

## Low-temperature ordered states of dysprosium, terbium, and holmium aluminum garnets

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The low-temperature ordered states of dysprosium, terbium, and holmium aluminum garnets have been investigated by the Luttinger-Tisza method, taking rigorously into account the dipole-dipole interaction. It is found that for each of these cases, in the absence of an external magnetic field, the lowest-energy configuration corresponds to antiferromagnetic ordering with three pairs of sublattices, in agreement with neutron scattering experiments. In the presence of external field along the [001] direction, the lowest-energy configuration turns out to be that of ferromagnetic chains in the  $x$  and  $y$  directions, with the nearest ferromagnetic chains pointing in the opposite directions, the same as that proposed by Landau and Keen. When the external magnetic field is along the [110] direction, there are two configurations which correspond to lowest energy; the individual chains are ferromagnetic but not all ferromagnetic chains are parallel, there being chains which point in opposite directions.

### I. INTRODUCTION

The garnets have been subjected to a large number of theoretical and experimental studies. It has been found experimentally that dysprosium aluminum garnet  $\text{Dy}_3\text{Al}_5\text{O}_{12}$  (DAG), terbium aluminum garnet  $\text{Tb}_3\text{Al}_5\text{O}_{12}$  (TAG), and holmium aluminum garnet  $\text{Ho}_3\text{Al}_5\text{O}_{12}$  (HAG) order antiferromagnetically at low temperatures. As far as theoretical treatments are concerned, no attempts have been made to consider the dipole-dipole interaction completely. The published treatments have mainly dealt with either the Ising-type interactions among the rare-earth ions (e.g., Norvell *et al.*<sup>1</sup>), in which only the dipole interaction between the nearest neighbors is considered, or using fewer sublattices than must be considered (e.g., Capel<sup>2</sup>). Since the dipole interaction is a long-range interaction, it appears desirable to calculate as to what kind of magnetic ordering will be energetically favored at low temperatures if the dipole-dipole interaction were to be rigorously taken into account.

It is the purpose of the present paper to discuss in detail the application of the Luttinger-Tisza (LT) method to derive the low-temperature ordered states of DAG, TAG, and HAG. The cases of both—when an external magnetic field is absent and present—will be considered.

### II. PUBLISHED RESULTS

#### A. Dysprosium aluminum garnet (DAG)

DAG has been studied both experimentally and theoretically by a large number of workers.<sup>1-31</sup> Ac-

cording to Gorton *et al.*<sup>3</sup> there is a “weak,” but rather complex interaction between the  $\text{Dy}^{3+}$  ions which leads to antiferromagnetic (AF) ordering at 2.5 K; further the  $\text{Dy}^{3+}$  ions are magnetically very anisotropic in the garnet structure. Ball *et al.*<sup>4</sup> found  $\lambda$  anomaly at  $2.49 \pm 0.01$  K (see also Ref. 5) and concluded that there was unusual type of AF ordering. They found that about 70% of the total interaction is dipole-dipole (see also Ref. 8). The  $\text{Dy}^{3+}$  ions may be described in terms of only one Kramer’s doublet (effective spin  $\frac{1}{2}$ ) with  $g_{\parallel} \approx 17.7$  and  $g_{\perp} \approx 0$  (see also Refs. 6 and 9) where the axes of different ions in the crystal lie along one of the  $x$ ,  $y$ , or  $z$  axes of the cubic unit cell, i.e., there is local anisotropy with the presence of at least six sublattices. Their calculations showed that one of the states, which may be described in terms of chains of antiparallel spins has a markedly lower magnetic energy, and that a model of purely magnetic dipole, nearest-neighbor magnetic and small exchange interactions (see also Ref. 7) do, in fact account for the observed magnetic behavior remarkably well. Ball *et al.*<sup>5</sup> found AF ordering with at least three mutually orthogonal pairs of antiparallel sublattices, where for different ions in the unit cell the “parallel” axis lies along either the  $x$ ,  $y$ , or  $z$  cubic axes, i.e., a model of three orthogonal, interpenetrating lattices of Ising spins. Ball *et al.*<sup>7</sup> refined further the measurement of  $T_N$  to be  $2.53(5) \pm 0.01$  K (see also Ref. 9). According to Hastings *et al.*<sup>9</sup> DAG has the garnet crystal structure with eight molecules in the unit cell. The structure belongs to the space group  $Ia\bar{3}d(O_h^{10})$  with unit cell parameter  $a_0 = 12.04$  Å. There are 24  $\text{Dy}^{3+}$  ions on the ( $c$ ) sites of the unit cell. As far as the  $g$  tensor is concerned

