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Critical behavior of the electrical resistivity in magnetic systems: Comments and theory

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Following recent theoretical advances on the critical behavior of the electrical resistivity ρ in magnetic systems, this paper points out that features of the critical region are not yet quantitatively explained. The need for a theory away from but close to the transition region is emphasized in view of extracting several parameters from the experimental data. A simple generalization of the Ornstein-Zernike correlation function, on both sides of T_c, based on Landau theory and previous work is proposed. This implies a magnetization-dependent amplitude of the correlation length below T_c. Different types of singularities are found as observed experimentally. Various features of $d\rho/dT$ away from the transition temperature can also be reproduced: In particular, an unexplained bump and dip in $d\rho/dT$ vs T (below T_c) usually obtained from data of ρ in localized spin systems can be found in our simple approach. Also, the existence of unequal magnitudes of $d\rho/dT$ above and below T_c now has a theoretical explanation.

The endeavor in two recent papers published in this journal by Alexander *et al.*,[1] and by Balberg and Helman[2] was to determine and to predict the power-law dependences governing the behavior of the electrical resisitivity, ρ, of magnetic (metallic or semiconducting) materials at various distances $\epsilon = 1 - T/T_c$ from the critical temperature T_c (or T_N). These papers have followed some important theoretical investigation by Geldart and Richard,[3-7] Kasuya and Kondo,[8,9] and others[10-15] on the influence of various parameters on the overall shape of $d\rho/dT$ in the critical region.

Although paving the way for detailed experimental investigations, these estimates[1-15] seem to be still far ahead of most of the experimental data, for by analyzing them (as obtained from various experimental techniques), one can hardly determine with the adequate accuracy (1) the value of the critical indices corresponding to $d\rho/dT$ and (2) the true size of the critical region. One might argue about the need of finer experimental work or "better" data treatment, although in view of recently published data[16] and their subsequent analyses,[17] it appears that even the most precise data currently available are not sufficient to fulfill our hope in discussing conclusively both fundamental questions (1) and (2).

An illustration of these difficulties is given by the different studies of ρ and $d\rho/dT$ on a typical metal like dysprosium.[18-22] They have failed, on one hand, to describe exactly how the transition takes place, and on the other hand, to allow for an unambiguous determination of the critical exponent(s) (see Table I).

In fact, such elaborate studies fall short of their goal because of (a) the nature of the transition itself between an antiferromagnetic phase and a paramagnetic one, (b) the varying dimension of the order parameter. A crossover between a dipolar and a XY system occurs close to T_N,[21] i.e., for $\log_{10}|\epsilon| \leq -2.3$.

For such a transition the critical exponent has been predicted to be very large,[13] although the region in which such an exponent has to be found is rather restricted. Hence the set of data points available for analysis is limited, and cannot be chosen for the specific purpose of obtaining a pleasant value (as done in the article by Balberg and Maman[22]). Furthermore, the smearing (or not) of band gaps appearing (or not)[15] below T_N does not seem to have been fully included in the analysis (even in Ref. 20).

In such a respect, it is interesting to notice the large value of the critical exponent found for $d\rho/dT$ in the case of a *ferromagnet*,[23] in which the electronic density-of-state fluctuations[15] likely play a very important role. This seems also to confirm *a posteriori* the *necessity* of considering two critical terms (and exponents) in doing a numerical analysis,[24] and casts some further doubt on the simple exponent-data analysis in dysprosium.

In view of these remarks, some attempt has been made to obtain a large number of $d\rho/dT$ data points with great precision, and in the truly critical region of the ferromagnetic-paramagnetic transition of an intermetallic compound TbZn.[16,17] Although below T_c the magnetic transition is complicated by a tetragonal distortion of the unit cell, a very reliable data analysis

TABLE I. Values of the critical exponent λ and of the Néel temperature T_N for dysprosium in various temperature ranges ($\epsilon = 1 - T/T_N$) according to various authors (references given).

