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ANISOTROPY OF EXCHANGE IN YTTERBIUM IRON GARNET

Kenneth A. Wickersheim and Robert L. White
General Telephone and Electronics Laboratories, Inc., Palo Alto, California

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A measurement has been made of the g tensors and of the exchange splittings of the Yb^{3+} ion in ytterbium iron garnet (YbIG) for both the ground-state doublet and for an excited state doublet. From these measurements one can conclude: (1) that the g tensor of Yb^{3+} is substantially the same in the iron garnet as in the gallium garnet[1]; (2) that the anisotropy in the exchange splittings is caused by anisotropy in the Yb^{3+}-Fe^{3+} exchange as well as in the Yb^{3+} g tensor. The purpose of this Letter is to describe the measurements leading to these conclusions, and to present a theory of exchange anisotropy which, considering its simplicity, fits the data remarkably well.

A spectroscopic study of the exchange splittings of Yb^{3+} in YbIG has already been reported in some detail.[2] The optical transition investigated is between the Γ_7 doublet of the ground $J=\frac{7}{2}$ state and the Γ_7 doublet of the excited $J=\frac{5}{2}$ state of the Yb^{3+} ion. These are the lowest-lying doublets of each J manifold. At the time of the original measurements only the g tensor of the $J=\frac{7}{2}$ doublet was known (from paramagnetic resonance in the isomorphic gallium garnet) and the substantial increase in anisotropy of the exchange splittings over the g tensor could have been due either to anisotropy in the exchange fields[3] or a difference in the Yb^{3+} g tensor in the iron garnet as compared to the gallium garnet. To remove this ambiguity and to give also further data on the upper state, a set of experiments was performed in which the effect of a large magnetic field upon the exchange splittings was observed. Fields up to 36 000 Oe were obtained using a PEM 12-in. electromagnet, and the spectra were taken photographically using the 30 000 line/inch grating of a Bausch and Lomb dual-grating spectrograph. From the change in splittings under an applied field, the g tensors for both upper and lower states were deduced. For the $J=\frac{7}{2}$ state the applied field acted to diminish the splittings; for the $J=\frac{5}{2}$ state the applied field increased the splittings.[4] The precision obtained in the g-tensor measurement was limited primarily by the optical linewidths encountered in the iron garnets.

The results of the measurements are given in Table I. Also given are the g values for Yb^{3+} in the gallium and aluminum garnets as determined by paramagnetic resonance and very recent optical Zeeman studies,[5] and both the experimental and theoretical exchange splittings.

In comparing the g tensors and exchange splittings in the iron garnet one finds that the two are markedly different, and in the $J=\frac{5}{2}$ state even topologically dissimilar. For $J=\frac{5}{2}$ the smallest exchange splitting is associated with the largest g value, the intermediate exchange splitting with the smallest g value, and the largest exchange splitting with the intermediate value. For the $J=\frac{7}{2}$ state the order of g values and of exchange splitting is the same, but the degree of anisotropy is much larger in the exchange splittings.

One can explain the above data only by assuming that the Yb^{3+}-Fe^{3+} exchange interaction contributes to the anisotropy, that is, has a magnitude which depends not only on relative spin orientation (as $S_1 \cdot S_2$) but also on the orientation of the spin vectors relative to the crystalline axes. That such an exchange is physically plausible can be seen as follows. The Yb^{3+} $4f$ wave

Table I. Principal values of g tensors and exchange splittings for ytterbium in the garnets.

State	Principal value	g' YbIG[a]	YGaG[b]	YALG[b]	Calculated cubic value	Exchange splitting (cm^{-1}) Experimental[c]	Theoretical
Lowest $^2F_{7/2}$ doublet (Ground state)	x	3.7 ± 0.15	3.78	3.87		30.6 ± 0.2	27.0
	y	3.6 ± 0.15	3.60	3.78		26.1 ± 0.1	26.8
	z	2.8 ± 0.1	2.85	2.48		11.8 ± 0.2	15.0
	(Average)	(3.2 to 3.5)	3.41	3.38	3.43	⋯	⋯
Lowest $^2F_{5/2}$ doublet	x	1.8 ± 0.2	1.98	2.46		5.6 ± 0.2	4.9
	y	1.8 ± 0.2	1.80	1.27		29.8 ± 0.2	31.2
	z	≤ 0.4	0.35	0.49		15.2 ± 0.1	14.1
	(Average)	(1.1 to 1.5)	1.41	1.38	1.43	⋯	⋯

[a] Measured in present experiments.

[b] Ground-state g values by paramagnetic resonance and confirmed by optical Zeeman effect. Excited state g values by optical Zeeman effect only.

[c] Slightly revised as compared to values of reference 2 on the basis of high-field experiments.

functions which are involved are markedly anisotropic, showing well-developed lobes for some states. Further, the $L \cdot S$ interaction is strong, considerably stronger than the crystalline field or exchange energies. Hence the spin component of such a $4f$ wave function will, when rotated, carry with it the orbital wave function, causing the lobes to protrude in altered directions and varying thereby the electronic overlap with neighbors.

