TRANSPORT PROPERTIES AT CRITICAL POINTS: THE RESISTIVITY OF NICKEL

Paul P. Craig*
Brookhaven National Laboratory, Upton, New York†

and

Walter I. Goldburg
University of Pittsburgh, Pittsburgh, Pennsylvania

and

T. A. Kitchens
Brookhaven National Laboratory, Upton, New York†

and

J. I. Budnick
Fordham University, New York, New York
(Received 26 October 1967)

The divergence of the temperature coefficient of resistivity of nickel at the Curie temperature is emphasized. Application of the modified Ornstein-Zernike spin-pair correlation function is shown to yield results which cannot be made consistent with both resistivity and susceptibility results.

The decrease in electrical resistance which is observed upon cooling a ferromagnet through the Curie temperature T_C was discussed many years ago by Mott[1] and has been reviewed recently.[2,3] The relationship between critical fluctuations and resistivity has received occasional theoretical attention, but the experimental result that the temperature coefficient of resistivity [$\alpha(T) \equiv d\rho/dt$, where ρ is the resistivity] is divergent at T_C appears to have been overlooked. We find $\alpha(T)$ to be positive both above and below T_C, and to diverge logarithmically above T_C with the strength of the divergence increasing within a few degrees of the transition. This is contrary to the predictions of Kim[4] and of de Gennes and Friedel.[5] These authors find the resistivity to exhibit a cusp at the Curie temperature, so that the temperature coefficient of resistivity changes sign at the transition. Although a logarithmic divergence of the resistivity is predicted, the sign of $\alpha(T)$ above the transition is negative, in disagreement with experiment.

The singular behavior of $\alpha(T)$ was apparently first noted by von Bohlen-Halbach and Gerlach,[6] whose results we reproduce in Fig. 1 [$\alpha(T)/\rho$ is plotted]. No critical discussion of these results seems ever to have been published. Although the data points are sparse, and the Curie temperature differs somewhat from present-day values, the solid curve is well represented some distance above the Curie temperature by an expression of the form $-A \ln|T-T_C| + B$. Experiments illustrating the logarithmic divergence both above and below T_C have recently been reported by Kraftmakher.[7] Divergent behavior is typical of equilibrium thermodynamic quantities such as specific heat near second-order phase transitions.[8] It now appears that divergences occur in transport quantities as well.

In order to characterize the critical behavior near T_C, we have directly measured $\alpha(T)$ in five-9's purity Ni, using an ac resistance bridge[9] (usually operated at 33 cps) in which two of the arms contained identical Ni samples

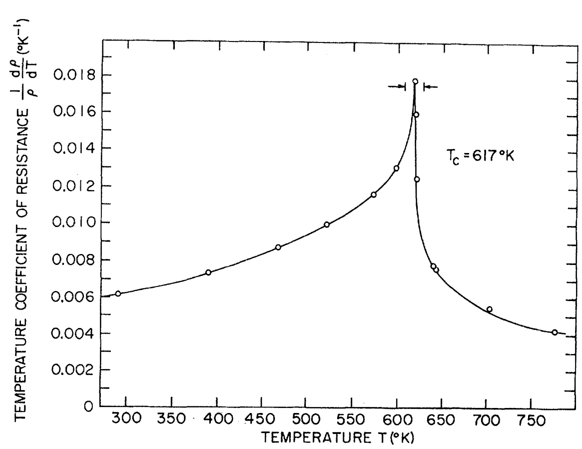

FIG. 1. The temperature coefficient of resistivity of Ni was measured over three decades ago by a student of Gerlach. The results, reproduced here, illustrate qualitative features (the divergence near T_C) relevant to transport theory today.

held at a fixed temperature difference of about 0.25°K. As the temperature of the samples was varied, $\alpha(T)$ was recorded versus T on an x-y recorder. A portion of a trace is shown in Fig. 2. The interval presented includes about 10°K above and below T_C. (For comparison, this interval is indicated by the vertical lines in Fig. 1.) Temperatures were measured with copper-Constantan thermocouples. No attempt was made to determine absolute temperatures accurately. Slight hysteresis of unknown origin was observed in a region including a few degrees about the Curie temperature. Similar behavior was reported by Gerlach.[6] The data reported here were obtained during cooling through the transition.

