Neutron-scattering study of interfacial roughening in twinned orthorhombic $Dy(As_{0.15}V_{0.85})O_4$

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In $Dy(As_{0.15}V_{0.85})O_4$, a structural example of a random-field Ising system, As/V substitutions depress the tetragonal-orthorhombic transition temperature and roughen the interfaces between twinned domains. The transverse broadening of diffraction peaks due to interfacial roughening in the orthorhombic phase has been studied by high-resolution neutron scattering. The observed broadening is interpreted with the use of a simple random-walk model that gives a value of 0.11 μm for the roughness parameter of the domain walls. Just above the transition temperature, a broad peak is visible whose width indicates short-range orthorhombic ordering with a similar length scale. Application of an ordering field shows that the interfaces become smoother as they are driven together, and are less rough than originally, when they reform after removal of the field.

I. INTRODUCTION

The effects of random fields on phase transitions, in particular for Ising systems, have been subjects of theoretical, experimental, and computational study for some twenty years. Imry and Ma[1] introduced a model for this system, the validity of which has been confirmed by subsequent analysis, in which random fields may pin the interfaces between domains of oppositely ordered spins and prevent long-range order (LRO).[2–4] Although it is now accepted that LRO is the equilibrium state in $d = 3$ dimensions, in practice systems may enter a nonequilibrium state characterized by microdomains whose walls are pinned for geological times by the random fields. A major focus of interest, then, is the interface between ordered domains that results from the competition between the spin-spin interactions and the Zeeman energy of spins in the random field. Spin-spin interactions alone would give smooth, mobile interfaces, but random fields favor a rough interface corresponding to the topology of the random field. As the system cools, it may be trapped in a metastable state with domain walls characterized by roughness on some length scale.

Notwithstanding the central role of domain size and roughness on the properties of random-field Ising (RFI) systems, rather little direct experimental information on domains has been obtained. The applicability of some experimental probes is unfortunately limited: most Ising systems are insulators so that electron microscopy is unsuitable, and the need to examine bulk as opposed to surface behavior is a disadvantage for x-ray studies.[5] Optical probes generally lack resolution for the study of domain topology, although speckle techniques can provide some information on the scale of a small fraction of the wavelength.[6] Neutron scattering has given important information on domain states in dilute antiferromagnetic (DAF) systems[7] through the line shapes of magnetic peaks after cooling in a random field. These studies provide estimates of domain sizes, but have not been able to confirm in a reasonably direct way that random fields do roughen domain walls, or to investigate the effects of temperature and ordering fields on wall roughness.

In recent work some structural phase transitions where random strain fields are introduced by impurity atoms have been identified as examples of the random-field Ising model.[8–11] Characteristic changes in critical exponents[10,11] and in equilibration behavior[8] in the mixed compounds $Dy(As_xV_{1-x})O_4$ compared to pure $DyVO_4$ were attributed to random-field effects.

For these systems, however, as well as for DAF systems, agreement with theoretical predictions, for example on critical exponents, is unsatisfactory. A better understanding of domain behavior in these systems would therefore be valuable, and might identify deficiencies in either the experiments or the theory. The domain topology of $Dy(As_xV_{1-x})O_4$ crystals differs from that in DAF crystals in an important respect: the low-temperature phase of the former, like those in many crystals undergoing tetragonal-orthorhombic transitions, has twinned orthorhombic domains whose configuration is essentially independent of composition, as long as random fields are not so large that the transition is suppressed. The domain walls are {110} planes, typically separated by a few mm. Random fields thus have the effect of roughening and pinning domain walls but they remain planar on average. The existence of twinned orthorhombic domains in the pure compounds implies long-range anisotropic elastic interactions that are not included in the basic Ising model, but interactions of this length scale are not expected to modify the basic picture of the random field effects.

In the course of neutron-scattering experiments on some $Dy(As_xV_{1-x})O_4$ compounds[9] we observed an effect that appeared to depend on roughness of the interfaces between orthorhombic twins. Diffraction peaks studied in high resolution showed additional transverse broadening below the transition, and this broadening was small for dilute samples and substantial for higher concentrations. An interpretation in terms of a mosaic-type broadening due to roughening of domain walls by atomic substitutions was proposed.[9] These experiments give only an average measure of wall roughness but do provide the first reasonably direct evidence that roughening by random fields does occur in these systems. The main purpose of the present paper is to describe results of a more systematic and detailed investigation of this transverse broadening and its implications for roughening of interfaces.

