Time-Reversed Antiferromagnetic States in Dysprosium Aluminum Garnet

J. F. Dillon, Jr., and E. Yi Chen
Bell Laboratories, Murray Hill, New Jersey 07974

and

N. Giordano and W. P. Wolf
*Becton Center, Yale University, New Haven, Connecticut 06520**
(Received 15 April 1974)

The field dependences of the magneto-optical rotation of the two time-reversed states of antiferromagnetic dysprosium aluminum garnet show significant differences. These are interpreted in terms of the spatial variation in the signs of the spin-spin interactions demanded by the crystal symmetry. The coexistence of the two time-reversed states has been studied by microscopy made possible by the rotation difference and also by a striking "decoration" of the domain walls by paramagnetic nuclei.

Largely because it appeared to approximate closely a simple two-sublattice Ising system, the antiferromagnet dysprosium aluminum garnet (DyAlG), with fields along [111], has been much studied over the past decade.[1] Recently in the course of magneto-optical studies of this crystal, we encountered a number of new and puzzling phenomena. These included (a) an unexpected dependence of the magnetization on past history[2] and (b) related microscope observations of magnetically active domains with curious properties seemingly unrelated to the accepted magnetic phase diagram. These phenomena can now be understood in the light of a recent discussion by Blume *et al.*[3] in which they pointed out that an applied field may induce significant antiferromagnetic order in DyAlG, so that the two time-reversed (TR) antiferromagnetic states become distinguishable in a field. In this paper we shall present magneto-optical observations of the TR states in DyAlG and a detailed interpretation in terms of the specific mechanism described in a separate publication.[4]

Following Blume *et al.*,[3] we show in Fig. 1(a) the revised phase diagram for DyAlG in a field H_i along [111]. Both positive and negative fields are included to emphasize the zero-field phase boundary separating the two TR antiferromagnetic states for $T < T_N$. The two TR states, A^+ and A^-, are characterized by different values of the order parameter $\vec{L}$, which is here the staggered magnetization $\vec{M}_b - \vec{M}_a$, where a and b are the two inequivalent types of sites discussed previously.[3,4] The variation of $\vec{L}$ with H_i for the two TR states at a temperature $T < T_c$ is shown schematically in Fig. 1(b). In zero field time-reversal symmetry demands that $\vec{L}(A^+) = -\vec{L}(A^-)$, but in a nonzero field TR symmetry only requires $\vec{L}(A^+, H) = -\vec{L}(A^-, -H)$. For a given field we can thus have $\vec{L}(A^+, H) \neq -\vec{L}(A^-, H)$. Correspondingly, the total magnetization $\vec{M} = \vec{M}_a + \vec{M}_b$ will show TR symmetry $\vec{M}(A^+, H) = -\vec{M}(A^-, -H)$, but for a given field we can have $\vec{M}(A^+, H) \neq \vec{M}(A^-, H)$ [Fig. 1(c)]. In the presence of a field only one of the two states will be stable but, in common with other first-order transitions, we may expect the possibility of a metastable state as the phase boundary is crossed. The essential feature, first recognized by Blume

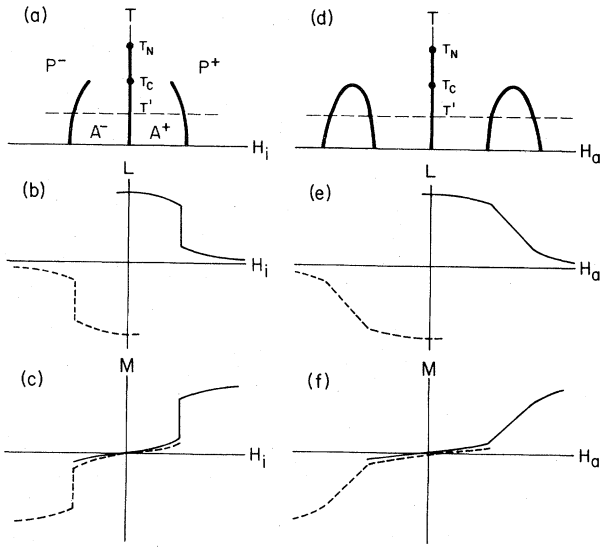

FIG. 1. (a) Revised phase diagram (schematic) for DyAlG (after Ref. 2). For low positive H_i the stable phase is A^+, and A^- is metastable. (b) Variation of the order parameter $\vec{L} = \vec{M}_b - \vec{M}_a$ for $T < T_c$. (c) Corresponding variation of the magnetization $\vec{M} = \vec{M}_a + \vec{M}_b$. (d)–(f) Similar plots in terms of the externally applied field $H_a = H_i + NM$.

