Anomalies of the thermoelectric power near magnetic transition points in gadolinium and terbium

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(Submitted January 17, 1975)
Zh. Eksp. Teor. Fiz. **69**, 611–616 (August 1975)

Theoretical and experimental investigations were made of the singularities in the temperature dependence of the thermoelectric power near magnetic-transition points. The temperature dependence of the thermoelectric power connected with critical scattering from long-range fluctuations is calculated near the Néel point. The obtained anomaly takes the form of a maximum (in absolute value) if $K_A \sim k_F$ (k_F is the Fermi quasimomentum and K_A is the value of the antiferromagnetism vector in the reciprocal lattice). If $K_A < k_F$, then an anomaly of the ferromagnetic type ('jump') should be observed at the Néel point. The calculations take into account, besides the critical scattering, also the presence of a gap, due to the magnetic splitting of the lattice, in the conduction-electron spectrum. Measurements were made of the thermoelectric power of single crystals of gadolinium and terbium in the critical temperature region. The observed anomalies have an anisotropic character—the anomalies of both metals are stronger in the basal plane than in the direction of the hexagonal axis. Anomalies of the type of maxima near the Curie and Néel points are observed in terbium in the basal plane.

PACS numbers: 72.20.Pa, 75.30.Jy

1. INTRODUCTION

Recent theoretical investigations have been devoted to the calculation of the anomalous resistance in metals, due to the critical scattering of the conduction electrons near the Curie point (T_C) and the Néel point (T_N).[1-5] The same cause is responsible for the anomalies in other kinetic phenomena, particularly the thermoelectric power. To be sure, the singularities of this anomaly in the thermoelectric power have as yet not been firmly established, owing to the contradictory nature of the experimental data. In [6,7] they observed an "oscillation" of the thermoelectric power of nickel (a maximum following a minimum on going through the Curie point). To the contrary, in [8] an inflection point was observed for nickel, and an oscillation for iron. According to the data of [9,10], on the other hand, the thermoelectric power of iron and cobalt has a discontinuity near the critical points. A discontinuity is observed also on the polytherm $\alpha(T)$ of antiferromagnetic chromium.[11] In [12] they investigated the temperature dependence of the thermoelectric power in single-crystal gadolinium and an anisotropy of the critical anomaly was observed, characterized by a discontinuity in the direction of the hexagonal axis and by an inflection point in the basal plane. Thus, experiment does not establish a unique form of the critical anomaly in the thermoelectric power. The reason for this is still not clear. It was shown in [13] that the critical anomaly in nickel depends strongly on the type of impurity. Thus, an inflection point is observed in the thermoelectric power of dilute alloys of nickel with iron or chromium, as against an oscillation in alloys with manganese. The latter is attributed by the authors to the fact that the impurity levels are located near the Fermi level.[1)] The shift of the Fermi level at the Curie point can lead in this case to a strong change of the density of states on the Fermi surface, and consequently to singularities in the behavior of the kinetic coefficients. But this means that the oscillation of the thermoelectric power is not connected with the critical scattering that is responsible for the anomalies in the heat capacity and in the kinetic coefficients. Calculations[14] of the thermoelectric power of ferromagnetic metals with allowance for the critical scattering have established the presence of a discontinuity of the thermoelectric power near T_C. Thus, to draw more definite conclusions concerning the character of the anomaly this problem must be investigated further from several points of view. We report here the results of an experimental study of the critical anomaly of the thermoelectric power in gadolinium and terbium, and we also calculate the thermoelectric power of antiferromagnetic metals in the critical region of temperatures.

