

spin flop. The change is gradual over roughly 2 kG, and close examination shows that different sections of the crystal flop before others; this is consistent with the domains expected from the demagnetization effect. The width of the transition is in reasonable agreement with a demagnetization correction of 2.9 kG for this particular sample. Experiments on samples with large demagnetization factors gave consistently broader transitions. It is clearly difficult to determine the beginning or the end point of a transition because of the fact that it might start in an area of the crystal not sampled in the photograph. Instead, the centre of the transition was estimated directly from the photograph by eye. This admittedly subjective procedure nevertheless gave results which were quite reproducible and accurate to within ± 0.5 kG, which was sufficient in view of the stress and demagnetization nonuniformities in the crystal. Values for the critical field H_c obtained by increasing the field through the transition were consistently larger than those obtained by decreasing the field. This effect was attributed to a hysteresis which arises because of the energy barrier between the metastable and stable potential minima of the system, as can be derived from the theory given in I. Experimental results on several different crystals at 4.2 and 1.4 K are plotted in Figs. 10 and 11, with a line connecting the two points which measure the hysteresis.

4. DISCUSSION

Symmetry considerations

Theoretical equations have been derived in I to account for the effects of stress and magnetic field on DyVO_4 . To apply this theory to the experimental results of the last section, it is necessary to relate σ , e and c of the theory to the true stresses, strains and elastic constants of the crystal. There are two conventions in the literature for describing these quantities in a tetragonal crystal, for there are two possible definitions of the tetragonal axes at 45° to each other in the basal plane [14]. We

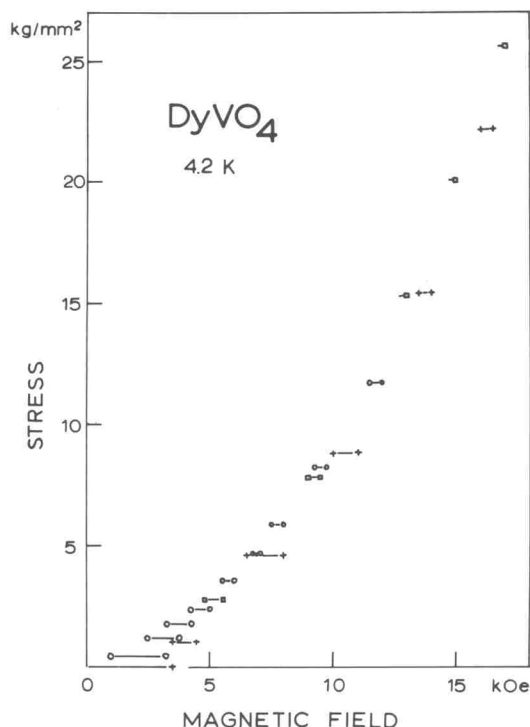


Fig. 10. Pseudospin flop fields in DyVO_4 at 4.2 K for various stresses and crystals with demagnetization factors of 3.14 (\circ), 3.72 (\square) and 4.15 ($+$). Lines connecting two points represent hysteresis between increasing and decreasing fields, as explained in the text.

adopt a set of axes abc which are parallel to the natural growth faces of DyVO_4 , and it has been shown that these axes coincide with those of the X-ray crystallographic unit cell. Taking the six elastic constants of a tetragonal crystal to be defined in this way, we may write the elastic energy of DyVO_4 as

$$\begin{aligned}
 U = & \frac{1}{4}(c_{11} + c_{12})(e_{aa} + e_{bb})^2 + c_{13}(e_{aa} + e_{bb})e_{cc} \\
 & + \frac{1}{2}c_{33}e_{cc}^2 + \frac{1}{2}c_{66}e_{ab}^2 + \frac{1}{4}(c_{11} - c_{12})(e_{aa} - e_{bb})^2 \\
 & + \frac{1}{4}c_{44}(e_{ac}^2 + e_{bc}^2) \quad (4.1)
 \end{aligned}$$

where the e 's are the strain components as defined, for example, by Kittel [15]. Each of these terms characterize distortions of a different symmetry type in the point group D_{4h} of the crystal; the first three are of type Γ_1^+ ; the others are Γ_3^+ , Γ_4^+ , and Γ_5^+ respec-