et al.,[3] is that DyAlG is unusual in that a field along [111] (and indeed in almost all other directions[5]) will couple to the order parameter, removing the degeneracy and hence the ambiguity between the two states, thus making it possible to follow a given state from one side of the zero-field phase boundary to the other.

DyAlG is not unique in this respect, but in the other systems which have been studied,[6] the field was always effective through an allowed weak ferromagnetic moment, with the result that the two TR states became distinct in both L and M, even in zero field.

Under actual experimental conditions, the situation is complicated by demagnetizing effects and in Figs. 1(d)–1(f) we show the corresponding behavior as a function of the applied field $H_a = H_i + NM$, where N is the demagnetizing factor. The only essential difference is the fact that the two phase boundaries in nonzero H are spread over a region of H_a, within which the antiferromagnetic and paramagnetic phases coexist in varying proportions.[1] This coexistence makes it possible to nucleate a small and controllable amount of one phase in the other and we have utilized this fact in our microscope observations.

In the first experiments measurements were made of the magneto-optical rotation, which is proportional to M under the conditions of our experiments.[2] The data were taken by an automatic technique described earlier.[2] The sample was 690 μm thick and roughly elliptical in shape with major and minor diameters of 16 and 7 mm. The effective demagnetizing factor was close to 4π and all field measurements were recorded as the corresponding internal fields. The results are shown in Fig. 2 as solid lines.

It can be seen that there are two distinct curves at each temperature, depending on the previous field history of the sample. The upper curve in each case was obtained by first exposing the sample to a large positive field, and, provided the field was never allowed to go negative, it was completely reversible. In terms of our Fig. 1 we interpret this as the magnetization of the state A^+. The lower set of curves were obtained by first exposing the sample to a large negative field and then making measurements in positive fields which were not allowed to exceed the critical field for the phase transition to the paramagnetic state. We interpret these curves as the behavior of the A^- state, which is metastable in low positive fields.

The difference in the response of the two TR

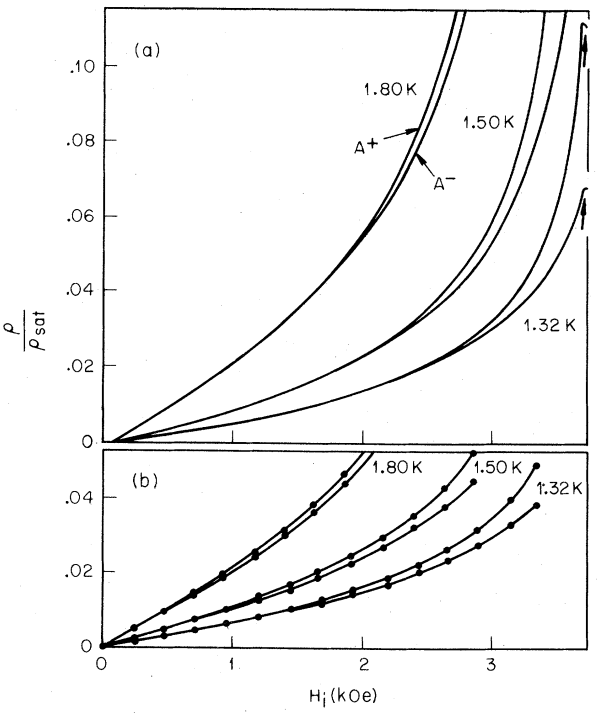

FIG. 2. (a) Automatic data plots of reduced magneto-optical rotation (ρ/ρ_{sat}) of two antiferromagnetic states of DyAlG versus internal field H_i at several temperatures. The upper curves (A^+) were obtained with a sample previously exposed to large positive H_i while the lower curves (A^-) were obtained following exposure to large negative H_i. The breaks in the 1.32-K curves indicated by arrows correspond to entry into the mixed phase; their nature must be discussed elsewhere. For negative fields the curves were entirely similar and consistent with the time-reversal requirement $\vec{M}(A^+, H) = -\vec{M}(A^-, -H)$. (b) Corresponding theoretical curves for a range of low magnetizations calculated according to the microscopic mechanism proposed in Ref. 4. Published interaction parameters were used.

