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Investigation of Magnetic Interactions in $TbAlO_3$ by Optical Spectroscopy

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High-resolution optical absorption spectra of $TbAlO_3$ have been obtained between 1.4 and 4.2°K, at 20°K and at 77°K. For $T>4$°K, all absorption lines show a triplet structure which is interpreted as arising from an interaction of each Tb ion with only two nearest neighbors. Below $T=3.95$°K, a temperature-dependent lineshift is observed giving a Néel temperature of $T_N=3.95\pm0.10$°K. Below the Néel temperature, the lineshift is proportional to $(1-T/T_N)^\beta$ with $\beta=0.32\pm0.08$. After correction for dipolar interactions, we find for the interactions along the c axis 2.4 cm^{-1}, and for those in the a-b plane 1.0 cm^{-1}. The magnetic moment of the ground state is derived as (9.0 ± 0.6) μ_B. The absorption spectra measured as a function of the external field show evidence for metamagnetic transitions, and a critical field of 8 kOe is derived.

1. INTRODUCTION

OPTICAL absorption and emission spectroscopy has become a useful tool for studying transparent magnetically ordered crystals. The sharp emission and absorption lines which are found in many transition-metal and rare-earth ionic compounds allow the study of magnetic phenomena with a resolution typically of about 1°K. The advantage of optical spectroscopy over conventional magnetic methods is that in principle one can deduce from the observed lines the energy-level schemes in the magnetically ordered state, which then can be used to interpret macroscopic magnetic properties such as magnetization and specific heat.

In optical measurements one measures the sum or difference of the properties of the ground state and an excited state. However, it is often difficult to relate, in a unique way, the observed lines to an energy-level scheme. This difficulty may be the main reason for the fact that to this date there are relatively few optical studies which have yielded completely new information not known from other measurements.

For the interpretation of optical spectra of rare-earth ions the molecular-field theory is usually adequate.[1] In this description one thinks of the magnetically ordered state as one with additional internal fields which split degenerate energy levels, thus giving rise to Zeeman patterns similar to those commonly observed with external magnetic fields.

On the other hand, the interpretation of optical lines in magnetically ordered compounds of transition-metal ions is most favorably done in terms of the spin-wave theory as becomes most apparent from the experiments of Greene *et al.*[2]

In the present paper a spectroscopic study of the metamagnet $TbAlO_3$ is reported. The orthoaluminates are isomorphic with orthoferrites. The orthoferrites show very interesting magnetic features.[3] This may possibly be due to the rare-earth–iron interaction. The iron-iron interaction can be studied very well in these compounds at high temperatures. To estimate the rare-earth–iron interaction it is desirable to know in addition the rare-earth–rare-earth interaction which can be obtained from a study of the orthoaluminates. In addition, the study of the orthoaluminates is interesting in itself for they form relatively simple systems (two magnetically inequivalent sites as compared with six in the garnets) and thus are good substances to learn more about the rare-earth–rare-earth exchange interactions in solids.

Magnetic measurements[4] and neutron diffraction[5] studies have shown that $TbAlO_3$ is antiferromagnetic below about 4°K. The magnetic structure contains four sublattices, with the magnetic moments confined to the a-b plane. Metamagnetic behavior was observed for magnetic fields applied in the a-b plane.[4] Our optical absorption data agree with these results. We observe a shift in the exchange splitting below 4°K, and anomalies in the Zeeman pattern associated with the metamagnetic transitions. The magnitudes of the exchange splittings are consistent with the observed Néel point.

A brief account of the present work has already been published.[6] It is one in a series of investigations to study by optical means the interactions which produce the ordering in the rare-earth sublattice. Results on $ErFeO_3$,[7] $HoFeO_3$, $DyFeO_3$, and $DyAlO_3$ [8,9] have already been reported.

2. EXPERIMENTAL RESULTS AND DISCUSSION

Single crystals of $TbAlO_3$ were grown by heating the mix Tb_2O_3:Al_2O_3:PbO:PbF_2:B_2O_3=36:8:130:90:6 g to 1300°C and cooling at 2°C/hr to 900°C. The ortho-

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[1] M. J. M. Leask, J. Appl. Phys. **39**, 908 (1968).

