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Observation of impurity-assisted depinning of domain walls

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Abstract. The depinning fields for the twinned orthorhombic domains in $DyVO_4$ and $Dy(As_xV_{1-x})O_4$ crystals have been determined from optical birefringence hysteresis measurements. In this Ising model system the depinning field in a mixed crystal ($x = 0.15$) is found to be much smaller than that in pure $DyVO_4$ at low temperatures. An impurity mechanism that assists the motion of domain walls appears to be more important than pinning by the random strain fields.

The domain properties of ordered phases have been of interest for many years, for both technological and fundamental reasons. Basic information on the displacement of domain walls by an applied field can be obtained from hysteresis plots, but real materials are sufficiently complicated that it is difficult to make detailed comparisons between theory and experiment. This paper presents results of measurements of ordering and hysteresis in a ferroelastic system where the phase transition and the domain configurations are different from, and simpler than, those usually studied. A notable observation is that the depinning field in this system is *reduced* at low temperatures by the addition of impurities that generate random fields. This result is the opposite of what is usually expected and observed, and may be characteristic of the particular case of an Ising model system with thin domain walls.

The compounds investigated are $DyVO_4$ and $Dy(As_{0.15}V_{0.85})O_4$. $DyVO_4$ and $DyAsO_4$ undergo tetragonal-to-orthorhombic transitions (D_{4h}^{19}–$I4_1/amd$ to D_{2h}^{28}–$Imma$) near 14 K and 11 K, respectively, driven by the coupling between the nearly degenerate Dy ground electronic levels and B_{1g} lattice distortions [1, 2]. This coupling can be described by an Ising Hamiltonian in a pseudospin representation, where the two spin states correspond to the two orientations of the orthorhombic distortion. In the mixed system the interactions are not diluted, but the As–V size mismatch generates random static strain fields at each site, one component of which has B_{1g} symmetry and couples to the order parameter. This system therefore provides a structural realization of the random-field Ising model [3]. The random fields are expected to pin the domain walls and affect both the critical and hysteresis properties.

The low-temperature phase has a domain structure, orthorhombic twinning, that occurs commonly in compounds with similar phase transitions, including copper oxide superconductors and ferroelectrics such as KDP. Domain walls along {110} planes minimize the elastic strain energy between regions of differing orthorhombic distortion orientations. The directions of the a and b axes of the basal plane of the orthorhombic unit cell in adjacent domains rotate away from each other in proportion to the order parameter $b - a$,

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and alternate through successive domains. This domain structure has been observed in optical reflection [4] experiments and also in optical and neutron diffraction experiments [5, 6]. Microphotographs [4] of $DyVO_4$ indicate an average domain width of about 2 μm, and domain walls extending over 0.1 to 1 mm, comparable to the sample size. A single domain is readily induced, either by a stress or a magnetic field parallel to a basal plane axis. In the latter case, the strong anisotropy in g_a, g_b in the orthorhombic phase favours the orientation for which the larger magnetic moment is parallel to the applied field [4]. Analysis of the ordering induced by a magnetic field B leads to the conclusion that B^2/T can be taken as the ordering field, at least to a good approximation [7, 8]. As these materials are very sensitive to residual stresses it is a great advantage to be able to apply a non-contacting ordering field.

The classic arguments developed to explain ferromagnetic domains [9], based on a comparison of the reduction in self-energy due to domain formation in a magnetized sample with the energy cost in creating the domain walls, apply also to ferroelectric [10] and ferroelastic [11] transitions. The Bloch argument [9] that leads to thick domain walls in the case of isotropic interactions does not apply to our anisotropic Ising system. Relatively thin domain walls are therefore expected, whose separation, shape, and width will be determined by material elastic properties and by sample size and shape [12]. Even in pure samples the domain walls will be pinned by potentials due to surface geometry and irregularities, and by internal defects such as vacancies and unavoidable impurities.