2. CRITICAL ANOMALY OF THERMOELECTRIC POWER IN ANTIFERROMAGNETS

Let us calculate the thermoelectric power of an antiferromagnetic metal near T_N with allowance for the scattering of the conduction electrons by phonons and spin inhomogeneities. Under the simplest assumptions of the theory (single-band model, existence of a relaxation time τ), the thermoelectric power is described by the formula (all the notation is standard, k_0 is Boltzmann's constant)

$$\alpha_s = \frac{\pi^2 k_0^2 T}{3e\varepsilon_F}\left(\frac{\partial \ln n}{\partial \ln \varepsilon_k} + \frac{\partial \ln \tau}{\partial \ln \varepsilon_k}\right)_{\varepsilon_k=\varepsilon_F}. \tag{1}$$

The relaxation time τ_s for the spin mechanism of scattering is determined from the expression

$$\frac{1}{\tau_s} = \frac{1}{\tau_0}\left[1 + \frac{1}{4k^4}\int q^2 (K_A+q)\Gamma_{K_A+q}\,dq\right]. \tag{2}$$

Here k and q are respectively the quasimomentum of the conduction electron and the quasimomentum transferred in the collision, K_A is the magnitude of the antiferromagnetism vector in the reciprocal lattice, Γ_{K_A+q} is the spin-spin correlation function, and τ_0 is the relaxation time with allowance for the spin correlation in one site. For the analysis of the critical contribution we write down the correlator in the form[15]

$$\Gamma_{K_A+q} = D(qR_c)/(R_0 q)^{2-\eta}, \tag{3}$$

where $R_c = R_0|t|^{-\nu}$ is the correlation radius, R_0 is a constant on the order of the lattice parameter, $t = (T - T_N)/T_N$, and ν and η are the critical exponents.

It is seen from formulas (1)–(3) that the critical temperature dependence of the thermoelectric power is determined by the function $D(qR_c)$. Its form depends on the relation between the quantities K_A and k_F. If $K_A \ll k_F$, the main contribution to the critical anomaly is made by scattering from the short-range order fluctuations (large q), since the long-range order fluctuations ($q \approx 0$) determine the small-angle scattering and are ineffective. This case occurs also in ferromagnetic metals ($K_A \approx 0$), where strong anomalies of the λ type are observed in $\partial\rho/\partial T$, as well as an inflection point in ρ. The thermoelectric power should experience a discontinuity in this case.

If $K_A \sim k_F$ then large-angle scattering can take place at $q \approx 0$. Under these conditions the critical anomaly is due to scattering by long-range order fluctuations. A peak-type anomaly then appears in the resistivity (and not in $\partial\rho/\partial T$).[2] Let us see what contribution this scattering makes to the thermoelectric power. Performing calculations similar to those made in[4], we represent the function $D(qR_C)$ in the form

$$D(x)=\lim_{x\to\infty} D(x)+E(x)=D_\infty+E_0/x^2, \qquad (4)$$

where D_∞ and E_0 are constants. We now calculate the relaxation time by integrating in (2) in the interval $0 < q < q_0$, where $q_0 \ll K_A$ is the temperature-dependent maximum quasimomentum transfer at which the effective contribution to the kinetic coefficients is determined by the critical scattering from the long-range order fluctuations. We have in place of (2)

$$\frac{\tau}{\tau_0}=\frac{K_A}{k^4}\frac{1}{R_0^{2-\eta}}\left\{D_\infty\frac{q_0^{\eta+1}}{\eta+1}-\frac{E_0}{R_0^{1+\eta}}\frac{|t|^{(1+\eta)\nu}}{x^{1-\eta}(1-\eta)}\right\}, \qquad (5)$$

where $x = q_0R_C \gg 1$ near T_N. Taking into account simultaneously the phonon scattering mechanism, and also the gap in the conduction-electron spectrum, which is produced in the antiferromagnetic region because of magnetic splitting of the lattice[16], we obtain from (1) and (5) an expression for the thermoelectric power

$$\alpha_s^\pm=\frac{\pi^2k_0^2T}{3e\varepsilon_F}\left[\frac{3}{2}-r_p^\pm+(r_{ph}-r_p^\pm)\frac{\rho_{ph}}{\rho}+(r_s-r_p^\pm-r_s')\frac{\rho_s}{\rho}\right]. \qquad (6)$$