Dysprosium $\frac{1}{\rho_c}\frac{d\rho}{dT} = A\,|\epsilon|^{-\lambda} + B$			
ϵ	λ	T_N	Ref.
$2\times10^{-2} < |\epsilon| < 3\times10^{-2}$	-0.25 ± 0.04	180.33	19
$1.5\times10^{-2} < \epsilon < 3.3\times10^{-2}$	$+0.40^{+0.02}_{-0.05}$		
. . .	. . .	180.4	20
$\epsilon > 5\times10^{-3}$	0.30		
$5\times10^{-2} < |\epsilon| < 3\times10^{-1}$	0.13 ± 0.01	183.15	21
. . .	. . .		
$4\times10^{-4} < |\epsilon| < 3\times10^{-2}$	0.04 ± 0.005	180.34	22
$4\times10^{-4} < \epsilon < 3\times10^{-2}$	0.04 ± 0.005		

above T_c can be and has been performed in order to distinguish various critical regimes. The data are consistent with a logarithmiclike dependence; we have shown, however, that the critical-exponent exact value *cannot* be determined unambiguously. In fact, our study has revealed that the *sign of the critical exponent* itself cannot be even determined from strict data analysis only. Such a conclusion seems also to have been reached in other cases as well.[25]

Therefore, it is safe to say that at most the experimental data have only led to a qualitative characterization of the presence of a singularity in $d\rho/dT$, but have not yet been described on a broad scale of temperature encompassing T_c (or T_N). The necessity of "controlling relevant parameters" well defined *away* from the critical region seems more and more important in order to use them as an input for better analysis of the critical region, hence in the control of the so-called "prefactors."

At the present stage of experimental research, it thus appears that one should not concentrate on obtaining precise answers to questions (1) and (2) but rather examine from a theoretical point of view the following questions. (3) What magnetic, electrical, "basic," . . . , properties of the systems are responsible for an experimentally observed shape of $d\rho/dT$ vs T? (4) What laws govern this shape and why? We present below some solution to these questions in a simplistic, practical, way.

Typically, one can wonder why, at the same distance from T_c, the value of $d\rho/dT$ corresponding to the ferromagnetic phase of a metallic ferromagnet is greater than that in the paramagnetic phase (see, e.g., Fote and Mihalisin,[26] Simons and Salamon,[27] Craig *et al.*,[28] Kawatra *et al.*,[29] and Bendick and Pepperhoff.[30] How does the spontaneous magnetization (the only relevant parameter in absence of anomalous expansion, lattice structure change, etc., influence the value (and behavior) of $d\rho/dT$? Another puzzle which we also discuss briefly below is why there occur minima or maxima in $d\rho/dT$ within the region of *one* of the phases, as seen, in particular, in localized spin systems.

Although there have been attempts to explain such features, theoretical considerations such as those in Refs. 1 and 2, which are based on asymptotic relations for the correlation function in the critical region, i.e., $q/\kappa >> 1$ or $q/\kappa << 1$ (q is the wave vector of the momentum transfer; κ is the inverse correlation length) cannot really answer the above questions, in a qualitative (and *a fortiori* in a quantitative) manner. Therefore it seems essential to provide experimentalists with some simple theoretical considerations and to point out what relevant parameters of the *materials* (and not of the critical region) lead to a particular behavior.

The most crucial problem is to find a proper model-independent correlation function $\Gamma(q,\epsilon)$ which should be sufficiently simple, and correct, in relatively wide intervals of momentum q.

For the *paramagnetic* phase ($T \geq T_c$) of an *isotropic* ferromagnet a suitably parametrized Ornstein-Zernicke correlation function satisfying the sum rule

$$\sum_{\vec{q}} \Gamma(q,\epsilon) = s(s+1) \qquad (1)$$

in $\vec{k}$ space can be conserved since it identically corresponds to the form used, in $\vec{r}$ space, by Geldart and Richard.[4–6]

For the *ferromagnetic* phase ($T \leq T_c$), a similar correlation function constructed from the *mean-field* correlation function of the Ising model[31] (as done in Ref. 2) has well-known shortcomings and cannot therefore be useful. It is, in our opinion, best to start from the Landau theory to which fluctuation terms are added,[32] or, in order to have "proper values" of the critical indices, from a refined Landau theory.[33,34] In so doing the above sum rule is slightly modified and includes on the right-hand side a magnetization (temperature-dependent) term. A uniform description on both sides of T_c is thus obtainable. This seems a trivial generalization of previous considerations,[4,5] but has interesting consequences.

