

# Ferrimagnetic Resonance Effects and Miscellaneous

## Resonance Measurements in Magnetic Garnets\*

G. P. RODRIGUE,† H. MEYER,‡ AND R. V. JONES

*Gordon McKay Laboratory, Division of Engineering and Applied Physics, Harvard University*

The effective  $g$  value, anisotropy constants, and line widths of single crystals of YIG, GdIG, SmIG, HoIG, ErIG, and YbIG have been measured over a range of temperatures from 1.5°K to 550°K by ferrimagnetic resonance. The temperature dependence of  $K_1$  in both YIG and GdIG is satisfactorily represented by the model of an individual ion subject to a crystalline field in addition to a Weiss molecular field. The  $g$  values of HoIG and ErIG appear to be in good agreement with the estimations of Kittel et al. for a ferrimagnetic system with one sublattice strongly damped. It has been found that SmIG and YbIG do not follow the predictions of the Kittel model, since different relative strengths of exchange and relaxation interactions seem to be involved.

### INTRODUCTION

THE general group of magnetic compounds exhibiting the garnet structure constitute a valuable test for the various models of ferrimagnetic interactions. Since the valence states and coordination numbers of these ions have been well established,<sup>1</sup> one should be able to make reliable predictions of the energy level schemes associated with each magnetic ion. Due to the many possible chemical substitutions, these isomorphous compounds provide a range of interactions between  $4f$  and  $3d$  magnetic ions in various combinations.<sup>2</sup> To gain insight into the interactions involved in the iron-garnet series (i.e.,  $5\text{Fe}_2\text{O}_3 \cdot 3\text{M}_2\text{O}_3$  usually labelled MIG) the ferrimagnetic absorption of single garnet crystals has been measured over a range in temperatures from 1.5° to 550°K. The resonances were observed in a conventional X-band reflection spectrometer.<sup>3</sup> The temperature of the microwave cavity was maintained by an electronic control system which provided, electrically, a heat flow to balance the heat lost to a refrigerant bath. The crystals were grown by the lead oxide flux method<sup>4</sup> and ground into highly polished spheres.<sup>5</sup> The results of preliminary measurements have been reported earlier.<sup>6</sup> A summary of more extensive measurements on yttrium-, gadolinium-, samarium-, holmium-, erbium-, and ytterbium-iron garnet are presented here.

If it is assumed that the resonance behavior of a

multisublattice system such as the garnets can be described by the ferromagnetic torque equation, then for various principal axes of the crystal the magnetic field required for resonance will be given in terms of the effective anisotropy constants,  $K_1$  and  $K_2$ , and the effective  $g$  value.<sup>7</sup> Wangness<sup>8</sup> has demonstrated the conditions under which this assumption is valid and has interpreted these effective resonance parameters in terms of the magnetic properties of the individual sublattices. For the case of two sublattices,  $A$  and  $B$ , and negligible damping, the effective resonance parameters are given by

$$g_{\text{eff}} = g_A g_B (M_A - M_B) / (g_B M_A - g_A M_B),$$
$$(K_1)_{\text{eff}} = (K_1)_A + (K_1)_B, \quad (1)$$

where the  $M$ 's specify the magnitudes of sublattice magnetizations. However, Kittel<sup>9</sup> has pointed out that for certain rare earth-iron garnets the damping of the rare earth sublattice is not negligible. In accounting for the preliminary resonance data on ErIG and HoIG, he found that although the rare earth ions contribute to the magnetization, and presumably to the free energy, the rare earth contributions to the gyroscopic characteristics of the magnetic system are damped out.<sup>10</sup> Under the assumption of large damping of the rare earth ions, the effective parameters become

$$g_{\text{eff}} = g_A (M_A - M_B) / M_A, \quad (K_1)_{\text{eff}} = (K_1)_A + (K_1)_B. \quad (2)$$

### YTTRIUM-IRON GARNET

The magnetization of the simplest iron garnet, YIG, has been interpreted by Pauthenet<sup>11</sup> in terms of a net

<sup>7</sup> C. Kittel, Phys. Rev. 73, 155 (1948); L. R. Bickford, Technical Rept. No. 23, ONR Contract N5 ori-07801, Laboratory of Insulation Research, M.I.T. (1949); J. F. Dillon, Jr., S. Geschwind, and V. Jaccarino, Phys. Rev. 100, 750 (1955).

<sup>8</sup> R. K. Wangness, Phys. Rev. 91, 1085 (1953); 93, 68 (1954); 95, 339 (1954); Am. J. Phys. 24, 60 (1956).

<sup>9</sup> C. Kittel, Phys. Rev. 115, 1587 (1959).

<sup>10</sup> It should be noted that Wangness had previously derived the appropriate expressions for the effective parameters of a ferrimagnetic system with damping, Phys. Rev. 111, 813 (1958).

<sup>11</sup> R. Pauthenet, Ann. Phys. 13, Series 1, 424 (1958).

\* Research jointly sponsored by an Air Force Cambridge Research Center contract and a National Security Agency contract.

† Present address: Sperry Microwave Electronics Division, Tampa, Florida.

‡ Present address: Department of Physics, Duke University.

<sup>1</sup> F. Bertaut and F. Forrat, Compt. Rend. 242, 382 (1956); M. L. Keith and R. Roy, Am. Mineralogist 39, 1 (1954); H. S. Yoder and M. L. Keith, Am. Mineralogist 36, 519 (1951).

