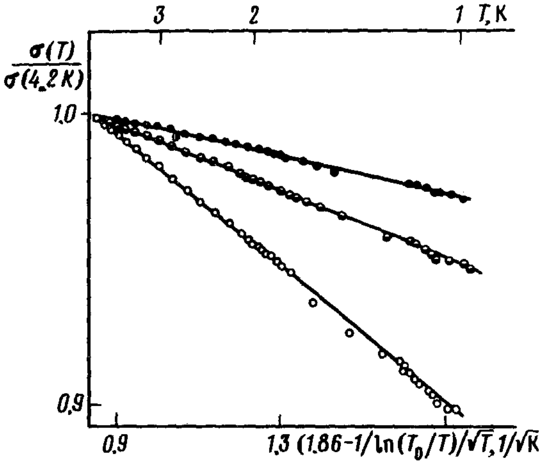

FIG. 2. Temperature dependence of the relative conductivity $\sigma(T)/\sigma(4.2\ \mathrm{K})$ of a polycrystalline Mo_2S_3 powder pressed into a tablet according to measurements at various cooling rates. ○—The sample is cooled from 300 to 30 K in 1 h (D_1); ◒—the sample is held at $T = 80$ K for 40 h before the measurements (D_2); ●—the sample is held at $T = 80$ K for 60 h before the measurements (D_3). $D_1/D_2 = 4$; $D_1/D_3 = 16$. The solid lines show the predictions of expression (2).

temperature dependence of χ of a polycrystalline Mo_2S_3 powder obtained at various cooling rates. It can be seen from the data on the susceptibility that the relaxation processes which are characteristic of single crystals also occur in the polycrystalline Mo_2S_3 powders.

We have suggested that the diffusion coefficient D should change during the freezing of the high-temperature phase. A change in D should lead to a change in the amplitude of χ_a and in the anomalous part of the electrical conductivity, σ_a, described by[4]

$$\sigma_a = -\frac{e^2}{2\pi^2\hbar}\sqrt{\frac{\hbar D}{kT}}\left(1.86 - \frac{1}{\ln(T_0/T)}\right) 4,91 \tag{2}$$

where 1.86 is the constant calculated from the value of λ_c (λ_c is determined independently from data on χ). Figure 2 shows curves of σ as a function of the parameter $[1.86 - (1/\ln(T_0/T))](1/\sqrt{T})$ in (2) found at various cooling rates. It can be seen from these results that the freezing of the high-temperature phase can change D by a factor of several tens. Since—except for χ_a—all of the temperature-dependent components of the magnetic susceptibility χ of the compound Mo_2S_3 are independent of the sample cooling rate, the difference $\chi_s - \chi_r$ (χ_s is the magnetic susceptibility of the polycrystalline Mo_2S_3 powder during slow cooling, and χ_r is that during rapid cooling) contains only the difference between anomalous parts:

$$\chi_s - \chi_r = \frac{3\xi(3/2)(g\mu_B)^2}{16\sqrt{2}\pi^{3/2}\sqrt{\hbar kT}}\left\{\frac{2}{\ln(T_0/T)} + 2\lambda_c\right\}\left(\frac{1}{\sqrt{D_s}} - \frac{1}{\sqrt{D_r}}\right). \tag{3}$$

Figure 3 shows an experimental curve of the difference $\chi_s - \chi_r$.

From the fact that σ_a and $\chi_s - \chi_r$ deviate from the behavior in (2) and (3), respectively, at temperatures above 12 K, we conclude that at 12 K the coherent length for the interacting electrons, $L_{\mathrm{int}} = \sqrt{\hbar D/kT}$, is comparable to the transverse dimensions of the quasi-one-dimensional filament in the compound Mo_2S_3, and the

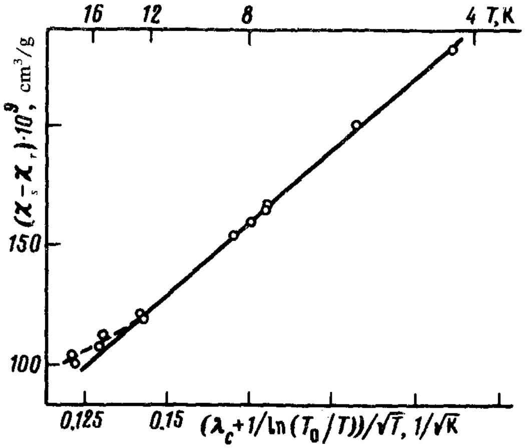

FIG. 3. Temperature dependence of the susceptibility difference $\chi_s - \chi_r$. The solid line is the prediction of expression (3). $D_r/D_s \gtrsim 5.6$.

quasi-one-dimensional approximation breaks down above 12 K. From the magnitude of the proportionality factor in the linear dependence of χ_r on the parameter $[(1/\ln(T_0/T)) + \lambda_c](1/\sqrt{T})$ we find the lower limit $D_r \gtrsim 0.1$ cm^2/s on the diffusion coefficient (taking paramagnetic impurities into account leads to an increase in D_r). Taking into account the value found for D_r, we find a lower estimate of the cross section (S_f) of the quasi-one-dimensional filament in Mo_2S_3: $S_f \gtrsim L^2_{\text{int}}$ (12 K) $= 120$ Å^2. The electron-electron interaction constant λ_c is found by straightening out the dependence of the difference $\chi_s - \chi_r$ on the parameter $[(1/\ln(T_0/T)) + \lambda_c](1/\sqrt{T})$; it turns out to be positive: $\lambda_c = 0.36$. The contribution of paramagnetic impurities was eliminated in this determination of λ_c.

In summary, the anomalous part of the magnetic susceptibility of the polycrystalline Mo_2S_3 powder which has been singled out in these experiments is due to an effect of the electron-electron interaction on the state density of conduction electrons in this compound. It has been established experimentally that the electron-electron interaction constant in Mo_2S_3 is positive (there is a repulsion between electrons), and the motion of the interacting electrons in Mo_2S_3 is quasi-one-dimensional.

We wish to thank B. L. Al'tshuler for interest in this study and for useful comments offered in a discussion.

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Translated by Dave Parsons