in a related context, that of spin-lattice relaxation). The Simanek-Orbach mechanism requires a noncubic mode of even inversion symmetry. A low-frequency resonance mode of the required symmetry could result from a reduction of the force constant accompanying the impurity; such a reduction is reasonable in that the lattice constant is substantially larger for SrO than for MnO. The impurity ion is stationary in a mode of even symmetry, and consequently there is no longer a distinction between strain and displacement of neighboring ions; thus, Eqs. (16) and (17) of Ref. 1 can be adapted to give a hyperfine temperature dependence of the form

$$A(T)=A_0+(A_1/\omega)\coth(\hbar\omega/2kT). \qquad (2)$$

The form of Eq. (2) is reasonable in that it is linear in T for high temperature ($\hbar\omega \ll kT$) and independent of T for very low temperature ($\hbar\omega \gg kT$). A proper test of this speculation would require additional low-temperature data. In fact, the choice between the alternative phonon spectra could be made only on the basis of a detailed investigation of $A(T)$ at low temperatures. The low-temperature form of Eq. (1) is

$$A(T)=A_0[1-\pi^4CT^4/15], \qquad (3)$$

and that of Eq. (2) is

$$A(T)=A_0+2(A_1/\omega)\exp(-\hbar\omega/kT). \qquad (4)$$

Finally, correction for thermal expansion might cause the SrO results to appear more reasonable although it would be difficult to explain this. In any case there is a clear need now to round out these results with pressure measurements for CaO and SrO in addition to detailed low-temperature measurements.

ACKNOWLEDGMENTS

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Rare-Earth–Iron Exchange Interactions in Europium Iron Garnet

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Recoilless absorption measurements of the 14.4-keV γ rays of Fe^{57} and the 21.6-keV γ rays of Eu^{151} in europium iron-gallium garnets ($Eu_3Ga_xFe_{5-x}O_{12}$) were carried out at 4.2°K for $x=0$, 0.66, 1.28, 1.60, 2.37, and 3.03. The iron concentrations in the octahedral and tetrahedral sites were determined from the Fe^{57} spectra. The results show that about 80% of the Ga ions occupy tetrahedral sites (~85% for $x=0.66$ and ~75% for $x=3.03$). The analysis of the Eu^{151} spectra shows that (88 ± 4)% of the exchange field acting on an Eu^{3+} ion in europium iron garnet is produced by the two nearest iron neighbors in the tetrahedral site, although the Eu-O-Fe angle for these ions is 92°, an angle often considered unfavorable for superexchange interactions. Excellent agreement between theoretical and experimental spectra is obtained assuming that the remaining 12% of the exchange field is produced by the 4 third nearest neighbors in the tetrahedral site.

INTRODUCTION

The main purpose of the present work was to study the rare-earth–iron exchange interactions in rare-earth–iron garnets and specifically the contributions of the various iron neighbors to the exchange field acting on a rare-earth ion in the garnets. The gallium-substituted europium iron garnet ($Eu_3Ga_xFe_{5-x}O_{12}$) system has the following advantages for the study of rare-earth–iron exchange interactions: (1) The magnitude of the magnetic hyperfine field (H_{eff}) acting on an Eu nucleus of an Eu^{3+} ion is very sensitive to the value of the exchange field (H_{exch}) acting on the ion. As explained in the next section, H_{eff} is approximately proportional to H_{exch} in ferrimagnetic compounds of europium. For any other rare-earth ion, the value of H_{eff} at magnetic saturation is almost independent of H_{exch}. (2) Well-resolved lines are obtained in the absorption spectra of the 21.6-keV γ rays of Eu^{151} in ferrimagnetic compounds of europium. (3) The Ga^{3+} ions strongly prefer the tetrahedral sites of the garnet and, therefore, by changing x, the relative concentrations of the iron ions in the tetrahedral and octahedral sites change. The hyperfine spectra are very sensitive to the value of x and from the analysis of the spectra obtained for absorbers with various values of x, it is possible to draw conclusions concerning the contributions of the various neighbors of an Eu^{3+} ion to the exchange field acting on it.

The crystal structure of the garnets has been dis-

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TABLE I. Relations between a rare-earth ion and its first, second, and third iron nearest neighbors.

