Effect of antiferromagnetic ordering on electron kinetic phenomena in dysprosium monosulfide

V. I. Novikov, V. P. Zhuze, V. M. Sergeeva, and S. S. Shalyt

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences
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Electron kinetic phenomena in the metal-like compound DyS were investigated experimentally. Pronounced anomalies of the electric conductivity, thermoelectric power, and thermal conductivity were observed near 38°K. The antiferromagnetic nature of the disordering in DyS near the indicated temperature is confirmed by investigation of the temperature dependence of the magnetization, the specific heat, and the magnetocaloric effect. An analysis of the experimental data on the temperature dependence of the electric conductivity reveals three components of the electric resistance, viz., residual, phonon, and magnetic. From the value of the latter component above the Neel point an estimate is made of the *s-f* exchange interaction energy ($I \approx 0.03$ eV); this leads to a tentative conclusion that the interaction in DyS is indirect. The character of the anomalies in the electron and lattice components near the Neel temperature can be assessed by analyzing the experimental data on the thermal conductivity of DyS. The role of the magnon component of the thermal conductivity at low temperatures is evaluated from the effect of a magnetic field on the thermal conductivity. From the formulas derived for the thermoelectric power by a theoretical study of a simple spherical model of the Fermi surface, with allowance for magnetic doubling of the crystal lattice constant and for formation of a gap in the related carrier energy spectrum, it is concluded that the gap is located near the Fermi surface itself. The temperature dependence of the thermoelectric power near the Neel point is consistent with the theory. A general analysis of the experimental data can explain the absence of a section of negative temperature dependence of the electric resistance from the experimental curve. A qualitative analysis is made of the experimental data on the effect of a magnetic field on the electric resistance, thermal conductivity, and thermoelectric power in the magnetic ordering region.

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If magnetic order exists in a crystal with n-type conductivity, then the kinetic coefficients determined by the electronic spectrum of the crystal can undergo abrupt changes at the magnetic-ordering temperature. If the order is antiferromagnetic, one of the causes of the ensuing anomalies in the temperature dependence of the kinetic coefficient is the magnetic doubling of the lattice constant and, as a consequence, the appearance of an energy gap in the carrier spectrum, and a splitting of the conduction band into two subbands.[1] The effect of this change in the electronic-spectrum structure of an antiferromagnet on its electric conductivity and thermoelectric power was considered theoretically by Irkhin and Abel'skiĭ [2-4]. It is obvious that from the results of an experimental study of the electric conductivity and of the thermoelectric power of a metallic antiferromagnet one can determine the main parameters of the indicated theoretical model, namely the width of the gap and its location in the Brillouin zone. Korenblit and Tankhilevich[5] investigated theoretically the magnon thermal conductivity of an antiferromagnetic metal at low temperatures.

The anomalies of the electronic kinetic coefficients are usually more strongly pronounced at low temperatures, and from this point of view greater interest attaches to substances with low ordering temperatures. However, to observe the temperature variation of the kinetic coefficients in the magnetic ordering region, when quasiparticles of a type peculiar to this state, namely magnons, can participate in the transport phenomena, one must select for the investigations an antiferromagnetic metal whose Neel temperature is not too low. From this point of view, a suitable object is dysprosium monosulfide, which is known to be a metal-like compound with one free electron per formula unit, and to have a crystal structure of the NaCl type. According to the available data, the magnetic susceptibility of DyS has a maximum at T = 32°K, which points to the antiferromagnetic character of the ordering that occurs in it.[6]

We have investigated experimentally, in the range from 1.6 to 120°K, the temperature dependences of the electric conductivity, the thermal conductivity, the thermoelectric power, the magnetization, and the magnetocaloric effect of a polycrystalline DyS sample of 6 mm diameter and 34 mm length, obtained by directed crystallization from the melt.[7] The electric conductivity was measured with a dc potentiometer, the thermal conductivity and the thermoelectric power were measured by the method of stationary heat flow through the sample, with the temperatures measured with an Allen Bradley carbon resistance in the 1.6–30°K range and with copper-constantan thermocouples between 25 and 120°K. The thermoelectric power of DyS was determined relative to pure copper. The magnetization was measured by a ballistic method, by introducing the magnetized sample into an induction coil placed in a homogeneous constant field. To refine the nature of the magnetic ordering, we measured the magnetocaloric effect and in this measurement it was important to determine the sign of this effect above and below the Neél temperature. To reveal qualitatively the singularity of the heat capacitance, which should accompany the magnetic ordering in the crystal, we followed the change of the sample temperature T with time t at a constant heat outflow. It was important in this case to determine the temperature at which a singularity appears in the heat capacity as estimated by such an approximate method.

