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ANALYSIS OF CRITICAL NEUTRON-SCATTERING DATA FROM IRON AND DYNAMICAL SCALING THEORY*

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Experimental three-axis spectrometer data of critical neutron-scattering data from Fe are reanalyzed and compared with the recent theoretical prediction by Résibois and Piette. The reason why the spin-diffusion parameter did not obey the prediction of dynamical scaling theory is indicated. Double-axis spectrometer data have previously been interpreted in terms of a non-Lorentzian susceptibility $\chi(q)$. It is shown that with proper corrections for the inelasticity of the scattering the data are consistent with a Lorentzian form of $\chi(q)$.

Critical scattering of neutrons from iron near the Curie temperature has been studied for more than twenty years. The experimental results have contributed considerably to the theoretical understanding of magnetic critical phenomena. In fact, Van Hove's pioneering papers[1] on the theory of critical magnetic phenomena were primarily inspired by the early observations of Squires,[2] and of Hughes and Palevsky.[3] When the inelasticity of the scattering was measured by Passell et al.[4] it was found, however, to be in apparent contradiction with Van Hove's theory. This led Marshall[5] to suggest that propagating modes may persist above T_c if the wave vector q is large compared with the inverse correlation range κ_1. Van Hove's theory, on the other hand, should only be valid in the opposite limit $q \ll \kappa_1$, the so-called hydrodynamic region. The experiments, which were in the intermediate region $q \approx \kappa_1$, therefore, could not be compared directly with Van Hove's theory. Halperin and Hohenberg[6] then showed how the static scaling concepts developed by Kadanoff[7] could be expanded to give a complete picture of magnetic critical dynamics. Some of the predictions of their theory have subsequently been verified by the recent comprehensive measurements of Collins et al.[8]

However, there are still difficulties in reconciling the critical scattering observed in iron with the theory. It is the purpose of this paper to point out how at least three of the difficulties can be resolved by use of the recent calculation of Résibois and Piette[9] of the linewidths of the inelastic scattering.

Triple-axis spectrometer data.—A triple-axis spectrometer allows the experimenter to observe the process in which incident neutrons with wave vector $\vec{k}_i$ are scattered to wave vector $\vec{k}_f$. It is convenient to describe the cross section for this process in terms of the wave vector transfer $\vec{q} = \vec{k}_i - \vec{k}_f + \vec{\tau}$ and the energy transfer $\hbar\omega = (k_i^2 - k_f^2)\hbar^2/2m$, $\vec{\tau}$ being any reciprocal lattice vector, including the origin. The essential part of the cross section is the product of the wave-vector–dependent susceptibility $\chi(q)$ and the relaxation function $F(q, \omega)$, normalized by $\int_{-\infty}^{\infty} F(q, \omega) d\omega = 1$ for any fixed q.

The simplest approximation for $\chi(q)$ is the Lorentzian form $\chi(q) \propto [q^2 + \kappa_1^2]^{-1}$. Small deviations from this have been anticipated by Fisher and Burford,[10] but it will be shown that the Lorentzian alone provides an adequate description of the experimental data.

In the hydrodynamic limit the relaxation function is given by

$$F(q, \omega) = \pi^{-1} \Lambda q^2 / [(\Lambda q^2)^2 + \omega^2], \quad q \ll \kappa_1. \tag{1}$$

Outside this region no accurate predictions of $F(q, \omega)$ have been made. Halperin and Hohenberg,[6] however, have introduced the very useful concept of the characteristic frequency $\Gamma(q)$, defined by

$$\int_{-\Gamma(q)}^{+\Gamma(q)} F(q, \omega) d\omega = \tfrac{1}{2}. \tag{2}$$

For a ferromagnet they find $\Gamma(q) \propto q^{5/2}$ in the limit $q \gg \kappa_1$, which they called the critical region, while outside this region

$$\Gamma(q, \kappa_1) = cq^{5/2} f(\kappa_1/q). \quad (3)$$

In the latter equation the temperature dependence of the characteristic frequency is explicitly indicated by its dependence on the inverse correlation range κ_1. The homogenous function $f(\kappa_1/q)$ is normalized to unity when $\kappa_1/q = 0$. In the limit $q \ll \kappa_1$ Eq. (1) implies $f(\kappa_1/q) \propto (\kappa_1/q)^{1/2}$ and thus the diffusion constant Λ in Eq. (1) should be proportional to $\kappa_1^{1/2}$.

