# Spectroscopic Study of the Ytterbium-Iron Exchange Interaction in Ytterbium Iron Garnet

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(Received January 26, 1961)

The exchange splittings of the ytterbium ground-state doublet and of an excited state ($J=5/2$) doublet have been determined from spectroscopic studies of single-crystalline, ytterbium-iron garnet. The splittings were observed at 77°K as a function of the orientation of the magnetization of the crystal. The splittings exhibit the functional form of the $g$ tensor of a two-level system. The principal values of the splittings (referred to local $x$, $y$, $z$ axes) are, for the ground state doublet, 11.6, 25.7, and 29.9 $cm^{-1}$ and, for the excited state doublet, 15.9, 5.8, and 29.9 $cm^{-1}$.

From the ground state splitting, using $g$ values for ytterbium in yttrium gallium garnet, the approximate principal values of the exchange field (assumed to act on the ytterbium spin magnetic moment only) were found to be 349 000 gauss, 611 000 gauss and 678 000 gauss. The principal values of the effective field (assumed to act on the total magnetic moment) are one fourth as large as those of the exchange field.

On the basis of the ground state exchange splitting, calculations of various macroscopic properties of ytterbium-iron garnet, as a function of orientation of magnetization and of temperature, have been carried out by others. The calculated properties are in generally good agreement with experiment, but the sharp, low-temperature magnetic anisotropy anomalies which have been observed in ytterbium-doped yttrium iron garnet are not explained by the spectroscopic data.

## INTRODUCTION

A PROBLEM of current interest involving the rare earth iron garnets concerns the nature of the iron-rare earth exchange interaction. Measurements of certain macroscopic properties of the garnets (such as magnetization and specific heat)[1] provide relevant information, but because such properties are related to single ion properties through sums or averages over a number of magnetically inequivalent sites, measurements of this sort do not normally yield a detailed picture of the exchange effects experienced by individual rare earth ions. Since the rare earth ions in solids exhibit sharp line spectra and since the iron garnets, in thin section, are sufficiently transparent to allow the observation of certain of the rare earth transitions, spectroscopy offers a means of examining on a single ion basis the iron-rare earth exchange interaction.

Preliminary optical studies[2] have shown that there exist in the absorption spectrum of ytterbium iron garnet readily observed exchange splittings. We have now completed a detailed study of these splittings including their behavior as a function of the orientation of the magnetization. From the results of this study, it has been possible to determine the approximate size and shape of the exchange field seen by the ytterbium ions. The spectroscopic results also enable one to reverse the usual procedure and calculate from the observed ground state exchange splittings many of the macroscopic properties of ytterbium iron garnet.

## BACKGROUND

Except for lutetium (which has no optical spectrum), ytterbium is spectroscopically the simplest rare-earth ion which enters the garnets as a major constituent. Trivalent ytterbium lacks one electron of having a closed $4f$ shell. The energy level scheme consists of a $^2F$ state split by a strong spin-orbit interaction and a weaker crystal field interaction. For the ion in a crystal field of low symmetry (e.g., for ytterbium on a $D_2$ site in the garnets) all orbital degeneracy is removed, but each level remains a Kramer's doublet, degenerate in the absence of magnetic interactions.

Spectroscopic studies of ytterbium in yttrium gallium garnet[3] have shown that the crystal field splittings of both the $J=7/2$ and $J=5/2$ states are relatively large (see Fig. 1). In particular, the ground state appears to be separated from the next nearest level of the $J=7/2$ manifold by more than 500 $cm^{-1}$.

The optical spectrum of ytterbium consists of a group of lines near one micron (approximately 10 000 $cm^{-1}$) in the near infrared. The transition from the ground state to the lowest $J=5/2$ level, falling near 10 300 $cm^{-1}$, is found to be especially sharp and strong in both the gallium[3] and aluminum[2] garnet. In an applied magnetic field, this sharp line splits into a Zeeman pattern from which the splittings of the terminal levels can (in principle) be deduced. Detailed Zeeman studies have not yet been completed.[4] The ground-state splittings of ytterbium in an applied magnetic field have been examined directly, however, by paramagnetic resonance techniques.[5,6]

In the iron garnets, the rare earths see via the surrounding oxygens an exchange field produced by the ferrimagnetically-coupled iron lattice. Preliminary experiments[2] have shown that in ytterbium iron garnet the exchange field splits the sharp ytterbium line