It is seen from Fig. 1 that the maximum of the speci-

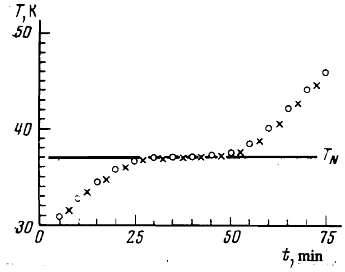


FIG. 1. Change of sample temperature in the case of constant heat outflow; this makes it possible to determine the position of the singularity of the specific heat on the temperature scale: X—without a field, ○—in a 10-kOe field.

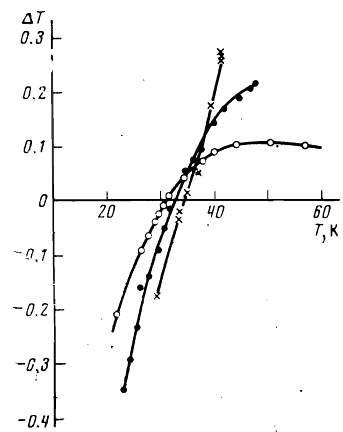


FIG. 2. Temperature dependence of the magnetocaloric effect for different values of the magnetic field: ○—5 kOe, ●—10 kOe, X—15 kOe.

fic heat turned out to be at 37°K. This value of the temperature practically coincides with the position of the maximum on the plot of the temperature dependence of the magnetization, which we measured (T = 38°K) and which differs significantly from the temperature of the maximum susceptibility as determined by Loginov, Serveeva, and Bryzhina[7] (T = 32°K). The correct value is T = 38°K obtained in our experimental setup, in which the thermometer was in direct contact with the sample, whereas under the experimental conditions of[6] the thermometer was only close to the sample, being separated from it by a gas gap.

The curves for the magnetocaloric effect in DyS, which are shown in Fig. 2, indicate that, as expected, the sign of this effect changes in the case of an antiferromagnetic ordering near the Neél temperature (T_N). The temperature at which the sign of the effect is reversed depends appreciably on the intensity of the magnetic field that is turned on and off to observe this phenomenon. We note that the position of the maximum of the specific heat, if it is determined from the heating curve (curve 1), does not depend noticeably on the field.

Figure 3 shows the temperature dependences of the resistivity ρ, the total thermal conductivity κ, and the thermoelectric power α of the investigated DyS sample. It is seen from this figure that all these three coefficients have near the Neél temperature T_N = 38°K strongly pronounced anomalies. To analyze the experimental data of Fig. 3 it is first necessary to separate from the total resistivity its four principal additive components: the residual ρ_0, phonon ρ_{ph}, magnetic ρ_S, and electron-electron ρ_{ee}.

The upper curve of Fig. 4 shows the experimentally measured temperature dependence of the total resistivity, from which it is necessary to separate its magnetic component ρ_S. To this end it is necessary to bear in mind that in the paramagnetic region, i.e., at $T > T_N$, the value of ρ_S should not depend theoretically on the temperature[8]:

$$\rho_s = \frac{3\pi}{2}\frac{m}{\hbar e^2 N_0}(g-1)^2 j(j+1)\frac{J^2(0)}{\varepsilon_F}, \tag{1}$$