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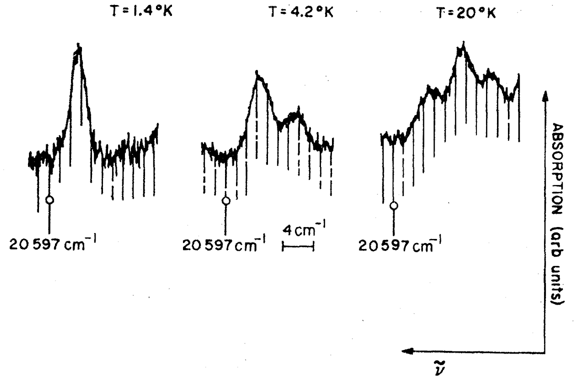

FIG. 1. Absorption line of TbAlO$_3$ at 20, 4.2, and 1.4°K.

aluminate structure[10] D_{2h}^{16} (*Pbnm*) and the lattice constants, $a=5.231$ Å, $b=5.305$ Å, and $c=7.425$ Å, were ascertained from powder diffraction patterns. Flux-free, colorless, and untwinned[11] crystals were selected for the measurements. For the optical measurements they were ground to plates with an area of 5×5 mm and a thickness varying from 0.1 to 2 mm. The spectroscopic measurements were performed with a 2-m Jarrel-Ash grating spectrograph. The crystals were placed directly in the cooling liquid (N$_2$, H$_2$, He). Below 4.2°K the temperature was deduced from the vapor pressure above the liquid helium. The absolute accuracy in this region is about ±0.5°K, whereas the relative accuracy is ±0.01°K.

The orientation of the crystals was checked by x-ray photographs. The alignment relative to the direction of the magnetic field is believed to be accurate to about ±3°.

A. Spectra without External Field

The absorption lines in the visible and ultraviolet region all show a triplet structure at 4.2 and 20°K. The spacing between two components of each triplet is (3.8 ± 0.5) cm^{-1}. Below (3.95 ± 0.10)°K an additional shift of the outer components of the triplets relative to their position at 4.2°K is observed; in addition the low-energy components lose intensity until at the lowest temperatures ($T<2.5$°K) only the highest-energy absorption line of each triplet is still present. In addition to the lineshift a narrowing of the lines is observed. The shift of the lines relative to their position at 4.2°K is (0.8 ± 0.3) cm^{-1}. A typical absorption line at 20, 4.2, and 1.4°K is shown in Fig. 1. In the absorption spectra taken at 77°K no additional lines are observed, the only change as compared to the 20°K spectra being a slight increase in linewidth.

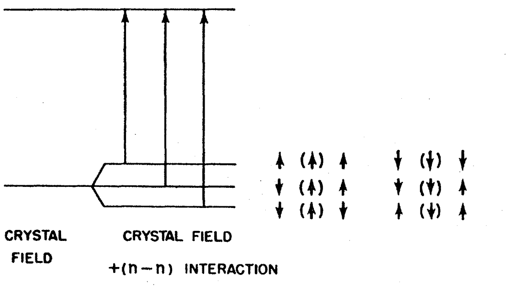

FIG. 2. Pseudo-energy-level diagram (energy-difference diagram) showing the origin of the triplets (according to Ref. 13). (↑) and (↓) designate the spin of the ion under question, whereas ↑ and ↓ are the spins of the neighboring ions along the c axis. Each level of the ground state is still doubly degenerate [spin up (↑) and spin down (↓)]; this degeneracy can be removed by an external magnetic field. All excited states are singlets.

The Tb^{3+} ion has the electronic configuration $4f^8$ with a $^7F_6$ ground state. This ground state is split by the crystal field of C_{1h} symmetry into 13 singlets. Since magnetic order occurs in this compound we must assume that the ground state produced by the crystal-field interaction is an accidental doublet. The triplet structure of the absorption lines is the same for all lines; this together with the temperature dependence suggests that it is due to a splitting of the ground state and that the crystal-field levels of the excited states are singlets. This simplifies the interpretation of the absorption spectra, for apart from second-order effects the structure of every absorption line reflects the ground-state energy levels. The observed triplet structure above 3.95°K is interpreted as being due to the interaction of each ion with only two neighbors. That this type of interaction gives rise to the observed structure was first pointed out by Bleaney *et al.*,[12] and then observed in optical spectra by Prinz.[13] A diagram which demonstrates the origin of the triplet structure associated with the interaction of a reference ion with two neighbors is shown in Fig. 2. In that diagram (↑) and (↓)