states to an applied field may be understood quantitatively in terms of two different mechanisms, both involving a crystallographic inequivalence between two types of sites for the Dy^{3+} spins. The mechanism proposed by Blume *et al.*[3] involves a difference in the effective magnetic moments and this may be estimated from previously determined g values and Van Vleck susceptibilities. An estimate of the effect of these differences on the behavior of the two TR states shows[7] that it should be very small, suggesting that some other mechanism is in fact dominant under most conditions. Such a mechanism was proposed recently by Giordano *et al.*,[4] and it involves a difference in the geometrical arrangement of the

spin-spin interactions. Values of the individual spin-spin interactions have been estimated from various thermodynamic measurements on DyAlG[8] and we can use these results directly in a low-field, low-temperature series expansion to predict M for both A^+ and A^-.

The calculation was made by including first-, second-, and third-nearest neighbors explicitly and the effect of all other neighbors in terms of a mean field, using the published values for the nondipolar interaction parameters and dipole sums calculated from the structure. Details of the calculation will be given elsewhere,[4,9] and we will note here only that it was necessary to include terms involving the simultaneous excitations of three nearest-neighbor spins before there was any difference between A^+ and A^-.

The results of the calculations are shown as the broken lines in Fig. 2. It can be seen that the agreement is quite satisfactory, especially at the lower temperatures where the approximation should be the best. It should also be noted that the calculated behavior is here based on a Hamiltonian which only includes Ising terms and that the difference between the two sublattices is an intrinsic property of this type of Ising model.

The substantial difference between the magneto-optical rotations of A^+ and A^- at elevated fields makes it possible to observe mixed antiferromagnetic states microscopically. As in Ref. 2, the apparatus consisted of a polarizing microscope whose optical axis lay along the axis of an electromagnet. The sample was immersed in superfluid helium in a Dewar with flat glass windows. Linearly polarized monochromatic light impinged on the sample. The analyzer was set to give the most revealing contrast.

In Figs. 3(a)–3(c) we show a sample which had been prepared in A^-, subjected to various field changes indicated schematically on the right. The temperature throughout was maintained constant at 1.32 K, as indicated by the hatched shading.

In Fig. 3(a) the field has taken the sample just into the mixed phase region and bright P^+ nuclei can be seen against the dark A^- background. If the field is now reduced back into the region where only A^+ is stable, the P^+ regions will transform into A^+ and in Fig. 3(b) we see the A^+ regions in A^-, as expected. The rotation contrast is here not as great as that between A^- and P^+ because of the much smaller difference in the magnetizations, but the coexisting domains are clearly visible. If the field is once again raised into the mixed phase region, P^+ is again nucleated and we can observe all three phases simultaneously [Fig. 3(c)]. It is interesting to note that P^+ is nucleated at the A^+-A^- boundary so that the antiferromagnetic domain walls have been effectively "decorated." That such nucleation would occur was noted by Jacobs and Lawrence,[10] and has recently been discussed by Mitsek and Gaidanskii.[11]

