PHYSICAL REVIEW B VOLUME 9, NUMBER 9 1 MAY 1974

Specific heat and resistivity of iron near its Curie point

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(Received 27 December 1973)

The specific heat and the temperature derivative of electrical resistivity of Fe have been measured simultaneously using an ac technique. The results for Fe demonstrate that the magnetic specific heat and the temperature derivative of the magnetic contribution to the resistivity are proportional both above and below the Curie point. The critical exponents are found to be $\alpha = \alpha' = -0.120 \pm 0.01$.

I. INTRODUCTION

An earlier work[1] presented new data for the specific heat of iron as part of a general survey of experimental data for Heisenberg-model magnets. These data were used in a test of universality and scaling for the magnetic order transformations in these solids. The specific-heat data for iron were taken simultaneously with a measurement of the temperature derivative of the resistivity. Since the resistivity data were deemed extraneous to the subject of the earlier work, their presentation and discussion has been reserved for this article.

Fisher and Langer[2] have predicted that the change in the temperature derivative of the electrical resistivity $d\rho/dT$ at a magnetic phase transition is related to the change in the specific heat C_P. In particular, they point out that in the region above the critical temperature T_c the dominant singularity in the resistivity is the same as that in the magnetic energy. Therefore, they predict that the critical parts of $d\rho/dT$ and C_P are proportional when $T > T_c$ (that they both vary as $|t|^{-\alpha}$, where $t \equiv T/T_c - 1$). The same conclusion also applies for the region below T_c if one considers only the coherent part of the scattering. However, since the spontaneous magnetization $M_0(T)$ is finite below T_c, there should also be a contribution to the resistivity from incoherent scattering which is proportional to $[M_0(T)]^2$. Thus, Fisher and Langer predict that incoherent scattering should contribute a term to $d\rho/dT$ which varies as $|t|^{2\beta-1}$.

Simons and Salamon[3] have shown that the behavior of β brass (a binary alloy) near its order-disorder transformation conforms to an adaptation of the Fisher and Langer results. They observe a direct proportionality between $d\rho/dT$ and C_P over a range of five decades in t above *and* below T_c. The temperature-dependent term from incoherent scattering is absent because the average effective potential for the alloy is temperature independent.

The correspondence between the predictions of Fisher and Langer and the available experimental results for ferromagnetic materials has not been as close.[4-7] Nickel has been extensively investigated[4-6,8] and the results seem to indicate that the contribution of incoherent scattering to $d\rho/dT$ below T_c is not significant. This conclusion is based on the fact that the critical exponents for $d\rho/dT$ appear to be in approximate agreement with those for the magnetic specific heat above *and* below T_c.

The present work will analyze new data for $d\rho/dT$ and C_P of iron in terms of the Fisher and Langer theory. Because the data for $d\rho/dT$ and C_P have been taken simultaneously, the functional dependence of the resistivity and specific heat may be found directly without resorting to a comparison of critical exponents which have been separately determined. In this sense the present work is free from a serious qualification of the earlier efforts[4-6] to compare experimental results for nickel with the predictions of Fisher and Langer.[2]

II. EXPERIMENTAL RESULTS

The samples used in this work were single crystals of 99.99% purity. The specific heat and the temperature derivative of resistivity were taken simultaneously using an ac technique. A more detailed account of the experimental procedure and apparatus has been presented in a previous work.[1]

The data are presented in Table I which lists the simultaneous value of T, $d\rho/dT$, and C_P. The data for $d\rho/dT$ and C_P have been normalized to the data of Kraftmakher and Romashina at 1075 K.[7,9] Figure 1 shows the behavior of $d\rho/dT$ as a function of temperature near T_c. The curve displays a great similarity to the behavior of C_P near T_c,[1] although the rounding in the peak of $d\rho/dT$ at T_c is somewhat more severe. Part of this effect may be brought about by the fact that C_P is derived from the temperature oscillations at one point on the sample, where $d\rho/dT$ is obtained from the potential difference between two widely separated leads, and hence is more subject to any temperature gradients which may exist.