The tetragonal-orthorhombic transition in $DyVO_4$ and related crystals is a cooperative Jahn-Teller transition driven by the coupling of nearly degenerate Dy electronic levels with lattice distortions. This phase transition is rather well understood; previous work gives details on the microscopic origin of the interactions and their description in terms of a pseudospin Ising model.[12] Lattice defects, whether of natural occurrence or due to impurity atoms, will locally induce distortions of orthorhombic symmetry that can be described by a site-random Zeeman field term in the Hamiltonian.[8] The mixed crystals $Dy(As_xV_{1-x})O_4$ have been studied for a wide range of compositions x by dielectric,[8] neutron-scattering,[9] ultrasonic,[10] and optical[11] probes to investigate effects of random fields on the phase transition, especially on the critical properties. As x or $(1-x)$ increases, the transition temperature T_D is depressed relative to the values 14.6 K for $x=0$ and 11.4 K for $x=1$. For x in the range $0.2 \leq x \leq 0.8$ approximately, no transition is observed. For the present experiment we chose a sample with $x=0.15$ (given as 0.154 in some previous reports[9–11,13]), as a good compromise between convenient temperatures, strong random field, and crystal size and quality.

II. EXPERIMENTS

The objectives of the experiment were to record the line shape of Bragg peaks (especially transverse to the scattered-neutron wave vector) as a function of temperature through and below the tetragonal-orthorhombic transition, and as a function of applied field at fixed temperatures in the ordered phase while a single domain is induced. The formation of twins in the orthorhombic phase is a complication in the study of Bragg peaks. Domain walls between regions corresponding to the two choices of orthorhombic distortion lie along {110} planes to minimize elastic energy, and as there are two orientations of {110} planes a given Bragg peak hkl may split into four peaks in general. To follow these peaks in reciprocal space requires time-consuming raster scans.[9] For peaks hhl, however, only three peaks are expected, as shown in Fig. 1, which is redrawn from a paper by McIntyre *et al.*[14] Since the splitting of such peaks is collinear, the transverse line shape can be recorded in a single scan at a great saving of beam time. For optimum intensity and resolution the {660} Bragg peaks were studied primarily.

Since the sample was fairly small (3x3 mm in the basal plane, 4 mm along the c axis), it was important to configure the spectrometer to conserve intensity as well as to obtain high resolution. Thus the sample, mounted with c axis vertical and perpendicular to the scattering plane, was used as a collimating element without any other collimators. A monochromator and analyzer (set for elastic scattering) were chosen to give a "W" geometry. The scattering angles of the monochromator, analyzer, and sample (660 planes) were roughly equal to each other and to 90°. The monochromator (Ge 335) and the analyzer (Si 135) had mosaic spreads of 0.06° and 0.12°, respectively. The experiment was carried out at the DUALSPEC C5 spectrometer at NRU, Chalk River, in a horizontal-field cryostat, with a neutron wavelength of 1.15 Å. Peak counts at the 660 Bragg peak were approximately 150 s^{-1}, with transverse and radial resolutions of approximately 0.045° and 0.062° full width at half maximum (FWHM).

The main features of the temperature dependence agreed qualitatively with expectations. Figure 2 illustrates the developments in the transverse line shapes of the 660 and $\bar{6}60$ peaks at three representative temperatures. Above T_D the line shape in both the transverse and radial directions was close to Gaussian. As the sample is cooled through T_D, the main peak broadens and additional peaks appear that move out from the main peak