$g_x \approx g_y \leq 1$ ;  $g_z = 18.2 \pm 0.2$  (see also Refs. 10, 17, and 32). In any case it seems a very good approximation to neglect both  $g_x$  and  $g_y$  as  $|g_x \pm g_y| < 0.5$ .<sup>17,33</sup> One additional possible complication which must be considered is the hyperfine interaction.<sup>34</sup> The dominant term has the form  $A_z S_z' I_z$  with  $A_z \approx 0.07$  K. Fortunately, it is possible to show<sup>11</sup> that the effect of this term does not alter the critical-point behavior of any Ising-like system.

According to Landau and Keen<sup>35</sup> (see also Refs. 16 and 17) the application of a magnetic field in arbitrary directions produces different effects at different sites. The cubic space group of DAG ( $O_h^{10}$ ) retains the threefold symmetry about [111] in going to the magnetic system. Because of this symmetry, all of the sites with a component of the applied field  $H_0$  along the spin direction become equivalent (e.g., for  $H_0$  along [111],  $+x$ ,  $+y$ , and  $+z$  are equivalent as are  $-x$ ,  $-y$ , and  $-z$ ). The application of a magnetic field along "special directions" other than [111] produces a transition from an antiferromagnetic configuration to the paramagnetic state (with the spins aligned along a  $+z'$  local axis) for either one or two pairs of sublattices while the other sublattices may become disordered or undergo transitions to a new antiferromagnetic or even ferromagnetic state.<sup>36</sup> DAG is highly unusual in that perfectly aligned paramagnetic spins are completely decoupled from all spins which are perpendicular to them. Based on these considerations Landau and Keen<sup>35</sup> proposed that for the case when the  $z$  spins are aligned by a strong magnetic field along [001], an antiferromagnetic arrangement of the  $x$  and  $y$  spins in which all  $x$ , or  $y$ , spins which have the same  $z$  coordinates constitute ferromagnetic chains; the two nearest  $x$ , or  $y$  ferromagnetic chains point in opposite directions. Further, Landau and Keen<sup>35</sup> conclude that the assumption of antiferromagnetic chains of  $z$  spins yields a positive internal energy at 0 K, i.e., the state is unstable, on the basis of dipolar interactions alone. In fact according to them, dipolar interactions predict the ordered state to be composed of ferromagnetic chains which are aligned in a three-dimensional ferromagnetic state.

### B. Terbium aluminum garnet (TAG) and holmium aluminum garnet (HAG)

Hammann<sup>37</sup> found by neutron-diffraction technique that both TAG and HAG order antiferromagnetically below  $T_N$  of 1.35 and 0.95 K, respectively, and have effective moments of  $5.7 \mu_B$  and  $5.8 \mu_B$ , respectively, at 0 K. He concluded that although a singlet lies lowest for both the  $Tb^{3+}$  and  $Ho^{3+}$  ions, the magnetic ordering is due to the interaction of the two lowest-lying singlets. He also found that the space group for both TAG and HAG is  $Ia\bar{3}d$  and both have  $a = 12.00 \pm 0.02$  Å (cubic). The position

of the rare-earth ions  $Tb^{3+}$  and  $Ho^{3+}$  in the unit cell are the same as those given in Table I for  $Dy^{3+}$  in DAG.