The following correlation function is obtained

$$\Gamma(K,\epsilon)=2^{2/3}(\tfrac{1}{3}\pi)^{4/3}\frac{r^{-2}\gamma(\epsilon)}{p^2K^2+\eta^{-2}(\epsilon)}\ , \tag{2}$$

where $p=2k_F\xi_0$ while η is the reduced (dimensionless) correlation length ($\kappa^{-1}\equiv\xi=\xi_0\eta$); $K=q/2k_F$ denotes the fluctuation wave vector in units of the (spherical) Fermi surface caliper, and $r=v^{+1/3}/\xi_0$ stands for the ratio of the spacing ($a\simeq v^{1/3}$) between magnetic ions to the amplitude of the correlation length. The lattice is supposed to be cubic, but generalizations are easily made, as in Ref. 34, e.g., to take into account Fermi-surface and fluctuation anisotropies.

As a consequence of the sum rule, the normalization factor is temperature dependent and different below and above T_c:

$$\gamma(\epsilon)=(1-m_0^2|\epsilon|^{2\beta})/(1-u)\ , \tag{3}$$

where

$$u=(\eta/\tilde r)^{-1}\tan^{-1}(\eta/\tilde r)\ , \tag{4}$$

and

$$\tilde r=r(6\pi^2)^{1/3}\ , \tag{5}$$

while m_0 is the amplitude of the magnetization, i.e.,

$$m_0^2=\begin{cases}\frac{10}{3}[1+s^2/(1+s^2)]^{-1}, & T<T_c\\ 0, & T>T_c\ .\end{cases} \tag{6}$$

One of the main points of this paper is to draw attention to the normalization factor (3), and hence to the asymmetry of the correlation function with respect to the point $\epsilon=0$. This is illustrated on Fig. 1.

The asymmetry of the Ornstein-Zernike-like correlation function (2) is of magnetic origin and results from the existence of the spontaneous magnetization itself below T_c. Notice that the asymmetry and its importance were pointed out in the first "modern" paper on the electrical resistivity near T_c.[35]

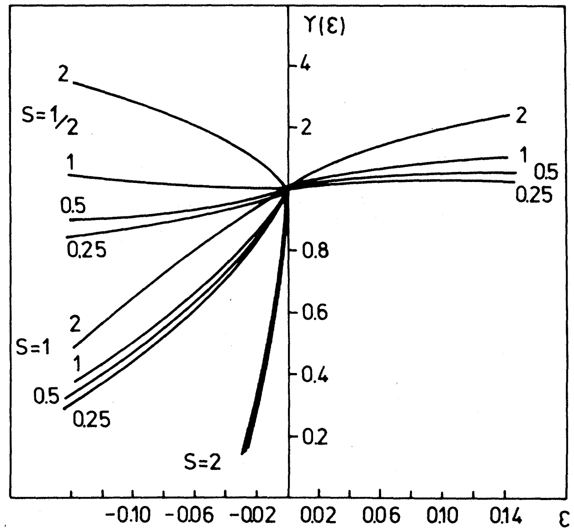

FIG. 1. Asymmetrical spin correlation function amplitude $\gamma(\epsilon)$ as given by Eq. (3), vs $\epsilon\equiv1-T/T_c$, near $\epsilon=0$, due to a magnetization-dependent amplitude obtained by satisfying the sum rule (1). Different values of the spin S and $r=v^{1/3}/\xi_0$ are considered. The critical exponent ν (2β) has been taken equal to $\frac{2}{3}$.