	Number of Fe neighbors	Site of Fe neighbors	Distance in units of cube edge ~12 Å	Number of O^{2-} common closest neighbors	Fe-O-M angle (deg)
1st Fe neighbors	2	d	$\frac{1}{4}$	2	92
2nd Fe neighbors	4	a	$\frac{1}{4}\sqrt{(5/4)}$	2	103
3rd Fe neighbors	4	d	$\frac{1}{4}\sqrt{(3/2)}$	1	122

cussed extensively by several authors[1-3] and we shall here summarize only those features necessary for the interpretation of our results. The unit cell of the rare-earth iron garnet contains eight formula units of $M_3Fe_5O_{12}$, where M Indicates a rare-earth ion. In each unit cell there are three types of sites which are occupied by metal cations: (1) 24 tetrahedral or d sites occupied by Fe^{3+} ions. (2) 16 octahedral or a sites occupied by Fe^{3+} ions. (3) 24 dodecahedral or c sites occupied by M^{3+} ions. The geometrical relations between a rare-earth ion and its first, second and third iron nearest neighbors are summarized in Table I. The other iron ions are much further away from the rare-earth ion (their distance is equal to or larger than $a\sqrt{(13/4)}$ and (4) their contributions to the exchange fields acting on a rare-earth ion are very probably negligible.

It is generally accepted that the superexchange mechanism (exchange via nominally nonmagnetic ions) is responsible for the ferrimagnetic properties of the garnets and ferrimagnetic insulators. The exchange interaction of two magnetic ions (A) through an oxygen ion between them was first conceived by Kramer[4] and has since been extensively studied.[5] In this interaction, the overlap of the $2p$ electrons (with dumbbell-shaped distribution) of the oxygen ions with the electronic distribution of the magnetic ions is an important feature. The interaction increases with the overlap and accordingly will be greatest for short A-O distances and for A_1-O-A_2 angles near 180°. As seen from Table I, the angle M-O-Fe is largest for the third iron nearest neighbors of a rare-earth ion (122°). Geller and Gilleo[2] therefore assumed that the M-Fe exchange interaction is strong only with the third iron nearest neighbors of a rare-earth ion in the garnets. Very recently, Levy[6] carried out calculations of the rare-earth–iron exchange interactions in the garnets assuming that the M-Fe exchange interaction is axial about the M-O axis. One of the main conclusions of his calculations is that the exchange interaction of the rare-earth ion in the garnet does *not* take place primarily with its two nearest iron neighbors in the tetrahedral sites.

The garnet system $Eu_3Fe_{5-x}Ga_xO_{12}$ was studied by LeCraw *et al.*[7] From spontaneous magnetization and ferrimagnetic-resonance measurements they could derive the sublattice magnetizations M_{Fe} and M_{Eu} at 4.2°K as a function of x. Their results prove that there is a strong preference of Ga^{3+} ions for the tetrahedral sites and that M_{Fe} goes through zero at $x\cong 1.2$ and reverses sign. By decomposing M_{Fe} for each x into its tetrahedral and octahedral components, they found that M_{Eu} is approximately proportional to the tetrahedral component of M_{Fe} and concluded that the c-d exchange interaction is much larger than the c-a exchange interaction. Geller *et al.*[8] and Anderson[9] arrived at the same conclusion from the analyses of the spontaneous magnetizations of various gadolinium garnets.

Hutchings *et al.*[10] have measured the resonance of single Fe^{3+} spins in the octahedral sites of europium gallium garnet. The Fe^{3+} g factor is shifted by the Eu-Fe exchange interaction. From the size of the shift, the magnitude of the exchange interaction between an Eu^{3+} ion and the Fe^{3+} ions at the octahedral sites could be estimated. The results of Hutchings *et al.* show that the contribution of a Fe^{3+} ion in the octahedral sites to the exchange field acting on its nearest Eu neighbors in europium gallium garnet (EuGaG) is about 0.1% of the total exchange field acting on an Eu ion in europium iron garnet (EuIG).

Although the works of LeCraw *et al.*,[7] Geller *et al.*,[8] Anderson,[9] and Hutchings *et al.*[10] indicate that the d-c exchange interaction is much stronger than the a-c exchange interaction, it is impossible to deduce from them the separate contributions of the first and third iron nearest neighbors (which occupy d sites) of a rare-earth ion in the garnets to the exchange field acting on it. The present experimental results prove that about 90% of the exchange field acting on an Eu ion in europium iron garnet (EuIG) is produced by the two nearest irons in the tetrahedral site, in disagreement with the qualitative conjectures of Geller and Gilleo[2] based on superexchange bonding angles and with the calculations of P. M. Levy.[6]