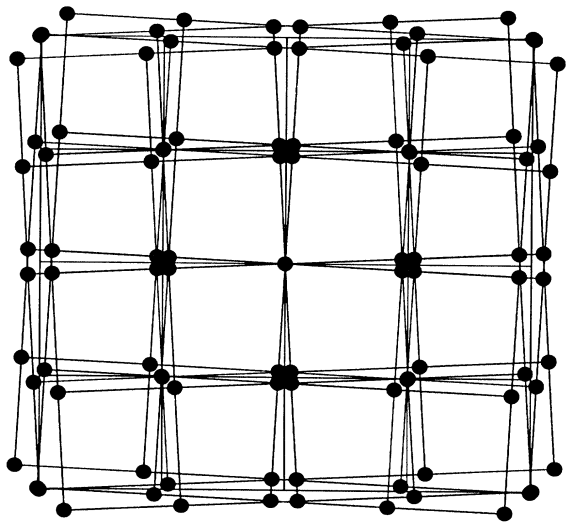

FIG. 1. The orientations of basal plane axes for the tetragonal lattice (horizontal and vertical) and the four orthorhombic lattices (tilted) resulting from twinning about (110) and ($\bar{1}10$) planes. Solid circles show reciprocal lattice points in the orthorhombic phase.

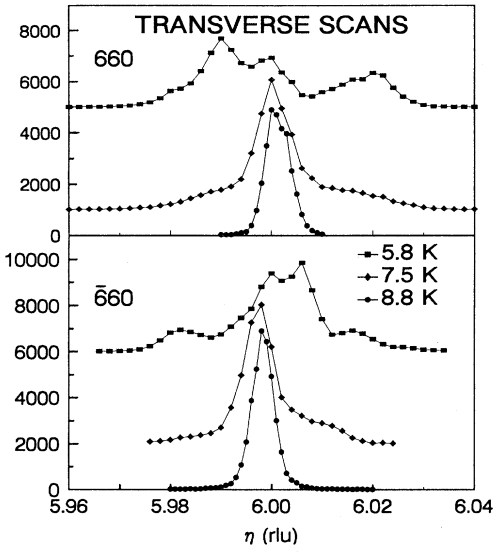

FIG. 2. Transverse line shapes at 660 and $\bar{6}60$ at temperatures above and below the tetragonal-orthorhombic transition. The horizontal scale shows the η (y) component of the scattered-neutron wave vector in reciprocal lattice units. (1 rlu = 0.885 Å^{-1}). Data at different temperatures are shifted vertically for clarity.

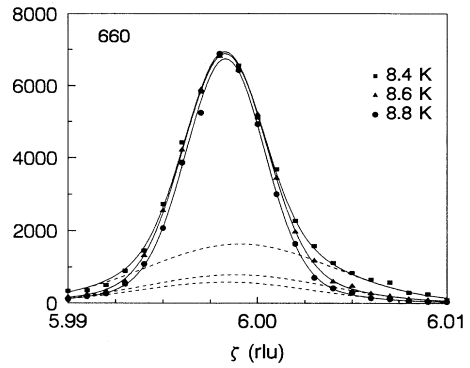

FIG. 3. Transverse line shapes at 660 just above the tetragonal-orthorhombic transition showing the growth of a broad peak attributed to short-range order. Solid lines show fits to the sum of two Gaussians while dashed lines show the fitted broad component. The horizontal scale shows the ζ (x) component of the scattered-neutron wave vector.

in directions perpendicular to the scattered-neutron wave vector. The expected three-peak structure (Fig. 1) is not clearly resolved because of the linewidths, but is recognizable. The intensities are not in the ratio 1:2:1 that would be expected for an ideally twinned lattice with all domain orientations equally probable. Evidently there are sample inhomogeneities that favor some orientations more than others. Note that the flattening of the top of the $\bar{6}60$ peak suggests that extinction is a significant effect; this is also implied by a significant growth in integrated intensities as the lines broaden at lower temperatures. It was found that when the sample was cooled through T_D to the same temperature at different times during the course of the experiment the diffraction line shapes were similar but not identical. This is consistent with a situation where the domain configuration is determined primarily by local defects and mounting stresses. The radial line shapes, as expected, are essentially unchanged over the same range of temperatures. The radial line width does increase slightly, by up to 15% at the lowest temperatures, but this may be a side effect of the dramatic changes that occur in the transverse direction.

From a detailed analysis of the transverse linewidths we deduced that the transition temperature was $T_D = 8.4 \pm 0.1$ K. Above this temperature small but significant precursive changes in the line shape could be detected, in the form of a broad peak developing underneath the main peak. A set of scans was carried out to follow the growth of intensity in the wings as T_D is approached from above, and Fig. 3 shows some of the data. The broad peak was not detectable above 9.5 K in these experiments.