Here $r_{ph} = 3/2$, $r_s = -1/2$, ρ_{ph} and ρ_s are the phonon and magnetic parts of the total resistivity ρ, and the signs + and − correspond to cases when the gap lies above or below the Fermi level; in addition, the influence of the gap is taken into account here by the terms with $r_p^\pm$, while the critical scattering is accounted for by the terms with r_s', with

$$r_s'=\frac{r_s}{s(s+1)}\frac{K_A}{k_F}\left[D_\infty\frac{q_0^{1+\eta}}{(1+\eta)R_0^{2-\eta}}-\frac{E_0|t|^{(1+\eta)\nu}}{(1-\eta)x^{1-\eta}R_0^3}\right], \qquad (7)$$

$$r_p^\pm=\pm\frac{1}{2}\frac{K_A}{|k_F-K_A|}\frac{p^2}{(k_F-K_A)^2}, \qquad (8)$$

where $p = m\Delta/2\hbar^2K_A$ and Δ is the width of the gap and is proportional to the sublattice magnetization. The form of the critical anomaly of the thermoelectric power is determined by formulas (6) and (7). Assuming the correlator to be symmetrical with respect to the Néel point, we obtain an anomaly of the maximum type in the thermoelectric power. The value of this maximum depends on the value of D_∞. Assuming, for example that $D_\infty = 4$,[15] $K_A \approx 2k_F$, $q_0 \approx 0.02K_A$, and $\rho_s \approx \rho_{ph}$ we obtain for the critical anomaly a maximum value close to 10% of the total thermoelectric power.

For the temperature coefficient of the thermoelectric power we can write down the expression

$$\frac{d\alpha_s^\pm}{dT}=\begin{cases}(A_1+A_2)|t|^{1-\alpha-\gamma}+A_3, & T<T_N\\ A_1|t|^{1-\alpha-\gamma}+A_3, & T>T_N.\end{cases} \qquad (9)$$

In this formula the terms with the coefficients A_1 and A_2 correspond to the contributions made to the thermoelectric power by the critical scattering and the gap, and A_3 corresponds to the contribution of the usual scattering mechanisms. These coefficients depend on the temperature, but in a narrow region near T_N, where the critical anomaly sets in, this can be disregarded. In formula (9), α and γ are the critical exponents.[15] Formula (9) at $T < T_N$ differs from the expression given[11] (with a reference to a private communication from Auslus and Kawasaki). The difference lies in the critical exponents in the gap term. The reason for this is that in the formula used in[11] the number of carriers is assumed to be proportional to the width of the gap Δ. We, on the other hand, use the result of[16] where, assuming an isotropic gap, the number of carriers is proportional to Δ^2 and, in addition, account is taken of the dependence of the relaxation time on the width of the gap. As a result, the increments to the resistance and to the thermoelectric power are proportional to Δ^2, and it is this which leads to the different critical exponents. In the case when $K_A \sim k_F$, the effect of the gap on the thermoelectric power is quite appreciable (see Fig. 1). The associated anomaly manifests itself in the entire antiferromagnetic region, whereas the critical anomaly appears only in a very narrow interval of temperatures near T_N. In the case of a large gap anomaly, the critical anomaly becomes smeared out and a small hump appears in the thermoelectric power rather than a peak. We note also that in addition to the critical anomaly due to scattering by the long-range spin fluctuations, there is in the thermoelectric power an additive anomaly of the ferromagnetic type. At small K_A one should therefore observe near T_N a jump of the thermoelectric power in place of a peak.

3. THERMOELECTRIC POWER OF GADOLINIUM AND TERBIUM NEAR THE MAGNETIC CRITICAL POINTS

Samples of Gd and Tb single crystals measuring $2 \times 2 \times 15$ mm were cut by the electric-spark method along the directions [11$\bar{2}$0] and 8° from the [0001] axis for Gd, and along [0001] and [10$\bar{1}$0] for Tb. The respective ratios of the room-temperature and liquid-helium resistivities were 51, 32, 28, and 25. The thermoelectric power was measured by a potentiometer method in a quasistationary regime (thermal drift rate 0.2°K/min). The error in the determination of the absolute values of the thermoelectric power did not exceed 10%, and the scatter of the points relative to the smoothed curve did not exceed 0.5%. The results of the experiments are shown in Fig. 2. The temperature dependence of the thermoelectric power of gadolinium agrees qualitatively with the results of[12], but the anomaly in the [11$\bar{2}$0] direction turned out to be more appreciable. The aniso-

