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Contributions to the Hyperfine Field in Europium Intermetallics*

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Mössbauer studies of $Eu_xYb_{1-x}Al_2$, $Eu_xYb_{1-x}Cu_2$, $Eu_xLa_{1-x}Al_2$, and $EuAl_4$ at low temperatures and in external fields yield the three major contributions to the hyperfine field. In Eu metal, $EuCu_2$, $EuAl_2$, and $EuAl_4$, these are (i) core polarization $H_{\rm core} \sim -340$ kOe, (ii) central-ion conduction-electron polarization $H_{\rm op}=190\pm20$, 140 ± 20, 105 ± 20, and 50 ± 20 kOe, (iii) neighbor contributions $H_n=-115\pm20$, 10 ± 10, -45 ± 10, 0 ± 10 kOe. The local-ion contribtution $H_{\rm core}+H_{\rm op}$ seems to be correlated to the isomer shift. The field of Eu^{2+} in $YbAl_2$, Eu^{2+} in $LaAl_2$, and Gd^{3+} in YAl_2 are -235 ± 20, -205 ± 10, and -140 kOe, respectively. This yields $+95\pm20$ kOe for the contribution to the hyperfine field of the additional conduction electron in $GdAl_2$ relative to $EuAl_2$ and -65 ± 10 kOe to the screening effect in Eu^{2+} in $La^{3+}Al_2$ relative to Gd^{3+} in $Y^{3+}Al_2$.

INTRODUCTION

The magnetic hyperfine field $H_{\rm hf}$ of $4f^7(S_{7/2})$ ions in metallic systems is commonly divided into three equally important contributions[1]:

$$H_{\rm hf}=H_{\rm core}+H_{\rm op}+H_n. \qquad (1)$$

Here $H_{\rm core}$ is the core-polarization field, $H_{\rm op}$ arises from conduction-electron polarization by the ion itself, and H_n includes all contributions from neighboring magnetic ions (conduction-electron polarization, covalency effects, etc.). Such an expression has been used to describe the experimental observations of Eu^{2+} hyperfine fields[1] in Eu_xYb_{1-x} and Eu_xBa_{1-x}, and of Gd^{3+} hyperfine fields[2] in Gd_xLu_{1-x}. Assuming $H_{\rm op}$ independent of x, the neighbor contribution H_n was removed by extrapolating the measured values of $H_{\rm hf}$ to $x=0$ in each of these systems. Then using the known value of $H_{\rm core}$, a value of $H_{\rm op}$ was obtained. Thus a complete decomposition of $H_{\rm hf}$ for the pure metal into its component parts was obtained.

Here we extend this approach to intermetallic systems. Mössbauer effect studies were performed for ^{151}Eu in compounds of the form $Eu_xYb_{1-x}Al_2$, $Eu_xYb_{1-x}Cu_2$, $Eu_xLa_{1-x}Al_2$, and $EuAl_4$. Analysis of the experimental results shows that, unlike the pure metals, neighbor effects are relatively small in the intermetallic compounds ($H_n=-45\pm10$ kOe for $EuAl_2$ and <10 kOe for $EuCu_2$ and $EuAl_4$). It is also found that the single-ion contribution to the hyperfine field ($H_{\rm core}+H_{\rm op}$) is monotonically correlated with the isomer shift.

EXPERIMENTAL RESULTS AND ANALYSIS

Data were collected using the 21.9-keV Mössbauer resonance of ^{151}Eu. The source was $^{151}Sm_2O_3$ and absorbers were powders having a total thickness of 40 mg/cm^2. In cases where the samples were magnetically split at 4.2 K, spectra were also taken at 1.4 K to obtain the saturation hyperfine field.

The samples $Eu_{0.05}Yb_{0.95}Cu_2$ and $Eu_{0.05}La_{0.95}Al_2$, which were paramagnetic at 4.2 K, were measured in external fields up to 44 kOe in order to obtain the hyperfine field. In such cases, the source was also placed in the magnet in order to obtain a favorable geometry. However, this results in an increase in the emission linewidth from 1.3 to only 1.6 mm/sec, which was confirmed by a separate measurement using a Eu_2O_3 absorber. The fact that only a small broadening occurs is due to the fact that in Eu^{3+} the hyperfine field from $4f$ electrons induced by the external field is of opposite sign and almost equal to the external field. In analysis of the spectra obtained for these two samples, we have assumed that the magnetic anisotropy is sufficiently low that the Eu magnetic moment lies along the external field direction. This is verified by the observation that best fits to the data were obtained by taking the intensity of $\Delta m=0$ transitions to be zero. These measurements were then extrapolated to saturation assuming a free-ion $^8S_{7/2}$ behavior for Eu^{2+},

$$H_{\rm hf}=H_{\rm ext}+H_sB_{7/2}\left(\frac{7\mu_BH_{\rm ext}}{kT}\right),$$

where $H_{\rm ext}$ is the external field, μ_B is the Bohr magneton, and H_s is the saturation hyperfine field.