This brings us to the first problem in comparing experimental data with dynamical scaling theory: Although Collins et al.[8] did show that spin waves below T_c do renormalize as predicted by dynamical scaling theory and that the linewidths at T_c do obey the predicted $q^{5/2}$ law, their analysis of the data above T_c failed to show $\Lambda \propto \kappa_1^{1/2}$. As a matter of fact there is only a weak indication in the data that the expected thermodynamic slowing down does occur.

The problem in extracting Λ from the measured ω scans at fixed q is to decide where the boundary for the hydrodynamic region is, or more explicitly, how large κ_1/q has to be before $f(\kappa_1/q)$ is reasonably close to its asymptotic form $\propto(\kappa_1/q)^{1/2}$. At the time when Collins et al. analyzed their data there was no theoretical prediction for $f(\kappa_1/q)$. To determine this boundary they therefore relied on a statistical χ^2 test of the data to indicate whether $F(q, \omega)$ at fixed q was of Lorentzian form, cf. Eq. (1). At each temperature the analysis did show a rather sudden increase of χ^2 with increasing q and this value of q was interpreted as being the boundary of the hydrodynamic region. However, there are no adequate theoretical predictions for $F(q, \omega)$ at general values of κ_1/q, and the χ^2 test does not, therefore, yield an unambiguous boundary for the hydrodynamic region.

The data of T_c did, however, confirm the $q^{5/2}$ law and yielded $c \approx 130$ meV Å$^{5/2}$. It is therefore possible to determine experimental values of the homogeneous function $f(\kappa_1/q)$ from the widths measured at temperatures above T_c.[11] By comparing these experimental values of $f(\kappa_1/q)$ with the recent predictions of Résibois and Piette[9] we can now conclude that the data of Collins et al. do not extend far enough in κ_1/q to enable a separate determination of Λ. The results of Résibois and Piette, shown in Fig. 1, refer to a Heisenberg Hamiltonian. But there is nevertheless a remarkable agreement with the experimental data for Fe as seen from the inset of Fig. 1. On the other hand, it is clear that κ_1/q has to be larger than ~2 before the hydrodynamic form $\Gamma(q, \kappa_1) = \Lambda q^2$ is valid. We must therefore conclude that the analysis leading to the values quoted for $\Lambda(T)$ by Collins et al. was not valid, and that it is very likely that data taken at sufficiently high values of κ_1/q would indeed have revealed $\Lambda \propto \kappa_1^{1/2}$.

Double-axis spectrometer data.—A double-axis spectrometer allows one to observe the process in which incident neutrons with wave vector $\vec{k}_i$ are scattered through a fixed angle θ. In most double-axis experiments, scattering around the forward direction was studied. It follows that the count rate at a scattering angle θ represents a cut of the cross section $S(q, \omega)$ in q-ω space, the relation between q and ω being

$$q(\omega) = \{2k_i^2 - (2m/\hbar)\omega + 2k_i^2[k_i - (2m/\hbar)\omega]^{1/2}\cos\theta\}^{1/2}. \quad (4)$$

In the following we shall approximate the shape of $F(q, \omega)$ by a Lorentzian with the proper characteristic frequency $\Gamma(q, \kappa_1)$ as given by Eq. (3). The scattering cross section $S(\theta)$ thus becomes

$$S(\theta) \propto \int_{-\infty}^{\hbar k_i^2/2m} [k_i^2 - (2m/\hbar)\omega]^{1/2} [\beta\hbar\omega/(1-e^{-\beta\hbar\omega})] \times [\kappa_1^2 + q^2]^{-1} F(q, \omega) d\omega, \quad (5)$$