[1] H. Meyer and A. B. Harris, J. Appl. Phys. **31**, 49S (1960).
[2] K. A. Wickersheim and R. L. White, Phys. Rev. Letters **4**, 123 (1960).
[3] R. Pappalardo and D. L. Wood, J. Chem. Phys. **33**, 1734 (1960).
[4] Zeeman experiments on ytterbium in yttrium aluminum garnet and in yttrium gallium garnet are currently being conducted at Johns Hopkins University under the direction of Professor G. H. Dieke.
[5] D. Boakes, G. Garton, D. Ryan, and W. P. Wolf, Proc. Phys. Soc. (London) **74**, 663 (1959).
[6] J. W. Carson and R. L. White, J. Appl. Phys. **31**, 53S (1960).

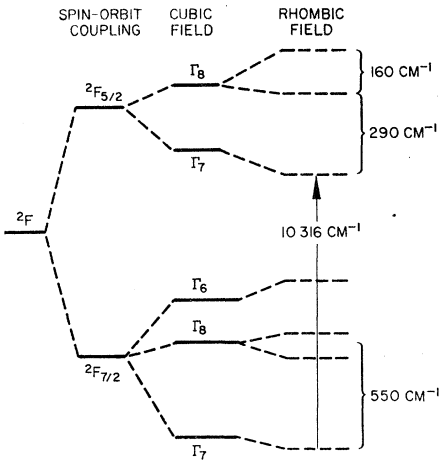


FIG. 1. Energy level scheme for ytterbium in yttrium gallium garnet (based on information contained in reference 3).

described above into an easily resolved pattern of absorption lines. The preliminary experiments also showed that it was possible at liquid nitrogen temperatures to vary the exchange splitting pattern by rotating the magnetization of the crystal (relative to the crystal axes) with an external magnetic field.

The experiments to be described were undertaken primarily for the purpose of determining the exchange splittings of the ytterbium ground-state doublet as a function of the orientation of the exchange field.

## EXPERIMENTAL PROCEDURES

In the experiments reported earlier,[2] spectra were obtained using a large grating spectrograph belonging to the University of California at Los Angeles. Because of the large $f$ number of this instrument and the low sensitivity of photographic emulsions available for use at one micron, exposure times of several hours were often required to obtain a single spectrum. For this reason, only a small amount of data could be obtained under a given set of experimental conditions. Furthermore, the samples of ytterbium iron garnet used in the early experiments were small, contained inclusions, and were not cut parallel to favorable crystal planes. In the optical experiments to be described, a number of improvements were made relative to the initial experiments.

Large single crystals of ytterbium iron garnet were grown from a lead oxide-lead fluoride flux.[7] Samples were prepared by sectioning the crystals parallel to good (110) faces. Clear, polished sections approximately 40 $\mu$-thick and several millimeters across were prepared by petrographic thin-sectioning techniques. The orientation of each finished section was checked by x-ray back reflection techniques. Each sample used was parallel to a (110) plane to within the accuracy of the x-ray measurement (i.e., to within a degree or so). The high-symmetry directions lying in the (110) plane were located to about the same accuracy.

During the spectroscopic studies, the sample was mounted on a holder designed to rotate the sample in its own plane through angles which could be measured to half a degree. The sample holder was constructed to fit into a small glass optical dewar which could in turn be positioned to support the sample in the center of a gap between the pole pieces of a magnet with the magnetic field perpendicular to the light path.

The current experiments were performed with the sample immersed in liquid nitrogen since at this temperature the ytterbium absorption lines are reasonably sharp, and it is still possible to rotate the magnetization with convenient external fields. (At lower temperatures, the crystalline magnetic anisotropy becomes large whereas the magnetization itself becomes quite small.) Another factor influencing the choice of the working temperature is the size of the ground state exchange splitting. Only at temperatures which are sufficiently high can absorptive transitions originating in the upper half of the ground state doublet be observed in the optical spectrum.

Exploratory experiments were performed using a 4-in. Varian magnet to provide fields which could be varied from zero to 8000 gauss. For fields below three or four thousand gauss, no significant Zeeman shifts were observed in the ytterbium spectrum. At liquid nitrogen temperatures, it was found that fields as low as 700 gauss were sufficient to saturate the ytterbium-iron garnet thin sections along any direction in the sample plane. With fields between these limiting values, colinearity of the applied and exchange fields was achieved for all sample orientations without the danger of Zeeman effects modifying the exchange splittings. In the final experiments, a large permanent (magnetron) magnet was used for convenience. Fields of up to 1500 gauss were obtained through the use of suitably shaped pole tips.