In addition and more remarkable is the spontaneous magnetization influence on the amplitude of the correlation function and of the correlation length ξ_0. Moreover, the latter amplitude in the ferromagnetic phase is exactly half that in the paramagnetic phase, as follows from Landau's theory,[32,33] i.e., if $\xi^{(+)}=\xi_0^{(+)}\eta_0^{(+)}\epsilon^{-\nu}$ corresponds with $\eta_0^{(+)}=1$ to the paramagnetic phase, in the ferromagnetic phase, one has $\eta_0^{(-)}=\sqrt{0.5}$. Comparison of Landau free-energy results to experimental data[32] leads indeed to a ratio $\eta_0^{(-)}/\eta_0^{(+)}\simeq\sqrt{0.5}$; this is "confirmed" by experimental data on the electrical resistivity.[16,17,36–40]

Since m_0^2 can also be rather accurately estimated from (3), the remaining *three* parameters[41] are $2k_F$, ξ_0, and v. They form the basis for characterizing the system and the behavior of $d\rho/dT$. Within the single-band model (also used in Refs. 1 and 2), the reduced spin-fluctuation-dependent resistivity becomes [as also obtained from Eq. (6) of Ref. 5]

$$R(\epsilon)=\gamma(\epsilon)\left[1-\frac{\ln(1+p^2\eta^2)}{p^2\eta^2}\right]\ , \tag{7}$$

where $p=2k_F\xi_0$. Its temperature derivative reads

$$T_c\frac{dR}{dT}=-\nu\,\mathrm{sgn}(\epsilon)\left\{\frac{R(\epsilon)}{1-m_0^2|\epsilon|^\nu}\left[\gamma(\epsilon)|\epsilon|^{-1}\left[\frac{1}{1+\eta^2\tilde r^{-2}}-u\right]+m_0^2|\epsilon|^{\nu-1}\right]+2|\epsilon|^{-1}\gamma(\epsilon)\left[\frac{\ln(1+p^2\eta^2)}{p^2\eta^2}-\frac{1}{1+p^2\eta^2}\right]\right\}\ . \tag{8}$$

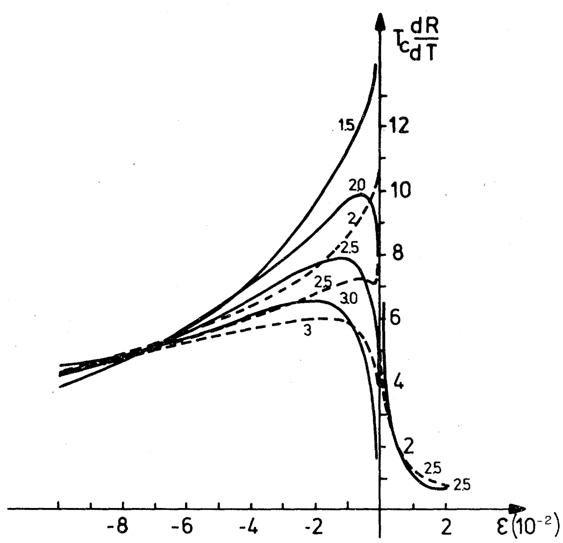

FIG. 2. Temperature dependence of $T_c dR/dT$ as a function of $\epsilon \equiv 1 - T/T_c$ near $\epsilon = 0$ for typical values of the parameter r, and $p = 2k_F\xi_0 = 1$, $m_0^2 = 2.6667$, $S = 1$, $\eta_0 = 0.3$(——), or $\eta_0 = 0.42$(– – –). Notice the structure in dR/dT below T_c for $r = v^{1/3}/\xi_0 = 2.5$ and $\eta_0 = 0.42$ at $\epsilon \simeq -10^{-2}$.