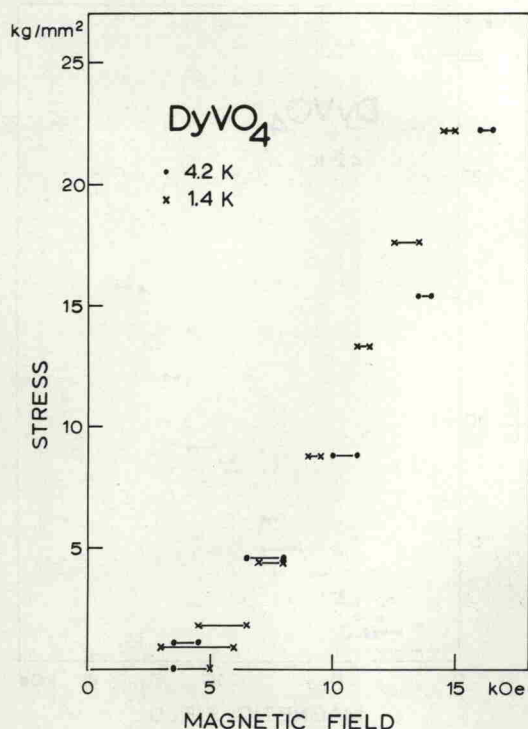


Fig. 11. Pseudospin flop fields in DyVO_4 at 4.2 and 1.4 K for a crystal with a demagnetization factor of $D = 4.15$. Lines connecting two points represent hysteresis as in Fig. 10.

tively (Koster *et al.*[10]; their xyz axes are at 45° in the basal plane to our abc axes).

The Jahn-Teller distortion in DyVO_4 is known to be of $(e_{aa} - e_{bb})$ or Γ_4^+ type, such that the reflection planes of the rare earth site group are preserved[5]. By contrast the TbVO_4 distortion is of an e_{ab} or Γ_3^+ type, such that the two-fold rotations in the basal plane are preserved. From the above equation it is clear that such strains do not couple to strains of any other symmetry, except in the presence of anharmonic forces. Assuming that e of the theory is defined precisely by $(e_{aa} - e_{bb})$ for DyVO_4 , the elastic constant c is $(c_{11} - c_{12})/2$.

The stress-strain equations of a tetragonal crystal show that $(e_{aa} - e_{bb})$ -type distortion can also be induced by uniaxial stress applied along a , or equivalently along b , that is, along the natural tetragonal crystal faces of DyVO_4 .

Such a stress σ_{aa} can be written $\frac{1}{2}(\sigma_{aa} - \sigma_{bb}) + \frac{1}{2}(\sigma_{aa} + \sigma_{bb})$, so that only one half of the measured stress is of a Γ_4^+ type. Therefore, in fitting to the theoretical equations of the previous theory, the experimental stress values must be divided by a factor of 2. The other half of the applied stress induces distortions of a Γ_1^+ type, which include both $(e_{aa} + e_{bb})$ and e_{cc} components because of the coupling terms in (4.1). In the absence of anharmonic forces, such strains will not modify the Γ_4^+ couplings at all, and therefore the theoretical equations derived earlier remain valid. Nevertheless these strains, which are of course linear in the applied stress and which have on-diagonal matrix elements, can also induce linear shifts in the energy levels. We interpret in this way the observed linear shift in the centre of gravity of the two absorption lines which represent transitions from the two ground levels to any given excited state. The fact that all the observed shifts are roughly the same suggests that the dominant shift comes from the ground rather than the excited states.

Molecular field theory

The basic equation relating the applied stress σ to the observed optical splitting Δ is (I-3-15):

$$\frac{\sigma}{\sigma_0} = \sqrt{\frac{\Delta^2 - \Delta_\infty^2}{\Delta^2}} \left(\frac{\Delta}{\Delta_0} - \tanh \frac{\Delta}{2kT} \right) \quad (4.2)$$

Δ_∞ , as given in Table 1, is determined directly from the optical spectrum above T_D , and Δ_0 has been determined by far-infrared and Raman spectroscopy to be $\sim 27 \text{ cm}^{-1}$ [6, 13]. Using this data, (4.2) predicts a co-operative ordering at $T_D = 18.9 \text{ K}$, whereas the observed value is only $14.00 \pm 0.05 \text{ K}$ [16]. This fact, suggests that there are large deviations from molecular field behaviour such as might be expected for a true Ising system with a small number of effective nearest neighbours[17]. Although molecular field theory accounts poorly for the critical behaviour in the vicinity