Gavignet-Tillard and Hammann<sup>38</sup> further investigated TAG by neutron diffraction at 0.31 K and by magnetization measurements at 0.36 and 4.2 K in fields up to 60 kG. They found that their results were consistent with mainly dipolar interactions, and two-singlet model, whose parameters are found to be 2.5 K for the energy splittings of the levels, and  $7.6 \mu_B$  for the only nonvanishing component of the magnetic moment. Gavignet-Tillard and Hamann<sup>39</sup> reported magnetic curves for TAG at 0.36 K with an external magnetic field applied along several crystallographic directions. They found that when the magnetic field is applied along [111], TAG is equivalent to a two-sublattice antiferromagnet. For the magnetic field along [001] direction in TAG, a large field polarizes the  $(\alpha-\alpha')$  sublattices, leaving  $(\beta-\beta')$  and  $(\gamma-\gamma')$  under the influence of their own interactions, which cause a reordering to another antiferromagnetic state of type AF' (the same as that found for DAG by Landau and Keen<sup>35</sup>). For the field in the [110] direction, contrary to DAG, no reordering of the  $(\alpha-\alpha')$  sublattices can be expected according to Gavignet-Tillard and Hammann.<sup>39</sup>

Theoretical considerations of Bidaux *et al.*<sup>40</sup> showed that the influence of splitting of energy levels for  $Tb^{3+}$  in TAG upon the metamagnetic transition suggests an analogy between an induced-moment system at 0 K and a Kramer's doublet at nonzero temperature.

## III. APPLICATION OF THE LUTTINGER-TISZA METHOD TO DAG

### A. Absence of external magnetic field

The Luttinger-Tisza method,<sup>41</sup> as applicable to the garnets studied in this paper, may be briefly described as follows. The crystal lattice is supposed to be divided into sublattices; each sublattice is generated from lattice translations  $\vec{l} = l\vec{a} + m\vec{b} + n\vec{c}$  applied to the various rare-earth ions situated in the unit cell. Thus, one considers 24 sublattices as there are 24 rare-earth ions in the unit cell of rare-earth aluminum garnets. The positions of the various ions in the unit cell of DAG are listed in Table I, which also contains the directions of spins as found by neutron scattering in the antiferromagnetic ordering at low temperatures. All the spins on any sublattice are assumed parallel. The configuration of spins is thus described by 72 numbers, the  $x$ ,  $y$ , and  $z$  components of the spins of the 24 spins in the unit cell. These can be considered as components of a vector  $\vec{S}$  in a 72-dimensional vector space. This configuration produces magnetic fields at each lattice point, which may

TABLE I. The relative orientations of the various spins in the unit cell, together with a number indicating the order in distance of the neighbor relative to the spin at  $(\frac{1}{2}, \frac{1}{4}, \frac{3}{8})$  taken as origin. The  $g$  factors are also included.

