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LETTER TO THE EDITOR

Bragg-regime diffraction of light by twinned orthorhombic domains

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Abstract. Optical Bragg-regime diffraction of light from the domain structure of a twinned orthorhombic crystal is reported for the first time. Diffracted beams were observed in $DyAsO_4$, $DyVO_4$, $TbVO_4$, and some mixed crystals below their orthorhombic phase transitions. The diffracted beams could be seen over a range of scattering angles, with peak intensity near 5° corresponding to an average domain spacing of about 3 μm. The temperature-dependent intensity was measured to obtain values for the order-parameter critical exponent.

Many crystalline compounds normally grow as twinned crystals, or adopt a twinned configuration when they undergo a symmetry-lowering phase transition. In a twinned crystal the unit cells in different regions, or domains, are identical in structure but differ in orientation in a regular manner related to the structure. Twinned domains have been observed by a variety of techniques including optical and electron microscopy, and x-ray and neutron diffraction. In this letter we describe the observation of optical Bragg-regime diffraction from domains in a simple orthorhombic twinned system, and show that it can provide information not easily obtained by other techniques. Our observations are similar in some ways to those reported by Kleeman and Ferré (1981) for optical Bragg diffraction from stripe domains in the transparent ferromagnet K_2CuF_4. The deflection of light by ferroelectric domain walls has been reported previously (Tsukamoto *et al* 1982), but there was no evidence for Bragg diffraction.

Our experiments have been carried out on several rare-earth vanadate and arsenate compounds that undergo tetragonal–orthorhombic phase transitions at low temperatures. Their properties have been studied in detail previously because they provide realizations of simple pseudospin Ising model structural phase transitions with interesting critical behaviour and dependence on the range of interaction (Gehring and Gehring 1975, Page *et al* 1984). These compounds are transparent and well suited to optical studies.

Below the transition temperature T_D a multidomain phase results from the two equivalent orthorhombic distortions of the parent tetragonal (D_{4h}) phase. The elastic energy associated with the interface between two domains is a minimum for a plane boundary containing the c axis and at 45° to the orthorhombic a and b axes. These domain walls tend to be regularly spaced through a large portion of the sample to

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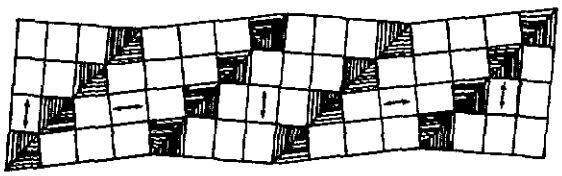

Figure 1. Schematic illustration of twinned orthorhombic domains separated by (110) planes. The crystal c axis is perpendicular to the plane of the figure. The arrows indicate the direction of the long orthorhombic axis in each domain.

minimize bulk sample strains. This twinning configuration is illustrated in figure 1, and is similar to that described by Jona and Shirane (1962) for some ferroelectric crystals.

Direct miroscopic observation of the domains in $DyVO_4$ was reported by Gehring and Rosenberg (1971) and in $TbAsO_4$ by Klein *et al* (1972). From their photographs the domains appear to be equally spaced, with a typical wall separation of several μm. This suggests the possibility of observing Bragg diffraction of visible light (wavelength $\simeq$ 0.5 μm) in this system if the domain geometry is sufficiently regular.

The samples were good quality single crystals grown by flux methods (Wanklyn and Watts 1984) at the Clarendon Laboratory, Oxford. The crystals were cut perpendicular to the crystal c axis with thickness $\sim$1 mm, polished on the basal plane faces, and checked for optical quality and alignment by observing the conoscopic figures with a polarizing microscope. For the experiments each crystal was first attached to a thin stainless steel foil with a pinhole of diameter about 0.8 mm, and then mounted on a sample holder with c axis horizontal in an optical helium flow cryostat. The sample could be rotated through 360° about the vertical axis. A well collimated uniform HeNe laser (633 nm) beam was transmitted through the pinhole close to the crystal c axis. A diffracted beam could be viewed directly on a screen or detected with a photomultiplier. The optical alignment, and the measurement of angular position were carried out by observing the back reflection from the crystal surface.

In all the crystals examined, a diffracted beam was easily seen in the orthorhombic phase for small incident angles θ. It disappeared above T_D, and its intensity increased at low temperatures to as much as 10% of the incident beam. The most convincing evidence that the diffracted beam originated from the periodic domain structure is the observation that it was always located in the plane perpendicular to the domain walls containing the incident beam. In $DyVO_4$ and $DyAsO_4$ the orthorhombic distortion axes are parallel to the a axes of the tetragonal unit cell and the domain walls are {110} planes. On the other hand for $TbVO_4$ the orthorhombic distortion has B_{2g} rather than B_{1g} symmetry (Gehring and Gehring 1975). Its orthorhombic principal axes are therefore rotated by 45° relative to those of $DyVO_4$ and $DyAsO_4$ and the domain walls are {100} planes. Thus for $DyVO_4$ and $DyAsO_4$ the planes containing the diffracted beams should be 45° to the tetragonal a axes while for $TbVO_4$ they should contain the a axes. This is precisely what was observed.