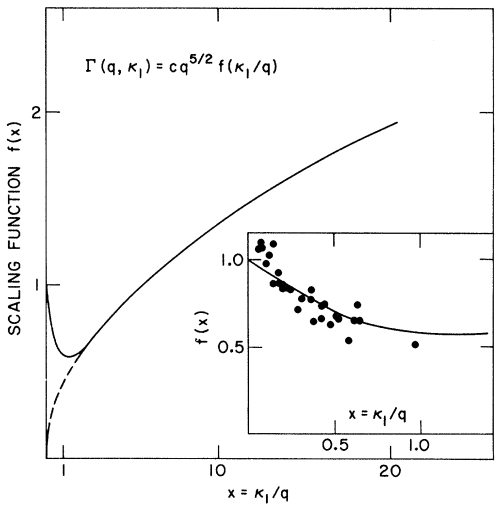

FIG. 1. The scaling function $f(\kappa_1/q)$ calculated by Résibois and Piette determines the linewidth of critical magnetic fluctuations relative to the linewidth at T_c. The hydrodynamic form $\sim(\kappa_1/q)^{1/2}$, shown as the dashed line, is valid only when $\kappa_1/q \gtrsim 2$. The inset shows the comparison with experimental data using an extended abscissa scale.

with q as given by Eq. (4). The first factor in the integrand takes into account the fact that $S(q,\omega)$ is proportional to k_f/k_i, the second factor is the detailed-balance factor with $\beta \equiv 1/kT$, and the last two factors are the susceptibility and the relaxation function, respectively.

The quasielastic approximation is obtained by letting $\Gamma(q,\kappa_1) \to 0$. It follows that

$$S_{el}(\theta) \propto \chi(q = k_i\theta) \propto [(k_i\theta)^2 + \kappa_1^2]^{-1}, \quad (6)$$

and thus κ_1 can be determined from double-axis spectrometer data within the limits of this approximation.

Two problems have arisen with the interpretation of double-axis data. We will show that they disappear when the effects of the inelasticity of the scattering are properly included. The first of these problems is the following:

Two groups[12,13] have independently shown that the intensity for varying temperatures at a fixed scattering angle has a flat maximum above T_c rather than at T_c and that this temperature shift increases with increasing scattering angle. This effect, which has also been observed in Ni,[14] has previously been interpreted in terms of Kocinski's theory[15] of critical fluctuations or in terms of the temperature shift, which is much smaller and which has a different dependence on θ from that observed. We have calculated the scattering cross section given by Eq. (5) for the value of $k_i \approx 5$ Å^{-1} used in the experiments, with $\Gamma(q,\kappa_1)$ given by Eq. (3) and with $f(\kappa_1/q)$ from Fig. 1.

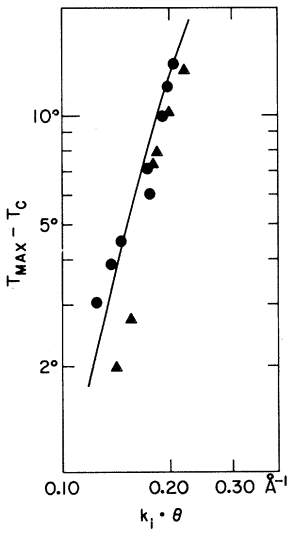

FIG. 2. The temperature for maximum scattered intensity at fixed angle versus $k_i\theta$. Circles (Ref. 12) and triangles (Ref. 13) are experimental data. The full line is calculated by including the inelasticity of the scattering.

The calculation displays a flat maximum above T_c and the calculated temperature shift versus scattering angle, shown in Fig. 2, agrees very well with the observed temperature shifts. Numerical analysis shows that the essential feature in the inelasticity which produces the temperature shift is the pronounced dip in $f(\kappa_1/q)$ when $\kappa_1/q < 2$. We emphasize that this is the region in which the data of Collins et al.[3] have already verified the predictions of Résibois and Piette.[9] Our interpretation for the observed temperature shift therefore does not require that $\chi(q)$ has other than Lorentzian form but is simply a consequence of the experimentally determined inelasticity of the scattering.

The second problem with which we are concerned is the generally observed deviation from a Lorentzian form[4,16,17] when $k_i\theta \gtrsim 0.10$ Å^{-1}. The conventional way of displaying this deviation is to plot the inverse intensity versus the square of the scattering angle. In this plot quasielastic scattering with a Lorentzian $\chi(q)$ [cf. Eq. (6)] will give a straight line. In Fig. 3 the data of Passell et al.[4] at $T = T_c + 30°$ are shown as an example. When corrected for inelasticity (which at that time it was believed could be described by $\Gamma(q,\kappa_1) = \Lambda q^2$] the data still showed a marked deviation from a straight line when $k_i\theta \gtrsim 0.08$ Å^{-1}. The authors therefore concluded that higher-order terms in $\chi(q)$ were necessary to account for the deviation, and using the expression $\chi(q) = [\kappa_1^2 + q^2 + \epsilon q^4]^{-1}$ they found $\epsilon \approx 29$ Å^2. In the molecular-field approximation for a Heisenberg magnet it is easy to determine ϵ in terms of the exchange interaction $J(r)$ and one can generally

