Coexistence of Two Length Scales in X-Ray and Neutron Critical Scattering: A Theoretical Interpretation

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(Received 20 December 1994)

The theory of critical phenomena in the presence of quenched disorder is applied to x-ray and neutron experiments which suggest two different length scales for critical fluctuations. Long-range random strains associated with defects generated near the sample surface can induce crossover to a "disordered" fixed point with different critical exponents, while the bulk retains the ordinary critical behavior. For Ho and Tb, it is proposed that the dominant defects are dislocation dipoles, resulting in a critical exponent for the second correlation length $\nu_s = 1$, in reasonable agreement with experiments.

PACS numbers: 64.70.–p, 61.10.Dp, 61.12.Bt, 75.40.Cx

X-ray and neutron critical scattering experiments in systems undergoing magnetic [1–3] or structural [4] phase transitions have recently revealed an unexpected feature, which appears to be at odds with the existence, in the critical region, of a single important length scale, given by the correlation length ξ, and characterized by the critical exponent ν, defined by $\xi \sim t^{-\nu}$, where t is the reduced temperature $t = |(T - T_c)/T_c|$. In contrast to this basic tenet of the theory of critical phenomena, the wave vector dependence of the scattering intensity is not at all well described by a single Lorentzian with a width proportional to the inverse of the correlation length; a much sharper feature appears near the center of the line, and a reasonable fit is only obtained by a superposition of a broad Lorentzian (referred to as the "broad" component), and a much sharper Lorentzian or Lorentzian squared component (the "sharp" component). For Ho [1] and Tb [2] the broad component has an exponent close to the theoretically predicted one for the magnetic spiral transition [5]. The second, much larger, length associated with the "sharp component" scales with temperature as $t^{-\nu_s}$, where the new exponent ν_s is different from and in most cases larger than the exponent ν associated with the first correlation length. Other important results of the experiments led to the conclusion that the sharp feature is related to a layer of the sample close to the surface [2], but with a thickness which can be as large as several μm, a surprisingly large depth for usual surface phenomena. Also, the dependence of the intensity of the sharp feature on the surface quality and mosaic and its enhancement by polishing and other forms of surface treatment [1] suggest very strongly a defect-related origin. Defects of unspecified nature, and the related long-range strain fields have been mentioned as a possible explanation [1], but no conclusive and quantitative analysis could be arrived at.

In the present paper we argue that the emergence of a second length scale is a consequence of the presence of quenched disorder in the neighborhood of the sample surface. In this part of the sample a crossover to critical behavior dominated by a "disordered" fixed point takes place, while the bulk displays the usual critical properties for the applicable universality class.

The theory of second order phase transitions in the presence of quenched disorder was worked out by Grinstein and Luther [6] for the case of short-range disorder. Later Weinrib and Halperin [7] (hereafter WH) extended it to the case of disorder with long-range correlations, which is more relevant to the experimental results of interest here. WH [7] analyze a situation in which the local critical temperature $T_c + \delta T_c(\mathbf{r})$ displays fluctuations about T_c due to the disorder in the system, fluctuations which are described by a correlation function $g(|\mathbf{r} - \mathbf{r}'|)$ that falls off slowly with distance, with a power law characterized by an exponent a:

$$\langle \delta T_c(\mathbf{r})\, \delta T_c(\mathbf{r}')\rangle = g(|\mathbf{r} - \mathbf{r}'|) \sim |\mathbf{r} - \mathbf{r}'|^{-a} \quad \text{for } |\mathbf{r} - \mathbf{r}'| \to \infty . \tag{1}$$

WH [7] showed that, if $a < d$, where d is the number of space dimensions, and if the pure, nondisordered system satisfies a modified "Harris criterion," i.e., if $2 - a\nu > 0$, then the critical behavior of the system is controlled by a disordered fixed point, with a new set of critical exponents, and in particular with a correlation length exponent:

$$\nu_s = 2/a . \tag{2}$$

(If $a > d$, the short-range disorder case, analyzed by Grinstein and Luther [6], is applicable instead, and different critical properties are predicted [8].)

If this picture is adopted for the near-surface region, and if the scattering probe has a penetration depth allowing one to sample both the superficial, disorder-dominated, region and the underlying bulk, the two length scales, and their different critical exponents, are then observed to coexist. The relative intensity will in fact

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depend on the penetration depth, which weighs the two components of the line shape differently. With x rays ($\sim$0.5 μm), mainly the sharp component will be detected, while the two features will coexist in neutron scattering (0.5 cm). This agrees with the observations in Ho [1]. Furthermore, they have shown that neutron experiments performed in a low-resolution configuration show little indication of a narrow component, explaining why it had not been detected before. Both components are clearly visible in high-resolution neutron-scattering scans.