Line shapes below T_D were also examined while the sample was progressively forced into a single domain. When an ordering field is applied the favored domains grow at the expense of the disfavored, and thus the interface between them is driven through a fluctuating random potential. The analysis of the diffraction linewidth during application and removal of the field will reveal any changes in the roughness of the interface. Hysteresis in both the domain configuration and the wall roughness may occur.

In this system the quantity B_x^2/T acts as an ordering field, at least to a good approximation,[11] where B_x is a magnetic field applied parallel to one of the basal

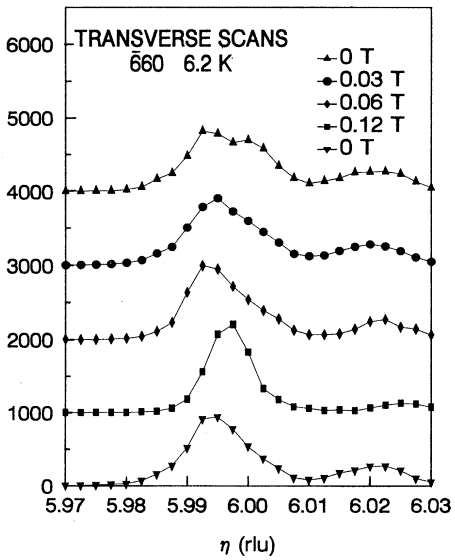

FIG. 4. Line shapes for the $\bar{6}60$ peak at 6.2 K as an ordering field is applied and removed. Triangles pointing up and down denote zero-field data before and after the field cycle, respectively.

plane axes. The field required to reach a single domain is about 0.15 T. For increasing fields the line shape changes from the broadened multipeak structure characteristic of twinned domains and approaches a single narrow peak appropriate to a single domain. Except for temperatures close to T_D there is significant hysteresis associated with the response of domains to the field, and to explore this we recorded spectra for both increasing and decreasing fields with the temperature fixed at 7.8 K, 6.2 K, and 4.7 K. Figure 4 illustrates the general behavior at 6.2 K.

In the orthorhombic phase the Dy ground state wave functions are altered to give a highly anistropic magnetic moment[12] with $g_a \approx 20$, $g_b \approx 0$. Magnetic interactions of Ising form result, and lead to antiferromagnetic ordering near 3 K for $DyVO_4$ and $DyAsO_4$. We investigated the magnetic transition temperature in our mixed sample to see if the random strains had an appreciable effect. Scans through the strong 100 magnetic peak, a forbidden peak in the chemical unit cell, were carried at low temperatures. The 100 peak appeared at about 2.9 K, indicating that magnetic ordering was not significantly affected by the random strains.

III. INTERPRETATION

In this section we present our results in more detail and analyze them in terms of domain formation and interfacial roughening. Each of the experiments performed will be discussed separately.

A. Line shapes at $T > T_D$

As shown in Fig. 3, transverse scans through the Bragg peak just above T_D reveal a broadening that appears to be a precursor of the displacements due to orthorhombic twinning below T_D. As indicated, the line shapes can be fit satisfactorily by the sum of two Gaussians, both centered at the 660 position, with widths in the ratio of 2.3:1 and nearly independent of temperature. The amplitude of the broad component is negligibly small above 9.2 K but increases rapidly as the temperature approaches T_D. Figure 5 shows the temperature dependence of the amplitude and width of the broad component.

It seems reasonable to associate this broad feature with short-range orthorhombic ordering. Since its width changes very little with temperature, the range of the ordering is likely controlled by the static random fields rather than by the scale of thermal fluctuations. That is, there is presumably some range, determined by the concentration of substitutional As impurities, over which the random fields favor, and induce, the same orthorhombic distortion. If the additional width of the broader line, amounting to 0.010 reciprocal lattice units (rlu), is attributed to the finite size of the induced distortion, that size would be about 0.075 μm, equivalent to 100 unit cells.