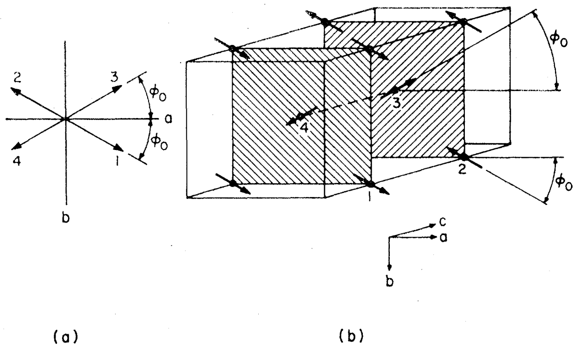

FIG. 3. Magnetic structure of antiferromagnetic TbAlO$_3$: (a) four-sublattice model deduced from magnetization data (Ref. 4); (b) magnetic unit cell (symmetry G_xA_y) from neutron diffraction (Ref. 5). Magnetic moments are confined in planes at $z=\frac{1}{4}$ and $z=\frac{3}{4}$.

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[12] B. Bleaney, R. J. Elliot, and H. E. D. Scovil, Proc. Phys. Soc. (London) **A64**, 933 (1951).

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represent the magnetic moment of the ion in question, while ↓ and ↑ represent the magnetic moments of the two neighbors with which the interaction takes place. In the perovskite structure each Tb ion is surrounded by six Tb neighbors, four in the *a*-*b* plane and two along the *c* axis, one above and one below (Fig. 3). According to Holmes *et al.*[4] and Bielen *et al.*[5] the four neighbors in the *a*-*b* plane are all on the same sublattice, but not the same as the reference ion. It is hard to see how two of the four neighbors could have a preferential position; we suggest, therefore, that the observed structure in the spectrum arises from the interaction of each Tb ion with its two neighbors above and below along the *c* axis. The observed interaction energy between two ions is 1.9±0.25 cm^{-1}. We can account for 0.66 cm^{-1} by dipolar interactions, thus leaving an exchange interaction of 1.2±0.3 cm^{-1} between two Tb ions along the *c* axis in this compound.

The additional shift observed below (3.95±0.10)°K we attribute to the onset of long-range order. A calculation of the magnetic dipolar fields in antiferromagnetic TbAlO_3 is described in the following section (Table II). We find that the dipolar contributions to the splitting *decrease* from 1.32 cm^{-1} above T_N, where only two neighbors contribute, to 1.12 cm^{-1} below T_N, in the presence of long-range order. The increase (0.8±0.3 cm^{-1}) in the splitting below T_N must be ascribed, therefore, to additional exchange interactions, totaling 1.0±0.3 cm^{-1} in the ordered state. It is natural to assume that these interactions arise from the coupling of a Tb ion with its four nearest neighbors in the *a*-*b* plane. This gives an exchange interaction of 0.25 cm^{-1} with each ion in the *a*-*b* plane as compared with 1.2 cm^{-1} with an ion along the *c* axis.

We have tried to make a careful measurement of the lineshift with temperature just below the Néel temperature. This measurement is difficult because the shift is small compared to the linewidth. The result of this measurement is shown in Fig. 4. If we assume that in the framework of the molecular-field theory the lineshift follows the sublattice magnetization, a temperature dependence of the form $[\Delta W(T)]/[\Delta W(T=0)]=D(1-T/T_N)^\beta$ is expected as was first demonstrated experimentally by Heller and Benedek.[14,15] Here $\Delta W(T=0)$ is the additional shift of the absorption lines between 4.2 and 1.4°K $[\Delta W(T=0)=0.8\ \text{cm}^{-1}]$. In the present experiment, values of $D=1.4\pm0.4$ and $\beta=0.32\pm0.08$ are derived.

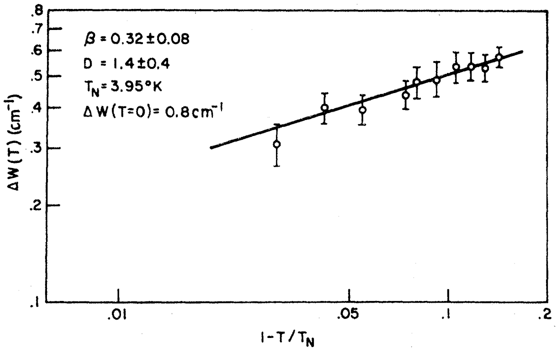

FIG. 4. The lineshift of a typical absorption line relative to its position at 4.2°K plotted as a function of $1-T/T_N$ on a doubly logarithmic plot.

