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Thermopower Anomaly in $GdNi_2$: Spin Scattering Model Versus Static Entropy Model*

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(Received 8 November 1972)

The thermopower anomaly in the vicinity of the ferromagnetic critical point of $GdNi_2$ has been measured and found to be of the form predicted by the spin disorder scattering model proposed recently by Thomas, Levin, and Parks. These results allow a vivid comparison between the above model and the idea that the anomalous thermopower measures the static electron entropy in metals, which has been recently discussed by Tang, Kitchens, and Craig.

In a recent Letter the critical thermopower of a metal in the vicinity of a second-order phase transition was worked out.[1] The basic approach was to take the formula for the thermopower derived by Mott and Jones,[2] $Q \propto |d\rho/d\epsilon|_{\epsilon_F}/\rho$, and assume that the critical portion of the resistivity ρ can be described by the Born scattering of the conduction electrons from localized critical fluctuations. The final result may be written in the form

$$\rho Q/T = A\rho_n + B\rho_c + C\,\Gamma(2k_F, T), \tag{1}$$

where ρ_n is the normal resistivity, ρ_c is the critical resistivity given by

$$\rho_c \propto k_F^{-4} \int_0^{2k_F} \Gamma(k,T) k^3\, dk, \tag{2}$$

$\Gamma(k,T)$ is the spin-spin correlation function, and A, B, and C are constants. Results were presented in Ref. 1 for the critical thermopower anomaly in β-brass. While the results appeared to be more compatible with the above analysis than the static entropy model,[3-5] which predicts a strict correspondence between the thermopower anomaly and the specific heat anomaly, data scatter due to the smallness of the anomaly precluded distinguishing between Eq. (1) and the less precise result,

$$\rho Q/T = A\rho_n + D\rho_c. \tag{3}$$

Note from Eq. (2) that $\Gamma(2k_F,T)$ and ρ_c are roughly similar.

Previous studies of $GdNi_2$ have revealed that it behaves in a unique manner, in that mean-field behavior appears to hold unusually close to T_c, which turn implies an unusually long spin-spin force range. This is manifested in the magnetic susceptibility as a decrease in the value of the critical exponent γ with inceasing temperature close to and above T_c,[6,7] and in the electrical resistivity as a dramatic change in the slope of $d\rho/dT$ close to and above T_c.[8,9] Because of the latter feature, $GdNi_2$ is a particularly appropriate system in which to test the two models discussed above, since the predictions of the two models are strikingly different.

The $GdNi_2$ sample used in the experiment was prepared by arc melting 99.99% Gd and 99.95% Ni in an argon atmosphere. The sample was annealed for 24 h at 1000°K, then cut into a rectangular shape, 16mm×2mm×3mm, suitable for the thermoelectric power (TEP) measurements. Typical values of the temperature difference used

in the experiment were 0.3°K close to T_c and 0.5°K away from T_c. The absolute TEP for $GdNi_2$ was determined by subtracting the TEP of copper, since copper voltage leads (spot welded to the sample) were used to monitor the TEP. Since the TEP of copper is only 7–8% that of the total TEP, this procedure introduces little error. The accuracy in measuring the TEP was approximately 0.5%.

The value of T_c, viz., $T_c = 74.2$°K, suggested by the peak in $d\rho/dT$ (see below) agrees exactly with the value determined from magnetic susceptibility measurements[7] made on a sample from the same initial boule of $GdNi_2$. The observed sharp drop in the magnetization at T_c (unpublished) and the sharpness of the peak in $d\rho/dT$ (Fig. 1) are indications that the sample is highly homogeneous.

The results for the thermopower are shown in Fig. 2, and for the quantities $d(Q\rho/dT)/dT$ and $d\rho/dT$ in Fig. 1. The derivative with respect to temperature of the measured TEP, as well as

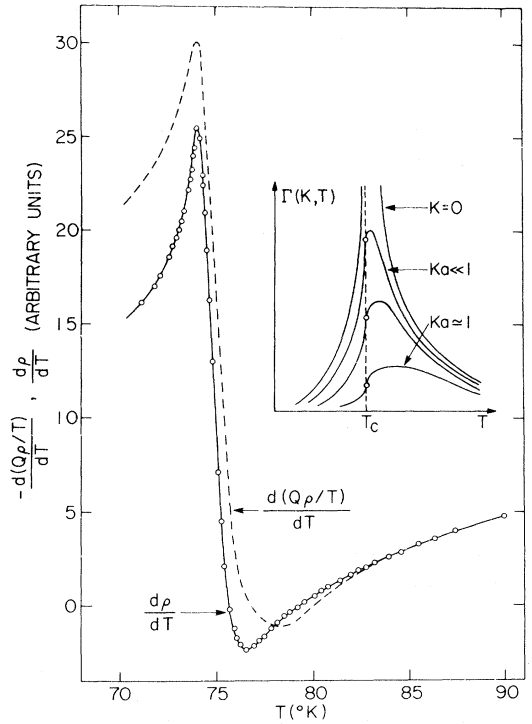

FIG. 1. Temperature derivatives of the resistivity ρ and the quantity $Q\rho/T$ of $GdNi_2$, where Q is the thermopower. The curves have been arbitrarily normalized and arbitrarily shifted relative to each other in the vertical direction. The line through $d\rho/dT$ data represents an eyeball fit. Inset shows expected form of spin-spin correlation function in the vicinity of T_c (after Fisher and Langer, Ref. 10).