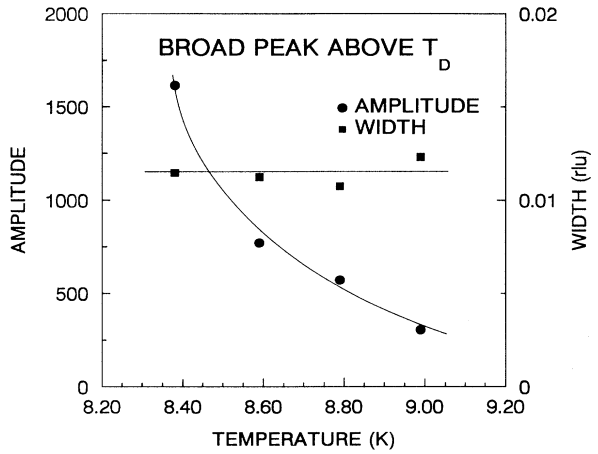

FIG. 5. Amplitude and width of the broad peak observed above T_D derived from fits as shown in Fig. 3. Lines are guides to the eye.

B. Line shapes below T_D

As already mentioned, for a normally twinned crystal there should be three peaks at the positions 660 and $\bar{6}60$: a central peak and two side peaks. The lowest-temperature scans in Fig. 2 show a recognizable three-line pattern, but (110) twinning planes predominate over $(\bar{1}10)$ so that the intensity ratios are not 1:2:1. Additional structure is also evident, implying that more complicated twinning patterns may be present. In some cases this structure shows sharp, resolution-limited features, suggesting that sizable single-domain regions occur. We will essentially ignore this extra structure, and focus attention on the widths of the main components, and their temperature dependence.

It must be emphasized that the additional transverse broadening that develops below T_D is a feature of samples with considerable As content and is expected to be very small for a pure $DyVO_4$ sample. Although we have not carried out the identical experiment for a pure sample, previous experiments on samples with a range of compositions[9] showed that the extra broadening increases with impurity concentration, and was quite small for a sample with only 0.5% concentration. This broadening is attributed to a mosaic of domains for which the misalignment of unit cell axes arises through the roughness of the interfaces between domains.[9]

Data such as those in Fig. 2 were analyzed by fitting multipeak Gaussian peaks to the observed peaks. The widths of the central and side peaks were similar, and both increased rapidly below T_D. The quality of the fits was only fair owing to the irregular line shapes arising from domain textural effects. Figure 6 shows an example of the fits obtained, in this case to the sum of four Gaussians. Figure 7 plots, as a function of temperature, the additional broadening, relative to the $T > T_D$ linewidth, of the central peak as well as one of the side peaks where it could be resolved. Notwithstanding the line structure due to texture effects, we see that there is a well-defined

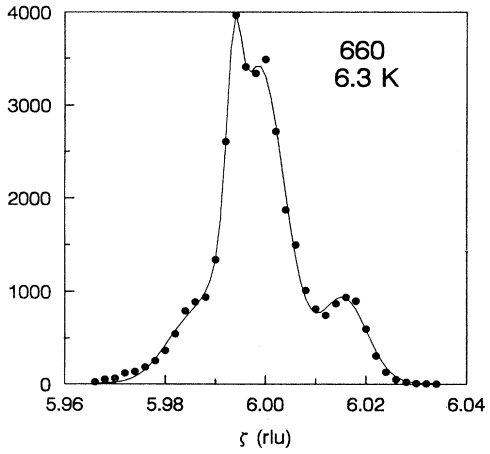

FIG. 6. Example of a fit to a peak in the twinned orthorhombic phase, in this case to four Gaussians of independent width, amplitude, and position.

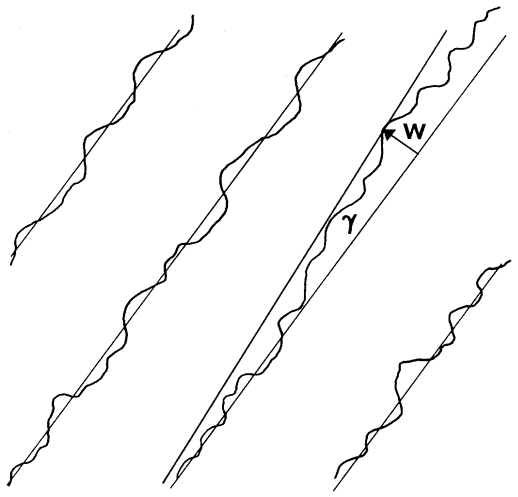

FIG. 8. Schematic illustration of twinned domain walls with roughness scale w and angular deviation γ. Straight lines show (110) planes in the undistorted lattice.

linewidth contribution that grows smoothly below T_D. Since this broadening occurs only in the direction transverse to the scattered-neutron wave vector, and not in the radial direction, we attribute it to a mosaic effect where alternating domains do not remain accurately parallel throughout the crystal due to the roughness of the interfaces.