For an ideal periodic structure, Bragg diffraction occurs at a unique angle $\theta = \sin^{-1}(\lambda/2d)$ where λ is the wavelength and d the separation between scattering planes. Instead we observe a diffracted beam for a range of θ values, in some samples from almost zero to 15° or more. However, in all cases the scattered beam intensity showed a clear maximum at some angle $\theta = \theta_B$ that we associate with Bragg diffraction corresponding to an average plane spacing d_B. Representative values for θ_B and d_B at temperatures well below T_D are as follows. For $DyAsO_4$: $\theta_B = 8.0°$; $d_B = 2.3$ μm (T_D = 10.9 K). For $DyVO_4$: $\theta_B = 7.2°$; $d_B = 2.5$ μm (T_D = 13.5 K). For $TbVO_4$: $\theta_B = 4.4°$; $d_B = 4.1$ μm (T_D = 32.4 K). The general similarity of these d_B values in spite of the

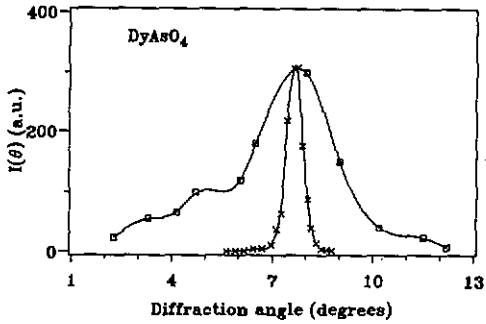

Figure 2. Intensity of the diffracted beam in $DyAsO_4$ at 7.4 K as a function of angle. The outer data (□) are for equal incident and diffraction angles θ. The inner data (×) are for a fixed incident beam angle. The solid curves are guides to the eye.

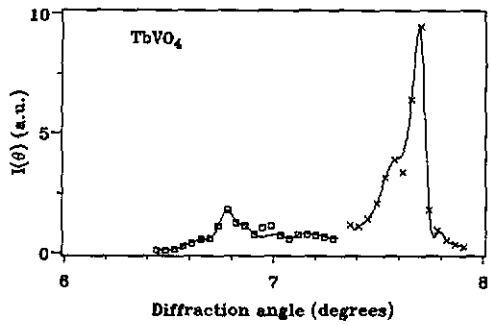

Figure 3. Diffracted beam intensity for $TbVO_4$ at 31.2 K with fixed incident beam angle and incident beam polarization (□) within the scattering plane and (×) perpendicular to it. The solid curves are guides to the eye.

differences in the range and strength of the interactions driving the phase transition (Gehring and Gehring 1975) implies that domain dimensions are insensitive to the rare-earth ion properties but depend on other factors such as elastic properties.

An example of the observed broad distributions in θ, $I(\theta)$, is given in figure 2 for $DyAsO_4$. So far the explanation for these distributions is not certain. Possible explanations include: (i) the domain wall separations d may be distributed randomly about mean value; (ii) d may be constant locally but may vary on a macroscopic scale due to sample strains and boundary effects; (iii) roughening of the domain walls and corresponding misalignment of domains may give a type of mosaic broadening (Graham *et al* 1991). Of these, mechanism (i) is the most likely in view of the expected magnitudes. A similar conclusion was reached in the case of the distributions observed in optical Bragg diffraction from ferromagnetic domains (Nitsche and Kleemann 1982a). The diffracted beam for arbitrary θ was moreover observed to be broadened relative to time incident beam as indicated in figure 2. Thus a detailed analysis of diffraction from twin domains should provide rather direct information on the distribution of domain sizes, and may also reveal something about the widths and texture of the walls.

In most cases two separate diffraction peaks of different intensity could be resolved when the incident laser beam was unpolarized. They were most clearly seen in $TbVO_4$ (figure 3), and were found to be plane polarized with orthogonal polarization directions and with an angular splitting independent of temperature. We identify them as the two rays arising from the birefringence n_c–n_a, which emerge at different angles as a result of reflections from the domain walls. We have not carried out a detailed analysis, but the observed angular separation $\leqslant 1°$ is consistent with the expected splitting of the ordinary and extraordinary rays (Gay 1967) for incident beam and angles near θ_B.

An advantage of the present system is that the multidomain structure can easily be forced into a single domain by application of an ordering (magnetic) field parallel to a basal plane axis (Gehring and Gehring 1975). We examined the effect of the ordering field as well as temperature on the domain configuration as portrayed by the distribution $I(\theta)$. In general the width of $I(\theta)$ is almost independent of temperature and ordering field: evidently the domain configuration is established near T_D and changes very little

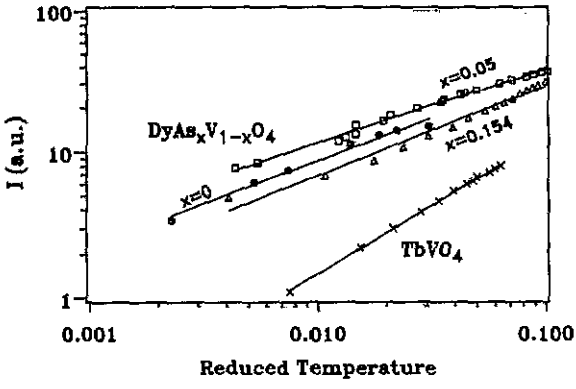

Figure 4. Log–log plot of temperature-dependent diffracted beam intensity versus reduced temperature $(1 - T/T_D)$ for pure and mixed samples as indicated. The slopes give the order-parameter critical exponent β.

with order parameter. The intensity of the peak initially increases slightly with application of an ordering field. Beyond a certain field the intensity decreases as domains start to flip and disappears when the sample becomes a single domain.