HYPERFINE INTERACTIONS IN Eu^{3+} IONS

The ionic ground state of Eu^{3+} is $^7F_0$, which is diamagnetic. The states $^7F_1$, $^7F_2$, and $^7F_3$ are at energies of 480, 1330, and 2000°K, respectively. The ionic magnetic moment, the magnetic hyperfine field H_{eff}, and the elec-

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tric field gradient q_{eff} are all zero for the $^7F_0$ state. But since the states $^7F_1$ and $^7F_2$ are relatively low, perturbations will mix them into the ground state and may produce nonvanishing magnetic moments and hyperfine interactions. Wolf and Van Vleck[11] succeeded in explaining the temperature dependence of the spontaneous magnetization of EuIG below its Curie temperature by taking into account the mixing of excited states into the ground state of Eu^{3+} by the exchange interaction and also by the thermal population of the excited states. They have shown that in first-order perturbation theory, the magnetic moment of Eu^{3+} in EuIG at 0°K is proportional to the exchange field $[H_{\text{exch}}(0°\text{K})]$ acting on the Eu^{3+} ion. In a similar way Gilat and Nowik[12] have calculated, as a function of temperature, the magnitude of H_{eff} and q_{eff} acting on the Eu nuclei in EuIG. They used first-order perturbation theory for the calculations of H_{eff}. For the calculations of electric-field gradients, second-order admixtures in the ionic eigenstates had to be taken into account, since the first-order perturbation calculations give a value of zero for the gradients. The experimental values[13,14] for H_{eff} acting on the Eu nuclei in EuIG, derived from recoilless absorption measurements of the 21.7-keV γ ray of Eu^{151} are in good agreement with the calculated values. In the approximation used by Gilat and Nowik, H_{eff} (0°K) is proportional to H_{exch} (0°K) and q_{eff} (0°K) is proportional to $H_{\text{exch}}{}^2$ (0°K). However these relations do not hold when the exchange interactions are relatively large. We have therefore carried out a high-order perturbation calculation of the dependence of H_{eff} (0°K) and q_{eff} (0°K) on H_{exch} (0°K) by diagonalizing the Hamiltonian $\mathcal{H}=E_J-2\beta_0(\mathbf{S}\cdot\mathbf{H}_{\text{exch}})$ within the 9 levels of $^7F_0+{}^7F_1+{}^7F_2$. The results obtained in this way for the dependence of H_{eff} (0°K) and q_{eff} (0°K) on H_{exch} (0°K) are shown in Fig. 1. For Eu^{3+} in EuIG, $\beta_0H_{\text{exch}}/k=22°\text{K}$[15] and the deviation from a linear relation between H_{eff} [0°K) and H_{exch} (0°K) for $H_{\text{exch}}\leq 22°\text{K}$ is about 4%. (This is the difference between the value of H_{eff} (0°K) in EuIG obtained by Gilat and Nowik[12] and the value obtained in the present calculations.] Crystal-field interactions have only a small effect on the hyperfine interactions in the Eu^{3+} ions of EuIG as in cubic symmetry the crystal field does not interact with the Eu^{3+} ion in the first excited state ($J=1$) and the point symmetry of the Eu ion site in EuIG is almost cubic. The value obtained experimentally for H_{eff} (0°K) in EuIG was 600±6 kOe.[14] A small part of this field may be due to core polarization. Freeman and Watson estimated the core-polarization contribution in the rare-earth to be 90 000 $\langle S\rangle$ Oe.[16] The value of $\langle S\rangle$ derived from the value of the magnetic moment of Eu^{3+} in EuIG is 0.7 and the calculated value of the contribution of the polarized core to H_{eff} (0°K) is about 60 kOe. This relatively small contribution of the polarized core is expected to be proportional to the magnetic moment of the Eu^{3+} ion and therefore to a good approximation proportional to the value of H_{eff} produced by the $4f$ electrons. It does not, therefore, distort the relation between H_{eff} and H_{exch} given in Fig. 1. [It can only change the scale of the ordinate (H_{eff}) in Fig. 1. This change has a negligible effect on the analysis of the present experimental results from which relative values of H_{exch} are determined.] By the use of Fig. 1 it is possible to derive the value of H_{exch} acting on an Eu ion when the value of H_{eff} acting on the Eu nucleus is known. Thus by the Mössbauer effect it is possible to prove the existence of magnetically inequivalent sites of the Eu^{3+} ions and to determine the values of H_{exch} corresponding to them. In the present case, the magnitude of H_{exch} acting on the Eu^{3+} ions in mixed europium iron-gallium garnet depends on the numbers of iron ions occupying the various neighboring sites. From the study of the hyperfine splittings, the contributions of the various neighboring iron ions to the exchange interactions acting on the Eu^{3+} ions in EuIG can be determined.