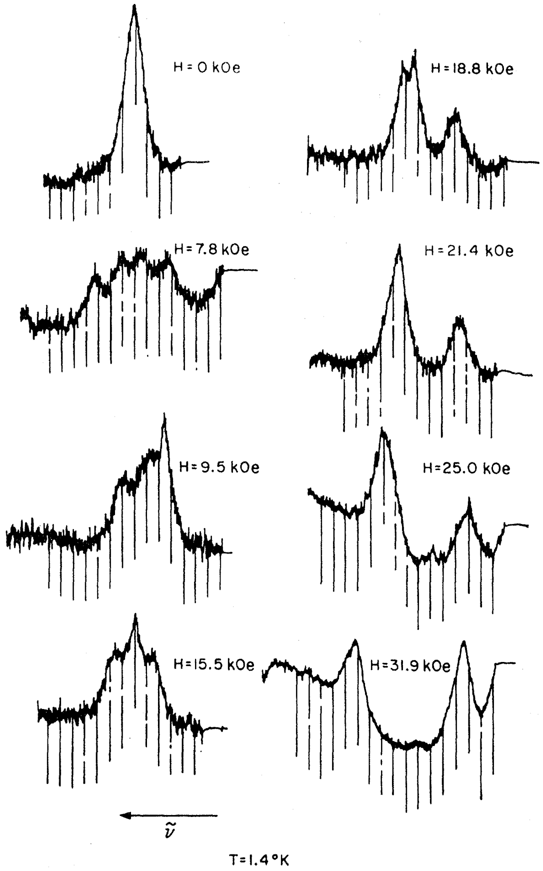

FIG. 5. Absorption line of TbAlO_3 at 1.4°K as a function of magnetic field; direction of magnetic field is along the direction of the magnetic moments in the ordered state.

Measurements of the emission spectrum at 77°K revealed transitions to excited crystal-field levels of the ground term $^7F_6$. We could find levels (or grouping of levels; the resolution of the emission spectra was only about 10 cm^{-1}) at 250 and 330 cm^{-1}. Crystal-field splittings ~200 cm^{-1} were estimated from the static susceptibilities at low temperatures.[4]

B. Measurements with External Magnetic Field

Measurements with external magnetic fields were performed at 4.2 and 1.4°K. The spectra at 1.4°K are much simpler because of depopulation of two of the triplet states. Only these spectra will be described in detail; those taken at 4.2°K give no additional informa-

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[14] P. Heller and G. B. Benedek, Phys. Rev. Letters **8**, 428 (1962).

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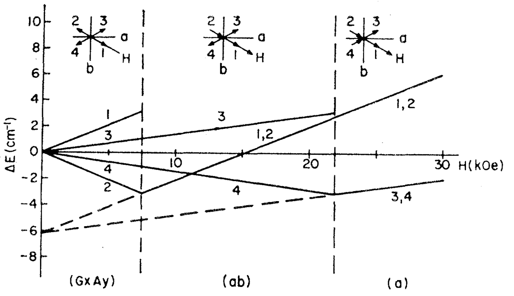

FIG. 6. Idealized Zeeman pattern in $TbAlO_3$.

tion. In addition the spectra below the λ point had the best signal-to-noise ratio and the sharpest lines thus giving the best resolution, which was essential for resolving some of the very complicated patterns. Figure 5 gives an absorption line at 1.4°K for various values of the external field along the direction of the magnetic moments.

For the immediate understanding of the Zeeman patterns in Fig. 5 we show in Fig. 6 a theoretical diagram of a typical absorption line. It is derived from the known magnetic structure[4,5] which is also indicated. Its full derivation will be discussed in the next section in detail.

In fields above 20 kOe generally two Zeeman lines are observed for every zero-field line (see Fig. 5). They are attributed to the Zeeman splitting of the ground state for the two magnetically inequivalent sites. Along the direction 35° off the a axis the maximum shift of one of these two lines is observed. This is to be expected from the magnetization[4] and the neutron diffraction[5] measurements which predict that the magnetic moments are at 36° and 34° from the a axis, respectively. The magnetic moment of the ground state was obtained from the high field splitting of two lines separated by more than 100 cm^{-1} from other lines. Thus, an approximate correction for second-order effects in the excited state could be made by taking the average splitting of the two lines. We obtain a value of $(9.0\pm0.6)\mu_B$ for the ground-state magnetic moment in good agreement with the value of the magnetization measurement $(9\mu_B)$ and the neutron diffraction measurement $(8.25\mu_B)$. Figure 7 shows the magnetic field dependence of a typical absorption line at 1.4°K. This figure reflects essentially the ground-state splitting. The interpretation of this pattern is done in terms of the known magnetic structure and will be described in the next section.

Experiments performed with the direction of the magnetic field along the c axis showed that within the accuracy of our experiment the magnetic moment in this direction is zero.