Absorption spectra were obtained photographically using the 30 000 line per inch grating of a Bausch & Lomb Dual Grating Spectrograph. The first order reciprocal dispersion in the region of interest was approximately 3.4 A/mm. A ribbon filament projection lamp was used to provide illumination. Hypersensitized I-M spectroscopic plates were used throughout the study. Exposure times of from 5–15 min were generally sufficient for the samples and optical system used.

For purposes of obtaining spectra as a function of exchange field orientation, the spectrograph slit height was reduced, and the sample was rotated relative to the saturating magnetic field in small, uniform steps. A spectrum was taken at each angular position, the plate being racked an appropriate distance between spectra.

[7] J. W. Nielsen, J. Appl. Phys. **31**, 51S (1960).

## INTERPRETATION OF THE FIELD-FREE SPLITTINGS

Before an interpretation of the angle-dependent spectra can be made, a starting point is required. The spectrum obtained with no external magnetic field provides an excellent reference point since this spectrum is relatively simple and is identical with the spectrum obtained from a sample saturated along a [111] direction. Our initial attempt to interpret the field-free spectrum led to ground state splittings which could not be reconciled (in detail) with specific heat data.[1] A second interpretation, compatible with the specific heat measurements, has now been established as the correct one through information provided by far-infrared studies. Some discussion of the factors leading to the initial interpretation as well as those leading to the final one seems desirable, since out of this discussion can come a better understanding of certain phenomena encountered in spectroscopic studies of the rare earth iron garnets.

In the presence of a magnetic field along a [111] direction, the rare earth ions in a rare earth gallium or rare earth aluminum garnet will be equally distributed on two sets of magnetically inequivalent sites.[8] In ytterbium iron garnet, with the exchange field oriented along a [111] direction (as occurs either when the crystal is saturated along a [111] or when there is no applied field, since the [111] is the easy direction) the same sort of site inequivalence exists. Each ytterbium transition originates and terminates on doublet levels which are split by the exchange interaction. Thus, for each electronic absorption line seen in the spectrum of ytterbium in the gallium or aluminum garnet, there should be seen in the field-free spectrum of ytterbium iron garnet two overlapping groups of lines consisting of four components each.

Figure 2 shows a portion of the field-free spectrum of ytterbium iron garnet obtained during the earliest experiments. The group of lines shown falls near 10 300 $cm^{-1}$ and consists of transitions originating in the ground-state doublet and terminating in the lowest of the $J=5/2$ doublets. In interpreting this pattern, we noted the differences in width of the various lines. Assuming the width of a line to be related to the width of one (or both) of the terminal levels, we initially grouped the diffuse lines together as belonging to one site and the sharp lines together as belonging to the other site. This grouping, together with liquid helium data and polarization data, led to an interpretation[2] which yielded the ground state and excited state splittings shown above the spectrum in Fig. 2. Although the average ground-state splitting could be fitted to specific heat measurements, the two markedly inequivalent splittings individually could not.[1]

[8] The site inequivalences can be visualized if a model of the garnet structure is examined. They can also be seen directly in the paramagnetic resonance data obtained from ytterbium in yttrium aluminum and yttrium gallium garnet.[5,6]

In our more recent spectra, all lines appear equally sharp, indicating that the original linewidth differences were spurious. The elimination of the false linewidth clue made possible a second interpretation of the spectrum which led to the splittings shown below the pattern in Fig. 2. This interpretation is clearly in better agreement with the specific heat data.

