Optical investigation of the temperature and order parameter dependences of interfacial roughening in a random-field system

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The pinning and roughening of structural domain walls by random fields was studied in $Tb(As_{0.15}V_{0.85})O_4$ by measuring the intensities of laser light Bragg-scattered from orthorhombic twin interfaces as the sample was repeatedly warmed and cooled through its structural phase transition. Determination of the temperature-dependent roughness with submicron precision as well as its correlation length has shown that the domain walls are rougher than in pure $TbVO_4$, has allowed the identification of a metastable microdomain state upon sample cooling, and has permitted investigation of the scaling of the roughness with order parameter. [S0163-1829(99)09013-X]

Interfacial topologies and dynamics in disordered systems have been studied in a variety of physical systems including pinned magnetic[1] and structural[2] domain walls, fluid interfaces in porous media,[3] and multicomponent chemical systems.[4] Much discussion has arisen regarding the interfacial dynamics of driven depinning[5] where the velocity plays the role of order parameter. By contrast, there has been little investigation of the distortion of stable (or metastable) interfaces by the underlying disorder in those random field systems where the order parameter is a function of temperature or applied field.[6] Although seminal theoretical papers[7,8] related the stability of long range order (LRO) in random field systems to the pinning and roughening of domain interfaces, experimental studies of such systems have focused on the existence of LRO and modified critical properties rather than on the interfacial properties. A primary reason is that in dilute antiferromagnets in a magnetic field (DAFF), which is the major physical realization of the random-field Ising model (RFIM), domains have very little contrast for most experimental probes and an ordering field cannot be applied. For the case of a structural RFIM, it has been possible to estimate the random-field-induced roughness by observation of the transverse broadening of neutron diffraction peaks and to examine its dependence on temperature and ordering field.[9] Further investigations of the evolution of equilibrium interfacial roughness in RFIM systems would be valuable in evaluating random field models and the associated critical behavior which is still not well understood.[10] In this context we report experimental results for the structural random-field system $Tb(As_{0.15}V_{0.85})O_4$ based on an optical interrogation approach which provides submicron resolution through speckle interference effects.

The structural phase transition in $TbVO_4$ from a tetragonal phase to a low-temperature twinned orthorhombic phase is driven by the coupling between the Tb electronic levels and the B_{2g} lattice distortions.[11] Below the transition temperature T_D, the order parameter σ is proportional to the B_{2g} strain. To minimize bulk sample strains, and the elastic energy of the interfaces separating the two equivalent orthorhombic orientations, the domain walls form regularly spaced {100} planes throughout the crystal. The random strain fields introduced by the As-V size mismatch in mixed crystals roughen and pin these domain walls, and depress the transition temperature.[12]

The essential periodicity in the domain structure of this and similar crystals enables the observation of the Bragg diffraction of visible light.[13] Whereas the diffraction angle $\theta_B = \sin^{-1}(\lambda/2d)$ is unique for a truly periodic arrangement of specular planes (where d is the interplanar separation and λ the probing wavelength), it spans a range of angles if there exists a distribution in d and local deviations from parallelism.[13] In the latter case, the consequent phase shifts induced in the scattered light are manifest as speckle in the diffracted light intensity.[14] Due to the optical field-averaging process inherent in the Bragg condition, these speckle features are sensitive to the changing interfacial topologies arising from order parameter evolution. In principle, domain wall topologies can be inferred from the speckle pattern, but this is unrealistic in practice for large scattering volumes. An alternative approach, which we have followed, is to extract average characteristics of an ensemble of such Bragg-scattering surfaces from fluctuations in the total scattered intensity.

Consider a planar monochromatic beam of light incident on an array of periodic rough planes such that the Bragg condition is satisfied. We define the roughness as the root-mean-square height deviation h from the mean planar position, and describe the scattered light as either specular or diffuse. Under the assumption that the roughness is normally distributed (but not sufficiently large to cause extinction of the specular component), the Bragg condition ensures that in the far-field free-space geometry only the specular component will be detected. Such a situation is equivalent to observing only specular scattering from a single rough surface. Taking advantage of the extensive research on electromagnetic wave scattering from rough surfaces,[15,16] we treat an ensemble of such equivalent surfaces to determine the mean character of the interfacial topology from the specularly scattered intensity.