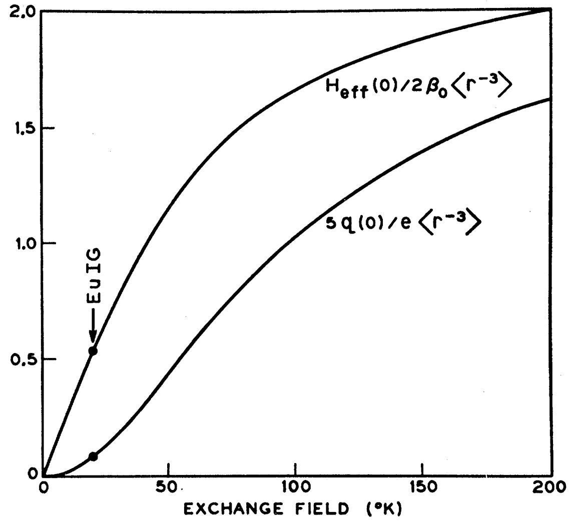

FIG. 1. The effective magnetic field and the electric field gradient acting on the nucleus of Eu^{3+} as a function of H_{exch}.

Experimental Details

The counting rate as a function of relative velocity between source and absorber was measured using an apparatus similar to that described by Cohen *et al.*[17] For the recoilless absorption measurements of the 14.4-keV γ rays of Fe^{57}, a Co^{57} source embedded in Pd was used and the detector was a xenon-methane proportional counter. For the recoilless absorption measurements of

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TABLE II. Summary of results derived from the analysis of the absorption spectra of the 14.4-keV γ rays of Fe^{57}.

x	T (°K)	N_d	N_a	$A=(3-N_d)/x$	$c=N_d/3$	Overall splitting d site (cm/sec)	Overall splitting a site (cm/sec)
0	77	3.00(4)	2.00(4)		1.00	1.525(29)	1.760(20)
0.66	77	2.44(4)	1.90(4)	0.85(6)	0.81(1)	1.505(20)	1.715(20)
1.28	20	1.91(5)	1.81(5)	0.85(4)	0.64(2)	1.490(20)	1.705(20)
1.60	20	1.66(4)	1.74(4)	0.83(3)	0.55(1)	1.470(20)	1.680(20)
2.37	4	1.17(6)	1.46(6)	0.77(3)	0.39(2)	1.440(20)	1.630(20)
3.03	4	0.785(45)	1.185(45)	0.73(2)	0.26(2)	1.390(20)	1.615(20)

the 21.6-keV γ rays of Eu^{151}, a Sm^{151} fission-product source in oxide form was used and the detector was a thin NaI(Tl) scintillation counter with a Be window. The sources were at room temperature in all the measurements. Measurements were carried out for six values of x ($x=0$, 0.66, 1.28, 1.60, 2.37, and 3.03). The samples of europium iron-gallium garnets from which the absorbers were prepared were those on which LeCraw *et al.* have carried out their ferrimagnetic-resonance measurements.[7] The absorbers were in powder form and their density was about 30 mg/cm² in the Fe^{57} absorption measurements and about 90 mg/cm² in the Eu^{151} absorption measurements.