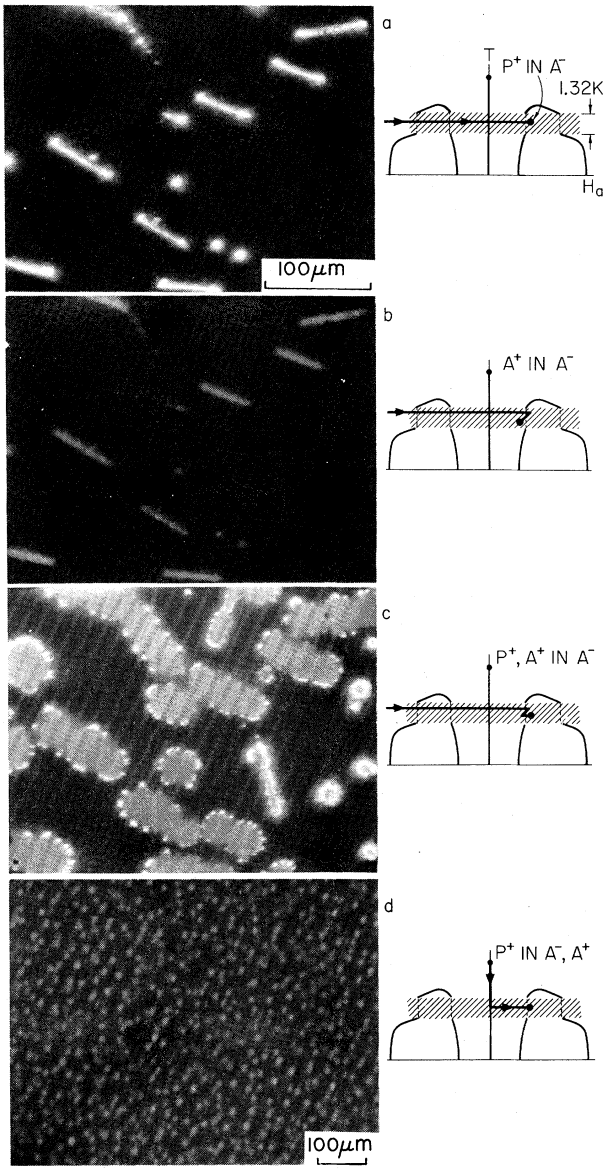

FIG. 3. Micrographs of the A^+, A^-, and P^+ magnetic phases obtained by various field and temperature treatments. All were taken at 1.32 K. For (a), (b), and (c) the sample was 690 μm thick; for (d), 190 μm. The field and temperature treatments used to obtain the four cases are indicated schematically on the right and are discussed in the text.

Neither this situation nor that of Fig. 3(a) is stable. In the course of minutes the P^+ slowly converts to a larger volume of A^+, corresponding to the lowest free-energy state. At higher temperatures the conversion is still more rapid, as one might expect.

Figure 3(d) shows the result of an experiment in which we cooled from above T_N in zero field ($<10^{-3}$ Oe) and at 1.32 K gently entered the mixed-phase region. The result of this procedure is a dense array of small needles of the paramagnetic phase. From experiments such as (c) above, we know that in the presence of an A^+-A^- mixture P^+ will nucleate on the domain walls, and we therefore interpret the present result as indicating an extremely finely divided mixture of A^+ and A^- in the initially cooled system. Cooling in larger fields produces various more complicated situations and we shall discuss these elsewhere.

These experiments have shown that it is possible to prepare a DyAlG sample either predominantly A^+ or predominantly A^- or else as various mixtures of the two. The distinctive rotation-versus-field curves of the two antiferromagnetic states has enabled us to study their properties directly. We have seen that two microscope techniques are effective for visualizing the TR domains. The first relies on the rotation difference at moderate fields, which provides a direct contrast when the sample is viewed between suitably set polarizers. This is applicable to all antiferromagnets in which the field couples to the antiferromagnetic order parameter.[6] The second technique makes use of the fact that the paramagnetic phase with its large rotation nucleates along A^+-A^- walls. We can thus "decorate" the antiferromagnetic walls with readily visible paramagnetic nuclei, an effect of still wider applicability. Further experiments are now in progress and will be reported elsewhere.

We would like to thank R. Alben, M. Blume, L. M. Corliss, and J. M. Hastings for a number of stimulating discussions.

---

*Work at Yale University supported in part by the U. S. Army Research Office, Durham, North Carolina.

[1]D. P. Landau, B. E. Keen, B. Schneider, and W. P. Wolf, Phys. Rev. B 3, 2310 (1971). This paper also gives extensive references to earlier work.

[2]J. F. Dillon, Jr., E. Y. Chen, and W. P. Wolf, in Proceedings of the International Conference on Magnetism, Moscow, U.S.S.R., 22–28 August 1973 (to be published).