3. INTERPRETATION OF THE ZEEMAN PATTERN

The Zeeman pattern (Fig. 7) may be fitted to a single-ion model for the magnetic behavior of Tb^{3+} in antiferromagnetic (af) $TbAlO_3$. Interactions between ions may be included with Weiss molecular fields. The data (Fig. 7) fall on straight lines, within experimental error. The second-order Zeeman effect may therefore be neglected. A calculation from the Van Vleck contributions to the static susceptibilities[4] predicts second-order shifts in energy for the ground state of less than 0.1 cm^{-1} at the highest fields used.

The proposed magnetic structures, projected on the a-b plane, are shown in Fig. 6 for each of the three distinct regions of the Zeeman pattern. At low fields the af state with point symmetry G_xA_y, in Bertaut's notation,[16] is stable.[5] In this state, the magnetic moments on the four Tb^{3+} ions in the unit cell are arranged as shown in Fig. 3. At intermediate fields we propose the state (ab), consistent with magnetization data,[4] in which the magnetic moments on two of the four ions in the unit cell are parallel to the applied field H. The state (a) is stable at high fields, giving a net magnetization along the a axis.

Detailed expressions for the energy shifts in the Zeeman pattern will be derived in following paragraphs. Lest the physics be obscured by details, we describe in this paragraph a more qualitative interpretation of the data. We consider only the strong interactions between neighbors (1 and 2 or 3 and 4) along the c axis. This approximation gives the Zeeman pattern in Fig. 6. In a weak magnetic field, the absorption lines shift according to the first-order Zeeman effect. The slopes are $\pm9\mu_B$ for ions 1 and 2 (parallel and antiparallel to H) and $\pm9\mu_B \cos 70°$ for ions 3 and 4. Let H^{af} be the Weiss molecular field parallel to any of the magnetic moments in the af state. At $H=H^{af}$, Zeeman and interaction energies balance to give a metamagnetic transition to state (ab). In this state, ions 1 and 2 are magnetically equivalent. Each has a magnetic moment of $9\mu_B$ in a molecular field $H'=-H^{af}$, giving the single line labeled 1, 2 in Fig. 6. The analogous transition for ions 3 and 4 occurs at $H=H^{af}/\cos 70°$. We have taken $H^{af}=7.5$ kOe

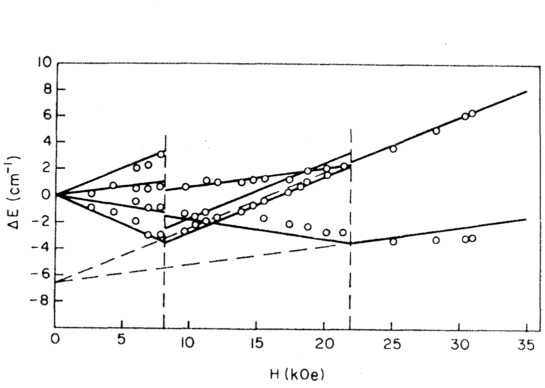

FIG. 7. Zeeman pattern of a typical absorption line of $TbAlO_3$ at 1.4°K. The magnetic field is directed at an angle of 35° off the a axis in the a-b plane. Sample is disk-shaped, with demagnetizing factor <0.001. Solid curves are derived from molecular-field theory (see text).

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[16] E. F. Bertaut, *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. III, p. 149.

TABLE I. Energy shifts in $TbAlO_3$.

	Magnetic state		
	G_xA_y	(ab)	(a)
Ion 1	$-\mu_0 H$	$-\mu_0 H+2\mu_0 H_{12}$	$-\mu_0 H+2\mu_0(H_{12}+H_{14})$
Ion 2	$+\mu_0 H$	$-\mu_0 H+2\mu_0(H^{\mathrm{af}}-H_{11})$	$-\mu_0 H+2\mu_0(H_{12}+H_{14})$
Ion 3	$-\mu_0 H \cos 2\phi_0$	$-\mu_0 H \cos 2\phi_0+2\mu_0 H_{14}$	$-\mu_0 H \cos 2\phi+2\mu_0(H_{12}+H_{14})$
Ion 4	$+\mu_0 H \cos 2\phi_0$	$+\mu_0 H \cos 2\phi_0+2\mu_0 H_{13}$	$-\mu_0 H \cos 2\phi_0+2\mu_0(H_{12}+H_{14})$
Unit cell	0	$-2\mu_0 H+2\mu_0(H^{\mathrm{af}}-H_{11})$	$-2\mu_0 H(1+\cos 2\phi_0)$ $+4\mu_0(H_{12}+H_{14})$

in Fig. 6 to give rough agreement with the observed Zeeman pattern (Fig. 7) and magnetization curves.[4]