where N_0 is the number of magnetic ions per cm^3, g is the Lande factor, j is the quantum number of the 4f shell, J(0) is the exchange-interaction integral at T = 0°K, ϵ_F is the Fermi energy, and m is the effective mass of the electron. The method of such a separation, which was used by us, is explained in Fig. 4: the dashed curve is a smooth continuation of the experimental $\rho(T)$ curve from the region $T > T_N$, where ρ_S is constant, while the curve $\rho_{ph}(T) + \rho_{ee}(T)$ is drawn from T = 0°K parallel to $\rho(T)$. If we determine the temperature dependence of the so-separated component $\rho_S(T) = \rho(T) - \rho_0 - \rho_{ph}(T) - \rho_{ee}(T)$, then, as seen from Fig. 4, this dependence is well approximated by the formula $\rho_S \propto T^5$ in the region 20—38°K (i.e., up to $T = T_N$!). For the contribution $\sim(\rho_{ph} + \rho_{ee})$ separated in this manner, we obtain from experiment a temperature dependence close to T^2. Since $\rho_{ph} \propto T^5$ and $\rho_{ee} \propto T^2$, the last result offers evidence that $\rho_{ee} > \rho_{ph}$ in this temperature region. At T < 20°K, the accuracy with which ρ_S is determined is not sufficient to analyze this dependence. The law $\rho_S \propto T^5$ for an antiferromagnetic metal was theoretically proved in a paper by Berdyshev and Vlasov[9] for the temperature region $T < T_N$.

From the value of ρ_S in the region $T > T_N$ we can estimate with the aid of (1) the energy I of the exchange s-f interaction, which turned out to be I ≈ 0.03 eV for DyS (m = 2.9m$_0$, N = 2.4 × 10^{22} cm^{-3}, g = 4/3, j = 15/2, ϵ_F = 1 eV[10]). Since DyS is a metal-like compound it follows that, assuming the main exchange-interaction mechanism to be indirect via the conduction electrons, we can estimate the magnetic-ordering temperature from the values of J(0) and ϵ_F:

$$k_0 T_N = \frac{J^2(0)}{\varepsilon_F}\frac{j(j+1)}{2}(g-1)^2.$$

As a result we get $T_N^{est} \approx 30$°K, i.e., a value close to the experimental T_N = 38°K.

We proceed now to analyze the thermal conductivity. In DyS at low temperature, three sorts of quasi-particles can take part in the transport of heat through the crystal: electrons, phonons, and magnons. Therefore the total thermal conductivity can be represented in the form

$$\varkappa = \varkappa_e + \varkappa_{ph} + \varkappa_s. \tag{2}$$

The electronic part of the thermal conductivity κ_e

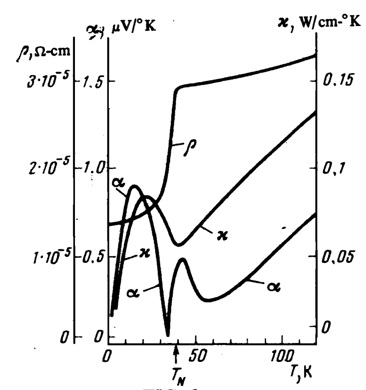


FIG. 3

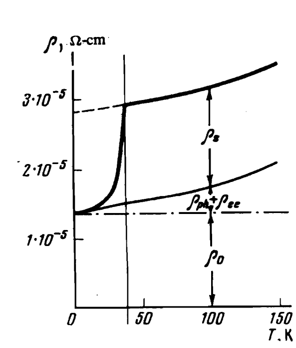


FIG. 4

FIG. 3. Temperature dependences of the resistivity ρ, the thermal conductivity κ, and the thermoelectric power α

FIG. 4. Graphic analysis of the resistivity $\rho = \rho_0 + \rho_{ph} + \rho_S + \rho_{ee}$.

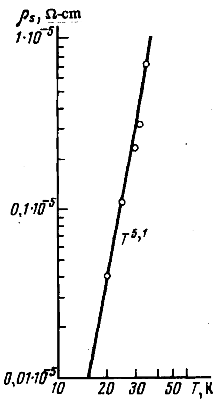


FIG. 5

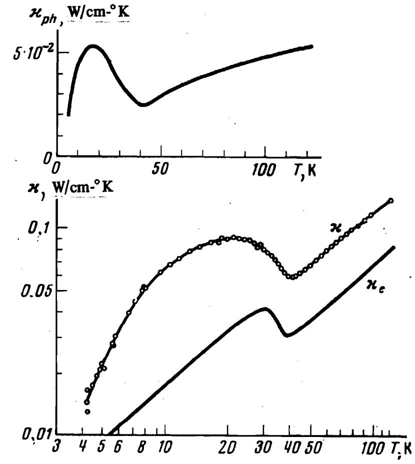


FIG. 6

FIG. 5. Temperature dependence of the magnetic part of the resistivity ρ_S at $T < T_S$.