The crystals were cut and polished perpendicular to their c axes, then individually mounted in an optical He-flow cryostat with the c axis nearly collinear with the incident linearly polarized laser light (543.5 nm), but oriented at a Bragg

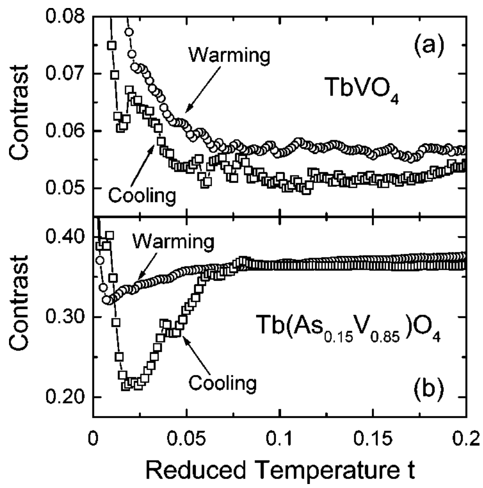

FIG. 1. Contrast vs temperature for (a) pure $TbVO_4$ and (b) the RFIM system $Tb(As_{0.15}V_{0.85})O_4$.

angle of $\theta_B = 3.8°$. Pinhole apertures against the polished faces allowed entrance and egress of the light, thus providing a uniformly illuminated and well-defined scattering volume. As the speckle pattern produced by the Bragg diffraction broadens with decreasing temperature (θ_B remains unchanged, however),[14] a wide-aperture photomultiplier tube, positioned in the far-field to exclude the zero-order beam, was used to detect the scattered light over the range of angles associated with the diffraction (rather than merely its peak position). This retained the scattering information from the entire distribution of interfaces and prevented bias in the self-averaging accomplished by the diffraction. A 12-bit PC-based data acquisition card was used to obtain data samples every 0.05 K (about once every second) as the temperatures were slowly varied. An ensemble of such specularly scattered intensities (approximately 100) was then acquired by repeatedly warming the samples past their transition temperatures and then cooling them down into another microscopically unique, but macroscopically identical, domain configuration. The topological properties of the domain walls may then be studied, both on warming and cooling, by determining the mean intensities $\langle I(t) \rangle$ over the measured temperature range (where $t = 1 - T/T_D$ is the reduced temperature), and calculating their standard deviations $s(t)$.

To determine whether the random fields do indeed roughen the domain walls, we extract the contrast $C(t) = s(t)/\langle I(t) \rangle$ for both the pure and mixed systems. Assuming the individual intensities in the ensemble to be normally distributed, the contrast will be equal to unity if the mean interfacial roughness is large compared to the wavelength; the contrast vanishes if the interface is smooth.[17] In Fig. 1(a), we note that for $TbVO_4$ the contrast is extremely low (yet increasing markedly near the transition temperature $t = 0$), although it remains nonzero, presumably due to the pinning of domain walls by surface defects and sample inclusions.[18] In addition, whether the contrast is determined upon cooling or warming the sample appears to make little difference, implying that the onset of LRO occurs in the same manner as its departure. In comparison, Fig. 1(b) shows that for $Tb(As_{0.15}V_{0.85})O_4$ the contrast is about six times larger, showing that, as expected, random fields do roughen and pin the domain walls. We also find, for temperatures immediately below the transition ($0.01 < t < 0.06$), a marked difference between contrasts extracted from sample-cooled and sample-warmed data. We argue that this is due to the formation of metastable microdomain states[7,8] upon initial cooling, before LRO is fully established, and further discuss this point below.

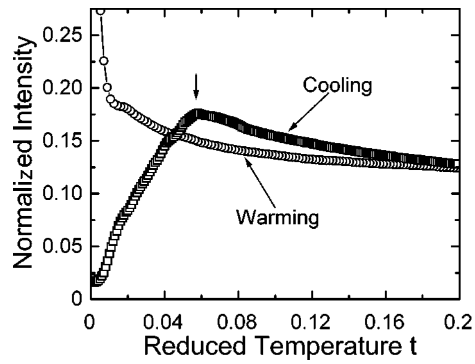

FIG. 2. Normalized intensity of light Bragg scattered from $Tb(As_{0.15}V_{0.85})O_4$ as a function of reduced temperature.