Now consider the effects of weak interactions between neighbors (1 and 4 or 2 and 3) in the *a-b* plane and long-range magnetic dipolar interactions. These interactions produce three changes in the idealized Zeeman pattern in Fig. 6: (i) The ratio of the critical fields is not exactly cos 70°, (ii) discontinuities occur for all of the Zeeman lines at the transitions, and (iii) the degeneracy of lines 1 and 2 is lifted in state (*ab*).

Let H_{ij} be the Weiss molecular field parallel to the magnetization of sublattice i due to the sublattice j in antiferromagnetic $TbAlO_3$. This field includes exchange and magnetic dipolar contributions

$$H_{ij}=H_{ij}^{\mathrm{ex}}+H_{ij}^{\mathrm{dip}}. \tag{1}$$

The total field on each ion in the af state

$$H^{\mathrm{af}}=\sum_{j=1}^{4} H_{ij} \tag{2}$$

is the same for all four ions in the unit cell. In Table I we have listed the shift in energy for each ion in a magnetic field H parallel to the magnetization of ion 1. The zero of energy is the af state at $H=0$. The magnetic moment per ion is μ_0 and the angle (35°) between the magnetic moments and the a axis is φ_0. In compiling the expressions in Table I, we have used the general condition of reciprocity

$$H_{ij}=H_{ji} \tag{3}$$

and the relations from symmetry

$$H_{11}=H_{22}=H_{33}=H_{44}, \tag{4}$$

$$H_{12}=H_{13}, \qquad H_{14}=H_{23}, \qquad H_{24}=H_{13}. \tag{5}$$

It may be observed that the expressions in Table I give the curves in Fig. 6 as a special case when $H_{11}=H_{13}=H_{14}=0$.

The magnetic dipolar contributions to H_{ij} are listed in Table II, for magnetic moments of $9\mu_B$ per ion. A lattice summation to evaluate these contributions was carried out with a computer for a spherical sample of radius $R_0=100$ Å. Following Bielen *et al.*,[5] we used crystallographic parameters for Tb ($x=-0.007$, $y=0.050$) as determined by Mériel for $DyAlO_3$. At $R_0=100$ Å, the change in H_{ij}^{dip} produced a 10% change in R_0 was less than 0.5%. We find a total dipolar field per ion of 2.66 kOe, as compared to 2.91 kOe determined by Bielen *et al.*[5] for magnetic moments of $8.25\mu_B$ per ion at angles ±34° from the *a* axis. We have not resolved this apparent discrepancy. We get the same result using Bertaut's technique[15] for calculating dipolar interactions.

The Lorentz fields in Table II sum to zero and do not, therefore, affect the energy in the af state. They must be included, however, to calculate the energy shifts and critical fields for a disk-shaped sample (Fig. 7).

For the exchange contributions to H_{ij}, we restrict ourselves to interactions for two nearest neighbors along the c axis (H_{12}^{ex}) and four nearest neighbors in the *a-b* plane (H_{14}^{ex}). Using the exchange splittings described in the previous section, namely 2.4 cm^{-1} for the exchange interaction along the c axis and 1.0 cm^{-1} for that in the *a-b* plane, we find

$$H_{12}^{\mathrm{ex}}=(5.7\pm1.4)\,\mathrm{kOe}, \tag{6}$$

$$H_{14}^{\mathrm{ex}}=(2.4\pm0.7)\,\mathrm{kOe}. \tag{7}$$

Expressions for the critical fields are obtained by equating the energy per unit cell in the states on either side of the transitions. These energies are listed in the last line of Table I. Note that one must be careful to include only once the interactions between a pair of ions. For the low- and high-field transitions, we find

$$H_{c0}=H^{\mathrm{af}}-H_{11} \tag{8}$$

and

$$H_c'=(H_{c0}-2H_{13})/\cos 70°, \tag{9}$$

respectively. Substituting from Eqs. (6) and (7) and Table II, we obtain

$$H_{c0}=(8.2\pm2.1)\,\mathrm{kOe}, \tag{10}$$

$$H_c'=(21.9\pm6.1)\,\mathrm{kOe}, \tag{11}$$

in good agreement with the anomalous points in the Zeeman pattern (Fig. 7) and with the value $H_{c0}=7.5$ kOe from magnetization data.[4] Since $2H_{13}$ is only 0.7 kOe (Table II), we have $H_{c0}/H_c'\approx\cos 70°$, as observed.