FIG. 6. Electronic (κ_e) and lattice (κ_{ph}) components of the thermal conductivity ($\kappa = \kappa_e + \kappa_{ph}$).

can be calculated from the experimentally measured function $\rho(T)$ (Figs. 3 and 4) with the aid of the Wiedemann-Franz relation, namely $\kappa_e = LT/\rho$, where $L(N, T, \rho_0)$ is a known function of the carrier density N, of the temperature T, and of the residual resistivity of the sample ρ_0.[11] Figure 6 shows plates of both the temperature dependence of the so-calculated function $\kappa_e(T)$, and the phonon part of the thermal conductivity, determined from the difference $\kappa_{ph} = \kappa - \kappa_e$. The curves of Fig. 6 show that in the case of antiferromagnetic ordering the anomaly in DyS is observed in both the electronic and the phonon components of the thermal conductivity. The former can be due to the fact that the scattering of the carriers by the inhomogeneities of the spin system becomes enhanced near T_N. The anomaly of the phonon thermal conductivity may be connected not only with the additional scattering of the phonons by the spin inhomogeneities, but with the anomaly of the spin specific heat near T_N.[12]

/ The magnon thermal conductivity κ_S in DyS can noticeably manifest itself below 10°K, inasmuch as at higher temperature the heat transport by the spin wave should be substantially restricted by magnon-magnon scattering. The appearance of the magnon component of the thermal conductivity can be observed on the plot of the change of the thermal conductivity $\Delta\kappa/\kappa_0$ in a magnetic field—see Fig. 7. The positive sign of this effect at $T < 10$°K can be attributed to the increase of the magnon component of the thermal conductivity in the magnetic field, a component that becomes noticeable in this temperature region. It can thus be assumed that above 10°K the thermal conductivity of DyS is due mainly to the electrons and the phonons.

As usual, the quantity altered most by magnetic ordering is the thermoelectric power. Abel'skiĭ,[4] in a theoretical paper dealing with the behavior of the thermoelectric power near the Neél temperature, explains how the formation of the gap in the carrier spectrum influences not only the concentration term of the thermoelectric power of an antiferromagnetic metal, as was considered in the past, but also the relaxation term. We shall therefore interpret our experimental results for DyS, which are shown in Fig. 3, by using the formulas of[4].

It is assumed that qualitatively correct results for the thermoelectric power can be obtained by considering a spherical Fermi surface with a gap Δ near this surface, and taking into account only the scattering of the electrons by the phonons and by the spin inhomogeneities. For such a model, the carrier spectrum takes the form

$$\varepsilon_{k\pm} = \frac{\hbar^2}{2m}[u^2+l^2\mp 2l(u^2+p^2)^{1/2}], \qquad (3)$$

and the resistivity and the total thermoelectric power are given by

$$\rho(T)=\rho_1(T)(1+p^2/u_F^2), \qquad (4)$$

$$\alpha^\pm = \frac{\pi^2}{3}\frac{k_0}{e}\frac{k_0T}{\varepsilon_F}\left\{\frac{3}{2}-r_p^\pm+\left(\frac{3}{2}-r_p^\pm\right)\frac{\rho_{ph}}{\rho}+\left(-\frac{1}{2}+r_p^\pm\right)\frac{\rho_s}{\rho}\right\}. \qquad (5)$$