III. DISCUSSION

It is possible to test the temperature dependence of $d\rho/dT$ versus the temperature dependence of C_P

by graphing $d\rho/dT$ vs C_P directly. Since Fisher and Langer predict a proportionality only between the critical contributions to C_P and $d\rho/dT$, one must eliminate the normal background components which depend on temperature. The normal phonon and electron contribution to C_P can be obtained from the behavior of C_P vs T in a region far from T_c. This procedure results in an estimation of a background term which is linear in t and is approximately equal to $100\,t$ over the range of experimental values.[1]

To a first approximation the background contribution to $d\rho/dT$ should be temperature independent. One expects this background to be composed primarily of the normal electron-phonon term. Interband scattering which would contribute temperature-dependent terms to $d\rho/dT$ is not important in iron.[10] It is, therefore, a good approximation to assume that the temperature dependence of $d\rho/dT$ near T_c arises predominantly from the critical contribution to ρ. Hence, one expects that $d\rho/dT$ will be proportional to $C_P - 100\,t$ at least above T_c.

The quantities $d\rho/dT$ and $C_P - 100\,t$ are plotted as a function of each other in Fig. 2. A least-squares fit to the data below T_c for $t < -10^{-3}$ indicates a slope of $(5.508 \pm 0.021) \times 10^{-3}$ and a linear regression coefficient[11] $r = 1.000$. The fit to the data above T_c gives a slope of $(5.513 \pm 0.045) \times 10^{-3}$ with $r = 1.000$. These results clearly show that over the temperature interval indicated, the specific heat and the temperature derivative of the resistivity are directly proportional. Furthermore, the identical slopes imply that the coefficients of the critical terms above and below T_c are proportionally the same for C_P and $d\rho/dT$. Since it has already been shown[1] that the coefficients of the temperature-dependent terms in C_P above and below T_c are nearly equal $(A^+/A^- = 1.036 \pm 0.015)$, the corresponding coefficients in $d\rho/dT$ must also be nearly equal. The linearity of the results implies that the critical contributions to $d\rho/dT$ and C_P are both proportional to $|t|^{-\alpha}$ where $\alpha = \alpha' = -0.120 \pm 0.01$. The numerical values of α and α' have been previously determined from the data for the specific heat.[1]

It is important to note that the determination of the temperature dependence of $d\rho/dT$ by the technique employed here is relatively insensitive to the choice of T_c. The linearity of the curves shown in Fig. 2 implies an identical temperature dependence for $d\rho/dT$ and C_P. The earlier determination[1] of the form of the temperature dependence of C_P was performed by merging the data above T_c with the data below T_c. Both data sets had been previously shown to be linearly related, indicating the validity of the scaling law $\alpha = \alpha'$. The appropriate value of T_c was determined by maximizing the linearity of

TABLE I. Specific heat and temperature derivative of resistivity of Fe. Results have been normalized to coincide with those in Refs. 7 and 9 for $T \gg T_c$.

T (K)	$\frac{d\rho}{dT}$ (μΩ cm/K)	C_P (J/mole K)	T (K)	$\frac{d\rho}{dT}$ (μΩ cm/K)	C_P (J/mole K)
1010.2	0.1859	52.553	1040.8	0.2610	70.384
1012.5	0.1879	53.187	1041.0	0.2595	71.517
1016.6	0.1920	54.151	1041.2	0.2593	72.474
1018.5	0.1940	54.788	1041.3	0.2421	69.504
1021.7	0.1974	55.804	1041.4	0.2140	64.181
1023.5	0.1995	56.354	1041.6	0.2027	61.691
1025.0	0.2019	56.861	1041.7	0.1951	60.337
1026.7	0.2045	57.587	1042.3	0.1778	57.017
1028.5	0.2079	58.308	1042.9	0.1680	55.483
1030.0	0.2112	59.079	1043.8	0.1592	53.869
1031.7	0.2144	59.890	1045.3	0.1492	52.535
1033.0	0.2180	60.704	1047.5	0.1395	50.986
1034.5	0.2223	61.516	1050.7	0.1297	49.474
1036.5	0.2304	63.283	1052.2	0.1256	49.067
1037.9	0.2378	64.663	1055.4	0.1190	48.340
1038.4	0.2407	65.179	1057.8	0.1153	47.926
1039.0	0.2448	65.958	1060.2	0.1122	47.511
1039.6	0.2509	67.127	1065.4	0.1052	46.810
1039.9	0.2533	67.648	1070.7	0.1002	46.415
1040.2	0.2579	68.517	1075.0	0.0965	46.203
1040.5	0.2604	69.256	1079.7	0.0927	46.029

the results (i.e., by maximizing r). A computer fit to the merged data then gave the numerical value of $\alpha = \alpha'$. The present technique relies only on the validity of the scaling laws and is therefore free from the uncertainties of earlier methods[4-6] which involved matching exponents whose determination was highly sensitive to separate choices of T_c.