TABLE II. Magnetic dipolar fields in $TbAlO_3$ ($T=0°K$).

	Spherical sample (kOe)	Lorentz field (kOe)	Total (kOe)
H_{11}^{dip}	0.89	1.70	2.59
H_{12}^{dip}	2.91	−1.70	1.21
H_{13}^{dip}	−0.24	0.58	0.34
H_{14}^{dip}	−0.90	−0.58	−1.48

Taking the mean values determined for H_{12}^{ex} and H_{14}^{ex}, and setting $\mu_0=9\mu_B$, we obtain the curves in Fig. 7, in reasonable agreement with the data. The data are inconclusive concerning the splitting of lines 1 and 2 in the region of intermediate field strength [state (ab)]. A small shoulder on the line, at about the predicted position, may be an indication of the splitting or of a slight misorientation of the sample. The predicted splitting is 1 cm^{-1}, as compared to a linewidth of 2.4 cm^{-1}. A similar comment may be made concerning the slight discontinuities in lines 2 and 3 at the transitions. The discontinuities are not ruled out by the data (Fig. 7), although the points do appear to fall on straight lines through the transition regions. We cannot eliminate the possibility that the magnetic moments on ions 3 and 4 are rearranged in state (ab) in such a way as to minimize (or eliminate) these discontinuities and the splitting of lines 1 and 2.

4. DISCUSSION—EXCHANGE CONSTANTS

Considerations of the exchange splittings and magnetic properties of $TbAlO_3$ have shown that neighboring ions along the c axis are more strongly coupled than neighbors in the a-b plane. For the exchange coupling of *pairs* of ions (Fig. 4), we find

$$E_{14}^{\mathrm{ex}}/E_{12}^{\mathrm{ex}}=0.21\pm0.12,$$

where E_{ij}^{ex} is the exchange energy in the af state for ions i and j. This result is in reasonable agreement with the value

$$E_{14}^{\mathrm{ex}}/E_{12}^{\mathrm{ex}}=(\cos 110°)/(\cos 180°)=0.34,$$

which would hold for equivalent Heisenberg (isotropic) interactions. On this basis, the reason for the preferential coupling of neighbors along the c axis is geometrical. These neighbors have antiparallel magnetic moments, whereas the moments in the a-b plane are locked at relative angles of 110° by the crystal field.

It might be noted that antisymmetric exchange of the Dzialoshinsky-Moriya[17] type cannot be ruled out in this structure. Such interactions would favor perpendicular orientations of the moments on ions 1 and 4 (Fig. 4). Antisymmetric interactions between Eu^{3+} and Fe^{3+} ions in the garnet structure have been proposed[18] to explain a large shift in the apparent value of the crystal-field parameter D. However, Tachiki and Sroubek[19] have argued that antisymmetric exchange will generally be small for rare-earth ions, because of the spin-orbit couplings involved. Anisotropy in the symmetric exchange is certainly to be expected in $TbAlO_3$. Even with anisotropy we would expect the maximum interaction for antiparallel magnetic moments, as we have proposed above.

A rough indication of self-consistency is provided by a calculation of T_N from the observed splittings. The magnetic structure in $TbAlO_3$ may be regarded as a collection of weakly interacting, antiferromagnetic Ising chains, with effective spin one-half. An expression for T_N in such a system has been obtained by Stout and Chisholm.[20] The strong intrachain interactions are treated exactly, whereas the weak interactions between chains are included in a molecular field. The expression for T_N in $TbAlO_3$ may be written

$$T_N=(\delta/k)\exp(\Delta/kT_N).$$

The parameter δ is the shift in the splitting below T_N, extrapolated to $T=0°K$, which results from interchain and long-range interactions. The parameter Δ is the splitting, at high temperatures, due to interactions of nearest neighbors on a given chain. Substituting $\Delta=3.8\pm0.5\ \mathrm{cm}^{-1}$ and $\delta=0.8\pm0.3\ \mathrm{cm}^{-1}$, we find

$$T_N=(4.2\pm1.0)°K$$

in good agreement with the observed value 3.95± 0.10°K.