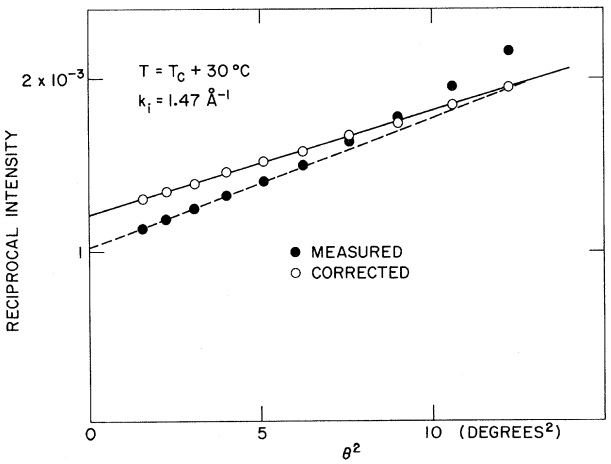

FIG. 3. The deviation from a Lorentzian form of critical magnetic-scattering data (filled circles) is accounted for by the inelasticity of scattering as the corrected data (open circles) show.

conclude that higher-order terms become increasingly important as the interaction range becomes large. It therefore seemed qualitatively reasonable that higher-order terms might be important in a band magnet like iron.

We have now reanalyzed the total set of data reported by Passell et al.[2] for $T = T_c + 2°$ up to $T = T_c + 60°$ with the proper form of the inelasticity and we find that the observed deviation from a straight line is entirely due to the inelasticity of the scattering. An example is given in Fig. 3 for $T = T_c + 30°$, the corrected data being shown as open circles. Furthermore, the goodness of fit as determined by the χ^2 test became substantially better with the present analysis than with the original analysis. It should be emphasized that this improvement in the value of χ^2 (typically a factor of 2-4) is obtained without introducing any additional parameter. The natural question arises: Is the reported value for the exponent γ in the power law $\chi(q=0) \propto [(T-T_c)/T]^{-\gamma}$ now obsolete? The answer is, no; $\gamma = 1.30 \pm 0.04$ is also found in the present analysis. However, the absolute values of κ_1 are about 10% higher than those originally reported.

Two other experiments[16,17] have also shown an apparent deviation from a Lorentzian $\chi(q)$ or the equivalent Ornstein-Zernike pair correlation function $e^{-\kappa_1 r}/r$. These experiments were carried out at larger incident-neutron wave vectors ($k_i \simeq 5$ Å^{-1}) and at first sight one might expect that the quasielastic approximation should be much better than in the case of the experiment of Passell et al. using $k_i = 1.47$ Å^{-1}. However, numerical evaluation of the integral in Eq. (5) shows that this is not the case. The observed deviation from the Lorentzian form is still mainly due to the inelasticity of the scattering. This effect was entirely neglected by Spooner and Averbach, and their conclusion that the Ornstein-Zernike pair correlation function is only valid for distances larger than ~15 Å is therefore highly questionable.

We note finally that the apparent deviation from a Lorentzian form is more pronounced in Ni than in Fe; Jacrot et al. reported a value of $\epsilon = 110$ Å^2.[18] We believe that this is due simply to the fact that linewidths in Ni generally are a factor of 2 larger than in Fe. One should thus not take the larger deviation from a Lorentzian form in Ni as evidence that the interaction range in Ni is substantially longer than in Fe.

The present reanalysis of critical scattering from Fe has eliminated most of the contradictions between experimental facts and current theoretical predictions based on the Heisenberg model. The key to reconciling theory with experiment is the recent calculation of the scaling function $f(\kappa_1/q)$ by Résibois and Piette. The measured linewidth data agree with their predictions but the data do not extend far enough into the hydrodynamic region to allow interpretation in terms of the Van Hove formulation. Observations of a shift in the peak of the scattered intensity to temperatures above T_c at fixed scattering angle are fully accounted for by the inelasticity of the scattering, and so is the apparent deviation of $\chi(q)$ from a Lorentzian. We conclude that there are no contradictions between current theory and the existing body of experimental data. However, further experiments are needed to examine the limiting case $q \ll \kappa_1$ described by Van Hove.