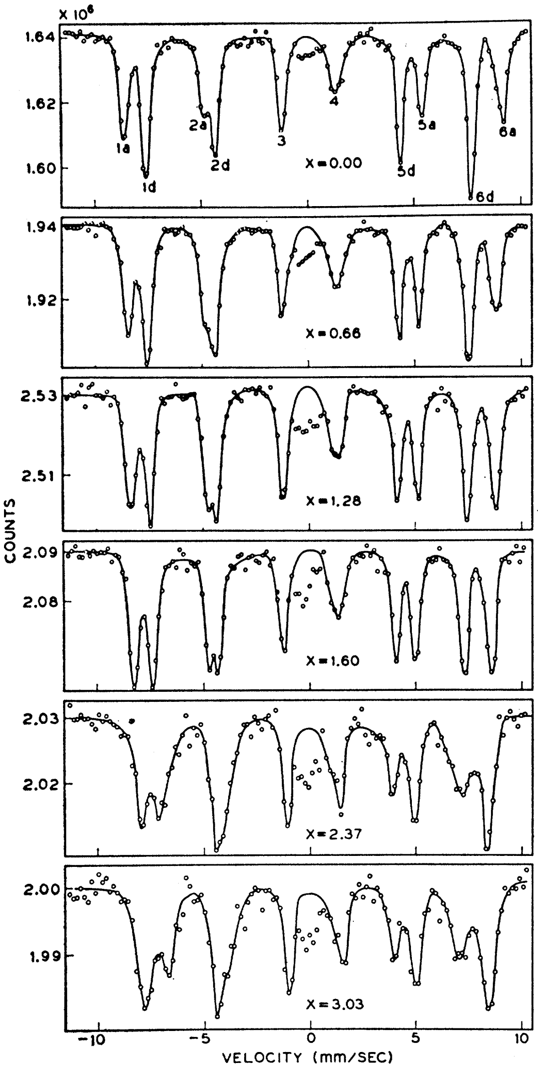

FIG. 2. Mössbauer spectra of Fe^{57} in europium iron-gallium garnets.

Absorption Spectra of the 14.4-keV γ Rays of Fe^{57}

The recoilless absorption spectra of the 14.4-keV γ rays of Fe^{57} in the six absorbers of europium iron-gallium garnet are shown in Fig. 2. The small peak seen in all the six spectra between lines 3 and 4 is due to the presence of a small amount of iron impurity in the beryllium window of the proportional counter, as was proved by carrying out a measurement without an absorber between the source and the window of the detector. The spectra are composed of two sets of six absorption lines, corresponding to the two inequivalent iron sites (a and d sites). Three pairs of lines are quite well resolved (1a-1d, 5a-5d, and 6a-6d) and thus the relative amounts of gallium ions occupying the two sites can be determined as a function of x from the intensities of these lines. The results derived from the analysis of the spectra are summarized in Table II. N_d and N_a denote the number of iron ions per formula unit in the d and a sites, respectively. The ratio (N_d/N_a) was determined from the experimental spectra in Fig. 2. As for each value of x, N_d+N_a should be equal to $5-x$, the values of N_d and N_a could be derived from the present experimental results. It is clearly seen from Fig. 2 that the value of (N_d/N_a) is a decreasing function of x. For $x=0$, $N_d/N_a=1.5$ and N_d becomes equal to N_a at $x=1.4$ approximately. For this value of x, the net magnetization of the iron sublattices is expected to vanish. The existence of such a magnetic compensation point at $x\sim1.3$ was shown previously by LeCraw *et al.* for the europium iron-gallium system by carrying out spontaneous magnetization and ferrimagnetic-resonance measurements[7] and by Lüthi and Henningsen for the yttrium iron-gallium system from spontaneous magnetization measurements.[18] The variation of the value of N_d/N_a with x is a result of the faster filling up of the tetrahedral iron d sites by gallium ions than of the octahedral a sites. The values of $A=(3-N_d)/x$ given in the fifth column of Table II, show that for low

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[18] B. Lüthi and T. Henningsen, *Proceedings of the International Conference on Magnetism, Nottingham, 1964* (Institute of Physics and the Physical Society, London, 1965), p. 668.

values of x ($x \leq 1.66$), about 85% of the Ga ions occupy tetrahedral sites, in agreement with the results obtained by Gilleo and Geller[19] and by LeCraw *et al.*[7] for the europium iron-gallium garnets and by Luthi and Henningsen[18] for the yttrium iron-gallium garnets. For high concentrations of gallium, the values of A are somewhat smaller. For $x=3.03$, about 75% of the Ga ions occupy the tetrahedral sites. The values of $c=N_d/3$ given in the sixth column of Table II measure the concentrations of the Fe ions in the tetrahedral sites. These values are essential for the analysis of the Eu^{151} absorption spectra.

In the two last columns of Table II, the over-all splittings (separation between the 1 and 6 extreme lines) for the two sites are given. The splittings for $x=0$ and $x=0.66$ were also measured at 4°K and were found not to differ by more than 1% from those obtained at 77°K. The splittings for $x=0$ are very close to those obtained previously for dysprosium iron garnet and yttrium iron garnet at 85°K[20] and for gadolinium iron garnet and erbium iron garnet at temperatures below 80°K.[21]