The displacement which is seen between the two curves in Fig. 2 is not found in a similar plot for β brass.[3] One might naturally assume that this displacement was related to the presence of a spontaneous magnetization $M_0(T)$ below T_c. The displacement, however, is constant for $t < -10^{-3}$. The magnetization, on the other hand, exhibits a temperature dependence which is given by $|t|^{\beta}$,[12] where $\beta \simeq 0.345$ for Heisenberg magnets.[13] It seems likely, therefore, that the mechanism which gives rise to the displacement is noncritical and depends only indirectly on the details of the temperature dependence of $M_0(T)$. One might speculate that the linear thermal expansion coefficient $\alpha_T \equiv (1/L)(dL/dT)$ contributes a vertical displacement of $d\rho/dT$ for $T \ll T_c$. This quantity exhibits critical behavior near T_c, but also undergoes a step change in its constant background term at T_c.[14] If one ignores the possible band-structure changes which might be brought about by the expansion of the lattice, the correction term in $d\rho/dT$ may be easily shown to be $2\rho\alpha_T$. This term, however, has a magnitude of approximately 2×10^{-3} $\mu\Omega$ cm/K near T_c, too small to account for the observed behavior. Other possible contributions to the step change in $d\rho/dT$ are the formation of magnetic domain boundaries or band-structure effects which affect scattering. But the exact explanation remains elusory.

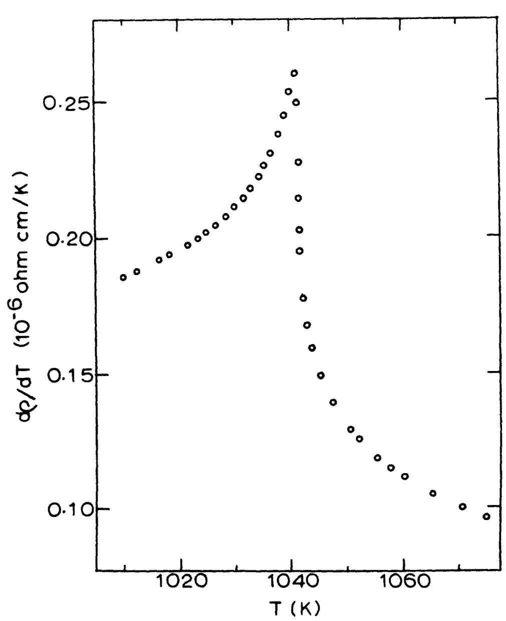

FIG. 1. Temperature derivative of the resistivity of iron plotted as a function of temperature near T_c. Data have been normalized to those in Ref. 7.

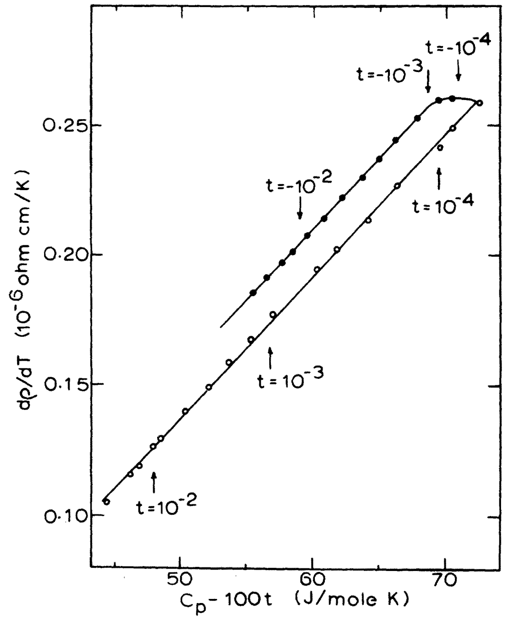

FIG. 2. Temperature derivative of resistivity plotted as a function of specific heat after the linear lattice background has been subtracted. The linearity of the curves suggests the same temperature dependence for $d\rho/dT$ and $C_p - 100t$.