The question of interpretation was settled unambiguously by far infrared data. In experiments performed at the University of California, Tinkham and Sievers[9] observed at 1.5°K, in ytterbium iron garnet two absorption bands at 23.4 and 26.4 $cm^{-1}$, respectively. Using the second interpretation of the optical spectrum, we have deduced from our recent (best) data at 77°K ground-state splittings of 22.1 and 25.3 $cm^{-1}$. In addition to establishing the second interpretation of the optical spectrum as the correct one, the far infrared data indicates that the ground state exchange splittings are slightly temperature sensitive between 1.5° and 77°K.[10]

There still exists the question of the differences in linewidth seen in the early optical spectra. We now believe that our early samples were more strained than our recent samples and that as a result the direction of the magnetization deviated about the [111] in the early samples. If we associate the width of a given transition not with the width of the individual terminal levels at a fixed orientation of the magnetization but rather with the joint behavior of the two levels as the orientation of the magnetization changes, we can explain the linewidths observed initially. In the next section, we will describe the behavior of the ground state and excited state levels. For certain transitions, both terminal levels shift in the same direction as the magnetization rotates. These transitions appear relatively sharp even in strained samples. For other transitions, the terminal levels diverge or converge as the magnetization rotates, and these appear broad in samples where the magnetization wanders about the [111] direction. The effect is indicated schematically in the lower half of Fig. 2.

## VARIATION OF EXCHANGE SPLITTING WITH EXCHANGE FIELD ORIENTATION

Figure 3 shows the behavior of the ytterbium spectrum as a function of the orientation of a (110) thin section relative to a saturating magnetic field. The fine, straight lines on the left side of the figure were caused by water vapor absorption in the light path. The diffuse absorption behind the water vapor lines belongs to the ytterbium spectrum, but the lines are too poorly defined to allow much interpretation.

[9] M. Tinkham and A. J. Sievers, III (private communication).

[10] The systematic 5% difference between the two sets of data would appear to be significant, since the joint error in the two experiments should be less than 1 $cm^{-1}$. The decrease in $M_{Fe}$ with increasing temperature would account for a 1 or 2% difference in the two sets of data. The remaining difference, if real, could be the result of a reduction of overlap with increase in lattice constant.

The exchange-split components of the 10 300 $cm^{-1}$ ytterbium line form the interesting pattern on the right side of the spectrum. This pattern is shown enlarged in the lower half of the figure. Indicated are the directions of the magnetization (and the exchange field) as determined by orientation of the sample relative to the applied field.

Using the correct interpretation of the field free or [111] spectrum as a starting point, we have unraveled the rotational pattern to obtain the exchange splittings of both the ground state and excited state doublets as a function of exchange field orientation. The deduced splittings are shown in Fig. 4. The scatter in the data can be attributed largely to the difficulty of making good frequency measurements on the absorption lines. Some lines fade for certain orientations, and a number of lines are incompletely resolved. Since the deduced splittings are obtained by taking frequency differences, measurement errors may be enhanced.

There is a distinct topological similarity between the ground state exchange splittings and the applied field splittings observed in paramagnetic resonance studies of ytterbium in the gallium and aluminum garnet.[5,6] This fact prompted us to make the trial assumption that the exchange splittings of a given doublet might

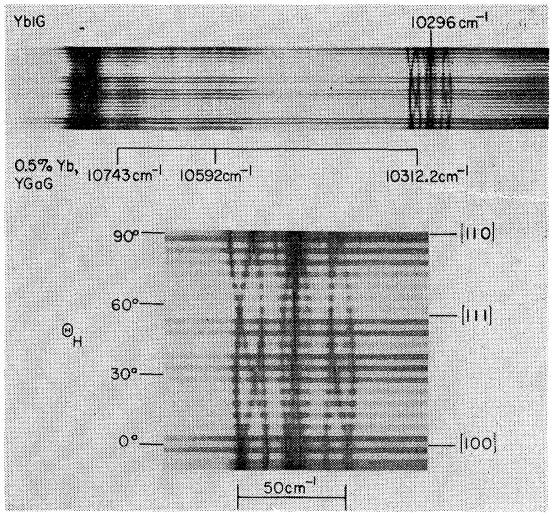


FIG. 3. Positive print made from a spectrographic plate taken using as a sample a thin (110) section of ytterbium iron garnet held at 77°K. The sample was rotated stepwise in its own plane relative to a saturating (1500 gauss) magnetic field, a spectrum being taken at each angular position. For comparison, the three electronic transitions of ytterbium (0.5%) in yttrium gallium garnet obtained at 77°K are plotted beside the ytterbium iron garnet spectrum.