of the resistivity, was obtained by computer analysis, whereby a least-squares fit by a polynomial was obtained through a fixed number of data points. The derivative at a given point was then obtained from analytical differentiation of the fitted polynomial. If a linear background term $\rho_n \propto T$ is assumed, the temperature derivative of Eq. (1) takes the form

$$d(\rho Q/T)/dT = A' + B\, d\rho_c/dT + C\, d\Gamma(2k_F, T)/dT. \quad (4)$$

The present results are especially interesting in that they distinguish at least qualitatively between Eq. (4) and the more approximate result,

$$d(\rho Q/T)/dT = A' + B'\, d\rho_c/dT \quad (5)$$

which was used in Ref. 1 in the analysis of the β-brass results. This can be appreciated by referring both to Eq. (2) and to the inset in Fig. 1, after Fisher and Langer,[10] which shows the qualitative behavior of the correlation function for different momenta. Roughly speaking, $\Gamma(2k_F, T)$ should resemble the lowest curve (since $2k_F \approx 2\pi/a$), whereas ρ_c, since it represents a convolution of $\Gamma(k, T)$ over a range of momenta, should resemble a higher curve. Mental differentiation of the curves in the inset implies the following qualitative differences between $d\rho/dT$ (or $d\rho_c/dT$) and $d(Q\rho/T)/dT$, which are borne out in the experimental results: (1) The anomaly in $d\rho/dT$ should be more articulated than that in $d(Q\rho/T)/dT$; (2) the minimum in $d\rho/dT$, which reflects the inflection point above T_c in the appropriate $\Gamma(k, T)$

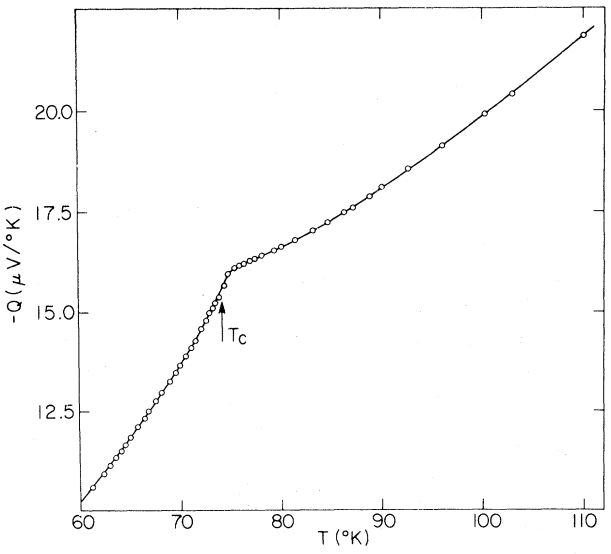

FIG. 2. Absolute thermoelectric power of $GdNi_2$.

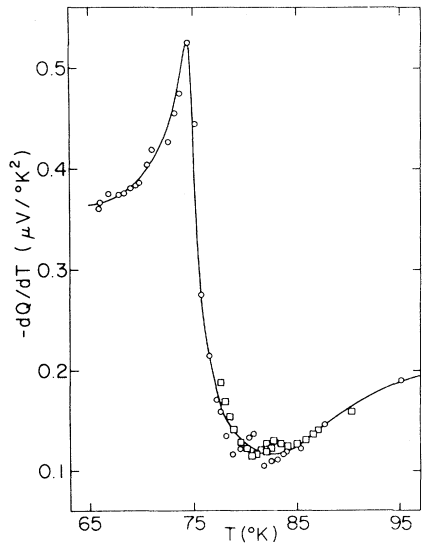

FIG. 3. Temperature derivative of absolute thermopower of $GdNi_2$. The circles correspond to points determined from 11-point polynomial fits to data, which span temperature intervals of approximately 2°K, for $|T-T_c| \lesssim 2$°K; and 27-point polynomial fits for $|T-T_c| \gtrsim 2$°K. The squares correspond to points determined from 39-point polynomial fits to data. The line through the data is an eyeball fit.

curves, should occur closer to T_c than that in $d(Q\rho/T)/dT$.

It is enticing to attempt to apply the following more provocative form of Eq. (1):

$$Q/T = \alpha + \beta\rho_n/\rho + C\,\Gamma(2k_F, T)/\rho, \tag{6}$$

where α and β are new constants, since, in principle, this can lead to the explicit determination of $\Gamma(2k_F, T)$ from measurements of both the thermopower and resistivity. In the present instance our attempt to do this was thwarted by our inability to determine sufficiently accurately the background term ρ_n.

