# Critical Resistivity of Antiferromagnetic Semiconductors*

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The resistivity of nearly stoichiometric iron oxide was measured in the temperature range $78 \leq T \leq 300$°K. The temperature derivative of the resistivity, $d\rho/dT$, in the vicinity of the critical temperature, $T_c$, was found to be proportional to $\epsilon^{-0.4}$, where $\epsilon = |(T - T_c)/T_c|$. The results are explained by incorporating a normalized Ornstein-Zernike correlation function into the de Gennes–Friedel formula for critical scattering.

In the last few years there has been an increasing activity in the study of transport properties in the vicinity of critical points in systems that undergo second-order phase transitions.[1,2] In particular resistivity anomalies associated with critical phenomena were observed experimentally[1-5] and examined theoretically.[6-9] It was found that for some ferromagnetic and antiferromagnetic metals the resistivity anomaly can be characterized by the singular behavior of the temperature derivative of the resistivity, $d\rho/dT$, which in the vicinity of $T_c$ is described by the critical exponents $\lambda$ and $\lambda'$.[10] Unlike these systems, no critical exponents were derived from experiments in antiferromagnetic semiconductors (AS) and the singular behavior of $d\rho/dT$ has not been examined theoretically for these materials.

Here we report the experimental results obtained in nearly stoichiometric $Fe_xO$ where $0.99 \leq x \leq 1.00$. To explain the results we suggest a normalized Ornstein-Zernike (OZ) correlation function where the normalization is based on the spin-correlation sum rule.

The present measurements were carried out on sintered bars of nearly stoichiometric $Fe_xO$. Previous transport measurements that were carried out on the nonstoichiometric ($x \lesssim 0.95$) materials did not show a resistivity anomaly.[11] However, in view of the gradual smearing out of the specific-heat peak[12] when $x$ decreases from 1.00, it was hoped that for $x$ approaching this value the critical behavior might be sampled by the resistivity as it is in similar materials.[13] Recently it was shown that by proper quenching a nearly stoichiometric compound can be prepared.[14] A similar method was followed here; its description as well as the magnetic properties of the resulting compound will be presented elsewhere.[15] The lattice constant of the material was found to be $a_0 = 4.326$ Å. Using the known relation[14] between $a_0$ and $x$ in $Fe_xO$, we established that $0.99 < x < 1.00$. The bars used for the measurements were $8\times2\times1$ mm$^3$. Four contacts were made by soldering indium after wetting the proper areas with indium amalgam. The resistivity was measured using a standard four-probe technique and the temperature was monitored by a copper-Constantan thermocouple. The voltage between the two voltage probes versus the thermocouple voltage was displayed on a recorder. The measurements were carried out in the temperature range $78 \leq T \leq 300$°K. Below 165°K the resistivity has an exponential temperature dependence with an activation energy of 0.09 eV. The continuously recorded resistivity in the temperature range from 165 to 215°K is shown in Fig. 1 by the curve $\rho_1$. Below 185 and above 189°K the resistivity decreases with increasing $T$, while between these temperatures the resistivity increases with $T$. This anomaly must be due to an intrinsic effect since changes such as in the dimensions of the sample at the critical region will cause an effect that is 2 orders of magnitude smaller than the observed one.[15] In Fig. 1 we have also included the curve $\rho_2(T) = \rho_0 \exp(0.09/k_B T)$ which represents the expected temperature dependence of the resistivity if no critical phenomena were present. Here $k_B$ is Boltzmann's constant and $\rho_0 = \rho_1(160°\text{K}) \exp(-0.09/160 k_B)$.[16]

To compare the results shown in Fig. 1 with theory it is useful to examine the quantity $P = (\rho_1 - \rho_2)/\rho_2$. If we assume that the observed anomaly is only due to critical scattering,[17] then $P = \tau_l/\tau_k$, where $\tau_l$ is the mean free time due to all the scattering processes apart from critical scattering. It is assumed hereafter that in the critical

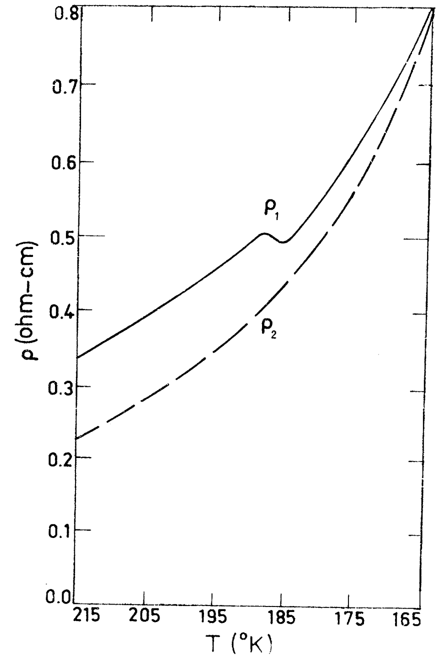


FIG. 1. Recorder trace of the resistivity dependence on temperature of nearly stoichiometric $Fe_xO$ (curve $\rho_1$) and the expected dependence in the absence of critical phenomena (curve $\rho_2$).