The observed "roughness broadening" can be related to an interfacial roughness parameter by means of a simple random-walk model. Suppose that the roughness parameter of an interface is w, leading to a misorientation of an interface from the (110) plane by the angle γ, as sketched in Fig. 8. Then γ represents an angular step on crossing the interface with the average value w/L. The probability that a given domain, n interfaces away from a reference domain, has a misorientation $m\gamma$ is[15]

$$P(m) = (\pi n/2)^{-1/2}\exp(-m^2/2n). \quad (1)$$

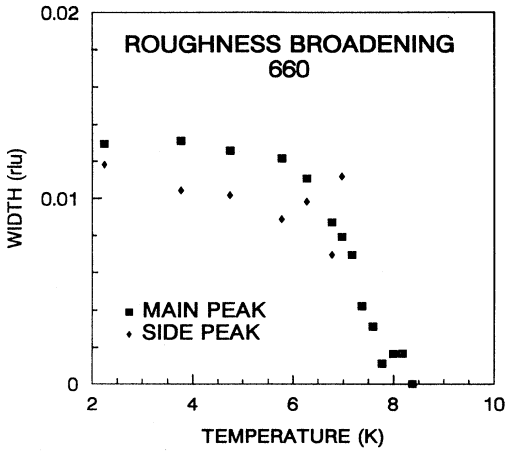

FIG. 7. Interfacial roughness contribution to the linewidth of the 660 peak as a function of temperature.

Assuming m, n are large numbers, we can identify the width of this distribution with the width of the observed mosaic broadening. The angular width of this broadening at the lowest temperature (see Fig. 7) can be taken as 0.0020 rad (FWHM). If the width of the scattering volume is L =3 mm and the average domain thickness is $t = 3$ μm,[6] we have $n \approx L/2t = 500$. Thus the FWHM of Eq. (1) corresponds to $2.355\sqrt{n}$ or 53 steps, and the average angular step size γ must be 0.0020/53 or 3.8×10^{-5} rad. The roughness parameter w is then $\gamma L = 0.11$ μm, apparently a reasonable value representing a few percent of the domain width.

The temperature dependence of the roughness broadening (Fig. 5) can be attributed in the simplest model entirely to the order parameter temperature dependence. This assumes that the roughness w, being controlled by the random fields, does not vary with temperature, so that the broadening depends only on the orthorhombic distortion.

More sophisticated models for the origin and roughness of interfaces in the RFI model have been presented in the literature.[16] The model of Andelman and Joanny,[17] specialized to the case of planar interfaces, was applied to earlier neutron-scattering data.[9] It gave a plausible interpretation of the dependence of broadening on As concentration for several samples, but the available data were not able to test other predictions fully. According to this model, the roughness parameter w should have a temperature dependence that goes as $\sigma^{2/3}$, where σ is the order parameter. Although σ is available from other experiments,[6] the data of Fig. 7 do not have the accuracy to determine whether the broadening is proportional to σ or $\sigma^{2/3}$.

It is interesting that the scale of the roughness of the interface between two domains is very similar in magnitude to the scale of short-range orthorhombic order observed just above T_D. In both cases they represent the range

over which an orthorhombic domain of one orientation is able to persist against thermal fluctuations on the one hand or a domain of opposing orientation on the other.

C. Ordering fields

Many of the basic ideas on domains and domain wall behavior that emerged through research on ferromagnetic domains[18] are applicable to structural domains. The domain configuration and the equilibrium wall separations minimize the total energy (elastic in this case) of the crystal. An ordering field drives the interfaces together, over energy barriers due to random strain fields, but the interfaces tend to return to a similar if not identical pattern when the field is removed. Hysteresis will be present to a greater or a lesser degree depending on the temperature, since thermal energy is needed to allow the interface to cross the barriers.