<sup>2</sup> M. A. Gilleo and S. Geller, Phys. Rev. 110, 73 (1958).

<sup>3</sup> J. O. Artman and P. E. Tannenwald, J. Appl. Phys. 26, 1124 (1955).

<sup>4</sup> J. W. Nielsen and E. F. Dearborn, J. Phys. Chem. Solids 5, 202 (1958).

<sup>5</sup> W. L. Bond, Rev. Sci. Instr. 22, 344 (1951).

<sup>6</sup> R. V. Jones, G. P. Rodrigue, and W. P. Wolf, J. Appl. Phys. 29, 434 (1958).

antiferromagnetic coupling of the ferric ions on the 24  $d$  sites with those on the 16a sites. Dillon has reported extensive measurements on the resonance behavior of YIG.<sup>12-14</sup> His work has dramatized the importance of small amounts of rare earth impurities in producing "anomalously" large line widths and anisotropies. The emphasis of the YIG work presented here has been directed towards the understanding of the "normal" or nonimpurity dependent contributions to the anisotropy energy. These are the contributions associated with the ferric ions in  $S$  states.

Several authors have suggested that the magneto-crystalline anisotropy of  $S$  state ions may be associated with the splitting of these states<sup>15-17</sup> by the local crystal-line field of the ion site. The spin Hamiltonian appropriate to this problem has been given by Bleaney and Trenam<sup>18</sup> in terms of the cubic field constant " $a$ " and the second and fourth order trigonal field constants  $D$  and  $F$ . If we assume that exchange forces which act on the magnetic ion are presented by a Weiss molecular field, and use the energy levels derived from the spin Hamiltonian, the partition function and, hence, the free energy of the system can be evaluated. Yosida and Tachiki,<sup>17</sup> and independently, Wolf<sup>16</sup> have carried out such a calculation and have evaluated the sublattice anisotropy energy in terms of functions of the sublattice magnetization. These functions,  $r(T)$  and  $t(T)$ , have been conveniently tabulated by Wolf<sup>16</sup> and the first order cubic anisotropy constant can be expressed as

$$K_1(T) = N[(a + (7/12)\gamma F)r(T) + (D^2/kT)\gamma t(T)], \quad (3)$$

where  $N$  is the number of magnetic ions per unit volume. The axial field contributions to the cubic anisotropy come about through the averaging of the local distortions over all of the nonequivalent sites of the cubic structure ( $\cos^4\theta = \gamma(l^2m^2 + m^2n^2 + n^2l^2)$ , where  $\theta$  and  $(l, m, n)$  denote the orientation of the magnetization relative to respectively the trigonal and cubic axes).

The experimental variation of  $K_1$  with temperature is presented in Fig. 1. The  $K_2$  term makes a negligible contribution to the anisotropy energy of YIG. The data for  $K_1$  does not show the rapid rise below 100°K, which was observed in the early work of Dillon,<sup>12</sup> and which must be attributed to rare earth impurities. The fitted curve in Fig. 1 represents a sum of appropriately weighted  $r(T)$  functions for the two types of ferric sites. The sublattice magnetizations given by Pauthenet<sup>11</sup> have been used in determining the  $r(T)$ 's. The value of the weighting factor leads to an estimate of the constant

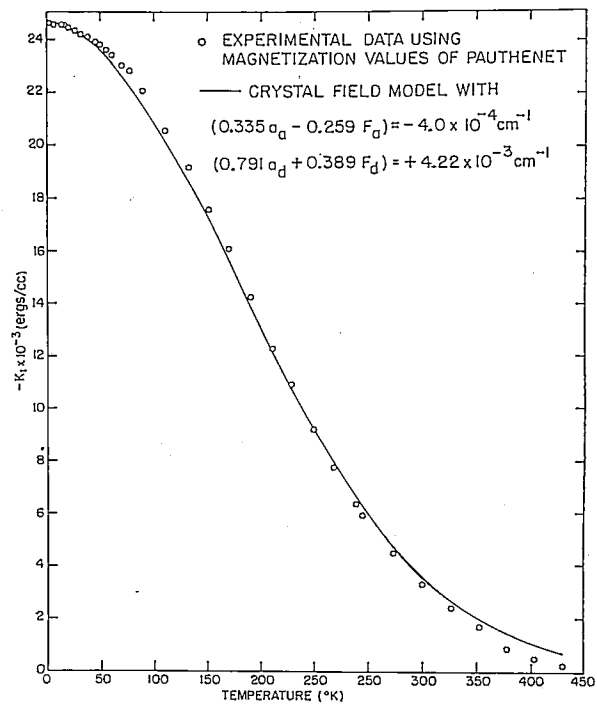


FIG. 1. Temperature variation of  $K_1$  in YIG. Cubic crystal-line terms have been used in the temperature fit with the indicated parameters.