The samples under investigation are transparent single crystals, allowing the use of birefringence experiments as a sensitive probe of the orthorhombic phase [7, 13]. The birefringence $\Delta n = n_a - n_b$ should be accurately proportional to the orthorhombic strain, but if the scattering volume includes many domains, as is usually the case, the net birefringence will be essentially zero. The basic experiment is therefore the measurement of Δn as a function of ordering field, which reveals the single-domain birefringence [13] and also gives information on how the domains progressively disappear and recover. The main elements of the birefringence apparatus [13] are a He–Ne laser source, with the beam propagating parallel to the c axis, linearly polarized at 45° to the orthorhombic axes, and an analyser with perpendicular orientation in front of the photomultiplier detector. A Soleil–Babinet compensator maintains a null output as detected with high sensitivity with the use of photoelastic modulation and lock-in detection. The change in birefringence is then determined from the displacement of the compensator, which is driven with a stepping motor under computer control. Flux-grown $Dy(As_xV_{1-x})O_4$ crystals with a range of compositions [14] were obtained from the Clarendon Laboratory, Oxford. Crystals with $x = 0$ and $x = 0.15$ were cut and polished with plane faces perpendicular to the c axis separated by about 1 mm, and mounted in a He flow cryostat with a axis vertical and c axis horizontal. To minimize stresses associated with the mounting of the sample we wrapped the sample in aluminium foil, with holes on the c axis faces to transmit the light, and attached the foil to the cryostat sample holder with grease at points well away from the sample.

The output of the experiments consists of the phase angle $\varphi = (n_a - n_b)2\pi l/\lambda$ between the wave components E_a, E_b in passing through the sample as a function of ordering field. Here l is the sample thickness and λ the vacuum optical wavelength. The field was increased in steps from zero until the sample birefringence saturated, usually in the range 0.1–0.2 T, then returned to zero. The usual measurement of hysteresis continues with inducing the opposite orientation until saturation, then returning to zero. However, in this case that would require a rotation of the applied magnetic field by 90°, which is not possible with the existing set-up. Application of the required magnetic fields and the operation of the compensator impose a rather slow time scale on our experiments. We can

detect time dependences in the range 1–100 s by observing the response to small changes in the ordering field, and will present some results, but they are not very quantitative.

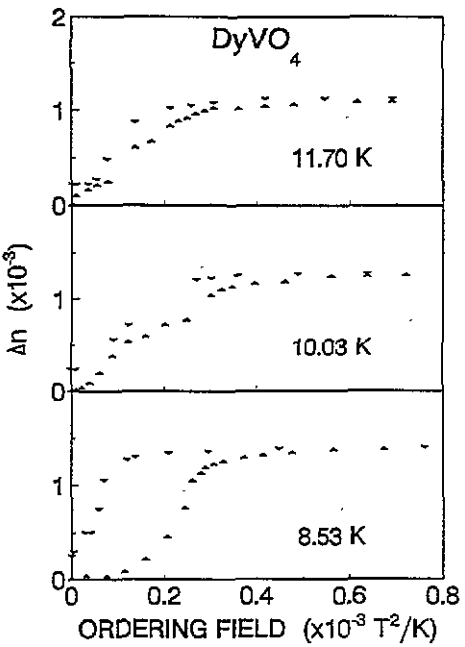

Figure 1. Induced birefringence for increasing and decreasing ordering fields in the twinned orthorhombic phase in pure $DyVO_4$.

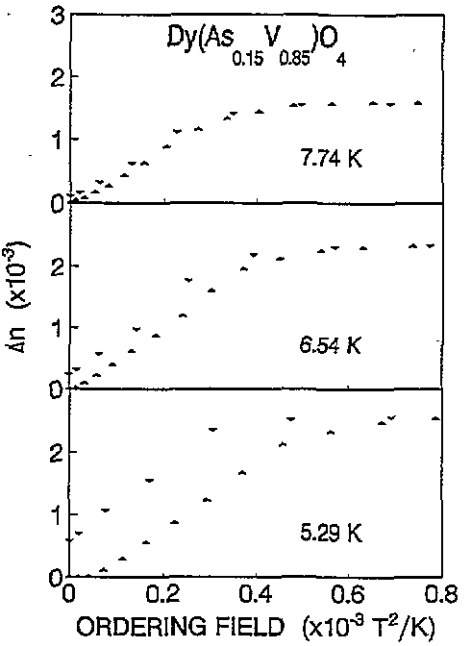

Figure 2. Induced birefringence for increasing and decreasing ordering fields in the twinned orthorhombic phase in $Dy(As_{0.15}V_{0.85})O_4$.