The problem of exchange in systems where the $L \cdot S$ coupling is large (so that the wave functions cannot be factored into purely spatial and purely spin components) is perhaps most easily handled through the concept of an exchange potential[6] which is different for spin-up and spin-down configurations. Pursuant to a suggestion by M. Tinkham, we have expanded the exchange potential for the Yb^{3+} in a harmonic series determined by the local site symmetry (D_2) and retained only the two leading (nonisotropic) terms. The resultant exchange Hamiltonian is then

$$H = g\beta[1 + aY_2{}^0 + b(Y_2{}^{+2} + Y_2{}^{-2})]S_1 \cdot \lambda M_{\text{Fe}}, \tag{1}$$

or alternatively

$$H = g\beta H_e\{1 + G_2{}^0[3L_z{}^2 - L(L+1)] + G_2{}^2(L_+{}^2 + L_-{}^2)\}S_1 \cdot S_0, \tag{2}$$

where S_1 is the Yb^{3+} ion spin; S_0 is a unit vector in the direction of the iron magnetization; L_z, L_+, and L_- operate on the orbital components of the Yb^{3+} wave functions only; and other symbols have their usual meaning. The z axis in this calculation was taken to be that local twofold axis which corresponds to the crystallographic S_4 axis.

In a somewhat less formal approach, one can regard the orbital part of expression (2) as a simple scheme for assigning a weighting factor to the orbitals according to whether they do or do not stick out toward their magnetic neighbors or the intermediate oxygen ions.

Wave functions for the relevant $J = \frac{7}{2}$ and $J = \frac{5}{2}$ doublets were obtained (in J, J_z quantization) by fitting the three components of the g tensors, as measured in the gallium garnet. Ambiguities of phase and sign were resolved by requiring that the wave functions go in the cubic limit to the appropriate form. These wave functions were then projected into L, L_z, S, S_z quantization and the exchange splittings calculated using (2). The three adjustable parameters H_e, $G_2{}^0$, and $G_2{}^2$ were then chosen for best fit to the six splittings, yielding

$$H_e = 500\,000 \text{ Oe},$$

$$G_2{}^0 = +0.0797,$$

$$G_2{}^2 = -0.145.$$

The exchange splittings calculated using these parameters are tabulated in Table I. It can be seen that this simple theory gives a remarkable fit to the data. In particular, the correct prediction of the topology of the $J = \frac{5}{2}$ state is noteworthy.

The quantitative success of this admittedly simplified theory in fitting the experimental splittings exceeded the expectations of the authors. Possibly the agreement will degenerate somewhat as the theory is developed in more general and sophisticated form. Nonetheless, the presumption seems strong that many of the complicated exchange processes in the rare earth compounds can be successfully and meaningfully treated, using the exchange potential formalism and viewpoint.

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[1] Though not the same as in the aluminum garnet (see Table I).

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[4] In both instances the exchange field is oppositely directed from the applied field. However, for the $J=\frac{7}{2}$ state the total magnetic moment μ_J, upon which the applied field acts, and the spin component of the magnetic moment μ_S, upon which the exchange field acts, are parallel, while for the $J=\frac{5}{2}$ state they are antiparallel.

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INFRARED ABSORPTION SPECTRUM OF SULFUR-DOPED SILICON

William E. Krag and Herbert J. Zeiger
Lincoln Laboratory,[*] Massachusetts Institute of Technology, Lexington, Massachusetts

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Sulfur, a group VI element, when diffused into silicon, gives rise to two donor-type impurity levels.[1] The positions of these donor levels have been found from photoconductivity and Hall measurements to be located 0.18 eV and 0.37 eV below the conduction band edge for the neutral and singly ionized sulfur atoms, respectively. We have observed the optical absorptions corresponding to excitation from the ground to the excited states of both the neutral and singly ionized impurities.

Since sulfur is a group VI element, substitutional sulfur has two extra electrons to contribute as donor electrons after the four tetrahedral bonds have formed. Neutral S in Si is, therefore, the analog of a helium atom, and S^+ is the analog of He^+. One portion of the neutral S spectrum is due to transitions from the ground state to a set of excited He-like p states; and one portion of the S^+ spectrum is due to transitions from the ground state to a set of He^+-like p states. Additional lines have been observed, which are probably due to transitions from the ground level to many-valley orbital triplet levels for both neutral S and S^+.

The measurements were made at ≈10°K on polished samples about 1 cm thick containing 10^{15}-10^{16} sulfur atoms/cm^3. A Perkin Elmer Model 12-C monochromator, modified for use with a 150 line/mm grating, and an NaCl fore-prism give us a resolution of ≈0.0005 eV. Atmospheric water vapor absorption lines were used for calibration. Table I lists all of the absorption lines seen in any of our samples with

Table I. Measured optical absorptions and their identification.

Energy (in eV)	Identification
Neutral sulfur transitions	
0.1555	$(1)(1) \rightarrow (1)(3)$
0.1602	
0.1624	
0.1750	$(1)(1) \rightarrow (1)(2p_0)$
0.1802	$(1)(1) \rightarrow (1)(2p_\pm)$
0.1809	$(1)(1) \rightarrow (1)(3p_0)$
0.1833	$(1)(1) \rightarrow (1)(3p_\pm)$
Singly ionized sulfur transitions	
0.2197	$(1) \rightarrow (3)$
0.2338	
0.3219	$(1) \rightarrow (2p_0)$
0.3425	$(1) \rightarrow (2p_\pm)$
0.3468	$(1) \rightarrow (3p_0)$
0.3563	$(1) \rightarrow (3p_\pm)$
0.3599	$(1) \rightarrow (4p_0)$