[3]M. Blume, L. M. Corliss, J. M. Hastings, and E. Schiller, Phys. Rev. Lett. 32, 544 (1974).

[4]N. Giordano, W. P. Wolf, J. F. Dillon, Jr., and E. Yi Chen, to be published.

[5]R. Alben (private communication) has pointed out that the presently assumed magnetic symmetry of DyAlG, *Ia3d*, predicts similar effects provided $H_x H_y H_z \neq 0$.

[6]A good review of previous work has been given by A. S. Borovik-Romanov, in *Elements of Theoretical Magnetism*, edited by S. Krupicka and J. Sternberk, translated by W. S. Bardo (CRC Press, Cleveland, Ohio, 1968), p. 193.

[7]R. Alben and N. Giordano, private communication.

[8]B. Schneider, D. P. Landau, B. E. Keen, and W. P. Wolf, Phys. Lett. 23, 210 (1966), and to be published; R. Bidaux, P. Carrara, and B. Vivet, J. Phys. (Paris) 28, 187 (1967).

[9]N. Giordano, thesis, Yale University (to be published).

[10]I. S. Jacobs and P. E. Lawrence, Phys. Rev. 164, 866 (1967).

[11]A. I. Mitsek and P. F. Gaidanskii, Zh. Eksp. Teor. Fiz. 62, 2252 (1972) [Sov. Phys. JETP 35, 1178 (1972)]; A. I. Mitsek, Phys. Status Solidi (b) 59, 309 (1973).

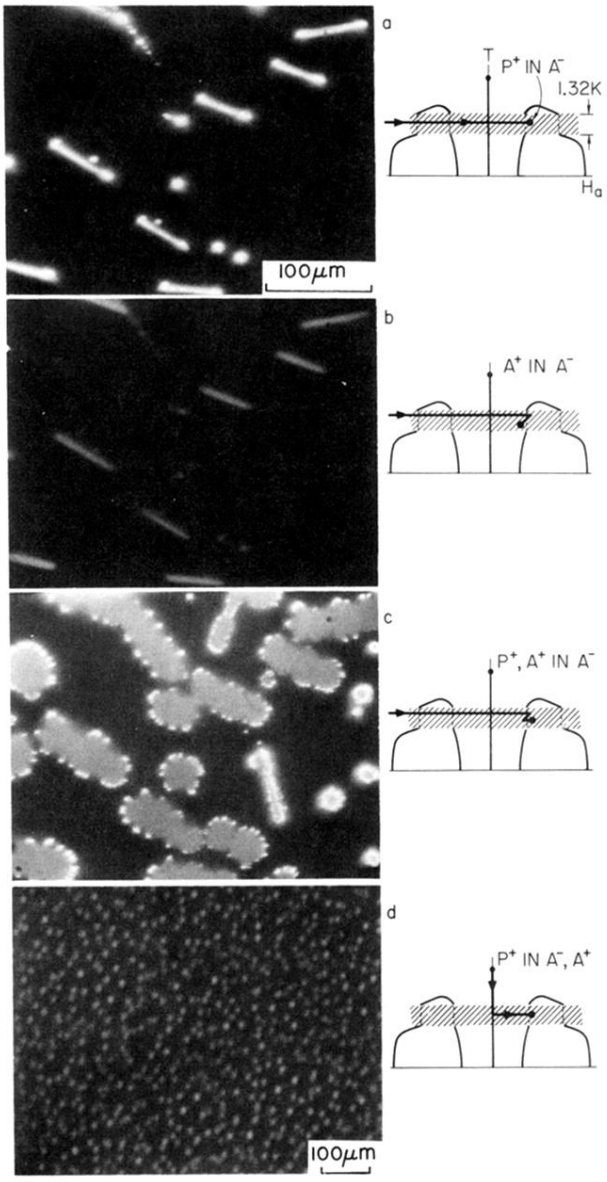

FIG. 3. Micrographs of the A^+, A^-, and P^+ magnetic phases obtained by various field and temperature treatments. All were taken at 1.32 K. For (a), (b), and (c) the sample was 690 μm thick; for (d), 190 μm. The field and temperature treatments used to obtain the four cases are indicated schematically on the right and are discussed in the text.