The samples $EuCu_2$ and $EuAl_4$ are known to be antiferromagnetic[3,4] at 4.2 K. Application of an external field tends to break the antiferromagnetic ordering, causing the spins to turn toward the $H_{\rm ext}$ direction. The hyperfine fields in turn then give information on H_n, which is affected by the nature of the spin alignment, as will be discussed below.

In $EuAl_4$, 90% polarization is reached,[3] but only 55% for $EuCu_2$.[4]

All spectra were analyzed using a least-squares computer program to obtain the magnetic hyperfine fields, isomer shifts, and linewidths. The results of this analysis are given in Table I, and some typical spectra are shown in Fig. 1.

DISCUSSION

The dependence of the saturation hyperfine field is shown in Fig. 2 as a function of concentration for $Eu_xYb_{1-x}Al_2$ and $Eu_xYb_{1-x}Cu_2$, as well as a value for the compound $Eu_{0.05}La_{0.95}Al_2$. For comparison we also give NMR results[6] previously obtained in $Gd_xY_{1-x}Al_2$. In all cases, the hyperfine field varies linearly with x, within experimental error, allowing one to obtain a value at infinite dilution by extrapolation to $x=0$. Since this value can have no neighbor contribution H_n, one has $H_{\rm hf}$ $(x=0)$ $= H_{\rm core} + H_{\rm op}$. Using a value of $H_{\rm core} = -340 \pm 20$ kOe, independent of structure, one then obtains $H_{\rm op}$. Finally, if $H_{\rm op}$ may be considered to be independent of concentration, then comparison with the hyperfine field for $x=1$ gives H_n for the pure-Eu compound. The last assumption of constant $H_{\rm op}$ implies essentially that the density of states does not change when these very similar compounds are mixed. In $EuCu_2$, little change is seen with x, indicating that the hyperfine field is solely due to the single-ion contributions $H_{\rm core} + H_{\rm op}$. In $EuAl_2$ one concludes that the neighbor contribution H_n is about -45 kOe. Values of H_s, $H_{\rm op}$, and H_n are summarized in Table II.

In addition, data has been obtained for the antiferromagnetic compounds $EuCu_2$ and $EuAl_4$ in an external field. Since only partial alignment of the spins is obtained (as indicated by the finite intensity of the $\Delta m = 0$ lines), these results must be treated differently. The observed hyperfine field in zero external field is given by

$$\begin{aligned} H_{\rm eff}(H_{\rm ext}=0) &= H_c + H_{\rm op} + H_n \\ &= H_c + H_{\rm op} + H_{n\uparrow} + H_{n\downarrow} . \end{aligned} \tag{2}$$

Here we denote $H_{n\uparrow}$ as the hyperfine field contribution from those atoms having magnetic moments parallel to the central magnetic moment, and $H_{n\downarrow}$ those antiparallel. In the presence of an external field, these moments will be pulled toward the direction of $H_{\rm ext}$, tending to become ferromagnetically aligned. If θ is the angle between the moment and the external field, then the average hyperfine field will be given by

$$\begin{aligned} H_{\rm eff}(H_{\rm ext}) &= H_c + H_{\rm op} + H_{n\uparrow} + H_{n\downarrow}\langle\cos(\pi - 2\theta)\rangle + H_{\rm ext}\langle\cos\theta\rangle \\ &= H_c + H_{\rm op} + H_{n\uparrow} - H_{n\downarrow}\langle\cos 2\theta\rangle + H_{\rm ext}\langle\cos\theta\rangle . \end{aligned} \tag{3}$$

If the field is sufficiently strong to completely

TABLE I. Results of spectral analysis.