In analyzing critical-region results, small errors in T_C can significantly affect the conclusions. We have, therefore, determined that T_C occurs within 0.2°K of the maximum of $\alpha(T)$ by simultaneously measuring $\alpha(T)$ and a quantity related to the magnetic susceptibility. This latter quantity was the high-frequency resistivity, which was measured at 40 kc/sec. Although the sample was noninductively wound, residual inductance produced reactance at high frequencies, and hence phase shifts near the Curie point where the sample susceptibility approaches infinity. (It was verified that no frequency-dependent effects occurred at low frequencies.) Thus, the curve of high-frequency resistivity versus T shows a peak at T_C.[9] The width of the transition measured in this way was about 1°K, in agreement with the $\alpha(T)$ results. The maxima in the two types of measurements, which we identify as T_C, occurred within about 0.2°K of each other. This discrepancy is too small to effect the data analysis significantly.

In order to analyze the data, we represent $\alpha(T)$ in a manner proposed by Fisher[10] for other divergent quantities:

$$\alpha(T) = (A/\lambda)(\epsilon^{-\lambda} - 1) + B. \tag{1}$$

Here $\epsilon \equiv |T - T_C|/T_C$, and A, λ, and B are constant or are only weakly temperature dependent. The Grüneisen function expressing phonon scattering yields $\rho(t) \sim T$, and hence $B = \text{const}$. When $\lambda = 0$, the divergence is logarithmic. Data analysis is simplified and quantities showing a slow variation with T are minimized in importance if we study

$$d\alpha(T)/d\epsilon = -A\epsilon^{-(\lambda+1)}. \tag{2}$$

Figure 3 is a log-log presentation of $d\alpha(T)/d\epsilon$ vs ϵ obtained through differentiation of Fig. 2 for $T > T_C$. For $\epsilon \gtrsim 5 \times 10^{-3}$, the slope $-(\lambda + 1)$ is unity, implying that $\lambda = 0$ and that there is a logarithmic divergence. However, for $\epsilon \lesssim 5 \times 10^{-3}$ the divergence becomes more rapid. While our experimental resolution precludes

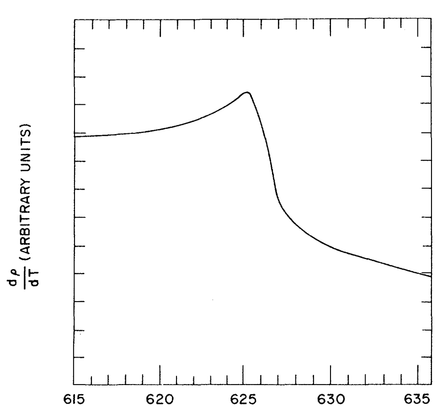

FIG. 2. Modern data on $\alpha(T) \equiv d\rho/dT$, the temperature coefficient of resistivity, in the immediate neighborhood of the Curie temperature. The region shown is approximately that indicated by the vertical lines in Fig. 1.

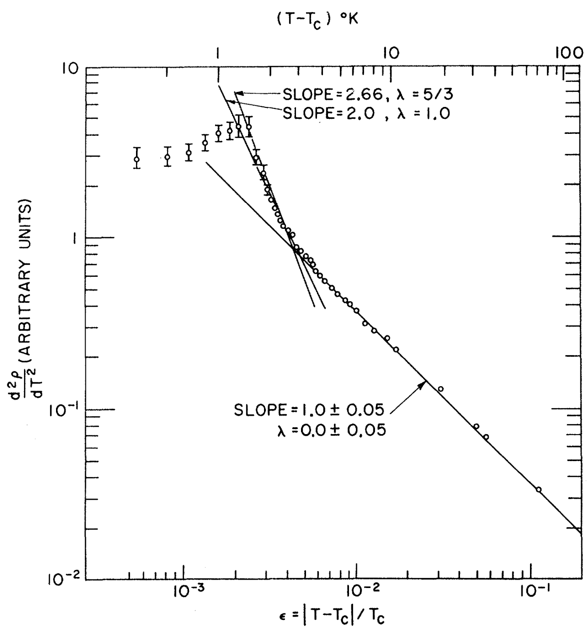

FIG. 3. The slope of the data of Fig. 2 above T_C, when plotted logarithmically versus $\epsilon \equiv |T - T_C|/T_C$, yields $\lambda + 1$, where λ is a critical exponent describing the divergence of $\alpha(T)$. A logarithmic divergence is found for $T - T_C \gtrsim 3$°K. The strength of the divergence increases near T_C.

precise statements about the behavior in this region, the data are consistent with $5/3 > \lambda > 1$, implying an approximately linear divergence of $\alpha(T)$.