The interfacial topology is not described solely by its roughness, but also by its roughness gradient, or interfacial correlation length ξ. Using Beckmann's facet model,[15] where the surface consists of horizontal facets (of area ξ^2) with a Gaussian height distribution, Pedersen[19] has calculated the contrast for our experimental geometry as

$$C(t) = \left\{ 1 - \left[1 - \frac{\xi^2(t)}{A} \{ 1 - \exp[\phi^2(t)] \} \right]^{-2} \right\}^{1/2}, \quad (1)$$

where A is the mean cross-sectional area of the sample along the c axis, and

$$\phi(t) = \frac{4\pi}{\lambda} h(t) \sin(\theta_B) \quad (2)$$

is the phase deviation due to surface roughness (measured in terms of the Bragg angle). It is thus apparent that the contrast data alone are insufficient to characterize the interfacial topology. For our system, however, a quantitative application of Rayleigh's criterion,[15,16] which uses the phase deviation to characterize the surface, has allowed us to extract the roughness from the mean intensity of the specularly scattered light, and thus the correlation length, from the contrast data. The intensity of light scattered from a rough surface in the specular direction, compared to that scattered from a smooth surface, is described as[15,16] $\langle I_{\mathrm{mixed}}(t) \rangle = \langle I_{\mathrm{pure}}(t) \rangle \exp[-\phi^2(t)]$ where $\phi(t)$ is defined in Eq. (2). The validity of this formulation for our system requires LRO as well as[16] $\xi \gg h$ to avoid surface self-shadowing (of concern due to the small Bragg angle). To determine the effect of random fields on interfacial roughening, we normalize the intensity of the light Bragg-diffracted from $Tb(As_{0.15}V_{0.85})O_4$ by that Bragg diffracted from $TbVO_4$. The experimental determination of this normalized intensity, whose uncertainty is approximately 15%, is shown in Fig. 2 as a function of the reduced temperature t. The warming data show a smooth increase in the normalized intensity, rising rapidly to unity as the transition temperature is approached. The cooling data do not display this smooth change in the normalized intensity, first rising

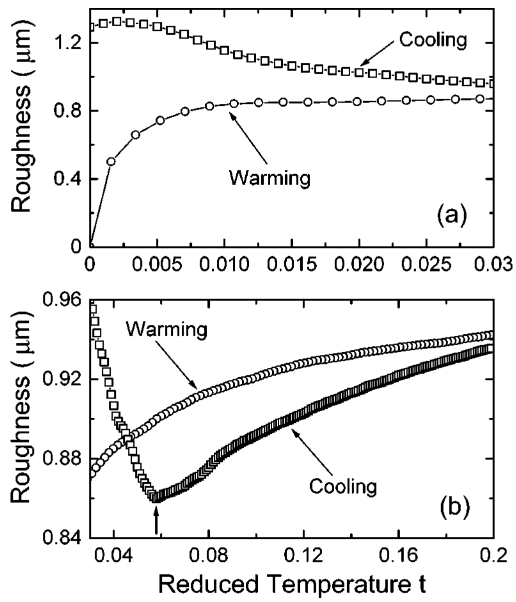

FIG. 3. Interfacial roughness in $Tb(As_{0.15}V_{0.85})O_4$: (a) near the phase transition, (b) at lower temperatures.

swiftly from near zero before peaking, then undergoing a moderate decline as the temperature is yet further reduced. Comparison with the cooling data in Fig. 1(b) reveals that the initial depression in the normalized intensity is also manifest in the contrast. These effects are attributed to incomplete development of LRO (i.e., the macroscopically twinned orthorhombic state) in this temperature regime. The orthorhombic phase nucleates randomly out of the parent tetragonal lattice, forming regions of local order (microdomains) that become pinned by the random fields. The normalized intensity is initially quite small because there is barely any preferred global alignment of the twin walls, hence little diffraction. These microdomain states are metastable in the sense that, although they persist for all experimentally observable times, as the temperature is reduced the increasing strains will steadily depin these regions and promote LRO. Similar behavior has been noted for DAFF systems.[10] At $t=0.057$ (black arrow, Fig. 2), LRO has been fully established, and the behavior of the normalized intensity now describes the mean interfacial roughening as the sample is cooled.