$(a + 7\gamma F/12)$ . However, in the garnets, as pointed out by Wolf<sup>16</sup> and Cooper,<sup>19</sup> the local cubic anisotropy energy associated with a given site is reduced when averaged over all sites since the local cubic axes are not parallel to the cubic axes of the unit cell. The octahedral  $a$  site is oriented with its three-fold axis along the various  $\langle 111 \rangle$  axes of the unit cell. By using the structural data of Geller and Gilleo,<sup>20</sup> the reduction factor associated with the cubic field of the  $a$  site is  $+0.335$  and for a  $\langle 111 \rangle$  distortion  $\gamma = +4/9$ . The twofold axis of the  $d$  site lies along the various  $\langle 100 \rangle$  axes of the unit cell. One finds for this case, a reduction factor of  $+0.78$  and for a  $\langle 100 \rangle$  distortion  $\gamma = -2/3$ . The weighting factors used in Fig. (1) are presented in Table (I) and are compared with the splitting constants determined by Geschwind<sup>21</sup> from measurements of the paramagnetic resonance of ferric ions in a yttrium-gallium garnet host crystal. From Geschwind's values of the  $D$  constant, one can estimate the second term in Eq. (3) to be about  $1/25$  of the first and, hence, the  $(D^2/k)$  term has been ignored in the temperature fit. If the  $F$  terms are neglected, one would need to invoke a particularly small and negative  $a_d$  to explain the observed data. The fact that the anisotropy can be explained in terms of a positive sign of the  $a$  constants for both the tetrahedral and octahedral sites is particularly satisfying. All reported paramagnetic resonance data<sup>18,21,22</sup> seems to be consistently in agree-

<sup>12</sup> J. F. Dillon, Jr., Phys. Rev. 105, 759 (1957).

<sup>13</sup> J. F. Dillon, Jr., Phys. Rev. 111, 1476 (1958).

<sup>14</sup> J. F. Dillon and J. W. Nielson, Phys. Rev. Letters 3, 30 (1959).

<sup>15</sup> G. T. Rado and V. J. Folen, Bull. Am. Phys. Soc. Ser. II, 1, 132 (1956).

<sup>16</sup> W. P. Wolf, Phys. Rev. 108, 1152 (1957).

<sup>17</sup> K. Yosida and M. Tachiki, Progr. Theoret. Phys. 17, 331 (1957).

<sup>18</sup> B. Bleaney and R. S. Trenam, Proc. Roy. Soc. (London) A223, 1 (1954).

<sup>19</sup> B. R. Cooper, Phys. Rev. 112, 395 (1959).

<sup>20</sup> S. Geller and M. A. Gilleo, J. Phys. and Chem. Solids 3, 30 (1957).

<sup>21</sup> S. Geschwind, Phys. Rev. Letters 3, 208 (1959).

<sup>22</sup> G. D. Watkins, Phys. Rev. 110, 986 (1958).

TABLE I. Yttrium-iron garnet.

| Site | Weighting factor<br>(i.e., coef. of $r(T)$ in Eq. 3)     | $g$    | $a$                                    | $F$<br>(Data of Geschwind <sup>21</sup> ) | $D$                       |
|------|--|--------|--|---|---------------------------|
| $a$  | $0.335a - 0.259F = -4.0 \times 10^{-4} \text{ cm}^{-1}$  | 2.003  | $+1.94 \times 10^{-2} \text{ cm}^{-1}$ | $+0.27 \times 10^{-2} \text{ cm}^{-1}$    | $-0.1356 \text{ cm}^{-1}$ |
| $d$  | $0.791a + 0.389F = +4.22 \times 10^{-3} \text{ cm}^{-1}$ | 2.0047 | $+0.65 \times 10^{-2} \text{ cm}^{-1}$ | $-0.42 \times 10^{-2} \text{ cm}^{-1}$    | $+0.0929 \text{ cm}^{-1}$ |

ment with the Watanabe<sup>23</sup> theory, which predicts a positive constant for all coordinations. However, the reported values required to fit the anisotropy of spinel ferrites have disagreed with this prediction.<sup>15,17,24</sup> It would appear that the  $F$  terms are perhaps important for the interpretation of the spinel data as well.<sup>24a</sup> It is not surprising that the  $a$  and  $F$  values required for the interpretation of the anisotropy of data in YIG should be somewhat different from those found for  $\text{Fe}^{3+}$  in yttrium-gallium garnet since the lattice constant of YIG is 0.8% greater than that of  $\text{YGaG}^2$ . The splitting constants should be sensitive functions of the lattice and oxygen parameters. For example, Kaminow has found that the anisotropy of YIG increases by 7% in a 0.5% volume change.<sup>25,26</sup> One may conclude that the normal anisotropy behavior of YIG can be well ac-

counted for in terms of the model of an individual ferric ion interacting with its local crystalline field and a Weiss molecular field.

It is seen from Fig. 6 that the effective  $g$  value for YIG is essentially independent of temperature. It varies only from  $2.006 \pm 0.002$  at 480°K to  $2.001 \pm 0.002$  at 1.5°K. These values are in good agreement with Geschwind's<sup>21</sup> results which are quoted in Table (I). Equation (1) combined with the sublattice magnetization data of Pauthenet<sup>11</sup> and the  $g$  values of Geschwind predicts that there should be a slight decrease in  $g_{\text{eff}}$  at low temperatures.