The crossover between the two types of behavior seems to occur when the range of the critical fluctuations (ξ) roughly equals the electronic mean free path (l). Near the transition, the fluctuations grow in size. If we assume that the crossover occurs when these two lengths become comparable, we may derive a value for the coefficient a_0 in the expression for the temperature dependence of the fluctuation size[8] ξ: $\xi = a_0\epsilon^{-\nu}$, where ν is a parameter usually determined by neutron scattering [see Eq. (4) below].[8] For our sample, $\rho(T_c) = 25\ \Omega$ cm,[11] implying an electronic mean free path $l \gtrsim 25$ Å. From Fig. 3, the crossover from logarithmic to approximately linear behavior occurs near $\epsilon = 5\times10^{-3}$. From these values we find that $a_0 \approx 0.9$ Å if $\nu = 0.63$,[8] and $a_0 \approx 1.8$ Å if ν has the classical value of $\frac{1}{2}$. Such values for a_0 are not atypical of those found and implied for ferromagnets and for liquid helium.[12,13]

We note that changes in critical exponents near $\epsilon = 0$ have heretofore been found below T_c in Ni[14] and in $YFeO_2$.[15] The mean-free-path criterion presented above suggests that crossover behavior is expected in resistivity and Mössbauer measurements, but not necessarily in specific-heat measurements. The absence of crossover in the specific heat of Ni,[16] either above or below T_c, thus offers support for this proposal.

It is instructive to re-examine the calculation of de Gennes and Friedel[5] and to generalize it slightly. These authors treat the conduction electrons as being scattered by the d spins and show that for $T > T_c$, the magnetic part of the differential scattering cross section as a function of momentum transfer κ is

$$d\sigma/d\Omega = \sum_r e^{i\vec{\kappa}\cdot\vec{r}}\langle \vec{S}_0 \cdot \vec{S}_r\rangle, \tag{3}$$

where $\langle \vec{S}_0 \cdot \vec{S}_r\rangle$ is the static spin-pair correlation function, and inelastic scattering has been neglected. In the work of de Gennes and Friedel, $\langle \vec{S}_0 \cdot \vec{S}_r\rangle$ is approximated using a molecular-field mode. The calculation can be carried out using the Ornstein-Zernike form

$$\langle \vec{S}_0 \cdot \vec{S}_r\rangle = (A/r)e^{-\kappa_1 r}, \tag{4}$$

where the inverse range vector is $\kappa_1 = 1/\xi = (1/a_0)\epsilon^{\nu}$. We convert the sum over lattice sites in Eq. (3) to an integral (a valid procedure near T_c, where ξ is large and the details of the lattice unimportant). Upon integrating over all solid angles and including the resistivity weighting factor $1-\cos\theta$,[5] the temperature coefficient of resistivity is finally found to vary with temperature as

$$\alpha(T) = \frac{T_c - T}{|T_c - T|} B\epsilon^{2\nu-1}\ln\epsilon + C. \tag{5}$$

Here the constant B is positive, and the factor $(T_c-T)/|T_c-T|$ has been included to show explicitly how the sign of $\alpha(T)$ reverses at the transition. The constant C includes both magnetic and phonon contributions to the resistivity. (We have assumed magnetic and phonon scattering to be additive.)

Equation (5) reduces to the result of de Gennes and Friedel when $\nu = \frac{1}{2}$, the classical value. It is in disagreement with experiment for all values of ν. In particular, if one takes $\nu = \frac{2}{3}$, the presently accepted value,[8,12,17] Eq. (5) implies that $\alpha(T)$ should show no divergence near T_c. It thus appears that the above approach, despite its apparent generality, is incapable of dealing with the experimental facts.

The region below T_c has not been discussed here, largely because the behavior appears more complex. The divergence is approximately logarithmic, and evidence is present for a break in the shape of the curves at about $T_c - T \approx 8$°K, where such behavior was seen in the Mössbauer measurements.[14]

Mott[1] has applied density-of-states arguments to a band-theory viewpoint, which omits fluctuations. Above T_c no magnetic resistivity contributions whatever are expected, while below T_c the theory predicts $\alpha(T) \sim \epsilon^{\beta-1}$, where β is the exponent (typically $\frac{1}{3}$) describing the temperature variation of the magnetization.[8] Molecular-field theory predicts a logarithmic divergence below as well as above T_c.[4,5] Although some progress on scaling laws for transport processes has been reported,[18] the results have yet to be applied to situations such as resistivity.

Improved measurements are in progress in the region below T_c. In addition, we are studying the thermal conductivity near T_c in an effort to understand the dramatic breakdown of the Wiedemann-Franz law.

We wish to thank L. P. Kadanoff for suggest-

ing to us the probable relevance of the temperature derivatives of transport quantities, and M. Blume, V. Emery, M. E. Fisher, P. C. Martin, and J. Langer for a number of comments.

Note added in proof.—Dr. M. E. Fisher and Dr. J. S. Langer have independently pointed out to us that while the Ornstein-Zernike model is a reasonable one for large ranges, at short ranges one may expect the resistivity to be controlled by factors involving the system energy. Should this occur, then $\alpha(T)$ is expected to vary approximately with the specific heat, as is qualitatively observed.