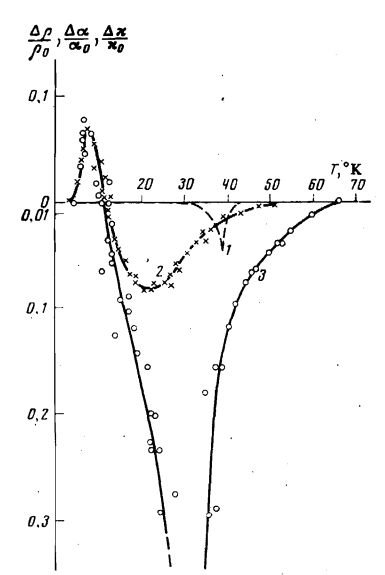
Here $u = |l - k|$, l is the value of the quasimomentum k at which the gap appears in the carrier spectrum, m is the carrier mass, $p = m\Delta/2\hbar^2 l$, $r_p^\pm = \pm\frac{1}{2}lp^2/u_F^3$, $u_F = |l - k_F|$, k_F is the quasimomentum on the Fermi surface, and $\rho_1(T)$ is the resistivity in the absence of the gap. The ratios ρ_{ph}/ρ and ρ_S/ρ were determined above in the analysis of the resistivity of DyS (Fig. 4). The dependence of the width of the gap Δ on the temperature is of the form[4,13]

$$\Delta = I(1-T/T_N)^\beta, \qquad (6)$$

where $\beta = \frac{1}{2}$ to $\frac{1}{3}$. Thus, the temperature dependence of the thermoelectric power near T_N is determined by two unknown parameters β and l/k_F, which can be determined by comparison with the experimental data. The best agreement for the thermoelectric power in the section 30—40°K was obtained at values $\beta = \frac{1}{3}$ and $l/k_F = 0.98$. The latter quantity shows that the gap in DyS occurs very close to the Fermi surface.

Irkhin[2] has shown that the formation of a gap in an antiferromagnetic metal can alter noticeably the temperature dependence of the resistivity near T_N: instead of a more or less smooth decrease of the resistivity with temperature, which is typical of metallic conductivity, the $\rho(T)$ curve may acquire a section with a negative derivative $d\rho/dT$. This phenomenon was indeed observed, particularly in single-crystal Dy.[14] In our case of DyS, however, no such section was observed on the $\rho(T)$ curve (Fig. 3). It can be shown that this fact

FIG. 7. Relative change of the resistivity (curve 1), of the thermal conductivity (curve 2), and of the thermoelectric power (curve 3) in a 15-kOe magnetic field.



agrees with the results of the foregoing analysis of the resistivity and of the thermoelectric power.

According to (4) we have

$$\frac{d\rho}{dT}=\frac{d\rho_1}{dT}(1+\gamma\Delta^2)+\rho_1 2\gamma\Delta\frac{d\Delta}{dT}, \qquad (7)$$

where $\gamma = (m/2\hbar^2 l u_F)^2$. The quantity $d\rho/dT$ can be negative in the case when the second term of (7), which contains the negative factor $d\Delta/dT$, exceeds the first. But the values of all the quantities in (7) were determined above, and for DyS the first term of the expression turned out to exceed the second, meaning that the derivative $d\rho/dT$ should be positive.

The three curves of Fig. 7 show how application of a magnetic field affects the resistivity ρ, the thermoelectric power α, and the thermal conductivity κ of the investigated DyS sample. In view of the low mobility of the carriers in the investigated polycrystal, the magnetoresistance is observed only near T_N, where the magnetic disorder changes abruptly, while the negative sign of the effect indicates that the carrier scattering due to the magnetic disorder of the crystal decreases in a magnetic field. The action of the magnetic field manifests itself in a stronger and more complicated manner in the thermoelectric power α.

As indicated above, the increase of the thermal conductivity in the magnetic field at $T < 10^\circ K$ can be attributed to the increase of the magnon concentration, and whereas in the first half of this section ($T < 5^\circ K$) the value of $\Delta\kappa/\kappa_0$ increases because of the increase of the magnon density with increasing temperature and because of the larger role they play in the thermal conductivity, in the second half of this section, where a decrease of $\Delta\kappa/\kappa_0$ is observed, the role of the phonon component of the thermal conductivity increases, and the increase of the magnon concentration with increasing temperature makes this thermal-conductivity component smaller, and by the same token decreases the entire influence of the magnetic field on the thermal conductivity.

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