MAGNETO-OPTICAL EFFECTS AND SHORT-RANGE ORDER IN AN ANTIFERROMAGNET

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Previous experiments on dysprosium aluminum garnet (DyAlG) have shown that it undergoes a transition to an ordered state at 2.49°K.[1] The ground-state spin configuration, deduced first by Ball et al.,[2] has recently been confirmed by neutron diffraction experiments,[3] which also show that the variation in sublattice magnetization with temperature has the usual form.

The present work consists of optical-absorption experiments on a single crystal of DyAlG in which the Dy^{3+} absorption lines have been studied as a function of temperature and magnetic field in the ranges 1.3-20.4°K and 0-23 kOe, respectively. Many of the lines are broad, but some are sufficiently narrow to display effects due to magnetic ordering at suitable temperatures. That these effects are magnetic in origin is clear from their behavior in a magnetic field. Details of the field experiments are not given here, but let it suffice to say that one obtains critical-field effects analogous to the observed magnetic-moment behavior.[4]

The spectrum at the lowest temperature (1.3°K) clearly shows two important features. Firstly, a few lines are split into doublets, the center of each doublet being shifted to the blue relative to the unsplit line at 20.4°K. Secondly, the unsplit lines are also shifted to the blue (such a shift has previously been reported by Hellwege et al.[5] The interpretation of these effects is straightforward. The excited states and the ground state of the ion interact with the internal field, and the Kramers' degeneracy is lifted. At 1.3°K only the lower component of the ground doublet is populated. If transitions to both components of the excited doublet are allowed, then a doublet absorption line which has been shifted to the blue will result. If transitions to only one component of the excited doublet are allowed, the absorption line will be unsplit and merely shifted, the shift depending upon the relative splittings of the ground and excited doublets. We have always observed shifts to higher energy, due, no doubt, to the dominance of the large ground-state g value (18.2). In Table I are listed the splittings Δ (where observed), and the shifts δ (between 20.4 and 1.3°K) of some of the lines of the DyAlG spectrum.

Of particular interest is the variation of shift δ with temperature. This is most easily measured on unsplit lines, and it is found that these all show the same form of variation. (The split lines broaden and overlap as the temperature is raised, making the measurement of either Δ or δ versus T difficult.) Figure 1 shows measurements made on a typical unsplit line compared with the integrated heat capacity and the neutron diffraction intensity. In Fig. 2 is shown the temperature dependence of two lines near 4032 Å which do split below T_N. These figures show that both the shifts and the splittings persist above the Néel temperature. Therefore, the internal field giving rise to these effects does not vary with temperature in the same manner as the sublattice magnetization.

An explanation of these results is that the shifts and splittings are attributable to the ef-

Table I. The splitting Δ (at 1.3°K) and the blue shifts δ (at 1.3°K, relative to the position at 20.4°K) of some of the absorption lines in dysprosium aluminum garnet. The effect of lattice expansion (between 4.2 and 20.4°K) on the position of the lines was checked by measuring shifts of lines in the spectrum of 1% Dy^{3+} in yttrium aluminum garnet. The observed shifts were at least 10 times smaller than the figures given above.

Wavelength (Å)	Splitting Δ (cm^{-1})	Blue shift δ (cm^{-1})
3022	2.2 ± 0.2	2.8 ± 0.3
3024		1.8 ± 0.3
3218	3.6 ± 0.3	2.7 ± 0.3
3382		3.0 ± 0.3
3385		2.3 ± 0.3
3515	3.4 ± 0.2	2.75 ± 0.3
3649		2.4 ± 0.2
4031	3.95 ± 0.1	2.65 ± 0.2
4033	1.85 ± 0.15	2.6 ± 0.2
4270		1.0 ± 0.1
4280		0.8 ± 0.1
4743		2.3 ± 0.3
4788	0.8 ± 0.1	2.7 ± 0.3

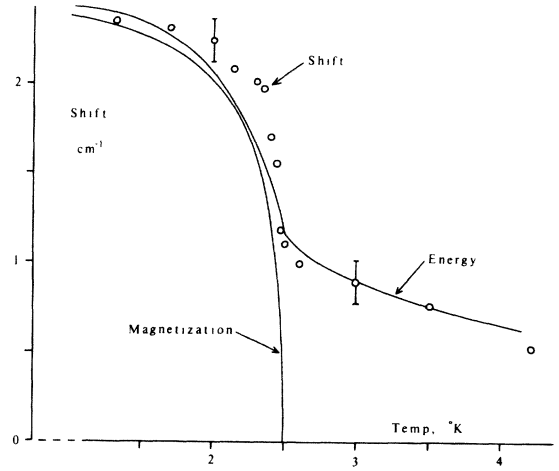

FIG. 1. The temperature dependence of the shift δ of a typical unsplit absorption line (at 3649 Å) is compared with the normalized magnetic energy (from specific heat data[1]) and the normalized sublattice magnetization (from neutron experiments[3]).

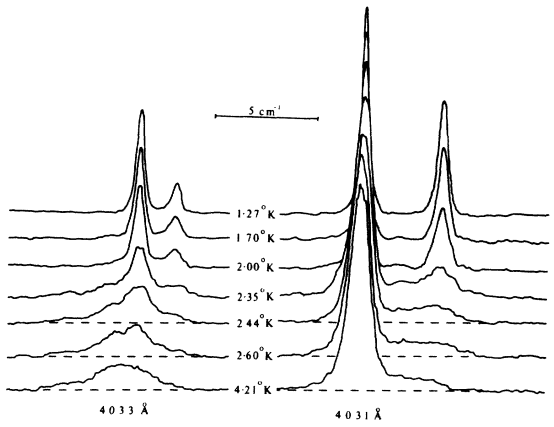

FIG. 2. The splittings Δ of two of the absorption lines in DyAlG as a function of temperature. (The shifts are not accurately represented in this figure.) It is seen that the splittings persist above the Néel temperature (2.5°K), and that they vary in the same way as the magnetic energy (see text).

fects of short-range order rather than long-range order, and that this kind of experiment measures a different kind of average, both in space and in time, from that measured in a neutron experiment.