Dy <sup>3+</sup> position	Order of neighbor relative to spin at $(\frac{1}{2}, \frac{1}{4}, \frac{3}{8})$	Spin direction	Type	$(g_x, g_y, g_z)$
$(\frac{1}{8}, 0, \frac{1}{4})$	2	-x	b	(18.2, 0, 0)
$(\frac{1}{4}, \frac{1}{8}, 0)$	2	-y	b	(0, 18.2, 0)
$(0, \frac{1}{4}, \frac{1}{8})$	4	-z	b	(0, 0, 18.2)
$(\frac{3}{8}, 0, \frac{3}{4})$	2	+x	b	(18.2, 0, 0)
$(\frac{3}{4}, \frac{3}{8}, 0)$	2	+y	b	(0, 18.2, 0)
$(0, \frac{3}{4}, \frac{3}{8})$	7	+z	b	(0, 0, 18.2)
$(\frac{5}{8}, 0, \frac{1}{4})$	1	+x	a	(18.2, 0, 0)
$(\frac{1}{4}, \frac{5}{8}, 0)$	5	+y	a	(0, 18.2, 0)
$(0, \frac{1}{4}, \frac{5}{8})$	4	+z	a	(0, 0, 18.2)
$(\frac{7}{8}, 0, \frac{3}{4})$	5	-x	a	(18.2, 0, 0)
$(\frac{3}{4}, \frac{7}{8}, 0)$	8	-y	a	(0, 18.2, 0)
$(0, \frac{3}{4}, \frac{7}{8})$	10	-z	a	(0, 0, 18.2)
$(\frac{5}{8}, \frac{1}{2}, \frac{3}{4})$	2	-x	b	(18.2, 0, 0)
$(\frac{3}{4}, \frac{5}{8}, \frac{1}{2})$	2	-y	b	(0, 18.2, 0)
$(\frac{1}{2}, \frac{3}{4}, \frac{5}{8})$	4	-z	b	(0, 0, 18.2)
$(\frac{7}{8}, \frac{1}{2}, \frac{1}{4})$	2	+x	b	(18.2, 0, 0)
$(\frac{1}{4}, \frac{7}{8}, \frac{1}{2})$	6	+y	b	(0, 18.2, 0)
$(\frac{1}{2}, \frac{1}{4}, \frac{7}{8})$	3	+z	b	(0, 0, 18.2)
$(\frac{1}{8}, \frac{1}{2}, \frac{3}{4})$	5	+x	a	(18.2, 0, 0)
$(\frac{3}{4}, \frac{1}{8}, \frac{1}{2})$	1	+y	a	(0, 18.2, 0)
$(\frac{1}{2}, \frac{3}{4}, \frac{1}{8})$	4	+z	a	(0, 0, 18.2)
$(\frac{3}{8}, \frac{1}{2}, \frac{1}{4})$	1	-x	a	(18.2, 0, 0)
$(\frac{1}{4}, \frac{3}{8}, \frac{1}{2})$	1	-y	a	(0, 18.2, 0)
$(\frac{1}{2}, \frac{1}{4}, \frac{3}{8})$	0	-z	a	(0, 0, 18.2)

also be described by their components at the 24 sublattices and represented by a vector  $\vec{F}$  in the 72-dimensional space. Since the magnetic fields depend linearly on the components of the spins, one may write  $\vec{F} = -\underline{A}\vec{S}$ , where  $\underline{A}$  is a  $72 \times 72$  matrix. The energy of the configuration per magnetic unit cell is  $-\frac{1}{2}\vec{S} \cdot \vec{F} = \frac{1}{2}\vec{S} \cdot \underline{A} \cdot \vec{S}$ , a quadratic form in the components of  $\vec{S}$ . This is to be minimized subject to the constraint  $s_{xr}^2 + s_{yr}^2 + s_{zr}^2 = 1$ ;  $r = 1, \dots, 24$ , where  $s_{xr}, s_{yr}, s_{zr}$  are the components of a representative spin of the  $r$ th sublattice; this set of constraints is known as the *strong constraint*, which also implies the *weak constraint*

$$\sum_{i=1}^{72} S_i^2 = 24.$$

Now the minimum value of  $\frac{1}{2}\vec{S} \cdot \underline{A} \cdot \vec{S}$ , subject to the weak constraint, is the lowest eigenvalue of  $\frac{1}{2}\underline{A}$ , and a configuration which corresponds to this value of energy is the corresponding eigenvector. If this eigenvector, or a linear combination of eigenvectors corresponding to the same lowest eigenvalue if degenerate, also satisfies the strong constraint, then this is the configuration of minimum dipole-dipole interaction energy which is being sought.

The interaction energy between two dipoles  $\vec{\mu}_1$  and  $\vec{\mu}_2$  separated by a distance  $\vec{r}$  is known to be

$$\frac{\vec{\mu}_1 \cdot \vec{\mu}_2}{r^3} - 3 \frac{(\vec{\mu}_1 \cdot \vec{r})(\vec{\mu}_2 \cdot \vec{r})}{r^5}.$$

TABLE II. Lattice sums for dysprosium aluminum garnet. The variables  $r$ ,  $x$ ,  $y$ , and  $z$  are the displacements and its components from an origin situated at an ion situated in the unit cell at  $(\frac{1}{8}, 0, \frac{1}{4})$  to each ion in one of the sublattices. The unit of length is 12.04 Å. The sublattices originate from each of the  $\text{Dy}^{3+}$  ion in the unit cell by the translations  $\vec{T} = l\vec{a} + m\vec{b} + r\vec{c}$ .