ABSORPTION SPECTRA OF THE 21.6-keV γ RAYS OF Eu^{151}

The absorption spectra of the 21.6-keV γ rays of Eu^{151} in the six absorbers of europium iron-gallium garnet at 4°K, are shown in Fig. 3. The spectrum for $x=0$ is similar to those obtained previously for EuIG[13,14] and other ferrimagnetic compounds of Eu. But the shapes of the spectra for $x \neq 0$ are quite different from the regular spectrum corresponding to a unique value of H_{eff}. It is seen from Fig. 3 that the shape of the spectra depends strongly on the value of x. The peaks at ± 2.3 cm/sec become relatively weaker as the value of x increases. Their intensity is approximately proportional to $[c(x)]^2$ where $c(x)$ is the concentration of the iron ions in the tetrahedral sites (see Table II). Peaks at ± 1.15 cm/sec which are not present in the spectrum corresponding to $x=0$, appear in the spectra of the absorbers with $x \neq 0$. Up to $x=1.60$, their intensity is an increasing function of x and for larger values of x it becomes a decreasing function of x. For large values of x, a peak at zero velocity appears in the absorption spectra. The relative intensity of this peak increases as the value of x increases or the value of $c(x)$ decreases. All these regularities in the relations between the shapes of the absorption spectra and their corresponding values of $c(x)$ indicate that the exchange field acting on the Eu ion in EuIG is mainly produced by the two nearest iron neighbors in the tetrahedral sites. We therefore calculated the absorption spectra assuming that all the exchange interaction acting on the Eu^{3+} ion is produced by the two iron nearest neighbors. Under this assumption, each absorption spectrum for $x \neq 0$ is a superposition of three sub-spectra. The first corresponds to a magnetic hyperfine field identical with that obtained for $x=0$ ($H_{\mathrm{eff}}{}^0$) and represents the Eu ions for which no Ga ions substitutes the two nearest iron neighbors. The statistical weight of this spectrum is $[c(x)]^2$. The second subspectrum corresponds to $H_{\mathrm{eff}}=0.52\ H_{\mathrm{eff}}{}^0$ and represents the Eu^{3+} ions for which a Ga^{3+} ion substitutes one of the two nearest iron neighbors. (For these Eu^{3+} ions the value of H_{exch} is expected to be half of the value of H_{exch} in pure EuIG and H_{eff} is expected, according to Fig. 1, to be equal to $0.52\ H_{\mathrm{eff}}{}^0$.) The statistical weight of this subspectrum is $2c(x)[1-c(x)]$. The third subspectrum corresponds to $H_{\mathrm{eff}}=0$ and represents Eu ions for which the two nearest iron neighbors are substituted by Ga ions. The relative intensity of this sub-spectrum is $[1-c(x)]^2$. The reconstructed spectra are shown in Fig. 4. In the calculation of the theoretical spectra a value of 0.53 was assumed for g_1/g_0, the ratio of the gyromagnetic ratios of the 21.6-keV level and the ground level of Eu^{151}, respectively,[14] and each single line was assumed to have a Lorentzian shape with a width of 3.75 mm/sec at half-height. This width was determined by measuring the absorption spectrum of the $x=3.03$ sample above its Curie temperature.

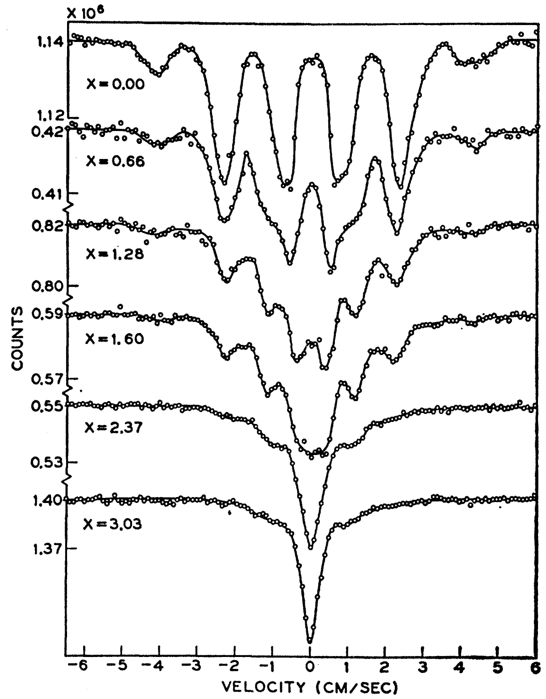

FIG. 3. Mössbauer spectra of Eu^{151} in europium iron-gallium garnets.