The highly polished samples used in the YIG experiments exhibited what might be termed "normal"<sup>14</sup> anisotropic behavior in the line width. The "normal" behavior, which has been observed for most of the ferrites, gives the broadest line width when the magnetic field is parallel to an easy axis and the narrowest when it is parallel to a hard axis. The temperature variation of the principal line widths is given in Fig. 2. The peak in this curve at 50°K has been studied extensively by the workers at Bell Telephone Laboratories.<sup>14,27</sup> Kittel

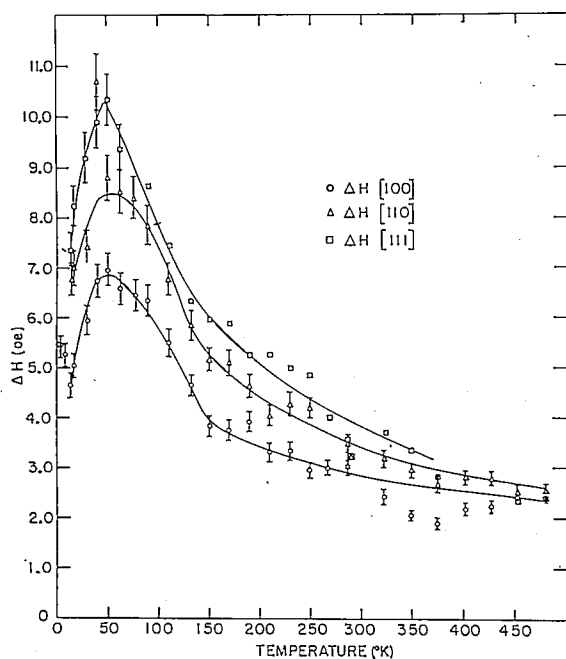


FIG. 2. Temperature variation of  $\Delta H$  in highly polished YIG crystals.

<sup>23</sup> H. Watanabe, Progr. Theoret. Phys. (Kyoto) 18, 405 (1957).

<sup>24</sup> V. J. Folen and G. T. Rado, J. Appl. Phys. 29, 438 (1958).

<sup>24a</sup> Note added in proof. V. J. Folen, J. Appl. Phys. 31, 166S (1960), notes this same possibility.

<sup>25</sup> I. Kaminow, W. Paul, and R. V. Jones, Bull. Am. Phys. Soc. Ser. II, 4, 177 (1959).

<sup>26</sup> The values quoted in this reference are in error and should be corrected to:  $-(K_1/M) = 44.7 \text{ oe} + (3.2 \times 10^{-4} \text{ oe/kg cm}^{-2})P$ .  $4\pi M = 1776 \text{ gauss} + (1.7 \times 10^{-3} \text{ gauss/kg cm}^{-2})P$ .  $K_2$  and  $g$  did not change significantly with pressure.

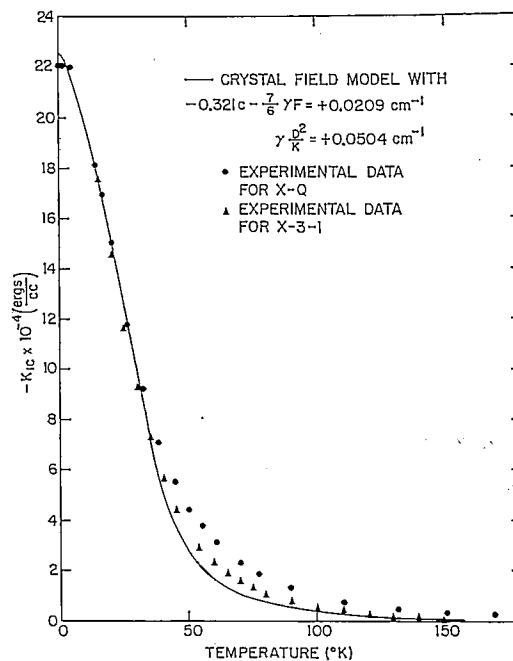


FIG. 3. Temperature variation of  $K_1$  in GdIG. Cubic and axial crystalline field terms have been used in the temperature fit.

<sup>27</sup> E. G. Spencer, R. C. LeCraw, and A. M. Clogston, Phys. Rev. Letters 3, 32 (1959).

TABLE II. Gadolinium-iron garnet.

| GdIG   |                           | Gd <sup>3+</sup> in CaF <sub>2</sub>                         |                                 |  |
|--|---------------------------|--|---------------------------------|--|
| Weighting factors ( <i>c</i> site)                             | $g_c$                     | $a$  | $g$                             |  |
| $\frac{7}{12} \gamma F = -4.18 \times 10^{-2} \text{ cm}^{-1}$ | $1.994 \pm 0.005$ (480°K) | $9.25 \times 10^{-3} \text{ cm}^{-1}$ (Low <sup>30</sup> )   | $1.991$ (Low <sup>30</sup> )    |  |
| $\frac{\gamma D^2}{k} = +5.04 \times 10^{-2} \text{ cm}^{-1}$  | $1.999 \pm 0.005$ (4.2°K) | $9.35 \times 10^{-3} \text{ cm}^{-1}$ (Ryter <sup>31</sup> ) | $1.9918$ (Ryter <sup>31</sup> ) |  |

and coworkers<sup>28</sup> have proposed that this maximum occurs at the point where the Weiss field acting on a given rare earth impurity becomes comparable with the damping forces on this impurity. Above the peak the  $\Delta H$  varies directly as the relaxation time,  $\tau$ , of the impurity and below inversely with  $\tau$ . The maximum value of 7 oe, would indicate an impurity content of the order of 0.1%. Such an impurity level would not affect the considerations of cubic anisotropy given above. However, such impurities did in several cases lead to the "giant anomalies" in the anisotropy energy as observed by Dillon<sup>18</sup> and discussed by Kittel.<sup>29</sup>

#### GADOLINIUM-IRON GARNET

The magnetization of GdIG has been attributed to the weak coupling of the rare earth ions on the 24 *c* sites with the ferric ions.<sup>11</sup> The problem is usually treated by assuming the coupling of the *a* and *d* sites is so strong that one may treat the iron sublattices as unaffected by the presence of the rare earth ions. Since the gadolinium ion is in a <sup>8</sup>S state, one may assume that it is not strongly damped,<sup>30,31</sup> and hence the early interpretation by Wangness<sup>8</sup> of ferrimagnetic resonance may be assumed to hold. The anisotropy constants and the *g* value associated with the *c* sublattice may be obtained from Eq. (1) by using the known values for the *a-d* iron sublattices determined from the YIG measurements.