Sublattice originating from	$\sum \frac{r^2 - 3x^2}{r^5}$	$\sum \frac{r^2 - 3z^2}{r^5}$	$\sum -\frac{3xy}{r^5}$	$\sum -\frac{3xz}{r^5}$	$\sum -\frac{3yz}{r^5}$
$(\frac{1}{8}, 0, \frac{1}{4})$	0	0	0	0	0
$(\frac{5}{8}, \frac{1}{2}, \frac{3}{4})$	0	0	0	0	0
$(\frac{5}{8}, 0, \frac{1}{4})$	-30.0849	15.0425	0	0	0
$(\frac{3}{8}, 0, \frac{3}{4})$	3.0350	-14.4964	0	0	0
$(\frac{1}{8}, \frac{1}{2}, \frac{3}{4})$	8.6704	-4.3352	0	0	0
$(\frac{3}{4}, \frac{7}{8}, 0)$	-11.9563	1.7258	-4.6903	-9.0518	-3.8114
$(\frac{3}{4}, \frac{3}{8}, 0)$	-1.8748	3.7495	2.7556	-2.4180	2.4180
$(\frac{1}{4}, \frac{1}{8}, 0)$	18.2486	-36.4971	-17.0216	33.6764	33.6764

For anisotropic spins, this may be generalized to  $P_{i\alpha j\beta} s_{i\alpha} s_{j\beta}$ ,<sup>42</sup> where

$$P_{i\alpha j\beta} = \frac{1}{4}\beta^2 \left( \frac{g_{i\epsilon\alpha} g_{j\epsilon\beta}}{r_{ij}^3} - \frac{3g_{i\alpha\lambda} g_{j\beta\mu} r_{ij\lambda} r_{ij\mu}}{r_{ij}^5} \right). \quad (1)$$

In Eq. (1),  $\beta$  is the Bohr magneton,  $s_{i\alpha}$  is the component of the spin  $i$ ,  $g_{i\epsilon\alpha}$  is the  $\epsilon\alpha$  component of its  $g$  tensor, and  $r_{ij\lambda}$  is the  $\lambda$  component of the separation between the ions  $i$  and  $j$ . Greek suffixes run over the indices  $x$ ,  $y$ , and  $z$  and the dummy suffix summation convention applies to repeated greek suffixes. Each row and column of  $\underline{A}$  may be indicated by a latin index which refers to the sublattice and a greek index which denotes the  $x$ ,  $y$ , or  $z$  axis. Then it is easily seen that  $A_{i\alpha j\beta} = \sum_k P_{l\alpha k\beta}$ , where  $k$  represents in turn each of the ions on the sublattice  $j$ , and  $l$  represents one (typical) ion of sublattice  $i$ .

The components of the  $g$  tensor for the various spins are listed in Table I. The elements of  $A$  are proportional to lattice sums. On account of the symmetry of the lattice not all the sums are independent. A set of linearly independent lattice sums is given in Table II; other lattice sums can be derived from these using the positions as given in Table I for the various ions in the unit cell. These sums were evaluated over all relevant lattice points within a sphere of radius 500 Å. Using these lattice sums the  $72 \times 72$  matrix  $\underline{A}$  was generated and diagonalized. It was found that the lowest-energy configuration is antiferromagnetic in three sublattices, as given in Fig. 1, and is