A comparison between the experimental spectra in Fig. 3 and the theoretical spectra in Fig. 4 indicates that there is a strong resemblance between them but

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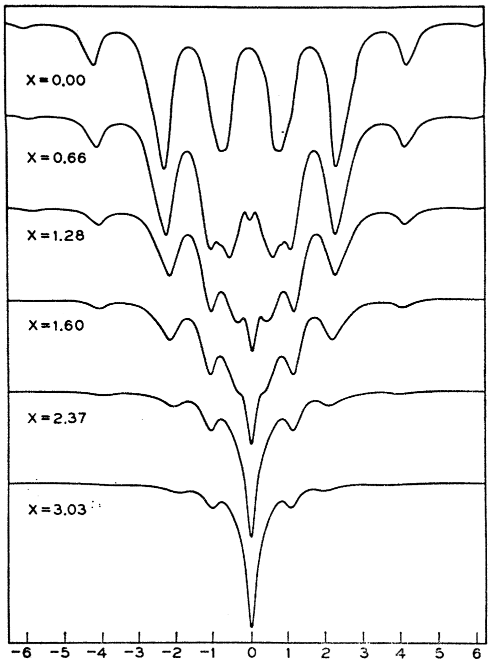

FIG. 4. Theoretical absorption spectra of Eu^{151} in $Eu_3Ga_xFe_{5-x}O_{12}$ assuming that the exchange field acting on an Eu ion is produced only by the two nearest iron neighbors.

that the fit is not perfect. The main differences between the two groups of spectra are: (a) The peak at zero velocity is much more intense in the theoretical spectra for $x=0.66$, 1.28, and 1.60 than in corresponding experimental spectra; (b) The width of the absorption line at zero velocity for $x=2.37$ and $x=3.03$ is considerably larger in the experimental spectra than in the theoretical ones; and (c) The agreement between the experimental and the theoretical spectra for $x=0.66$ at velocity regions around $+1.2$ cm/sec and -1.2 cm/sec is quite poor. Since the ESR measurements of Hutchings *et al.*[10] indicated that the exchange interactions between the Eu^{3+} ions and Fe^{3+} ions in the octahedral sites in europium gallium garnets are very weak, it is natural to try to get better agreement between the present experimental results and theory by assuming that part of the exchange field acting on an Eu ion in EuIG is produced by the four third iron neighbors in the tetrahedral sites. Theoretical spectra were calculated assuming various relative contributions of the first and third iron neighbors to the exchange field. Each reconstructed spectrum is a superposition of 15 subspectra. Each subspectrum characterized by the integers m and n $[S(m,n)]$ where $m=0$, 1, 2 and $n=0.1$, 2, 3, 4 represents the Eu ions for which the numbers of the Fe^{3+} ions occupying the first- and third-nearest sites are m and n, respectively. If H_1 and H_3 are the exchange fields (at an Eu ion) produced by a single Fe^{3+} ion occupying the first- and third-nearest sites, respectively, then the whole exchange field in "state" (m,n) will be given by $H_{\text{exch}}(m,n)=mH_1+nH_3$. To a very good approximation (see Fig. 1) H_{eff} (m,n) can also be expressed in the form $mH_{\text{eff},1}+nH_{\text{eff},3}$. The statistical weight of each subspectrum $S(m,n)$ is given by

$$W(m,n)=\binom{2}{m}[c(x)]^m[1-c(x)]^{2-m}\binom{4}{n}[c(x)]^n \times[1-c(x)]^{4-n},$$

where

$$\binom{2}{m} \text{ and } \binom{4}{n}$$

are binomial coefficients. These reconstructions show that excellent agreement between the theoretical and experimental spectra is obtained assuming that $\beta_0H_1/k=(9.7\pm0.4)°K$ and $\beta_0H_3/k=(0.6\pm0.2)°K$ and that therefore $(88\pm4)\%$ of the exchange field acting on an Eu ion in EuIG is produced by the two nearest iron neighbors and the remaining $(12\pm4)\%$ of the exchange field is produced by the four third iron neighbors and is in the same direction as the exchange field produced by the two iron nearest neighbors. The reconstructed spectra calculated under these assumptions are shown in Fig. 5. The reconstructed spectra which were calculated assuming that the exchange field acting on an Eu ion in EuIG is mainly produced by its second and third iron nearest neighbors do not show any resemblance to the experimental spectra.