A subtraction of the first order anisotropy constant of YIG from that of GdIG yields the contributions to  $K_1$  from the rare earth sublattice. The experimental variation of this ( $K_1$ )<sub>0</sub> is plotted in Fig. 3 where the magnetization data of Pauthenet<sup>11</sup> have been used. Also the fact that Gd<sup>3+</sup> is in an *S* state enables one to apply the crystalline field model of anisotropy discussed in connection with Eq. (3).<sup>15-17</sup> In comparison with YIG, one would expect that the *D* term would be more important for GdIG even if the constant has the same value in both cases. At  $T=0^\circ\text{K}$ , this term varies as  $D^2/g\beta H_{\text{eff}}$  where  $H_{\text{eff}}$  is the Weiss molecular field at the

given ion. The Weiss field at the ferric ions is of the order of ten times that at the gadolinium ions and therefore the *D* contribution to the anisotropy of the gadolinium sublattice cannot be ignored. The experimental points in Fig. 3 have been fitted by a weighted combination of the  $r(T)$  and  $t(T)$  functions from Wolf's paper<sup>16</sup> and magnetization data taken from Pauthenet.<sup>11</sup> The *c* site has three two-fold axes which are permuted in orientation along respectively two  $\langle 110 \rangle$  axes and one  $\langle 100 \rangle$  axis of the unit cell. Since the local cubic axes are not aligned with the unit cell axes, the local contributions to the cubic anisotropy will be again reduced.<sup>16,19</sup> In this case, the reduction factor will be  $-0.321$ . The appropriate direction of the distortion axis of the *c* site is not clear to the authors. The weighting factors required for Fig. 3 are presented in Table (II) and are compared with typical *a* constants taken from paramagnetic resonance data on Gd<sup>3+</sup> in a calcium fluoride cubic host crystal.<sup>30-32</sup> A positive sign for the *a* constant of Gd<sup>3+</sup> in GdIG would not be unreasonable. However, the noncubic terms for this case become quite important.

The temperature variation of the  $g_{\text{eff}}$  of two GdIG samples is shown in Fig. 4. On referring to Eq. (1),

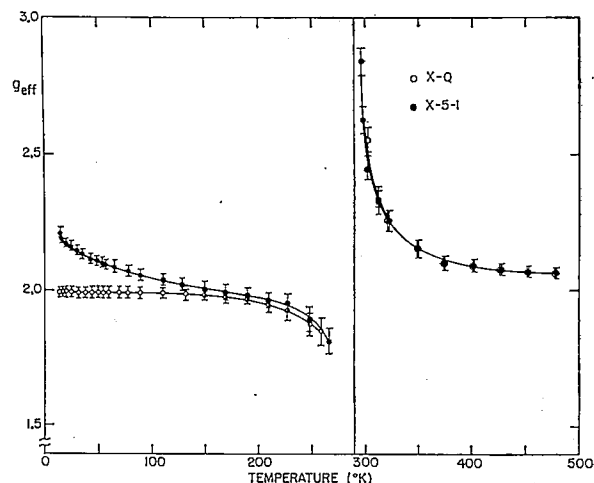


FIG. 4. Effective *g* factors as a function of temperature in GdIG.

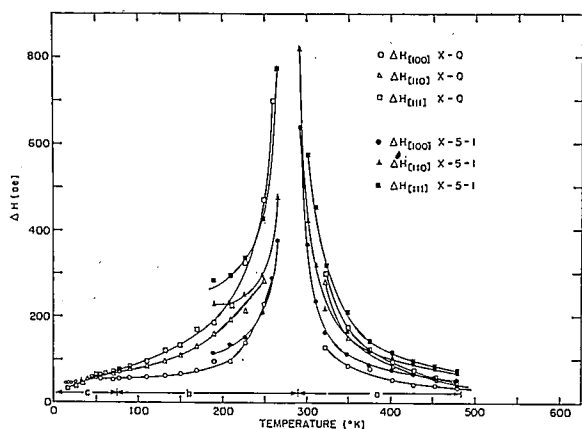
<sup>28</sup> P. G. deGennes, C. Kittel, and A. M. Portis, Phys. Rev. 116, 323 (1959).

<sup>29</sup> C. Kittel, Phys. Rev. Letters 3, 169 (1959); also Phys. Rev. (to be published).

<sup>30</sup> W. Low, Phys. Rev. 109, 265 (1958). (The *c* used in this reference is 2*a* of reference 18).

<sup>31</sup> C. Ryter, Helv. Physica Acta 30, 354 (1957).