In order to arrive at a quantitative understanding of this system, a Monte-Carlo method[6] has been used to compute the magnetic energy and sublattice magnetization as a function of temperature. If dipolar interactions alone are present (with $g_{\parallel}=18.2$, $g_{\perp}=0$, and $a_0=12.0$ Å the internal field H_d is 4.65 kOe at 0°K), this method shows that the Néel temperature would be 1.63°K. In order to obtain agreement with the experimental value of T_N, a nearest-neighbor exchange interaction was introduced into the calculation. The required exchange interaction is found to be 52% of the nearest neighbor dipolar interaction and is assumed to have the same form.[7] The internal field H_{d+e} is then increased to 6.42 kOe, and the ground-state splitting to 5.46 cm^{-1}. From Table I it is seen that the average of twice the mean shift of the split lines is 5.4 cm^{-1}, in excellent agreement with the predictions of our Monte-Carlo calculations. The computed variation of energy with temperature has relevance here, since a direct check can be made with the integrated experimental heat capacity. Agreement is very good, and since 81% of the energy at 0°K is due to nearest-neighbor interactions, the energy can be interpreted as a measure of the short-range order. This strengthens our conclusion that optical experiments on this substance are observations of the effect of short-range order. Full details of the Monte-Carlo calculation will be published later.

Further, a spin-wave analysis of the low-temperature specific heat,[1] assuming an Ising model, gives a ground-state splitting of 4.9 cm^{-1}. The introduction of some isotropic exchange would increase this number, so here again there is good agreement with both the optical experiments and the Monte-Carlo calculation.

There is one other feature of this system that calls for comment. It is seen from Table I that the shifts δ of the split lines are constant (within experimental error) from line to line. This result is somewhat surprising when one considers that the local single-ion excitation is not a proper description of the system. A dipolar (or exchange) Hamiltonian can transfer the excitation from one ion to another, giving a change in energy which depends on the matrix elements of the Hamiltonian between the ground and excited states, and hence on the detailed nature of the excited state. For the lines reported here this effect is apparently small. This model suggests that, if one were able to observe similar splittings for impurity rare-earth ions in DyAlG, effects arising from excitation transfer degeneracy will certainly

not occur. Experiments of this type using DyAlG doped with Er^{3+} are being carried out.

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MAGNETIC BIREFRINGENCE OF EuSe

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Magnetically ordered EuSe exhibits very large magneto-optical effects in a wavelength region where the material is exceptionally transparent. We have previously reported Faraday rotation and absorption of EuSe in the paramagnetic region.[1] This paper describes measurements of Faraday rotation (magnetic circular birefringence) and Cotton-Mouton phase difference (magnetic linear birefringence) made at 4.2°K where EuSe is ferromagnetic in high magnetic fields and antiferromagnetic in low magnetic fields.

The Curie point measured in an applied field and the Néel point measured in zero field by neutron diffraction are both approximately 7°K.[2] Neutron diffraction also has shown that the zero-field magnetic structure consists of a basic component having the second kind of fcc antiferromagnetic ordering and an additional square-wave-modulated ferromagnetic component. Application of a moderate field (1750 Oe at 1.9°K) aligns the modulated component, causing the appearance of a partially saturated ferromagnetic moment, and a high field (8000 Oe at 1.9°K) eliminates the basic antiferromagnetic component, leaving pure ferromagnetism.

EuSe has the NaCl crystal structure and is semiconducting with a low Hall mobility.[3] A sharp optical absorption edge is observed at about 670 mμ. Some values of room-temperature absorption coefficient (neglecting reflection losses) are 293 cm^{-1} at $\lambda = 660$ mμ, 89 cm^{-1} at $\lambda = 700$ mμ, and 66 cm^{-1} at $\lambda = 750$ mμ. Due to the strong absorption at shorter wavelengths, even very thin bulk crystals could not be measured at wavelengths less than 600 mμ. Absorption measurements made on evaporated thin films of EuSe showed[1] that the absorption edge observed with bulk crystals of 670 mμ is actually the long-wavelength edge of a broad absorption band with a peak at 460 mμ. It has been suggested that this absorption peak at 460 mμ is due to a charge-transfer transition $(4f^7)(4f^7) \rightarrow (4f^6)(4f^7 5d)$ between neighboring Eu^{++} ions.[4]

The magnetic birefringence was measured on a single crystal approximately 2 mm square, with a thickness of 157 μ (±5% error). The sample was directly immersed in liquid helium. The light source is a Bausch and Lomb monochromator with a tungsten lamp and an output of about 60 Å half-width. The remainder of the experimental apparatus includes a 12-in. magnet with axial hole, 150-cps light chopper, Glan-Thomson prisms, several lenses, Soleil compensator, RCA 7102 photomultiplier, and a lock-in detector.

In the Faraday configuration H is applied parallel to the light beam, and the two eigenmodes of propagation in the crystal are right- and left-handed circularly polarized light (for magnetization M parallel to H). The difference in index of refraction for these modes $\Delta n_F = n_r - n_l$ gives rise to Faraday rotation, and the difference in extinction coefficients $\Delta k_F = k_r - k_l$ results in circular dichroism. In the Cot-