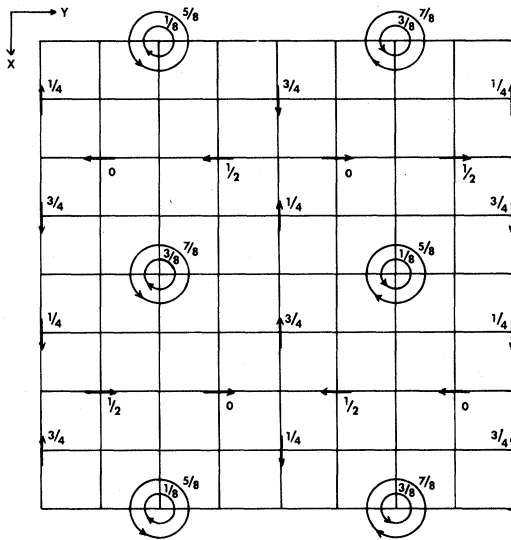


FIG. 1. Spin arrangements in dysprosium aluminum garnet.  $Dy^{3+}$  ions are in special positions 24 (c) of space group  $Ia\bar{3}d$ . The numbers in the figure give the height above the  $z=0$  plane. Spins pointing along the positive  $z$  axis are indicated as current loops in a right-hand sense.

the same as that found experimentally by neutron scattering by Norvell *et al.*<sup>1</sup> The energy of this configuration is  $-1.4075$  K. (It should be noted that the configuration where all  $x$  spins are ferromagnetic in  $x$  direction, all  $y$  spins are ferromagnetic in  $y$  direction, and all  $z$  spins are ferromagnetic in  $z$  direction has the energy  $-0.5940$  K.)

#### B. Presence of external magnetic field

For the presence of a strong external field along the special directions  $[110]$  and  $[001]$  the lowest-energy configurations can again be determined using the Luttinger-Tisza method and the fact that the spins along which there is a nonzero component of external field are effectively rendered paramagnetic and decoupled from the remaining spins.<sup>35</sup> Thus for an external field in the  $[001]$  direction one need consider only the 16 sublattices as generated by the  $x$  and  $y$  spins in the unit cell (Table I), since the  $z$  spins are effectively decoupled out. Similarly for external field in the  $[110]$  direction, one need consider only the eight sublattices as generated by the  $z$  spins in the unit cell (Table I) as the  $x$  and  $y$  spins are effectively decoupled out. The right-hand side of the weak constraint becomes equal to 16 and 8 for the cases of 16 and 8 sublattices, respectively. The  $(48 \times 48)$  and  $(24 \times 24)$   $A$  matrices for the external field direction along  $[001]$  and  $[110]$  axes were constructed and diagonalized. The results are as follows.

$[001]$  direction: The lowest-energy configuration is as given in Fig. 2 and is exactly the same as that pro-

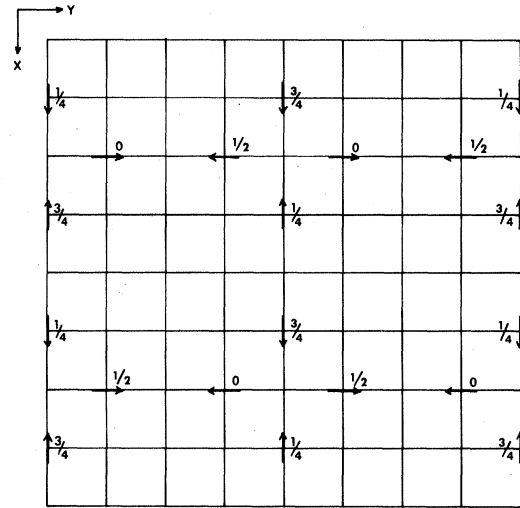


FIG. 2. The lowest-energy configuration of spins for the presence of a large external magnetic field in the  $[001]$  direction. For notation, positions, etc., see the caption of Fig. 1.

posed by Landau and Keen. The energy of this configuration is  $-0.8034$  K. When the external field is either in the  $[010]$  as in the  $[100]$  direction the lowest-energy configuration has the same energy but the spin directions are correspondingly different. (The energy of the ferromagnetic configuration in which two of  $x$ ,  $y$ , or  $z$  spins are ferromagnetic is  $-0.3980$  K.)

$[110]$  direction: There are two lowest-energy configurations as given in Figs. 3 and 4. Each has the energy  $-0.5729$  K. (The ferromagnetic configuration, taking into account the demagnetization factor of  $-0.0619$  K, has the energy  $-0.1990$  K, and thus does not correspond to the lowest-energy configuration.)