The roughness $h(t)$ extracted with the use of the above expressions for $\phi(t)$ is plotted in Fig. 3 as a function of temperature. Figure 3(b) shows that as the sample is warmed up, the domain walls undergo a moderate degree of smoothing, from 0.94 μm at $t=0.2$ to 0.88 μm at $t=0.04$, before rapidly rolling off to vanish at transition [Fig. 3(a)]. Sample-cooled data immediately below transition ($t<0.057$; black arrow) presumably does not represent true interfacial roughening, but is in some sense a measure of the misalignment of the domain states with respect to LRO. For $t>0.057$, this roughening is less, by about 5% at the point of establishment of LRO, than that found from the sample-warming data, although this difference is steadily reduced as the temperature is further lowered.

The mean intensity data were combined with the contrast data to determine the interfacial correlation length using Eq. (1); the results are shown in Fig. 4. The correlation length is

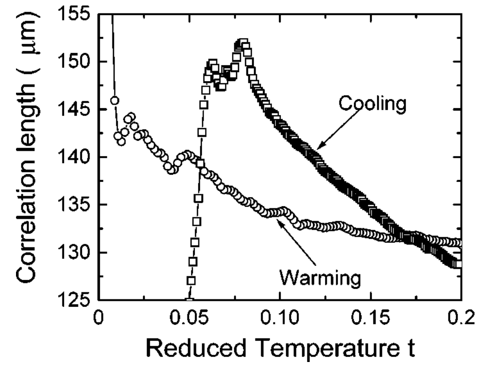

FIG. 4. Dependence of the interfacial correlation length ($\propto$ roughness gradient) on reduced temperature.

large, up to $\sim$15% of the sample dimensions. As the sample is warmed, ξ shows a gentle increase then diverges to sample dimensions upon approaching transition. The cooling data (once LRO has been established) are significantly larger initially but the change is much more rapid than for warming, so that at the lowest temperatures the correlation length is smaller for sample-cooled than for sample-warmed data. Finally, $h/\xi\sim 10^{-2}$ so surface self-shadowing has no significant effect despite the small Bragg angle.[16]

Since the intensity of the scattered light varies as the square[13] of the order parameter σ the scaling behavior of the roughness may be determined from a log-log plot of h against $I_{\rm rf}^{1/2}$, where $I_{\rm rf}$ is the Bragg-scattered light intensity. This is shown for the rf system in Fig. 5, for both warming (a) and cooling (b) data. The warming data reveal that, very close to the transition temperature, the roughness is consistent with a $\sigma^{2/3}$ dependence, as earlier predicted.[8] However, over most of the temperature range the roughness is only weakly dependent on order parameter, being proportional to $\sigma^{0.084}$. The reason for this is not understood. The cooling data (once LRO has been achieved) show a somewhat stronger $\sigma^{0.18}$ dependence. Interestingly, the disappearance of the metastable domains upon cooling also scales with order parameter, with a rather abrupt transition to the LRO scaling regime.

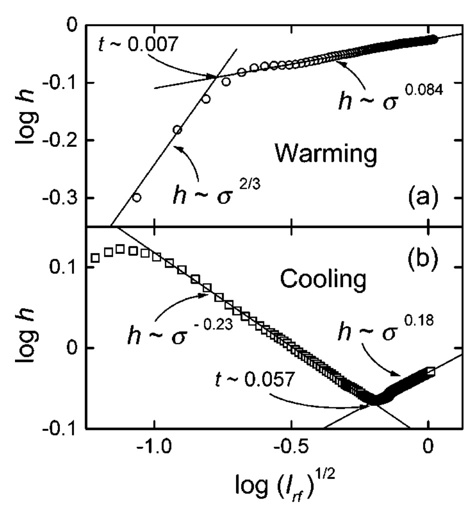

FIG. 5. Scaling behavior of roughness with order parameter: $\log h$ vs $\log I_{\rm rf}^{1/2}$ for (a) warming, (b) cooling data.

In summary, measurements of the Bragg-scattered light as a function of temperature for both sample warming and cooling have enabled quantitative determination of the roughness and interfacial correlation length, and the identification of a metastable microdomain state for a structural RFIM system. Over most ($0.057 < t < 0.17$) of the temperature range investigated, the domain walls appear slightly rougher upon sample warming than otherwise, while at very low temperatures the opposite may be true. Near transition, upon sample warming, the interfacial properties approach those of pure $TbVO_4$. We hope these experimental results will further motivate inquiry into interfacial topologies and their connection to critical properties of RFIM systems.

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