Compound	T (°K)	$H_{\rm ext}$ (kOe)	$H_{\rm exp}$ (kOe)	H_s[a] (kOe)	Isomer shift (mm/sec)	2Γ (mm/sec)
$EuAl_2$[b]	1.8	0	−278(10)	−280(10)	−9.7(3)	...
$EuAl_2$	4.2	0	−270(5)	...	−9.5(1)	2.9(1)
$Eu_{0.75}Yb_{0.25}Al_2$	4.2	0	−266(5)	−275(10)	−9.4(1)	2.8(1)
$Eu_{0.5}Yb_{0.5}Al_2$	1.4	0	−255(5)	−260(10)	−9.4(1)	3.6(1)
$Eu_{0.25}Yb_{0.75}Al_2$	4.2	0	unresolved	...	−9.3(1)	6.0(2)
$Eu_{0.25}Yb_{0.75}Al_2$	1.4	0	−236(10)	−250(15)	−9.1(3)	4.4(6)
$Eu_{0.05}La_{0.95}Al_2$	4.2	0	0	...	−9.3(2)	2.9(2)
$Eu_{0.05}La_{0.95}Al_2$	4.2	44	−150(5)	−205(10)	−9.2(2)	2.9(2)
$Eu_{0.05}La_{0.95}Al_2$	1.4	0	unresolved	...	−9.1(4)	8.0(2)
$EuCu_2$	1.4	0	−194(10)	−200(10)	−9.1(3)	3.9(2)
$EuCu_2$	4.2	0	−183(5)	...	−9.0(1)	3.3(2)
$EuCu_2$	4.2	44	−156(5)	...	−9.0(1)	3.9(2)
$Eu_{0.75}Yb_{0.25}Cu_2$	4.2	0	−175(5)	...	−8.8(1)	3.8(2)
$Eu_{0.75}Yb_{0.25}Cu_2$	1.4	0	−190(5)	−195(10)	−8.9(1)	3.7(2)
$Eu_{0.5}Yb_{0.5}Cu_2$	4.2	0	−168(5)	...	−8.7(1)	4.0(2)
$Eu_{0.5}Yb_{0.5}Cu_2$	1.4	0	−191(5)	−195(10)	−8.9(1)	2.8(2)
$Eu_{0.25}Yb_{0.75}Cu_2$	4.2	0	unresolved	...	−8.8(1)	6.0(2)
$Eu_{0.25}Yb_{0.75}Cu_2$	1.4	0	−198(5)	−205(10)	−8.7(1)	3.5(2)
$Eu_{0.05}Yb_{0.95}Cu_2$	4.2	44	−150(10)	−210(15)	−8.0(1)	2.9(2)
$EuAl_4$[b]	1.8	0	−290(10)	−290(10)	−11.4(3)	...
$EuAl_4$	4.2	0	−278(5)	...	−11.6(1)	2.6(2)
$EuAl_4$	4.2	44	−254(5)	...	−11.6(1)	3.3(3)

[a] Obtained assuming T_c to be linear in x and a $J=\frac{7}{2}$ Brillouin function behavior for $H_{\rm eff}(T)/H_s$.

[b] Reference 5.

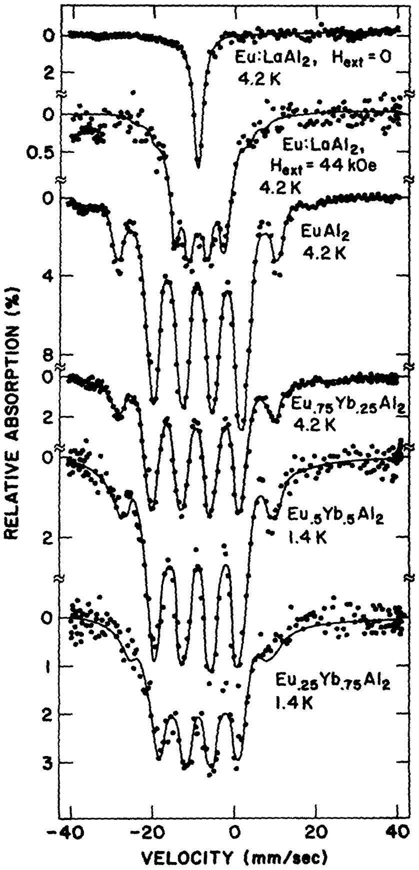

FIG. 1. ^{151}Eu hyperfine spectra obtained in various Eu intermetallic compounds.

overcome the antiferromagnetic coupling, then one will find a hyperfine field $H_c+H_{op}+H_{n\uparrow}-H_{n\downarrow}$, corresponding to $\theta=0$. The negative sign on $H_{n\downarrow}$ indicates that its direction has been reversed by the external field. In general, however, only partial polarization is obtained. In a $M1$ transition like the present case, the intensities of the absorption lines are dependent on θ through

$$\mathcal{I}(\Delta m=0)=\tfrac{3}{2}\sin^2\theta,$$

$$\mathcal{I}(\Delta m=\pm 1)=\tfrac{3}{4}(1+\cos^2\theta).$$

Comparison of these intensities in a given spectrum thus allows one to measure $\langle\cos^2\theta\rangle$. From experimental magnetization data, one may obtain $\langle\cos\theta\rangle$ as a function of H_{ext} since $\mu_{exp}/\mu_{sat}=\langle\cos\theta\rangle$. Thus the angular factors of Eq. (3) may be determined. Comparison with Eq. (2) then gives a value for $H_{n\downarrow}$. However, since $H_{n\uparrow}$ appears in both (2) and (3) in the same form, it cannot be obtained. In Table II we give a value for H_n in EuAl$_4$ under the assumption $|H_{n\downarrow}|\approx|H_{n\uparrow}|$. In a simple antiferromagnetic structure, one would anticipate $|H_{n\downarrow}| > |H_{n\uparrow}|$ since $H_{n\downarrow}$ will arise predominantly from the first-neighbor shell, and $H_{n\uparrow}$ only from more distant neighbors. In general, such a conclusion is not justified and the assumption of $|H_{n\downarrow}|\approx|H_{n\uparrow}|$ is merely an attempt to obtain an estimate of H_n. It does seem clear from these results, however, that H_n is not very large in EuAl$_4$.