<sup>32</sup> J. M. Baker, B. Bleaney, and W. Hayes, Proc. Roy. Soc. (London) A247, 141 (1958). (The authors of this reference were unable to interpret the paramagnetic spectrum in terms of a cubic term.)

FIG. 5. Temperature variation of  $\Delta H$  in GdIG.

one sees, as indicated in earlier work,<sup>33-35</sup> that the compensation point for the angular momentum must occur at a temperature higher than that of the magnetization in order to account for the dispersive shape of the curve near the compensation point ( $\sim 295^\circ\text{K}$ ). By using Eq. (1) and magnetization data,<sup>11</sup> one finds that the  $g$  value of the  $c$  sublattice,  $g_c$ , is essentially independent of temperature for the purer crystals. It ranges from  $1.994 \pm 0.005$  at  $480^\circ\text{K}$  to  $1.999 \pm 0.005$  at  $4.2^\circ\text{K}$ , remaining approximately constant through the compensation point. The higher values previously reported by Calhoun and coworkers<sup>35</sup> may probably be attributed to impurities. For comparison, the  $g$  values of  $\text{Gd}^{3+}$  from paramagnetic resonance in calcium fluoride have been given in Table II.<sup>30,31</sup>

The line width of GdIG (Fig. 5) shows the "normal" anisotropy mentioned above and an enormous broadening at the compensation point. This broadening is probably associated with the rapid rise in the anisotropy field  $K_1/M$  and the breakdown of dipolar narrowing near this point.<sup>36</sup> Below the compensation point, the line width decreases monotonically with temperature reaching a value of about 45 oe at  $14^\circ\text{K}$ . One might suspect that the GdIG line width at low temperatures would approach the YIG value of less than 5 oe, since neither the  $\text{Fe}^{3+}$  nor  $\text{Gd}^{3+}$  ion is strongly coupled to the lattice. The large value of  $\Delta H$  near  $T=0^\circ\text{K}$  could possibly be explained in terms of the overlap of two curves of the type observed for the temperature variation of  $\Delta H$  in YIG (Fig. 2). As noted above, the maximum of such a curve occurs at the temperature where the damping force on the impurity becomes comparable with the coupling between the impurity and the major contribution to the magnetization. For GdIG, the  $\text{Gd}^{3+}$  sub-

lattice at low temperatures is the major contributor and the coupling is much less.<sup>11</sup> Hence, the temperature at which the peak occurs decreases. Therefore, the peaks from the  $\text{Gd}^{3+}$  and the  $\text{Fe}^{3+}$  sublattices should wash out to give the fairly flat region  $c$  in Fig. 5. The "giant anisotropy anomalies"<sup>13,29</sup> were also observed at the lowest temperatures.

#### ERBIUM AND HOLMIUM-IRON GARNET

As mentioned earlier Kittel and coworkers<sup>9,28</sup> have constructed a theory of ferrimagnetic resonance for particular garnets in order to account for early data on HoIG, ErIG, and DyIG.<sup>33,6,34</sup> The essence of this theory lies in the observation that at temperatures near  $300^\circ\text{K}$  the rare earth ions, which have a nonvanishing orbital angular momentum will individually be more strongly coupled to the lattice than to the other ferric and rare earth ions. The calculation of resonance under these conditions leads to Eq. (2) for the  $g_{\text{eff}}$  and to an approximate expression for the line width<sup>28</sup>

$$\Delta H/H = H_{\text{ex}}(\gamma\tau). \quad (4)$$

This expression applies above the temperature at which the damping field (represented by  $1/\gamma\tau$ ) and the exchange field (represented by  $H_{\text{ex}}$ ) are comparable.

Figure 6 presents the experimental temperature variations of the  $g_{\text{eff}}$  in both polycrystal and single crystal samples of HoIG and ErIG. The curves both have a downward slope near room temperature as predicted by Eq. (2) and the magnetization data.<sup>11</sup> In fact, one would always expect a positive slope of the  $g_{\text{eff}}$  curve near room temperature since there all measured rare earth garnets have  $(M_A - M_B)/M_A$  increasing with temperature. The dotted curve indicates the shape of the variation predicted by Eq. (2). The 5% discrepancy in the results could be easily attributed to errors in the magnetization values. The "damped  $c$  sublattice" model<sup>9,28</sup> seems to give a rather good account of the behavior of ErIG and HoIG  $g$  values.

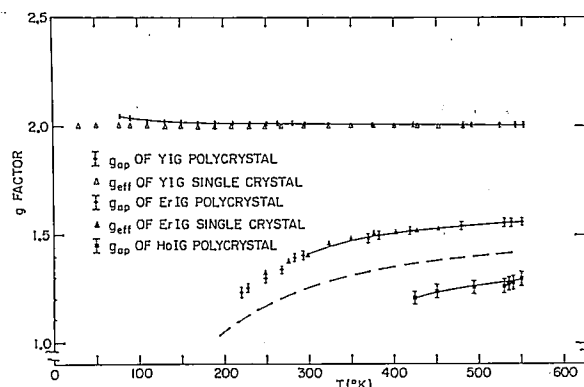


FIG. 6. Effective  $g$  factors as a function of temperature in YIG, ErIG, and HoIG. Upper curve YIG. Middle two curves ErIG. (Dashed curve theoretical. See text.) Lower curve HoIG.

<sup>33</sup> G. P. Rodrigue, J. E. Pippin, W. P. Wolf, and C. L. Hogan, *Inst. Radio Engrs. Trans. PGMTT* 6, 83 (1958).