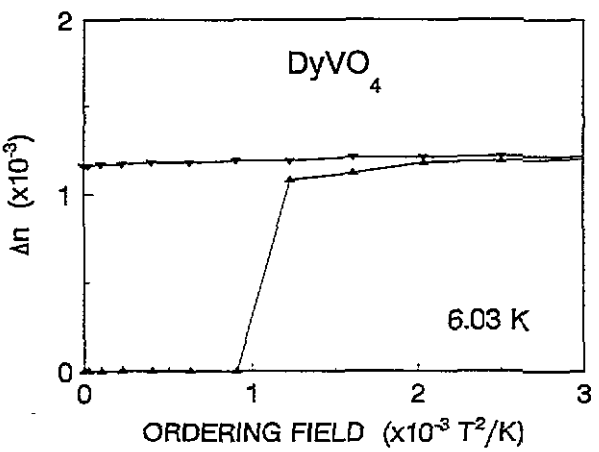

Figure 3. Data as in figure 1, at a lower temperature. Note change in scale of the horizontal axis. Solid lines connect the data points.

Measurements of induced ordering as described were carried out for fixed temperatures close to and below the transition temperature T_D. Below the critical regime, for which results have already been reported for the susceptibility and isotherm critical exponents [13], the

response is dominated by domain alignment effects. The results of runs at three temperatures are shown graphically in figures 1 and 2 in the form of Δn as a function of the ordering field B^2/T. Data for increasing and decreasing fields are indicated by triangles pointing up and down, respectively. At lower temperatures the hysteresis becomes much more pronounced and the sample ultimately appears to switch abruptly from the initial multidomain state to a single domain. We did not make many measurements in this regime but figure 3 shows an example of strongly hysteretic behaviour in $DyVO_4$. Figures 1–3 resemble the familiar hysteresis loops seen in many materials [9, 10]. To describe our results, we fitted straight lines to the quasi-linear region of data for increasing and decreasing fields, and extracted values for parameters F_c, S, and R, as given in table 1. The depinning field F_c is the intercept on the field axis of the line fitting the data for increasing fields, the slope S is the average of the slopes for increasing and decreasing data, and the parameter R is the separation between the intercepts of the two fitted lines on the birefringence axis. R and F_c can evidently serve as rough indications of the area and the coercive field of the full hysteresis loop for each sample, at least for well-behaved cases.

Table 1. Hysteresis parameters obtained from fits of figures 1–3.

Sample	T (K)	T/T_D	S (K T^{-2})	R (10^{-3})	F_c (10^{-3} T^2 K^{-1})
$DyVO_4$	11.70	0.84	4.1	0.14	0
	10.03	0.72	3.5	0.20	0.02
	8.53	0.61	8.4	1.1	0.14
	6.03	0.43	—	—	1.0
$Dy(As_{0.15}V_{0.85})O_4$	7.74	0.93	4.6	0.17	0.021
	6.54	0.78	5.3	0.36	0.024
	5.29	0.64	5.5	0.91	0.059

The slope S of the linear portion of the hysteresis plots is associated with the widening of domains that are favoured by the field and the narrowing of those that are not, in close analogy with ferromagnets. In the simplest model [9], the wall displacement, induced by the ordering field is balanced by a linear restoring force, and in steady-state conditions, the wall displacement and hence the birefringence is proportional to the applied field [9]. Defects that pin the walls will cause non-linearities by preventing any displacement for small fields, and by inhibiting motion even for large fields. If pinning is not large, the slopes of the quasi-linear response should be similar for the two samples and expressible in terms of magnetic and elastic coupling parameters. The data for the 0.15 sample show that S is fairly constant with temperature (we have data at two other temperatures, 5.6 K and 7.1 K, where $S = 5.2$), supporting the validity of the expression B^2/T for the ordering field used in the hysteresis plots. Values of S for the pure sample are similar in magnitude, but are less accurate because of noisier data. There is a tendency for S to increase at the lowest temperatures, especially in $DyVO_4$ (see figure 3) and this is understandable since domains walls become more strongly pinned and then move abruptly.