We note in passing that the modified Harris criterion suggests that ν_s is always larger than ν. Indeed, if

$$2 - a\nu > 0, \quad \text{then } \nu_s = 2/a > \nu . \tag{3}$$

In order to test the applicability of these ideas to a specific system and possibly to identify the nature of defects which are responsible for the correlated disorder, it is necessary to establish the correspondence between the nature of defects and the value of a. We consider a mechanism by which defects induce a local compression or expansion of the lattice, thereby affecting the local critical temperature (to an extent which can be estimated from the pressure dependence of T_c and from the compressibility of the system) [9]. For example, a substitutional impurity with a size misfit with respect to the host induces a radial strain field which decreases with distance from the defect as r^{-3}; the same dependence occurs for the long-range strain produced by a dislocation loop, while a single dislocation can be associated with a r^{-1} decay law. Let us consider in general a random distribution of defects of the same type, with a characteristic exponent a' for the power-law decay of the associated strain field. The assumed randomness in the distribution of defects implies that the only origin for spatial correlations of the type given in Eq. (1) is in the individual defect properties, i.e., it is associated with the exponent a'. Considering first the case of point defects, from the assumed large-distance behavior of $g(|\mathbf{r} - \mathbf{r}'|)$ and of $\delta T_c(\mathbf{r})$ we easily find for the leading behavior at small k of the respective Fourier transforms

$$\tilde{g}(k) \sim k^{a-d}, \quad \delta T_c(k) \sim k^{a'-d}, \tag{4}$$

where $a, a' < d$ is assumed. But since the Fourier transform of $g(|\mathbf{r} - \mathbf{r}'|)$ is the square modulus of the transform of $\delta T_c(\mathbf{r})$, we readily obtain that $(a - d)/2 = a' - d$, or $a = 2a' - d$. If instead of point defects, line defects are considered, such as dislocations, the strain field becomes independent of the coordinate parallel to the dislocation line. The Fourier transform is performed in the remaining dimensions only, so that d in Eq. (3) is to be replaced by $d - 1$. In general we obtain

$$a = 2a' - d_{\text{eff}}, \tag{5}$$

$$d_{\text{eff}} = d \quad \text{for point defects,}$$

$$d_{\text{eff}} = d - 1 \quad \text{for line defects, etc.}$$

Equation (5) is the link which allows us to establish, for a given defect type, whether the associated strain field is relevant, in the renormalization group sense, i.e., if the modified Harris criterion is satisfied. If so, it determines the modified critical exponent for the defective system, which we propose to identify with ν_s.

This analysis will be now applied to recent detailed results for the magnetic spiral phase transition in Ho and Tb. It is important to notice that in these systems, where the critical exponent of the broad component, i.e., of the perfect bulk crystal, is less than 0.6 [1,2], the modified Harris criterion is satisfied for all values of a up to 3, the value beyond which the short-range disorder model and the unmodified Harris criterion apply. In other words, long-range strain fields are expected to be relevant for the critical behavior. Furthermore, as these crystals are hexagonal, with a ratio of the crystallographic parameters, $c/a \sim 1.58$ [1,2] at room temperature, not too different from the ideal close-packed one, 1.633, the easy-glide plane is the basal plane [10]. Surface treatments such as polishing, will induce slipping parallel to the surface. With a moderate mosaicity, it is plausible to assume that the dominant kind of defect consists in *edge dislocation dipoles,* nearly parallel to the surface (if the latter is orthogonal to the c axis), but otherwise randomly oriented in the basal plane (see Fig. 1). Notice that the following discussion by no means requires that no other types of defects are present, but only that the others have a shorter range of spatial correlations, as discussed by WH.

The parameter a' of the dislocation dipole depicted in Fig. 1 is readily established using the results of dislocation theory [10]. In fact, if we denote by $\pm x$ the direction of the Burgers vectors of the dipole, and by $\pm x_0$ the intersection of the two dislocation lines with the x axis, the trace of the strain tensor at a point belonging to the (xy) plane and located at a distance r from the origin and an angle θ from the x axis is [to lowest order in $(x_0/r) \ll 1$]

$$\varepsilon_{xx} + \varepsilon_{yy} = 2A(x_0/r^2)\sin 2\theta , \tag{6}$$

where $A = (b/2\pi)[(1 - 2\sigma)/(1 - \sigma)]$, b is the Burgers vector length and σ the Poisson ratio. Therefore $a' = 2$, and according to Eqs. (5) and (2), $a = 2$ as well, so that