Therefore for the qualitative description of $d\rho/dT$, beside η_0 and m_0, the values of p and r are the essential parameters. Notice that the ratio p/r can be considered as an effective number of the current carriers in the nearly-free-electron single-band approximation, since $p/r = 2k_F v^{1/3}$. This emphasizes the difference between metals and semiconductors, but also between ferromagnets and antiferromagnets near the critical temperature.[1,13–15,24,42–45]

The dependence of dR/dT on ϵ for representative values of p, r, and η_0 is shown on Fig. 2. The gradual lifting of the curves for $\epsilon < 0$ with respect to those for $\epsilon > 0$ is noticeable. As shown above, this results from the existence of the spontaneous magnetization itself.

Furthermore, following the above considerations based on the Landau theory and the generalized Ornstein-Zernike correlation function (2), we stress that a maximum and a minimum of $d\rho/dT$ in the *ferromagnetic* phase can be predicted for a reasonable range of the parameters (see Fig. 2). Such behavior is found experimentally[30,36–40] but seems to have been difficult to obtain and understand in previous theoretical work (because of the natural restriction of theoretical investigations to the critical region only). We have thus pointed out that such complex behavior can be extracted from an expression like Eq. (7) by considering only the singular term (without reference to any anomalous expansion,[36] and the like[10]), but considering the full temperature dependence of usual parameters. Such a structure results from a competition between the one- and two-body correlation function. (The various types of possible singularities are compiled in Table II.)

It is of interest to substantiate these remarks by comparing the position of the minimum obtained from experimental data to that theoretically predicted. For physically reasonable values of the parameters

TABLE II. The predicted type of singularity in $d\rho/dT$ near a ferroparamagnetic critical point in a metallic magnet according to the temperature range. The origin of the singularity is given.

T range	Type of singularity of $d\rho/dT$	Reason of existence
	(a)	
$\epsilon < 0$	$-\lvert\epsilon\rvert^{\nu-1}$	large-q correlations accounted for through sum rule
$\epsilon > 0$	$+\lvert\epsilon\rvert^{\nu-1}$	
	(b)	
$\epsilon < 0$	$\lvert\epsilon\rvert^{2\nu-1}\ln\lvert\epsilon\rvert^{-2\nu}$	small-q correlations accounted for through generalized Ornstein-Zernicke correlation function
$\epsilon > 0$ (only for $\nu \leq \frac{1}{2}$)	$-\lvert\epsilon\rvert^{2\nu-1}\ln\lvert\epsilon\rvert^{-2\nu}$	
	(c)	
$\epsilon < 0$	$\lvert\epsilon\rvert^{\nu-1} = \lvert\epsilon\rvert^{2\beta-1}$	effect of spontaneous magnetization
$\epsilon > 0$		

(Fig. 2) the minimum should occur at $\epsilon = 10^{-3}$; a typical experiment[39] leads to $\epsilon = 10^{-2}$.

Notice that we find the possibility of both signs for the singularity for some value of r and η_0; a maximum below T_c followed by an unphysical negative divergence may occur. These discrepancies are likely due to the inaccurate account of the large-q correlations, so important at T_c, and have already been partially discussed in previous theoretical work.[1–12] One can hope that a better choice of the parameters will remove such a spurious prediction. Nevertheless, such an improvement, and hence a quantitative explanation of the various effects on $d\rho/dT$ near T_c (even including, e.g., anomalous lattice deformation[17,37]) still calls for a good large-q correlation function.

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[41]In previous theoretical work, *two* free parameters were in practice available, i.e., k_F and $v^{1/3} \simeq a$. A natural assumption was $\xi = a$. There seems no evident basis for accepting such an assumption. On the contrary, as indicated here, following Landau theory in Ref. 32, the *amplitude* of the correlation length is magnetization dependent. There-

fore instead of one dimensionless parameter $(k_F a)$, a second dimensionless parameter has to be considered, i.e., a/ξ_0. This consideration is quite relevant in more complicated systems: antiferromagnets, uniaxial ferromagnets, . . . (see, e.g., Ref. 34, and further work unpublished).
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[46]After this paper was accepted for publication, K. D. received a copy of the paper by G. Malmström and D. J. W. Geldart, Phys. Rev. B 21, 1133 (1980), where other values of λ given in Table I are discussed.