#### IV. EXTENSION OF DAG RESULTS TO THE LOW-TEMPERATURE ORDERING OF TAG AND HAG

As discussed by Gavignet-Tillard *et al.*<sup>43</sup> for both TAG and HAG one can consider  $g_z \neq 0$ ,  $g_x, g_y = 0$  for  $z$  spins (similar  $g$  values for  $x$  and  $y$  spins) even though for these garnets one has the situation of two lowest-lying singlets, rather than of a lowest-lying Kramer's doublet as is true for DAG. Thus there is complete parallelism of TAG and HAG cases to that of DAG insofar as the application of the LT method is concerned especially when the crystal structures of TAG, HAG, and DAG are exactly the same and the unit-cell dimensions " $a$ " are almost the same. Therefore, the low-temperature orderings expected for TAG and HAG should be exactly the same as those for DAG with or without the presence of an external

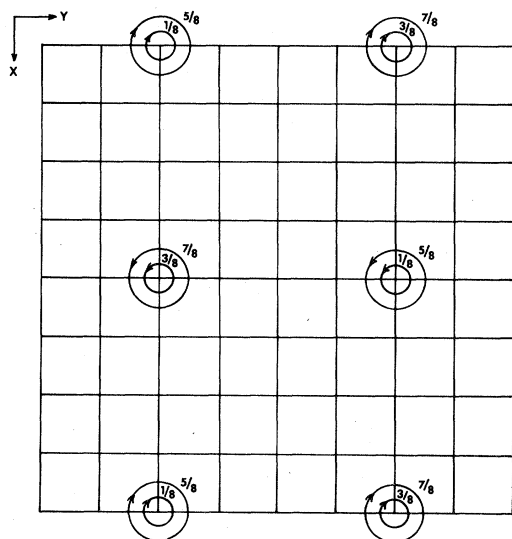


FIG. 3. One of the two lowest-energy configurations of spins for the presence of a large external magnetic field along the  $[110]$  direction. For notations, positions, etc., see the caption of Fig. 1.

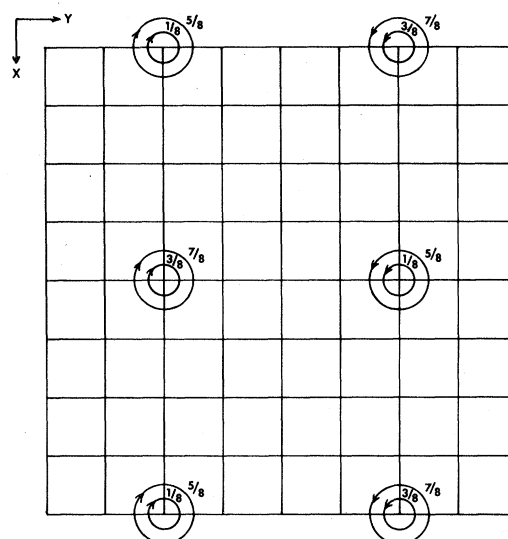


FIG. 4. Another lowest-energy configuration of spins for the presence of a large external magnetic field along the  $[110]$  direction. For notation, positions, etc., see the caption of Fig. 1.

field; these have already been described in Sec. III above.

### V. CONCLUDING REMARKS

The dipole-dipole interactions have here been taken into account rigorously for DAG, TAG, and HAG by the application of the LT method. The resulting predicted low-temperature ordered states in the absence of external field are exactly the same as those found experimentally. When the external field is in the  $[001]$ ,  $[010]$ , or  $[100]$  direction the predicted ordering is in agreement with that proposed by Landau and Keen.<sup>35</sup> As for the direction of external field along  $[110]$ ,  $[101]$ , or  $[011]$  there is no ordering available for comparison, either on the basis of ex-

periment, or on the basis of other theoretical considerations. For these directions, then the predicted orderings of this paper remain to be confirmed by further experimental and/or theoretical work.

As far as the low-temperature ordered states of other garnets are concerned, it is expected that for those garnets for which the  $g$  factors are similar to those for DAG, one should expect the same orderings as predicted in this paper provided their crystal structures are the same as that for DAG.

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