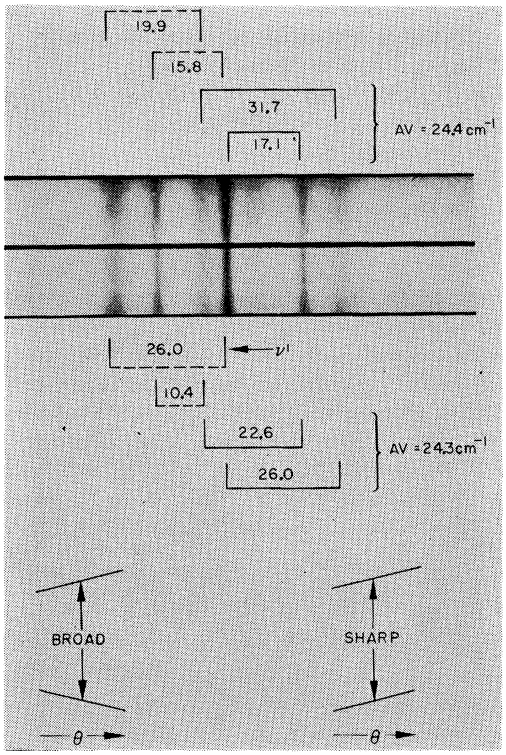


FIG. 2. Portion of an early spectrum of ytterbium iron garnet obtained photographically at 77°K with no applied magnetic field. Two possible interpretations of the pattern are shown, with the ground state splittings indicated by the solid brackets. In the lower half of the figure is shown a linewidth controlling mechanism thought to be operating in the sample from which this spectrum was obtained.

behave in the same general way as the $g$ tensor of a two-level system.[11] That is, we assumed that the splittings have the form

$$\Delta E=[(\Delta E)_x^2 l^2+(\Delta E)_y^2 m^2+(\Delta E)_z^2 n^2]^{\frac{1}{2}},$$

where $\Delta E_x$, $\Delta E_y$, and $\Delta E_z$ are principal values and $l$, $m$, and $n$ are the direction cosines of the exchange field relative to the principal axes of the system.

For the problem at hand, namely for the ytterbium ion in the garnet with the exchange field constrained to a (110) plane, there occur at most four inequivalent sites, the exchange splittings of a doublet being given by

$$(\Delta E)_1=[(\Delta E)_x^2\cos^2\Theta+(\Delta E)_y^2\sin^2\Theta]^{\frac{1}{2}},$$

$$(\Delta E)_2=[(\Delta E)_x^2\cos^2\Theta+(\Delta E)_z^2\sin^2\Theta]^{\frac{1}{2}},$$

$$(\Delta E)_3=\Bigg[(\Delta E)_x^2\frac{\sin^2\Theta}{2}+(\Delta E)_y^2\left(\frac{\cos^2\Theta}{2}+\frac{\sin^2\Theta}{4}-\frac{\sin\Theta\cos\Theta}{\sqrt{2}}\right)+(\Delta E)_z^2\left(\frac{\cos^2\Theta}{2}+\frac{\sin^2\Theta}{4}+\frac{\sin\Theta\cos\Theta}{\sqrt{2}}\right)\Bigg]^{\frac{1}{2}},$$

[11] See for example, K. D. Bowers and J. Owen, *Reports on Progress in Physics* (The Physical Society, London, England, 1955), Vol. 18, p. 316.

TABLE I. Principal values of the exchange splittings obtained from the spectrum of $Yb^{3+}$ in ytterbium iron garnet and of the effective and exchange fields deduced using the $g$ values for $Yb^{3+}$ in yttrium gallium garnet.

| State | Principal value | $\Delta\nu'=\Delta E/hc$ ($cm^{-1}$) | $g'$ | $H_{eff}=\Delta E/\beta g'$ (gauss) | $H_{ex}=[g_J/2(g_J-1)]H_{eff}$ (gauss) |
|---|---|---|---|---|---|
| Ground state (lowest $J=7/2$ doublet) | $x$ | 11.6 | 2.85 | 87 200 | 349 000 |
| | $y$ | 25.7 | 3.60 | 153 000 | 611 000 |
| | $z$ | 29.9 | 3.78 | 169 000 | 678 000 |
| Excited state (lowest $J=5/2$ doublet) | $x$ | 15.9 | ... | ... | ... |
| | $y$ | 5.8 | ... | ... | ... |
| | $z$ | 29.9 | ... | ... | ... |

$$(\Delta E)_4=\Big[(\Delta E)_x^2\frac{\sin^2\Theta}{2}+(\Delta E)_y^2\Big(\frac{\cos^2\Theta}{2}+\frac{\sin^2\Theta}{4}+\frac{\sin\Theta\cos\Theta}{\sqrt{2}}\Big)+(\Delta E)_z^2\Big(\frac{\cos^2\Theta}{2}+\frac{\sin^2\Theta}{4}-\frac{\sin\Theta\cos\Theta}{\sqrt{2}}\Big)\Big]^{\frac{1}{2}},$$

where $\Theta$ is measured from the [100] direction in the (110) plane. Similar relationships have been used in calculating the paramagnetic resonance curves for the ground state of ytterbium in yttrium gallium and yttrium aluminum garnet.[12] Just as in the paramagnetic resonance studies, the principal values can be taken directly from data obtained in the (110) plane.