The most striking result from these experiments is that the depinning field F_c in the mixed sample is much smaller than that for the pure sample at low temperatures. Close to T_D, F_c is larger for the mixed sample; this is difficult to see from the figures because of the poor quality of data for the pure sample but is consistent with other experiments. For example, recent optical speckle diffraction experiments [15] on the same samples show

that the domain walls in the mixed sample are pinned very close to T_D but this is not the case for pure $DyVO_4$. In both samples F_c increases as the temperature is reduced, but this increase is very rapid in the pure sample and relatively slow in the mixed sample. A comparison of figures 1 and 3 implies that in $DyVO_4$ the depinning mechanism changes from one that proceeds in small steps to one that occurs in one large step, while in the mixed sample depinning proceeds smoothly down to low temperatures. Although we have no birefringence measurements on the $x = 0.15$ sample below 5.3 K, other experiments such as optical diffraction [15] and neutron scattering [6] indicate that near 4.7 K F_c is still relatively small (roughly 0.2×10^{-3}) and depinning proceeds smoothly.

As expected, we find that R also increases at lower temperatures, but the differences between the two samples are minor. In considering the behaviour of both F_c and R, it is important to recognize that all samples, whether nominally pure or mixed, will contain strong-pinning centres such as vacancies, interstitials, and surface irregularities. The mixed crystals will also have a high density of pinning centres, presumably of weaker strength, arising from the random strain fields around substitutional atoms. We suggest that the approach to and return from a single domain is controlled by the deeper traps, and these have a similar density in pure and mixed crystals, giving comparable values of R. On the other hand the depinning field F_c should depend on the overall density of pinning centres. It should therefore be smaller for pure $DyVO_4$, as is observed near T_D. At low temperatures F_c will rapidly become large as long-range order is well established and thermal fluctuations are small compared with most of the pinning energies. The effect of impurities in reducing F_c relative to the pure sample must involve a new mechanism (see below).

In general, the response of all samples to an applied field was rapid (< 1 s) at temperatures above $T^* \simeq 0.9$. At lower temperatures where hysteresis is evident, it was still difficult to detect any time dependence: either there was no detectable response to a field, or the response was completed within a few seconds. What this implies is that the response of the sample switches quickly from unobservably slow to quite rapid. To study this in more detail, we used a chart recorder to show changes in the birefringence as a function of time, $\Delta n(t)$, while small ordering fields were applied, for the mixed sample at 6.5 and 5.3 K. For fields much less than F_c no response was detectable at the level of the noise present ($\Delta n \simeq 5 \times 10^{-6}$). Weak responses observed near F_c reached equilibrium at about 50 s. At larger fields, the response showed two distinct components: a rapid response, followed by a slow response with a 50 s time scale. The fast component became more important at higher fields and eventually dominated. This behaviour resembles that seen in other disordered materials where a characteristic field separates regimes of equilibrium and non-equilibrium response [16]. The time scales of the observed responses were essentially the same at 6.5 K and 5.3 K; that is, the main effect of reducing the temperature is to increase the field F_c at which a response is observable. We were not able to obtain useful data of this nature for the pure sample for comparison, because data tended to be noisier and less reproducible.

The central result of this study, the effect of random fields in reducing the depinning threshold, is supported by data on other mixed samples that we have examined. Birefringence measurements on a sample with $x = 0.05$ gave values of F_c at 6.0 K that were also much lower than in pure $DyVO_4$, but we have not presented them because it appears that the sample was poorly aligned. Other types of measurement [6, 15] that can give estimates of F_c are all consistent with the same behaviour. Many mechanisms for domain pinning and coercivity are analysed in the literature, but a general finding is that pinning increases with disorder [17]. It is interesting that an analysis of depinning for the random-field Ising model by Nattermann [18] predicts that the depinning field is an exponentially small function of random-field strength for the specific case of three dimensions and thin

domain walls, conditions that should be satisfied in our system. This ineffectiveness may permit other mechanisms that assist domain wall motion at higher impurity concentrations to come into play.

For some ferroelectrics, it has been found that the addition of certain impurities reduces the coercive fields [19, 20], but these are complex situations where the substitution of ions of different valence creates vacancies that may relieve stresses associated with, for example, the presence of both 90° and 180° domains, or the proximity of a phase boundary.

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