The data of Fig. 4, taken at 6.2 K, show the expected steady progression towards a single domain. The lines corresponding to particular domain orientations can be seen to be narrowing for increasing fields. This implies that the interfaces become smoother as they are pushed together, although the disappearance of some interfaces would reduce the number of "steps" in the random-walk effect, and hence the widths. Data taken at 4.7 K (not shown) are very similar, but hysteresis is more pronounced: for example, the spectrum is unchanged when a field of 0.03 T is applied. This observation may be compared with studies[19] using other techniques that identify a depinning field, that is, the minimum ordering field required to initiate motion of domain interfaces, in this system. Our experiments imply that the depinning field is less than 0.03 T at 6.2 K and greater than 0.03 at 4.7 K, consistent with the earlier data for a sample with the same concentration of As impurities.[19]

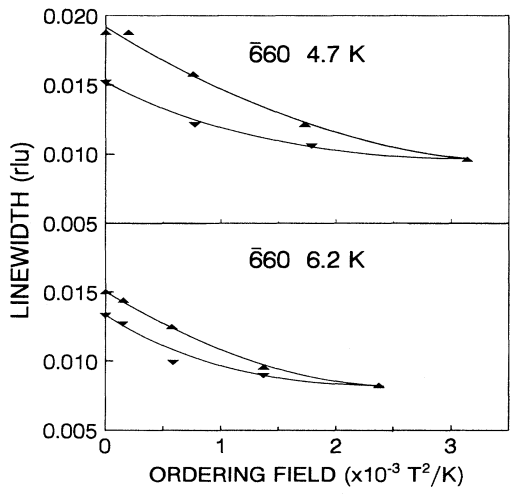

FIG. 9. Changes in $\bar{6}60$ linewidth for application and removal of ordering fields at 6.2 K and 4.7 K. Triangles pointing up and down refer to increasing and decreasing fields, respectively. Solid lines are guides to the eye.

Linewidths were determined from Gaussian fits for different fields and are shown for both 6.2 K and 4.7 K in Fig. 9. These data also show quite clearly that the linewidths are narrower after the field returns to zero. The difference is more marked at 4.7 K, consistent with the general increase of hysteresis at lower temperatures. Data taken at a temperature of 7.8 K (not shown) were qualitatively consistent with that for lower temperatures. The initial spectrum consisted of a strong peak, still quite narrow, with broad, poorly resolved structure (similar to that for 7.5 K in Fig. 2). For increasing fields the broad structure narrowed and weakened, but changes in the strong peak were insignificant. Hysteresis was not detectable at this temperature.

IV. CONCLUSIONS

High-resolution neutron-scattering measurements provide a reasonably direct probe of the roughness of planar interfaces such as those between orthorhombic twinned domains. Data taken over a range of temperatures support a picture where the local random fields due to As impurities induce short-range orthorhombic distortions of about 0.1 μm size that give a broad peak in the scattering just above T_D. These ordered regions tend to persist when domain walls form, and set the scale of the roughness of the walls. At lower temperatures the interfacial roughness causes the twinning angles to depart from their average value, giving a mosaic-type broadening to the diffraction peaks. From the observed broadening and a simple random-walk model, the roughness scale of the interfaces in this sample was estimated to be 0.11 μm. This can be compared with the separation between domain walls of approximately 3 μm, and the thickness of a typical wall in this type of system[20] of about 3 nm.

An ordering field greater than the depinning field drives interfaces together to create a single domain. The data imply that the interfaces become somewhat smoother under the action of the applied field, and when the domains reform after its removal the interfaces are likewise smoother than originally. Pronounced hysteresis effects become apparent as the temperature is lowered.

The antiferromagnetic 100 peak was observed to appear at about 2.9 K, indicating that the random strain fields do not significantly depress the magnetic ordering temperature relative to the pure compound. This is not surprising, since the orthorhombic distortion, when it occurs, will overwhelm the random strains. All the Dy spins in a given domain will then have the same lattice structure and anisotropy axis. A much different situation obtains when the random fields are large enough to suppress the tetragonal-orthorhombic transition, leaving the Dy spins exposed to random local distortions and thus to competing anisotropy axes. Preliminary experiments on a sample with 35% As concentration[21] show that antifer-

romagnetic ordering occurs at a much lower temperature in this case.

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