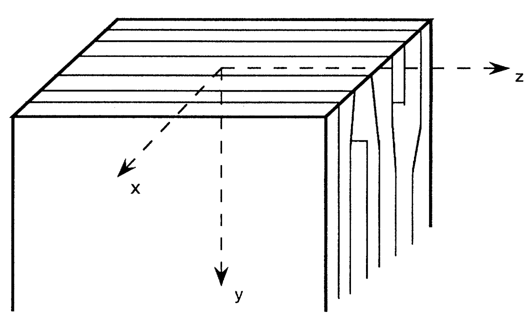

FIG. 1. Schematic view of an edge-dislocation dipole, with dislocation lines parallel to **z** and Burgers vectors **b** parallel to **x**, the surface of the crystal being the (xy) plane.

$\nu_s = 1$. This value is to be compared with values for the sharp component (extracted from fits to the observed line shapes) which range between 0.9 and 1 for Ho [1] and around 1.3 for Tb [2].

The same remarks apply to the other long-range-disorder fixed point exponents [7]: $\alpha = 2(a - d)/a$, $\beta = (2 - \varepsilon)/a + O(\varepsilon^2)$, $\gamma = 4/a + O(\varepsilon^2)$, and $\eta = O(\varepsilon^2)$. Note that, for $a = 2$ and $d = 3$, $\alpha < 0$ (as it should), while $\beta \approx \frac{1}{2}$ and $\gamma \approx 2$. According to Ref. [1], the value of γ is between 2 and 5 in Ho. It would be worthwhile to have accurate experimental determinations of these exponents and their crossover range. Of course, one should not forget that the WH fixed point has only been studied in the vicinity of $d = 4$ and $a = 4$, although $\nu_s = 2/a$ is a more general result.

We will now see that the stress field produced by the dislocation dipoles can be large enough to account for the local fluctuations of T_c yielding the observed experimental behavior. The proper way to evaluate the require density of defects per unit area, N, would be via the crossover temperature. In the absence of an unambiguous experimental determination of this quantity, we identify the characteristic size of critical temperature fluctuations due to disorder, δT_c, as the range above T_c where the narrow-component signal emerges. For Ho and Tb, δT_c is of order 1 K [1,2]. We may write $\delta T_c = \langle(\delta T_c)^2\rangle^{1/2} = \sigma_0(\partial T_c/\partial p)$, where $\sigma_0 = \langle\sigma_0^2\rangle^{1/2}$ is the rms compressional stress due to the dipoles, and $\partial T_c/\partial p$, the pressure dependence of T_c, about 0.5 K (kbar)$^{-1}$ in Ho and 1 K (kbar)$^{-1}$ in Tb [11].

As can be seen in Fig. 2, the stress field is very large close to the dipoles, but decaying with distance until it reaches the field of the neighboring dipole. A rough estimate of σ_0 can be obtained as follows: the far-field contribution is provided by the mean square of the stress calculated from Eq. (6) (where $D = \mu b/[2\pi(1 - \sigma)]$ replaces A, μ is the shear modulus) over the region in the (xy) plane with linear dimension given by the characteristic half distance between two neighboring defects $r_0/2 = N^{-1/2}/2$,

$$\langle\sigma_0^2\rangle_{\text{far}} = (1/\pi)(8Dx_0/r_0)^2 \int_0^{2\pi} d\theta \times \sin^2(2\theta) \int_{x_0}^{r_0/2} dr(1/r^3) = 32(D/r_0)^2, \quad (7)$$

where $2x_0$ is the dipole separation, typically a few hundred angstroms [10]. The near-field contribution between b and x_0 is approximated by simply taking twice the mean-square stress of a single dislocation, which results in an additional contribution to Eq. (7) given by

$$\langle\sigma_0^2\rangle_{\text{near}} = (4/\pi)(D/r_0)^2 \int_0^{2\pi} d\theta \times \sin^2\theta \int_b^{x_0} dr/r = 4(D/r_0)^2 \ln(x_0/b). \quad (8)$$

With typical values for the parameters, $\mu = 250$ kbar, $\sigma = 0.3$, $b = 3$ Å, and $x_0 = 100$ Å, a density of $N \sim 10^9 - 10^{10}$ cm^{-2} yields a characteristic stress $\sigma_0 \sim 1-2$ kbar, so that the overall δT_c is indeed of order 1 K in both Ho and Tb, in acceptable agreement with the experiment. This value of the defect density at the damaged "skin" of the sample is not unreasonable.