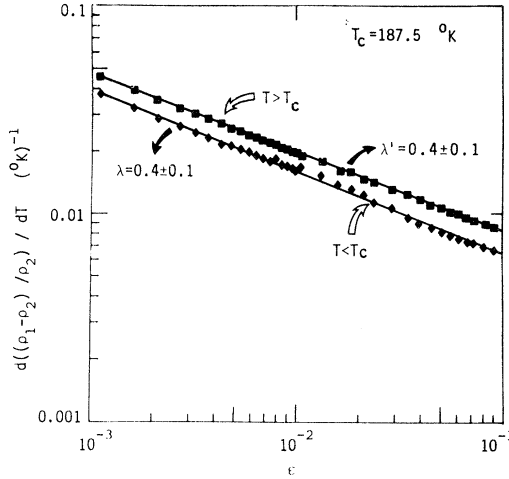


FIG. 2. Log-log plot of the temperature derivative of $(\rho_1-\rho_2)/\rho_2$ as a function of the reduced temperature $\epsilon$. The critical temperature $T_c$ is the temperature for which the derivative has its maximum value.

region $\tau_l$ can be considered to be temperature independent.

Since the critical scattering is characterized[6,8] by $d(1/\tau_k)/dT$ we have differentiated $P$ with respect to temperature. It was found that $dP/dT$ is positive for the temperature range shown in Fig. 1, and that the maximum of $dP/dT$ is at 187.5°K. This temperature was identified as $T_c$. The results both above and below $T_c$ are shown on a log-log scale in Fig. 2. The obtained linear dependence is in accordance with a power-law behavior. The values of the critical exponents derived from the slopes of the straight lines are $0.4\pm0.1$ for both $\lambda$ and $\lambda'$. The experimental values should be considered reliable since the linear behavior of log$P$ extends over 2 orders of magnitude of $\epsilon$ and down to $\epsilon = 10^{-3}$ (this criterion was suggested by Kadanoff *et al.*[18]).[19]

The description of the spin fluctuations by a normalized OZ-type correlation function offers a plausible explanation of the observed power-law dependence and the observed sign of the differential resistivity. Let us consider a two-sublattice AS with a reciprocal-magnetic-lattice vector $\vec{Q}$. In the first-order perturbation and within the elastic approximation the mean free time $\tau_k$ for scattering of carriers by the ion spins in an AS can be written as

$$1/\tau_k = (\pi/2\hbar)N(E_k)\Omega J^2\chi_{AF}(0), \tag{1}$$

where $E_k = \hbar^2k^2/2m$ is the kinetic energy of the carrier. Here $k$ is the carrier wave vector and $m$ its effective mass. $N(E_k)$ is the density of states for a given carrier spin, $\Omega$ is the volume per ion spin, $J$ is the exchange coupling constant between the carrier and the ion, and $\overline{\chi}_{AF}(\vec{q})$ is the Fourier transform of the static spin-spin correlation function[8] of the antiferromagnet. Equation (1) is the de Gennes–Friedel equation[6] when one considers the fact that $q < 2k \ll Q$ for a semiconductor and that for an antiferromagnet $\chi_{AF}(q)$ is a slowly varying function[20] of $q$ (for $q \ll Q$). However,[21] $\chi_{AF}(\vec{q}-\vec{Q}) = \chi_F(\vec{q})$ is the correlation function of the same magnetic lattice, but with an exchange coupling constant $-J$. Hence,

$$(1/\tau_k) = (\pi/2\hbar)N(E_k)\Omega J^2\chi_F(\vec{Q}). \tag{2}$$

In the Ornstein-Zernike approximation[18,22] $\chi_F(\vec{q})$ is given by

$$\chi_F^{OZ}(q) = [S(S+1)/r_1^2(\kappa^2+q^2)], \tag{3}$$

where $1/\kappa = r_1\epsilon^{-\nu}$ is the correlation length, $r_1$ is a constant that depends on the magnetic structure,[23] and $\nu = \frac{2}{3}$. It is known[22] that $\chi_F^{OZ}(q)$ describes well the asymptotic behavior of $\chi_F(\vec{q})$ for $q \ll \kappa$. Outside this region the behavior of $\chi_F(\vec{q})$ can be assessed by using the sum rule $\int_{BZ}\chi(\vec{q})dq^3 = S(S+1) - \langle S\rangle^2$ where the integral extends over the Brillouin zone (BZ). This sum rule is a consequence of the definition[8] of $\chi_F(\vec{q})$. We then approximate $\chi_F(\vec{q})$ by a normalized $\chi_F{}^{OZ}(q)$ which, near the critical point, is given by

$$\chi_F{}^N(q) = 2\pi^2[S(S+1) - \langle S\rangle^2]/\Omega[\Lambda - (\pi/2)\kappa](\kappa^2+q^2). \tag{4}$$

$\chi_F{}^N(q)$ was derived using Eq. (3), the above sum rule, and approximating the Brillouin zone by a sphere of radius $\Lambda$ which has the same volume as that of the BZ.