For the purpose of discussing the possible applicability of the static entropy model discussed in Refs. 3–5, the quantity $-dQ/dT$ for the $GdNi_2$ data is shown versus T in Fig. 3. This model focuses on the relationship, $dQ/dT = -dS^*/e\,dT$, where S^* is the transport entropy, and proposes that in the critical region S^* is formally the same as the static entropy S, related to the heat capacity of the electrons by $C_e = T\,dS/dT$; hence,

$$dQ/dT = -C_e/eT. \tag{7}$$

The rationale for the latter hypothesis comes from the apparent similarity[11] of the specific heat and thermopower in the critical region of nickel[3,5] and a-axis gadolinium.[4,5]

The results in Fig. 3 are inexplicable in terms of the static entropy model, which demands that the critical portion of dQ/dT be a monotonically decreasing function above T_c, since the critical part of C_e is expected to mirror the functional form of the critical specific heat of the localized spin system. The nonmonotonicity (dip) observed is clearly a property of the critical portion of the TEP and not the result of a superposition of a monotonically decreasing critical part and a monotonically increasing noncritical part. The latter possibility would require the existence of a much more rapidly varying background term than indicated by the results for $T \ll T_c$ (see Figs. 1 and 3). The spin scattering model discussed above explains the observed anomaly throughout the relatively large temperature interval spanned by the data. Hence, we conclude that the proper description of the critical thermoelectric power is found in Eq. (1) and not in the static entropy model. This does not rule out the possibility that $T\,dQ/dT$ and C_e are of rather similar functional form[12] in the immediate vicinity of T_c, since C_e and $d\rho_c/dT$ are expected to diverge with the same exponent[10,13] in the limit $|\epsilon| \to 0$, where $\epsilon = (T - T_c)/T_c$ and since, roughly, from Eq. (5), $dQ/dT \sim d\rho_c/dT$, provided that $\rho_n(T_c) \gg \rho_c(T_c)$.

We are grateful to Paul Horn and Konrad Schürmann for helpful discussions and to the authors of Ref. 5 for providing a preprint of that paper before publication.

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*Work supported in part by the U. S. Office of Naval Research under Contract No. N00014-67-A-0398-0010.

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Inequality Relating the Ground-State Energies of Two and Three Bosons

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(Received 11 October 1972)

We report sufficient conditions for the validity of the inequality $E_0(3) \leq 3E_0(2)$ for the ground-state energies of two and three nonrelativistic identical bosons interacting via spherically symmetric pair potentials.

Let $E_0(2)$ and $E_0(3)$ be the ground-state energies for two and three nonrelativistic identical bosons interacting by spherically symmetric pair potentials. We have established sufficient conditions for the relation

$$E_0(3) \leq 3E_0(2) \tag{1}$$

to be valid. Our conditions are stated in terms either of properties of the pair potential $V(r)$ or of properties of the ground-state solution[1] $\varphi(r)$ of the two-body Schrödinger equation for particles of mass m,

$$-(\hbar^2/m)\nabla^2\varphi + V(r)\varphi = E_0(2)\varphi. \tag{2}$$

Any of the following conditions are sufficient to prove Eq. (1). (i) $V(r)$ has one minimum and no other local extrema; that is, there is some separation R (R may be zero) for which the following conditions are valid:

$$\begin{aligned} &dV/dr \leq 0, \quad 0 \leq r \leq R; \\ &dV/dr \geq 0, \quad R \geq r < \infty. \end{aligned} \tag{3}$$

(ii) $F(r)$ defined by

$$F(r) \equiv r\varphi^2(r) \tag{4}$$

has one maximum. (iii) $F(r)$ defined by Eq. (4) satisfies the following conditions:

$$\begin{aligned} &F(r) = 0, \quad r \leq \sigma; \\ &dF/dr \leq 0, \quad r \geq 2\sigma. \end{aligned} \tag{5}$$

The proofs are based on the use of the Jastrow-type trial function for three particles,

$$\psi(1,2,3) = \varphi(r_{12})\varphi(r_{23})\varphi(r_{31}) \tag{6}$$

(the r_{ij} are the interparticle separations), in a Rayleigh-Ritz upper bound for $E_0(3)$. We first obtain

$$E_0(3) \leq 3E_0(2) - (3\hbar^2/4m)\int d^3r_1\, d^3r_2\, d^3r_3\, \varphi^2(r_{23})[\nabla_1\varphi^2(r_{12})]\cdot[\nabla_1\varphi^2(r_{13})] \times\left[\int d^3r_1\, d^3r_2\, d^3r_3\, \varphi^2(r_{23})\varphi^2(r_{12})\varphi^2(r_{13})\right]^{-1}. \tag{7}$$

After some algebra we obtain

$$E_0(3) - 3E_0(2) \leq (9\hbar^2/8m)J/N, \tag{8}$$

where

$$J = \int_0^\infty dr \int_0^\infty ds\, F(r)F(s)(d/dr)F(r+s), \quad N = \int_0^\infty dr \int_0^\infty ds \int_{r-s}^{r+s} dt\, F(r)F(s)F(t). \tag{9}$$