---

*Also at State University of New York, Stony Brook, New York.

†Work supported under the auspices of the U. S. Atomic Energy Commission.

[1]N. F. Mott, Proc. Roy. Soc. (London) 156, 368 (1936); N. F. Mott and K. W. H. Stevens, Phil. Mag. 2, 1364 (1957).

[2]T. Kasuya, Progr. Theoret. Phys. (Kyoto) 16, 58 (1956).

[3]B. Cole, Advan. Phys. 7, 40 (1958).

[4]Duk-Joo Kim, Progr. Theoret. Phys. (Kyoto) 31, 921 (1964).

[5]P. G. de Gennes and J. Friedel, J. Phys. Chem. Solids 4, 71 (1958). The arguments are slightly extended and experimental comparisons are made by R. J. Weiss and A. S. Marotta, J. Phys. Chem. Solids 9, 302 (1959).

[6]W. Gerlach, Physik. Z. 24, 953 (1932). Results near T_c were reported later [H. Bittel and W. Gerlach, Ann. Physik 33, 661 (1938)] which are consistent with our finding that the strength of the divergence increases near T_c. Very close to T_c, $\alpha(T)$ linearly approaches a finite limit. Whether this is a real effect or a result of instrumental resolution (we believe the latter) is not certain.

[7]Ya. A. Kraftmakher, Fiz. Tverd. Tela 7, 8 (1965) [translation: Soviet Phys.—Solid State 9, 1199 (1967)].

[8]L. P. Kadanoff, W. Götze, D. Hamblen, R. Hecht, E. A. S. Lewis, V. V. Palciauskas, M. Rayl, J. Swift, D. Aspnes, and J. Kane, Rev. Mod. Phys. 39, 395 (1967).

[9]The infinite susceptibility of ferromagnets at T_c complicates the use of the ac bridge to measure resistivity. Spurious effects were avoided by the use of a low frequency and noninductively wound samples. When used with inductively wound samples or with high frequencies, the ac bridge offers a simple and precise method of determining Curie temperatures (J. I. Budnick, P. P. Craig, and T. A. Kitchens, to be published).

[10]M. E. Fisher, private communication.

[11]S. Arajs, J. Appl. Phys. 32, 97 (1961). The absolute resistivity reported here, and the approximate values for $\alpha(T)$ which may be derived from it, are in agreement with our results, but both Arajs's and our magnitudes for $\alpha(T)$ are substantially lower than those of Gerlach (Ref. 6). This discrepancy is thought to relate to higher purity of modern samples. It does not appear to affect the nature of the divergence of $\alpha(T)$.

[12]L. Passell, K. Blinowski, T. Brun, and P. Nielsen, Phys. Rev. 139, A1866 (1965); D. Cribier, B. Jacrot, and G. Parette, J. Phys. Soc. Japan Suppl. 17, 67 (1962).

[13]J. S. Langer and M. E. Fisher, Phys. Rev. Letters 19, 560 (1967).

[14]D. G. Howard, B. D. Dunlap, and J. G. Dash, Phys. Rev. Letters 15, 628 (1965).

[15]G. Gorodetsky, S. Shtrikman, and D. Treves, Solid State Commun. 4, 147 (1966).

[16]P. Handler, D. E. Mapother, and M. Rayl, Phys. Rev. Letters 19, 356 (1967).

[17]P. Weiss and R. Forrer, Ann. Phys. (Paris) 5, 153 (1926), as reanalyzed by J. S. Kouvel and M. E. Fisher, Phys. Rev. 136A, 1626 (1964).

[18]B. I. Halperin and P. C. Hohenberg, Phys. Rev. Letters 19, 700 (1967).

TEMPERATURE AND FREQUENCY DEPENDENCE OF THE AMPLITUDE OF THE RADIO-FREQUENCY SIZE EFFECT IN GALLIUM*

P. H. Haberland† and C. A. Shiffman‡
Department of Physics, Center for Materials Science and Engineering,
Massachusetts Institute of Technology, Cambridge, Massachusetts
(Received 30 October 1967)

In recent years a number of workers have used the radio-frequency size effect (RFSE) to determine the dependence of the mean collision time on temperature for well-defined groups of carriers on the Fermi surface.[1-4] The underlying assumption used in the analysis of the experimental data has been that the strength of a resonance is proportional to $\exp(-1/\omega_c\tau)$, where ω_c is the cyclotron frequency and τ the mean collision time. This Letter reports measurements of the strength of the RFSE in very pure gallium which cannot be explained in such simple terms. We have found resonances whose strength S increases monotonically with tem-