<sup>34</sup> B. Dreyfus, *Compt. Rend.* 241, 552 (1955).

<sup>35</sup> B. A. Calhoun, W. V. Smith, and J. Overmeyer, *J. Appl. Phys.* 29, 427 (1958).

<sup>36</sup> A. M. Clogston, *J. Appl. Phys.* 29, 3, 334 (1958); S. Geschwind and A. M. Clogston, *Phys. Rev.* 108, 49 (1957); E. Schlömann, *J. Phys. Chem. Solids* 6, 242 (1958).

The temperature variation of the  $K_1$  values of ErIG are presented in Fig. 7. It was found that the second order term  $K_2$  is generally negligible over the temperature range considered. It would be difficult to make any definite statement about the interpretation of the rapid temperature rise of  $K_1$  until the distribution of the energy levels of the  $I_{15/2}$  state of  $\text{Er}^{3+}$  has been made clear. This distribution will be primarily dependent on the magnitude of the zero field splitting of the  $\text{Er}^{3+}$  manifold by the crystalline field of the  $c$  site. Although White and Andelin,<sup>37</sup> and independently, Ayant and Thomas<sup>38</sup> have to some extent clarified the level disposition, there still remain several undetermined parameters in the calculations. Therefore, an evaluation of the system's free energy in terms of these energy levels would not seem meaningful at present.

The dependence of  $\Delta H$  on  $T$  for ErIG and HoIG has been plotted in Fig. 8 and Table III lists the approximate slopes of  $\log \Delta H$  vs  $\log T$ . For both ErIG and HoIG the line width seems to be, roughly, inversely proportional to  $T^2$ . deGennes, Kittel, and Portis<sup>28</sup> have assumed that  $H_{ex}$  varies as  $(1/T)$  for these garnets. From Eq. (4), our experimental results would imply a  $(1/T)$  dependence for  $\tau$  at temperatures near 300°K.

#### SAMARIUM- AND YTTERBIUM-IRON GARNET

The magnetization of both YbIG and SmIG exhibit unusual characteristics. The temperature variation and absolute magnitude of the magnetic moment of SmIG is virtually identical with that of YIG.<sup>11</sup> The negligible contribution of  $\text{Sm}^{3+}$  ions to the magnetization of SmIG is probably attributable to the small effective moment of the  $\text{Sm}^{3+}$  ions in the ground state.<sup>39</sup> A calculation of the magnetization of YbIG below  $T=80^\circ\text{K}$  based on Pauthenet's data<sup>11</sup> reveals particularly large exchange coupling constants for  $\text{Yb}^{3+}$ . This calculation yields  $n'=29$  ( $c$ - $c$  coupling) and  $n=76$  ( $ad$ - $c$  coupling) compared to much smaller values calculated by Pauthenet for other rare earths (see Fig. 8 in reference 11). It seems clear from these considerations that the assumed sublattice dynamics of the Kittel model<sup>9</sup> may not be appropriate to SmIG and YbIG, since the exchange frequency may no longer be very much less than  $1/\tau$ .

TABLE III. Approximate slopes of linear portions of  $\log \Delta H$  vs  $\log T$  curves.

| Garnet        | YIG          | GdIG          | YbIG | SmIG | HoIG | ErIG |
|---------------|--------------|---------------|------|------|------|------|
|               | (above 50°K) | (below 300°K) |      |      |      |      |
| 100 direction | -0.84        | +4.8          | -1.8 | -1.3 |      | -1.8 |
| 111 direction | -0.87        |               | -4.0 |      |      | -2.1 |
| Polycrystal   |              |               | -2.0 |      | -2.1 | -2.0 |

<sup>37</sup> R. L. White and J. P. Andelin, Phys. Rev. 115, 1435 (1959).

<sup>38</sup> Y. Ayant and J. Thomas Compt. Rend. 248, 1955 (1959).

<sup>39</sup> J. H. Van Vleck, *The Theory of Electric and Magnetic Susceptibilities* (Oxford University Press, New York, 1932).

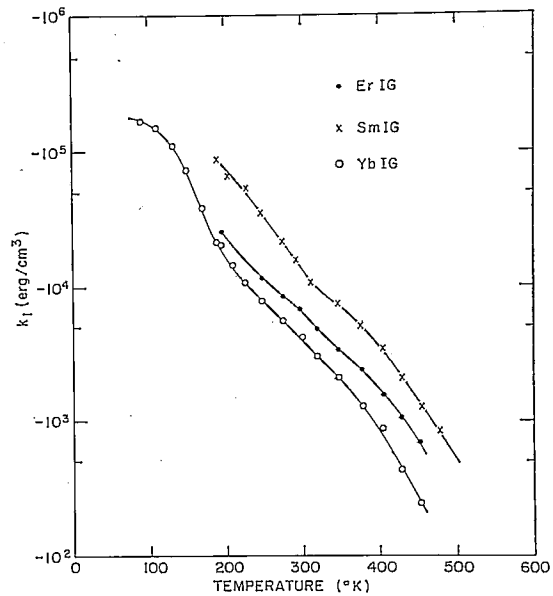


FIG. 7. Temperature variation of  $K_1$  in YbIG, ErIG, and SmIG.