IV. CONCLUSIONS

The analysis of the data for the specific heat of Fe,[1] and the data for C_P vs $d\rho/dT$ (present work), shows that there is a proportionality between the contributions of spin fluctuations to $d\rho/dT$ and C_P. The proportionality above T_c persists over the entire region studied $10^{-5} \lesssim t \lesssim 3\times10^{-2}$. The linear dependence of $d\rho/dT$ on C_P is preserved even into the region where the peaks are rounded $t \lesssim 10^{-4}$. The data below T_c indicate a proportionality which persists over a range $8\times10^{-4} \lesssim |t| \lesssim 3\times10^{-2}$. If one assumes the usual power-law divergence for specific heat,[1] then the following form of temperature dependence is indicated for C_P and $d\rho/dT$:

$$C_P^+ = A^+(t^{-\alpha} - 1)/\alpha + B^+ + Ct, \quad T > T_c$$

$$C_P^- = A^-(|t|^{-\alpha'} - 1)/\alpha' + B^- + Ct, \quad T < T_c$$

$$\frac{d\rho^+}{dT} = a^+(t^{-\alpha} - 1)/\alpha + b^+, \quad T > T_c$$

$$\frac{d\rho^-}{dT} = a^-(|t|^{-\alpha'} - 1)/\alpha' + b^-, \quad T < T_c$$

where $\alpha = \alpha' = -0.120 \pm 0.01$,[1] $C = 100.0$ J/mole K,[1] and $A^+/A^- = 1.036 \pm 0.015^1 \cong a^+/a^-$.

The results above T_c are in complete agreement with the predictions of Fisher and Langer. However, the behavior below T_c shows no indication of the predicted dependence of $d\rho/dT$ on incoherent scattering, which would contribute a term proportional to $[M_0(T)]^2 \propto |t|^{2\beta-1} \cong |t|^{-0.3}$ for Heisenberg ferromagnets. One must conclude that the dominant scattering mechanism giving rise to critical behavior is the same above and below T_c. This mechanism involves the scattering of conduction electrons from short-range static spin fluctuations in the long-range magnetic order. Fisher and Langer have shown that this mechanism leads to a relation for the characteristic scattering time

$$\tau_0/\tau = \Gamma(0, T) + (8k_F^4)^{-1} \int_0^{2k_F} \hat{\Gamma}(K, T) K^3 \, dK , \qquad (1)$$

where Γ is the static spin-spin correlation function and $\hat{\Gamma}$ is its Fourier transform. The leading term $\Gamma(0, T)$ represents the contribution of incoherent scattering which is unity for $T > T_c$ and is proportional to $[M_0(T)]^2$ for $T < T_c$. The present work and the earlier investigations in Ni[4-6] have shown that the assumption[2] that the temperature dependence of the first term would dominate the temperature dependence of $d\rho/dT$ for $T < T_c$ is incorrect. It seems instead that the major temperature dependence arises from the temperature dependence of $\hat{\Gamma}(K, T)$ near $K = 2k_F$ both above and below T_c.

Richard and Geldart[15] have recently suggested that, if the mobile charge carriers can be described by a single band lying inside the first Brillouin zone, there exists a sum rule for the oscillatory electronic function

$$\Phi(\vec{R}) = f(\vec{R})\, p(\vec{R}) ,$$

(see Ref. 2) such that

$$\sum_{\vec{R}} \Phi(\vec{R}) = 0 . \qquad (2)$$

This sum rule arises quite simply from the fact that any perfectly periodic potential cannot contribute a resistance. The imposition of an extra periodic potential resulting from magnetic ordering causes a change in the electronic wave function, which may alter the cross sections of existing scattering mechanisms, but does not itself contribute an extra term to the resistivity.[15] Richard and Geldart show that the sum rule, Eq. (2), requires that the magnetic resistivity below T_c must have the form

$$\frac{\rho(T)}{\rho_0} = 1 + \sum_{\vec{R} \neq \vec{0}} \langle \vec{S}_{\vec{R}} \cdot \vec{S}_{\vec{0}} \rangle \frac{\Phi(\vec{R})}{S(S+1)} . \qquad (3)$$

This expression states that the contribution of incoherent scattering ($\vec{R} = 0$) is temperature independent. The sum rule requires that the temperature-dependent term [see Fisher and Langer,[2] Eq. (10)] in the full expression for incoherent scattering must be zero. The experimental results for Fe and Ni[4-6] are in accordance with these predictions.

ACKNOWLEDGMENTS

The author gratefully acknowledges many helpful discussions of this work with M. B. Salamon and H. Ashworth.

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