Using $\chi_F{}^N(Q)$ rather than $\chi_F(Q)$ in Eq. (2), taking the derivative of $1/\tau_k$ with respect to temperature and keeping only the terms that diverge at $T=T_c$, we get

$$d(1/\tau_k)/dT = A\epsilon^{\nu-1} \text{ for } T>T_c \tag{5}$$

and

$$d(1/\tau_k)/dT = B\epsilon^{2\beta-1} - A\epsilon^{\nu-1} \text{ for } T<T_c, \tag{6}$$

where $A=[\pi^2\hbar kJ^2S(S+1)\nu]/(4mT_cE_QE_\Lambda r_1)$ and $B=(2S\Lambda r_1 A)/[\pi(S+1)]$. The term $B\epsilon^{2\beta-1}$ arises from the sublattice magnetization[18,22] $\langle S\rangle$. For $T<T_c$, $\langle S\rangle = S\epsilon^\beta$ with $\beta=\frac{1}{3}$, while for $T>T_c$, $\langle S\rangle = 0$.

The effect of the other interactions of the carrier with the lattice (phonons, impurities, defects) on $\tau_k$, which would arise from higher-order perturbations, can be accounted for phenomenologically[8] by replacing $\kappa$ by $\kappa + 1/l$ in $\chi_F{}^N(q)$, where $l$ is the carrier mean free path in the absence of critical phenomena. Using this procedure it is found that $A$ and $B$ in Eqs. (5) and (6) have to be multiplied by $\{1+(\pi/\Lambda l)[1-(4/\pi^2)\times(\Lambda/Q)^2]\}$ and $[1+(\pi/2\Lambda l)]$, respectively. We have neglected here terms of higher order than $(\Lambda l)^{-1}$.

The divergence given by Eqs. (5) and (6) is in accord with the above experimental results both below and above $T_c$. Further, above $T_c$ the positive sign of $dP/dT$ is in accordance with Eq. (5). Below $T_c$, the experimental positive sign of $dP/dT$ indicates that in our case the positive term arising from the sublattice magnetization dominates. Thus, both above and below $T_c$, the resistivity anomaly can be interpreted in terms of the present model.

In conclusion, we found that the normalized OZ correlation function $\chi_F{}^N(q)$ leads to a good description of the resistivity anomaly of nearly stoichiometric $Fe_xO$ in the temperature range $10^{-1} \geq \epsilon \geq 10^{-3}$. The present work indicates that $\chi_F{}^N(q)$ can be a useful extension of $\chi_F{}^{OZ}(q)$ to $q$ regions where the unnormalized function is not applicable.

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[11]See, for example, I. Bransky and D. S. Tannhauser, Physica (Utrecht) 37, 547 (1967), and Trans. Met. Soc. AIME 239, 75 (1967). From these transport studies it is apparent that the nearly stoichiometric material is $p$-type, and this seems to indicate that the carriers in the present material are holes (I. Balberg and H. L. Pinch, to be published).

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[16]The results to be presented below are not sensitive

(Ref. 15) to the choice of 160°K as the point where $\rho_1 = \rho_2$.

[17]We do not expect any critical effect due to changes in the carrier concentration. In nearly stoichiometric iron oxide the carriers are thermally activated from shallow donor levels (0.09 eV). Any change in band structure due to critical fluctuations will shift the donor levels together with the corresponding band, because shallow donor states can be accurately described in terms of states of the single nearest band.

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[19]It should be noted that in the very close vicinity of $T_c$ the ferromagnetic susceiptibility for $q \neq 0$ is expected to be proportional to $\epsilon^{1-\alpha}$, where $\alpha$ is the critical exponent of the specific heat ($\alpha \lesssim 0.1$) [M. E. Fisher and R. J. Burford, Phys. Rev. 156, 583 (1967)]; however, the present experimental results indicate that in the range of $\epsilon$ studied here, the OZ-type susceptibility is still applicable. The temperature where the transition from the OZ-type behavior to the $\epsilon^{1-\alpha}$ behavior takes place is system dependent. See, for example, P. M. Horn, R. D. Parks, D. N. Lambeth, and H. E. Stanley, Phys. Rev. B 9, 316 (1974).

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[23]When $T_c = \Delta$, where $\Delta$ is the paramagnetic Curie-Weiss temperature, then $r_1 = a_0/\sqrt{6}$ and the OZ correlation function of an fcc antiferromagnet of the second kind can be approximated by $[S(S+1)/r_1^2]/[\kappa^2 + (\vec{q} - \vec{Q})^2]$, where $a_0$ is the lattice constant (see Ref. 19). In $Fe_xO$ it was found experimentally (Ref. 15) that $\Delta \simeq 180$°K while $T_c = 187.5$°K.