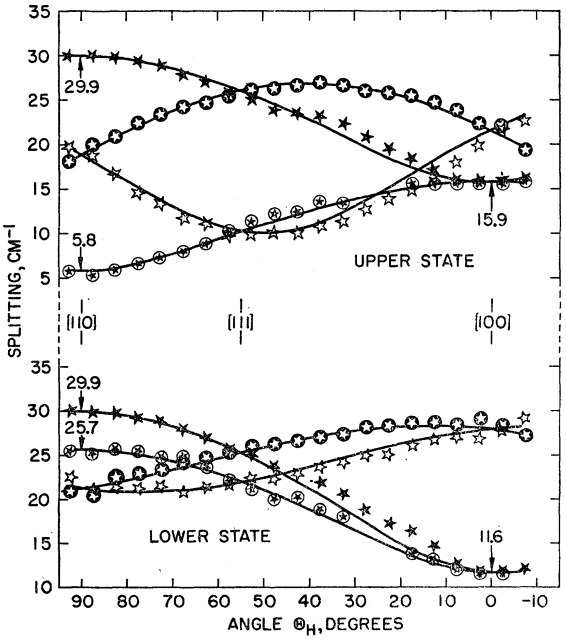


FIG. 4. Exchange splittings of the ytterbium ground state doublet and of the lowest of the $J=5/2$ doublets plotted as a function of the orientation of the exchange field in the (110) plane. (The splittings were deduced from the spectrum shown in Fig. 3.)

[12] J. W. Carson (private communication).

The principal values used in calculating the exchange splittings are listed (in $cm^{-1}$) in Table I and shown in Fig. 4. The calculated splittings are indicated by the smooth curves in Fig. 4. Although there is some scatter in the data, the general agreement of the observed and calculated curve is sufficiently good that we are willing to accept the calculated curves as a true picture of the azimuthal behavior of the exchange splittings.

## CALCULATION OF THE EXCHANGE FIELD

We have assumed that the ytterbium exchange splittings can be written in the same functional form (with the same principal axes) as the $g$ tensor of a rare earth doublet in the garnets and have demonstrated empirically that this assumption is valid. We would now like to argue that this result is to be expected even if the exchange field is anisotropic, provided only that the principal axes of the exchange field coincide with those of the $g$ tensor (which is to be expected on symmetry grounds).

We can write for the exchange interaction term in the Hamiltonian of the system

$$\mathcal{H}=-\boldsymbol{\mu}\cdot\mathbf{H}_{eff},$$

where $\boldsymbol{\mu}=\beta\mathbf{g}'\cdot\mathbf{S}'$ is the total magnetic moment of the ion ($\mathbf{g}'$ being the paramagnetic resonance $g$ tensor for the doublet in question and $\mathbf{S}'$ being an effective spin of 1/2) and where $H_{eff}=-\lambda\cdot\mathbf{M}_{Fe}$ ($\mathbf{H}_{eff}$ being an anisotropic effective field[13] written in terms of the net magnetization of the iron lattice, $\mathbf{M}_{Fe}$, and an anisotropic Weiss constant, $\lambda$).