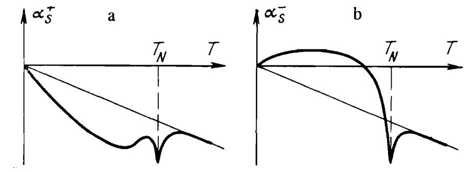

FIG. 1. Temperature dependence of the thermoelectric power of an antiferromagnetic metal ($l < 0$, $k_A \sim k_F$); a) gap above the Fermi level, b) gap below the Fermi level.

tropy of the thermoelectric power of terbium was apparently never investigated before. We observed in Tb along the [10$\bar{1}$0] axis anomalies of the maximum (in absolute value) type on going through T_N and T_C. These anomalies were much less pronounced along the hexagonal axis.

It is known from investigations of the resistivity of terbium that the gap exerts no significant influence on the $\rho(T)$ dependence. If it is assumed that its influence on the thermoelectric power is also small, then the anomalies near T_N and T_C can be associated with critical-scattering effects. An interesting feature of terbium is that the maxima of its thermoelectric power are observed in the basal plane and there are none in the direction of the hexagonal axis, along which the antiferromagnetism vector is directed. The possible explanation of this singularity is that the critical scattering by long-range order fluctuations is effective in the K_A direction if $K_A \approx 2k_F$ (see Fig. 3). On the other hand if $K_A \approx k_F$ the large-angle scattering takes place at an angle to the vector K_A and an anomaly of the ferromagnetic type should apparently be observed along the c axis. It is possible that this latter case is realized in terbium. We note that in the ferromagnetic gadolinium ($K_A = 0$) the stronger anomaly is also observed in the basal plane and not in the direction of the hexagonal axis. In addition, the assumption that the thermoelectric power depends little on the gap is not sufficiently well founded. The point is that the influence of the gap on the thermoelectric power [16] and on the ordinary Hall effect [17,18] is much stronger than on the resistance, and the available experimental data [19m, 20] seem to offer evidence of a strong gap anomaly in the usual Hall coefficient of Tb.

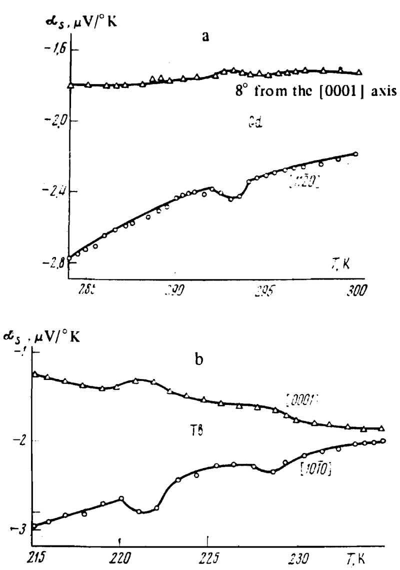

FIG. 2. Thermoelectric power of single-crystal Gd (a) and Tb (b) near the magnetic critical points (the directions are indicated on the plots).

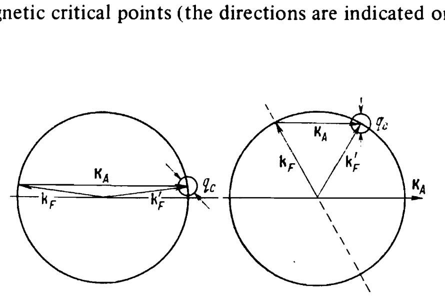

FIG. 3. Large-angle scattering processes in the case of a spherical Fermi surface at $q \approx 0$: a) $K_A \approx 2k_F$, b) $K_A \approx k_F$.

We note finally that anomalies of the maximum type are observed in terbium not only near T_N but also in the region of T_C, although no second-order phase transition occurs here. It is possible that the anomaly is due to the vanishing of the gap. It is not excluded, incidentally, that the cause of the anomaly is the vanishing of one of the branches of the spin excitations. [21]

We note in conclusion that additional investigations are needed to determine more fully the causes of the observed anomalies. In particular, in the calculation of the thermoelectric power it is necessary to take into account the anisotropy of the effect.

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Translated by J. G. Adashko
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