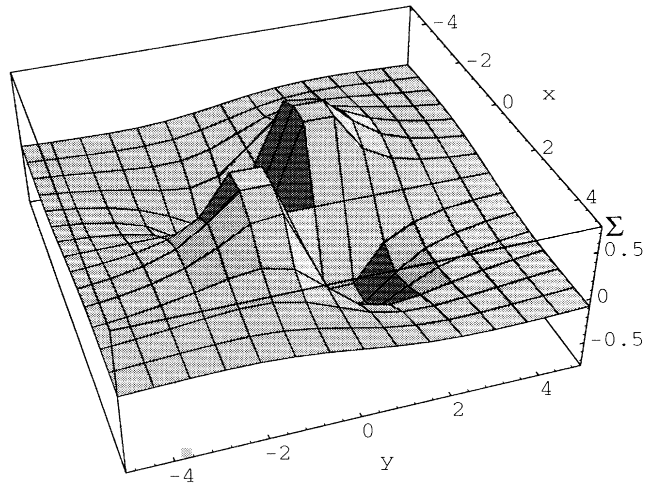

FIG. 2. Profile of the trace $\Sigma = \sigma_{xx} + \sigma_{yy}$ of the stress tensor in the plane (xy) perpendicular to an edge-dislocation dipole. The stress tensor is expressed in units of D, and x and y are in units of x_0 (see text), so that the two dislocation lines intersect the (xy) plane at $(1,0)$ and $(-1,0)$. The divergent stress at those points is cut off to simulate the physical effect of the dislocation cores.

There have been extensive discussions about the spatial origin of the sharp component. Technically, neutron scattering measurements in Tb show that this component lies in some skin of the sample, and not in the bulk [2]; the narrower the beam, the greater the signal. We believe that the narrow component comes from the region damaged during the polishing, a few μm deep and from the even deeper region affected by the resulting long-range strains. It is important to notice that the correlation length is observed to be nearly isotropic parallel and perpendicular to the surface. This is explained by the very large depth in which the disorder-induced critical behavior is present, larger than the correlation length at all temperatures $T > T_c$ except very close to T_c. On the other hand, the crystallographic structure, favoring the creation of defects in the basal plane, accounts for the large enhancement of the narrow component intensity in those faces which are orthogonal to the c axis [2].

In conclusion, we have shown that quenched long-range disorder can be at the origin of the second length scale observed in x-ray and high-resolution neutron scattering studies. We argued that this length scale can be attributed to new critical behavior in the near surface region, while the bulk displays the critical behavior of the pure system. The more quantitative analysis performed for Ho and Tb, strongly suggests dipole dislocations introduced in the basal plane by the polishing procedure, as the cause of this change of the behavior in the sample skin. These two length scales in the critical fluctuations have been observed in a variety of materials: with magnetic, structural, second order smooth or sharper transitions. It would be worthwhile to perform an analogous study for these different systems, looking for the most relevant kind of defects in each case, trying to account for the same type of phenomenological critical behavior. The differences between experimental values for the correlation-length exponents in Ho [1] and Tb [2] should be further analyzed. Comparison with other critical systems displaying two length scales, particularly perovskites [4], is presently under investigation. While we believe that the explanation in terms of disorder-induced modifications of critical behavior near the surface is applicable in general, the microscopic nature of the defects involved is certainly different for each class of systems. In the case of perovskites an additional modification of the theory may be necessary because of the weakly first order character of the transition.

On the theoretical side, there is at least one intriguing point. Whatever the universality class to which the pure system pertains, long-range disorder (when relevant) seems to define a "super-universality-class" fixed point with the same exponent $\nu_s = 2/a$ for a given correlation power a. Furthermore, since for perfectly self-similar disorder (i.e., same a for all large-length scales), a is, in general an integer, (e.g., $a = 2$ for dipoles), the superuniversal exponent ν_s will itself assume only the values 2, 1, or $\frac{2}{3}$ in three dimensions [see Eq. (5)]. This of course does not hold for ordinary, short-ranged disorder.

The present explanation for the coexistence of the two length scales, not only is in agreement with the standard theory of second order transitions, but furthermore suggests that the recent results on Tb and Ho provide, to our knowledge, the first example of critical behavior modified by quenched long-range disorder, according to Weinrib and Halperin's theory.

We acknowledge suggestions by Ph. Nozières, F. Louchet, and J. Imry, discussions with N. Bernhoeft, S. Langridge, J. E. Lorenzo, and D. Gibbs and T. R. Thurston, and useful correspondence with G. Parisi, B. I. Halperin, and R. Cowley.

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