It is important to establish whether the diffracted beams originate in volume (i.e. Bragg regime) diffraction or in plane diffraction. The former is suggested by the absence of higher-order diffracted beams. Previous work (see Moharam *et al* 1980) has shown that a parameter ρ may be used to distinguish between 'thick' and 'thin' diffraction gratings. The Bragg-regime diffraction limit was found to be characterized by $\rho \geqslant 10$, where $\rho = \lambda^2/\Lambda^2 n_0 n_1$. Here Λ is the grating period ($=d$ in our case), n_0 is the sample refractive index, and n_1 the amplitude of the periodic variation of the refractive index. Substituting approximate values $n_0 = 2$, $n_1 = 2 \times 10^{-3}$ (Kasten 1980), and $\Lambda = 2.5\ \mu$m, we obtain $\rho \approx 16$, well within the Bragg diffraction regime.

This optical diffraction technique also provides a very convenient method for accurate measurement of the order parameter. The strength of the scattering at the interfaces depends on the degree of ordering, and previous arguments (Nitsche and Kleeman 1982b) have shown that the temperature dependent intensity is proportional to the square of the order parameter. We have collected data for the temperature dependence of the intensity of the Bragg peak to determine both T_D and the order parameter exponent β through the relation $I(\theta_B, T) \propto (1 - T/T_D)^{2\beta}$. Figure 4 shows a log–log plot of Bragg-peak intensity as a function of temperature for $DyVO_4$, $TbVO_4$, and also for some mixed $Dy(As_xV_{1-x})O_4$ crystals. For the pure compounds $DyVO_4$ and $TbVO_4$ our results for β are 0.31 ± 0.03 and 0.47 ± 0.03 in good agreement with previous results (Harley and MacFarlane 1975). For the mixed compounds, surprisingly, figure 4 shows that β appears to be unaffected by the random strain fields introduced by As/V substitutions (Taylor and Reza 1991).

It is generally appropriate to describe Bragg diffraction in terms of the reciprocal lattice. In this case, where the repeat distance perpendicular to the domain walls d is orders of magnitude greater than the tetragonal unit cell, the reciprocal lattice takes a very simple form. For optical waves where the momentum h/λ is very small, the only accessible points of the reciprocal lattice are those separated by $2\pi/d$ along the normal to the domain walls. Thus only one Bragg peak is seen for a given orientation of twins, but samples are usually multiply twinned with domain walls rotated by 90° (Gehring and Rosenberg 1971). For incident light bisecting the angle between domain walls, and Bragg condition may be satisfied for both sets of domains and two peaks may be seen.

An unexplained feature of our observations is that we only detect Bragg diffraction when the incident beam is close to the crystal *c* axis. Attempts to observe the same peaks with the incident beam near one of the *a* axes were unsuccessful even though the Bragg condition is the same.

We anticipate that future work with an improved detection system will provide important information on domain wall properties in these and similar twinned systems.

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References

Gay P 1967 *An Introduction to Crystal Optics* (London: Longmans & Green)
Gehring G A and Gehring K A 1975 *Rep. Prog. Phys.* **38** 1
Gehring K A and Rosenberg H M 1971 *Phys. Status Solidi* b **47** K75
Graham J T, Taylor D R, Noakes D R and Buyers W J L 1991 *Phys. Rev.* B **43** 3778
Harley R T and MacFarlane R M 1975 *J. Phys. C: Solid State Phys.* **6** L451
Jona F and Shirane G 1962 *Ferroelectric Crystals* (London: Pergamon)
Kasten A 1980 *Z. Phys.* B **38** 65
Kleemann W and Ferré J 1981 *Phys. Rev.* B **24** 1568
Klein L, Kahle H G, Schopper H C and Walter H 1972 *Int. J. Magnet.* **3** 17
Moharam M G, Gaylord T K and Magnusson R 1980 *Opt. Commun.* **32** 14
Nitsche W and Kleemann W 1982a *J. Magn. Magn. Mater.* **28** 163
—— 1982b *Opt. Commun.* **41** 1
Page J H, Taylor D R and Smith S R P 1984 *J. Phys. C: Solid State Phys.* **17** 51
Taylor D R and Reza K A 1991 *ICM91 (Edinburgh)* submitted
Tsukamoto T, Hatano J and Futama H 1982 *J. Phys. Soc. Japan* **51** 3948
Wanklyn B M and Watts B E 1984 *Mater. Res. Bull.* **19** 825