Starting with the above relationship, it is easily shown that the resultant splitting of the doublet is given by

$$\Delta E=\beta M_{Fe}[(g_x'\lambda_x l)^2+(g_y'\lambda_y m)^2+(g_z'\lambda_z n)^2]^{\frac{1}{2}}=[(\Delta E)_x^2 l^2+(\Delta E)_y^2 m^2+(\Delta E)_z^2 n^2]^{\frac{1}{2}}.$$

This result, aside from verifying our earlier assumption, shows that it is possible, given the appropriate $g$ tensor, to deduce from the exchange splittings the

[13] The relationship between the effective field (assumed to act on the total magnetic moment of the ion) and the exchange field (acting on spin only) is discussed by W. P. Wolf and J. H. Van Vleck, Phys. Rev. 118, 1490 (1960). Although the exchange field has the greater physical significance, the derivation is presented here in terms of the effective field because of the direct analogy in this form with the ordinary Zeeman effect.

principal values of the effective field itself. There is some question about what $g$ tensor to use since there are no paramagnetic resonance data on ytterbium in ytterbium iron garnet. If we use the ground state $g$ values obtained for ytterbium in yttrium gallium garnet, we are probably making a small error, yet this is the best approximation available.[14]

The principal values of the effective field have been calculated from the $g$ values of ytterbium in yttrium gallium garnet. The exchange field is simply related to the effective field through the relation[13]

$$H_{ex}=[g_J/2(g_J-1)]H_{eff}.$$

For the ground state of ytterbium, the Landé $g$ factor $g_J$ is equal to 8/7, so that the exchange field is four times as large as the effective field. The principal values of both the exchange field and the effective field are given in Table I.

We have almost no data, other than the exchange splittings, for the excited ($J=5/2$) doublet of ytterbium. It is hoped that the Zeeman studies mentioned earlier will provide for this state the equivalent of the paramagnetic resonance data now available for the ground state. Given such information, we can determine whether, for example, the ion in the excited state sees the same exchange field as the ion in the ground state.

Until information about other states, and possibly other ions, is available, it would seem premature to speculate in detail about the form of the exchange field. It is worth noting, however, both the large size and the marked anisotropy of the exchange field. Wolf[15] has suggested that the iron-rare earth exchange interaction might show an anisotropy over and above that of the $g$ tensor itself, but he speculated that for ytterbium, where the ground state $g$ tensor is not very anisotropic, the exchange field might also be expected to be fairly isotropic. According to our experimental results, the exchange field is considerably more anisotropic than the $g$ tensor.

## CALCULATION OF MACROSCOPIC PROPERTIES

Wolf[15] has also pointed out that anisotropy in either the $g$ tensor or in the exchange field will lead, in general, to anisotropy in the net rare earth sublattice magnetaiztion and also to anisotropy in the magnetic contribution to the energy of the system. Calculations of a number of macroscopic properties of ytterbium iron garnet have now been carried out.[16] These calculations use a single-ion model and are based on the optically observed ground state exchange splittings. Since the calculations will be described elsewhere, we will note only a few features of the results here.

Most of the properties calculated show significant anisotropy. Insofar as experimental checks are available, the calculated magnetic and thermal properties of ytterbium iron garnet are in good, general agreement with experimental data. Only the small, sharp magnetic anisotropy anomalies observed at very low temperatures by Dillon and Nielsen[17] in ferromagnetic resonance studies of ytterbium-doped yttrium iron garnet (and more recently observed by Teale, Pearson, and Hight[18] in torque measurements of the same materials) do not show up in calculations based on the spectroscopic data.[19]

The last observation may have special significance. The currently accepted explanation for the sharp anisotropy anomalies observed in the rare earth doped iron garnets involves the crossing or near-crossing of rare earth energy levels as a function of exchange field orientation.[20] For the individual ytterbium ions, at least, such crossings or near-crossings of the exchange-split ground state levels do not occur. The next nearest ytterbium levels lie some 500 $cm^{-1}$ above the ground state, and even if near-crossings of these excited levels were to occur, their populations at 1.5°K should be vanishingly small.

## ACKNOWLEDGMENTS

We are indebted to the following persons: R. L. White for providing the initial impetus for this study and for many helpful discussions; M. Tinkham for instructive discussions of the exchange interaction problem and for providing, with A. J. Sievers, III, unpublished far-infrared data; R. A. Lefever for essential support through the growth of high-quality crystals; A. B. Chase for the preparation of the polished thin sections used as samples; and J. W. Torpy and J. W. Reed for assistance with various other phases of sample preparation and orientation.

[14] The paramagnetic resonance studies[5,6] have shown that the $g$ tensor for $Yb^{3+}$ in yttrium gallium garnet does not differ greatly from that for $Yb^{3+}$ in yttrium aluminum garnet even though there is a significant difference in the lattice constants of the two crystals. Since, on the basis of ion size and lattice constant, ytterbium iron garnet is quite similar to yttrium gallium garnet, it seems likely that the $g$ tensor of $Yb^{3+}$ is quite similar in the two crystals.