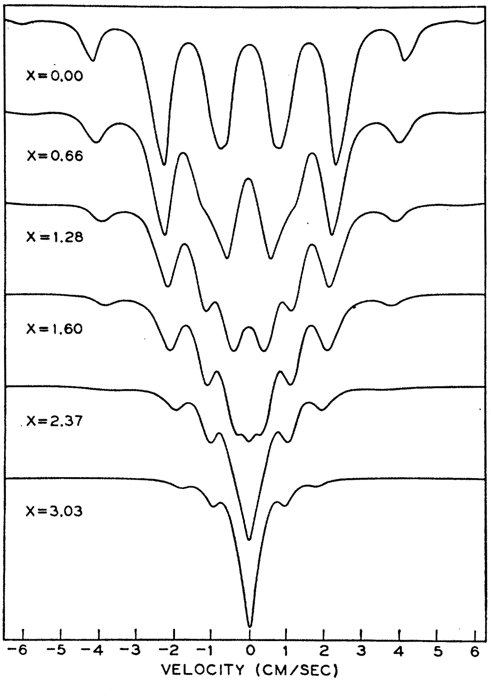

FIG. 5. Theoretical absorption spectra of Eu^{151} in $Eu_3Ga_xFe_{5-x}O_{12}$ assuming that 88% of the exchange field acting on an Eu ion is produced by the two nearest iron neighbors and the remaining 12% is produced by the 4 third iron nearest neighbors.

The small differences between the experimental spectra shown in Fig. 3 and the reconstructed spectra of Fig. 5 are probably produced by the following factors: (1) In the reconstruction of the spectra it was assumed that all the Eu ions in EuIG are magnetically equivalent and that the value of H_{eff} is unique for all of them. According to the calculations of Eicher[22] which include also crystal-field interactions, the values of H_{eff} for the two magnetically inequivalent sites which exist in EuIG when the magnetization is along the [111] direction differ by a few percent. (2) The quadrupole interactions were neglected in the theoretical reconstructions, as they are known to be very small.[12–14] (3) For each sample used, the gallium content x is not unique and there may be a spread of about 10% in the values of x corresponding to different parts of the same sample. In the reconstruction a unique value of x was assumed for each sample.

It should be emphasized that although the present experimental results indicate that about 88% of the exchange field acting on an Eu ion in EuIG is produced by the two nearest iron neighbors, it is impossible to conclude from this alone whether the remaining 12% are produced by the second iron nearest neighbors in the octahedral sites with a ferromagnetic coupling, by the third iron nearest neighbors in the tetrahedral sites or by contributions from both kinds of irons. The ESR measurements of Hutchings *et al.*[10] were carried out on crystals of europium gallium garnet containing Fe^{3+} ions in small concentrations (0.01% to 3%). From these measurements they concluded that the *c-a*, Eu-Fe exchange interaction in europium gallium garnet is very small. But as pointed out by Hutchings *et al.* it is not certain whether the *c-a* exchange interaction is the same for europium gallium garnet containing Fe^{3+} ions in small concentrations and for EuIG. The exchange interaction between any Eu^{3+}-Fe^{3+} pair may depend, to some extent, on the nature of the ions occupying other nearby sites. In the garnets each a site shares a common O^{2-} neighbor with one c site and with one d site ion. In EuIG the d site is occupied by a Fe^{3+} ion, there is a strong *c-d* superexchange interaction through this oxygen and it is possible that this could affect the much weaker interaction between the c and a sites which simultaneously involves the same O^{2-} electrons. But if we assume that in EuIG and europium iron-gallium garnets too, the *c-a* interaction is very small (<1% of the total Eu-Fe interaction), the conclusion from the present measurement is that the four third iron nearest neighbors of an Eu^{3+} ion in EuIG produce about 12% of the exchange field acting on the Eu ion.

CONCLUSIONS

The present measurements show that recoilless-absorption measurements in ferrimagnetic compound of Eu can yield detailed information about the exchange fields acting on the Eu ions in these compounds. The main conclusion from the present work is that about 90% of the exchange field acting on an Eu ion in EuIG is produced by the two nearest iron neighbors in the tetrahedral site, although the Fe-O-Eu angle for these irons is 92°, an angle often considered unfavorable for superexchange.[23] This result has at present no rigorous theoretical explanation and is in disagreement with recent calculations which assume that the M-Fe exchange interaction is axial about the M-O axis.[6]

Note added in proof. Geller *et al.* [Phys. Letters **12**, 495 (1966)] have recently deduced from magnetization measurements that the variation of the Eu^{3+} ion moment in the system $\{Eu_{3-x}Ca_x\}[Fe_2](Fe_{3-x}Si_xO_{12})$ is not a linear function of the d-site Fe^{3+} ion concentration. The deviations from linearity in the region $0 \leq x \leq 1.6$ were about 10%. The present measurements do not prove the existence of such deviations but they are not sensitive enough to rule out the possibility of their existence.

ACKNOWLEDGMENTS

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[22] H. Eicher, Z. Physik **179**, 264 (1964).

[23] See Ref. 5 for a discussion of the dependence of superexchange on the A_1-O-A_2 angle.