5. SUMMARY

This study of the low-temperature optical absorption spectra of $TbAlO_3$ has provided detailed information on the exchange interactions and the magnetic behavior of the Tb^{3+} ion in the orthoaluminate structure. The triplet structure observed in the absorption spectra at temperatures as high as $20T_N$ showed that each Tb^{3+} ion interacts most strongly with only two of its six neighbors. By calculating the dipolar contributions to the interactions an exchange interaction of (1.2 ± 0.3) cm^{-1} was found between two ions with their moments being antiparallel. Below $(3.95\pm0.10)°K$ a shift of the high-frequency component of each triplet was observed indicating the onset of long-range order at that temperature. Just below the Néel temperature the line-shift had a temperature dependence of $D(1-T/T_N)^\beta$ with $\beta=0.32\pm0.08$ and $D=1.4\pm0.4$. The magnitude of the temperature-dependent shift gave a value of

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(1.0 ± 0.3) cm^{-1} for the exchange interaction of each Tb^{3+} ion with its four nearest neighbors in the a-b plane. A magnetic moment of $(9.0\pm0.6)\mu_B$ per ion at an angle of 35° from the a axis was obtained from the high-field Zeeman splitting.

Metamagnetic behavior produces anomalies in the Zeeman pattern at about 8 and 20 kOe. Good agreement was obtained between the experimentally determined exchange interactions, the positions of the metamagnetic anomalies, and the observed Néel point.

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Absence of Magnetic Ordering in One and Two Dimensions in a Many-Band Model for Interacting Electrons in a Metal*

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The Bogoliubov inequality is used to prove the absence of spontaneous magnetic ordering in one and two dimensions for a many-band model for electrons in a metal interacting via the Coulomb and the exchange interactions. The discussion follows closely Mermin and Wagner's recent proof of a similar theorem for the Heisenberg model of an interacting spin system.

USING an inequality due to Bogoliubov, Mermin and Wagner[1] have proved rigorously the absence of both ferromagnetism and antiferromagnetism in one- and two-dimensional spin systems described by the Heisenberg Hamiltonian. This theorem concerning the absence of magnetic ordering in one and two dimensions can be proved not only for systems described by the Heisenberg Hamiltonian, but for many others as well. Wegner[2] has shown that it applies to ferromagnetism in systems of locally interacting electrons and nuclei, and Hamilton[3] has shown that it applies to spin density waves in an electron gas. Much of the above work was stimulated by Hohenberg's[4] discussion of a similar theorem for one- and two-dimensional superfluids and superconductors.

Wegner proves the nonexistence of ferromagnetism in systems of electrons for all potentials V, describing the interaction of the electrons among themselves and with nuclei, that satisfy the condition

$$[C', V]=0, \tag{1}$$

where

$$C'=\sum_i \exp(i\mathbf{k}\cdot\mathbf{r}_i)\,(s_{ix}+is_{iy}) \tag{2}$$

and $\mathbf{r}_i$ and $\mathbf{s}_i$ are the position and spin of the ith electron. Potentials that are local (i.e., that do not contain the electron momenta $\mathbf{p}_i$) and do not contain the spin coordinates $\mathbf{s}_i$ satisfy this condition. Since V can contain a periodic potential, the proof applies, for example, to electrons in energy bands interacting via Coulomb forces.

The purpose of this paper is to demonstrate the absence of ferromagnetism and antiferromagnetism in systems of electrons described by the Hamiltonian (3), which is often used in practice[5] to discuss magnetic ordering in metals. Although the interaction, described by the terms H_C and H_{ex}, is usually assumed to represent approximately the effects of a full Coulomb, or a screened Coulomb interaction, it is not in general a local interaction. For this reason, a separate proof is required to establish that model Hamiltonians of the type (3) also do not admit magnetic ordering in one and two dimensions.

We consider systems described by the Hamiltonian

$$H=T+H_C+H_{\text{ex}}+H_Z, \tag{3}$$

where

$$T=\sum_{\mathbf{l}\mathbf{l}'r\sigma} T_r(\mathbf{l}-\mathbf{l}')\,c_{\mathbf{l}r\sigma}{}^{\dagger}c_{\mathbf{l}'r\sigma}, \tag{4}$$

$$H_C=\sum_{ll'\sigma\sigma'} K(ll')\,n_{l\sigma}n_{l'\sigma'}, \tag{5}$$

$$H_{\text{ex}}=-\sum_{l\neq l'} J(ll')\Big[\sum_{\sigma} n_{l\sigma}n_{l'\sigma}+2S_+(l)\,S_-(l')\Big], \tag{6}$$

$$H_Z=2h\sum_l \cos(\mathbf{q}\cdot\mathbf{l})\,S_Z(l)\,. \tag{7}$$

The nuclei in the crystal are assumed to form a Bravais

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