[15] W. P. Wolf, Proc. Phys. Soc. (London) **74**, 665 (1959).

[16] J. W. Henderson and R. L. White (private communication).

[17] J. F. Dillon, Jr., and J. W. Nielsen, Phys. Rev. **120**, 105 (1960).

[18] R. W. Teale, R. F. Pearson, and M. J. Hight, paper presented at the 6th Annual Conference on Magnetism and Magnetic Materials, New York, November 14–17, 1960.

[19] Both temperature and concentration extrapolations were made in arriving at this conclusion. The anomalies show up only at very low temperatures whereas the majority of the spectroscopic data were obtained at 77°K. However, the general agreement of the field-free splittings obtained optically at 77°K and the far infrared data obtained at 1.5°K indicates that the exchange splittings are not strongly temperature dependent. We have checked the effect of dilution by comparing the total optical splitting observed in ytterbium iron garnet with that observed for 1% ytterbium in yttrium iron garnet. Although there is a decided dependence of the splitting on lattice constant (the total splitting decreasing by about 7% with a lattice constant increase of about 0.7%) the total change is not large enough to alter the conclusion.

[20] C. Kittel, Phys. Rev. **117**, 681 (1960).

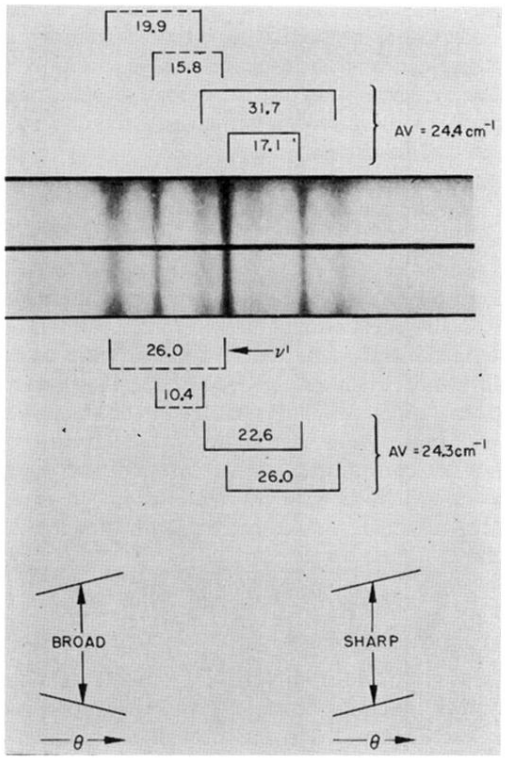


FIG. 2. Portion of an early spectrum of ytterbium iron garnet obtained photographically at 77°K with no applied magnetic field. Two possible interpretations of the pattern are shown, with the ground state splittings indicated by the solid brackets. In the lower half of the figure is shown a linewidth controlling mechanism thought to be operating in the sample from which this spectrum was obtained.

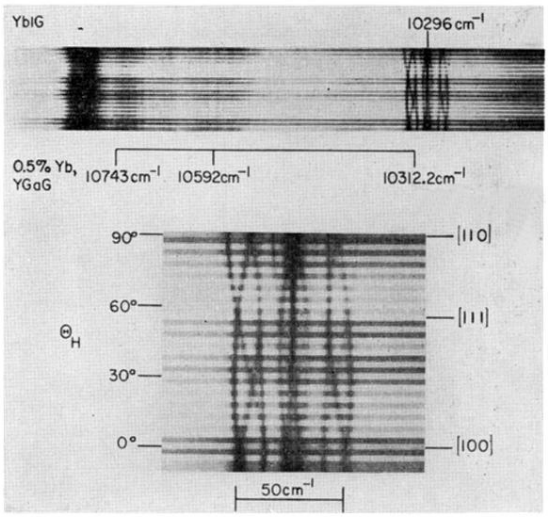


FIG. 3. Positive print made from a spectrographic plate taken using as a sample a thin (110) section of ytterbium iron garnet held at 77°K. The sample was rotated stepwise in its own plane relative to a saturating (1500 gauss) magnetic field, a spectrum being taken at each angular position. For comparison, the three electronic transitions of ytterbium (0.5%) in yttrium gallium garnet obtained at 77°K are plotted beside the ytterbium iron garnet spectrum.