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ORDER PARAMETER AND PHASE TRANSITIONS OF STRESSED $SrTiO_3$

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EPR spectra of Fe-V_O pairs were used to study how the order parameter of $SrTiO_3$ varies with uniaxial stress applied to a (111) face. A second-order cubic-trigonal phase boundary appears above the stress-free transition temperature T_a. A first-order tetragonal-trigonal phase boundary is found below T_a. An independently determined Landau potential describes the results.

In this Letter we report effects of applied uniaxial stress on the structure of strontium titanate ($SrTiO_3$), a solid with a structural transition in the free state. We observe the variation of the order parameter with stress and temperature, construct phase boundaries from its behavior, and account for our results with the Landau theory.

Recent investigations have revealed the close connection between the cubic-to-trigonal and cubic-to-tetragonal transitions in $LaAlO_3$ and $SrTiO_3$, respectively, which occur in the absence of applied stress.[1-3] In each case the transition, with critical temperature T_a, results from an instability of an optic-mode displacement representing a rotation $\pm\vec{\varphi}$ of nearly rigid TiO_6[4,5] or AlO_6[6,7] octahedra.[2] Whether the trigonal ($R\bar{3}c$) or the tetragonal ($I4/mcm$) structure is realized depends on the ratio of fourth-order parameters in the Landau theory,[3] if these parameters are adjusted for interaction with strain.[8] In the lattice-Hamiltonian theory of Pytte and Feder[9] this issue is decided by fourth-order terms in the oxygen-ion potential and correlation functions, together with strain interactions.

While the parameter values place $LaAlO_3$ within the trigonal phase, both theories show that $SrTiO_3$ is a borderline case slightly favoring the tetragonal structure.[8,9] Indeed, studies by Burke and Pressley[10] of Cr^{3+}-impurity fluorescence-line splittings under uniaxial stress showed that 24.7 kg/mm^2 (1 kg/mm^2 = 98 bar) applied along a [111]-pseudocubic axis at 4.2°K, suffices to induce the trigonal phase.

The electron paramagnetic resonance (EPR) technique[2,11] used in this work has the advantage of providing direct measures of components of the order parameter $\vec{\varphi}$, obviating the crystal-field parameter fitting required to analyze effects of stress on Cr^{3+} fluorescence.[12] Another advantage of EPR is that accurate measurements are possible at higher temperatures, close to T_a, where Cr^{3+} fluorescence lines become broad.

In the experiments a stress p was applied along the axes of cylindrical samples parallel to a cubic [111] direction. The sample diameter was typically 1 mm, its height about 3 mm. Special care was necessary to polish parallel end planes and use Teflon sheets there to ensure homogeneous strain in the sample and reduce hysteresis effects at the first-order transitions. The nominal Fe_2O_3 content was 0.03%. The variable-temperature cryostat allowed the application of stress only perpendicular to the external magnetic field H. Under this geometry ($p \parallel [111]$) the EPR spectrum of Fe^{3+} substitutional for Ti^{4+} is insensitive to the octahedral rotation $\vec{\varphi}$ in the trigonal phase. However, the spectrum of the lower symmetry Fe^{3+}-V_O pair center,[13] i.e., a Fe^{3+} with a nearest-neighbor oxygen vacancy, does vary with φ_{111}. It has been used throughout this work taking into account the smaller rotation of this center, $\overline{\varphi}(Fe^{3+}\text{-}V_O) = \varphi/(1.59 \pm 0.05)$. This newly determined coefficient differs from the value 1.4 deduced earlier from Ref. 11.

Figure 1(a) shows the variation of a group of EPR lines with [111] stress for H parallel to a $[1\bar{1}0]$-pseudocubic direction at 78°K. It corresponds to but one line for $T > T_a$ and $p = 0$, which results from Fe^{3+}-V_O pairs with symmetry axis at 45° to H, i.e., parallel to [100] and [010].[13]

For zero stress there are four lines. The outer two are both due to pairs located in [001] domains. These pairs lie on two inequivalent