The temperature variation of the  $g_{\text{eff}}$  for these garnets, as shown in Fig. (9), does not particularly follow the prediction of Eq. (2). Both materials show a slight decrease in  $g_{\text{eff}}$  with decreasing temperature and indicates some of the Kittel interaction.<sup>9</sup> However, the magnitude of the decrease for YbIG is by no means large enough. The rapid rise at approximately 200°K does not fit the behavior of Eq. (2) at all. The rise for YbIG could be explained by using Eq. (1) and assuming that the magnetic compensation point, which is at approximately  $T=0^\circ\text{K}$  for YbIG, lies below the angular momentum compensation point. If one considers the splitting of the  $\text{Yb}^{3+}$  state in a cubic field<sup>37,40</sup> this assumption is not

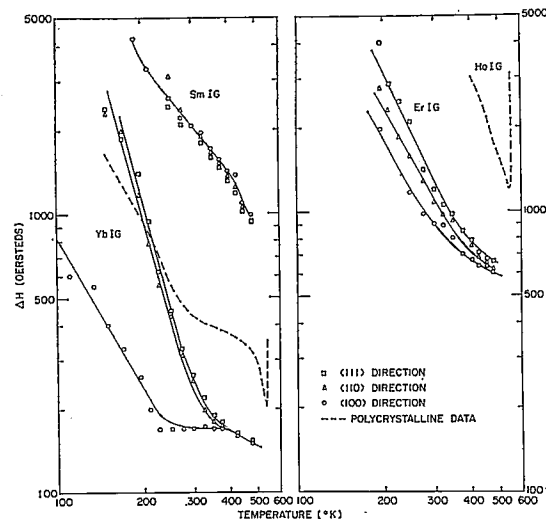


FIG. 8. Temperature variation of  $\Delta H$  in YbIG, SmIG, ErIG, and HoIG.

<sup>40</sup> Y. Ayant and J. Thomas, Compt. Rend. 248, 387 (1959).

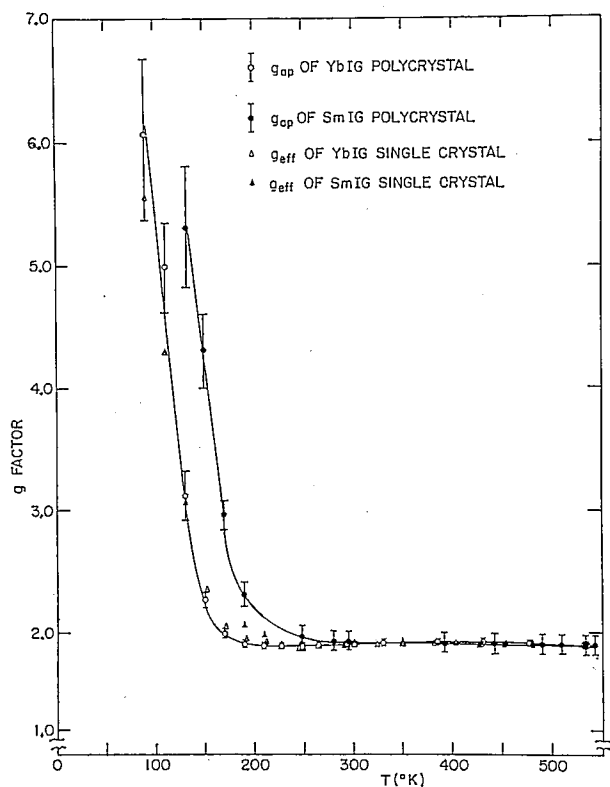


FIG. 9. Effective  $g$  factors as a function of temperature in SmIG and YbIG.

unreasonable since it appears that the angular momentum of  $\text{Yb}^{3+}$  will approach compensation much more rapidly than will the magnetic moment. Equation (1) indicates that  $g_e$  stays at a fairly constant value of

approximately 3.9 near room temperature and drops to a value of approximately 1.0 at low temperatures. If the lowest two levels of  $\text{Yb}^{3+}$  in a cubic field are the only ones populated at low temperatures then the  $g$  tensor should be isotropic and equal to the Landé  $g$  value of  $8/7$ . Since paramagnetic resonance measurements of  $\text{Yb}^{3+}$  in YGaG yield a fairly isotropic  $g$  value<sup>41</sup> at low temperatures, it seems that one is justified to first approximation in using the results of the cubic field calculations. No explanation for the  $g_{\text{eff}}$  behavior of SmIG is offered.

As in the case of the ErIG anisotropy data presented above, further clarification of the appropriate energy level systems must be available before one could hope to explain the temperature dependence of the  $K_1$ 's of SmIG and YbIG, presented in Fig. 7. Again, the  $K_2$  term did make a significant contribution to the anisotropy over the range of temperatures measured.

From Fig. 8 and Table III it may be noted that the variation in the YbIG line width along the hard and easy axes have considerably different temperature dependencies. For SmIG, there is a striking lack of anisotropy in the line width. There seems to be no definitive interpretation of these observations at the present time.

#### ACKNOWLEDGMENTS

The authors would like to express their appreciation for the interest and participation of Dr. C. L. Hogan and Dr. W. P. Wolf in the early phases of this work. Discussions with Professor N. Bloembergen have been invaluable.

<sup>41</sup> D. Boakes, G. Garton, D. Ryan, and W. P. Wolf, Proc. Phys. Soc. (London) (to be published); J. W. Carson and R. L. White, J. Appl. Phys. 31, 53S (1960).