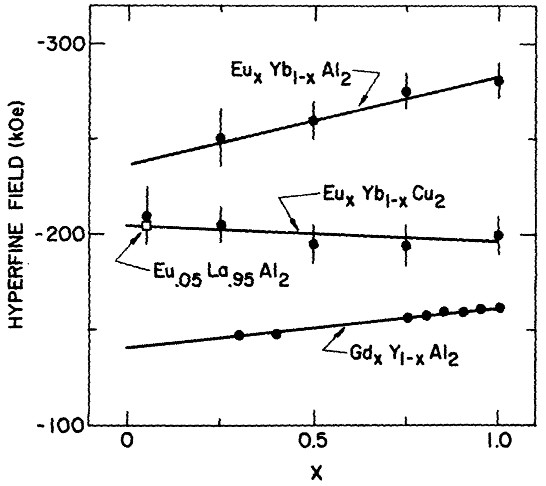

FIG. 2. Variation of the hyperfine field with concentration in Eu intermetallic compounds. The data for Gd$_x$Y$_{1-x}$Al$_2$ are taken from Ref. 6.

From investigation of the results presented in Table II, one may draw a number of conclusions.

(i) The values of H_n vary widely in the alloy systems. This quantity should depend on a number of factors, such as band structure, crystal structure, degree of covalency, etc. However, not surprisingly, one can see here a strong dependence simply on interatomic distance. Thus, in the Eu-Al alloys, H_n is largest in pure-Eu metal, smaller in EuAl$_2$ and very small in EuAl$_4$.

(ii) The single-ion contribution to the hyperfine field, $H_{core}+H_{op}$, is correlated with the isomer shift, as shown in Fig. 3. Extrapolation of the experimental values to the region of observed isomer shifts for pure-ionic-Eu^{2+} compounds gives a hyperfine field very near to the experimental core polarization value of − 340 kOe.

The hyperfine field and the isomer shift do not

TABLE II. Values of H_s, H_{op}, and H_n.

Compound	Isomer[a] Shift mm/sec	H_s kOe	H_{op} kOe	H_n kOe	$H_{n\downarrow}$ kOe
Eu metal[b]	−8.3(3)	−265(5)	+190(20)	−115(20)	
EuCu$_2$	−8.9(3)	−200(10)	135(20)	+5 ± 5	3 ± 5
EuAl$_2$	−9.5(3)	−280(10)	105(20)	−45(5)	
EuAl$_4$	−11.5(3)	−290(10)	50(20)	5 ± 10[c]	5 ± 5

[a] With respect to Eu in Sm$_2$O$_3$.

[b] Reference 1.

[c] Estimated assuming $|H_{n\uparrow}|\approx|H_{n\downarrow}|$.

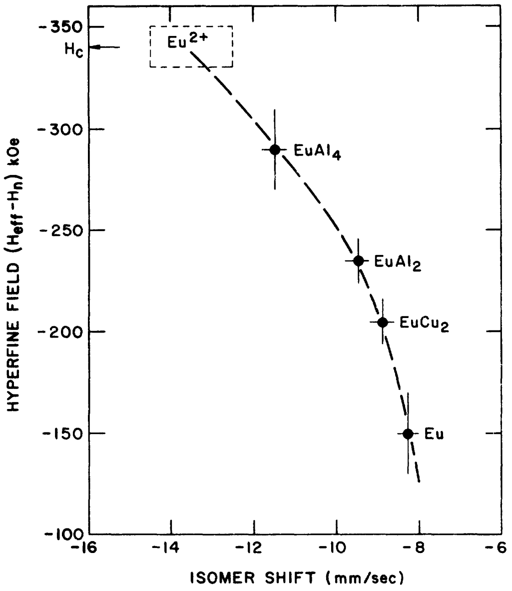

FIG. 3. Correlation between the isomer shift and the single-ion contribution to the hyperfine field, $H_{core}+H_{op}$. All isomer shift values are relative to ^{151}Eu in Sm_2O_3.

measure the same quantities. H_c+H_{op} is proportional to the spin density at the nucleus and the isomer shift is proportional to the charge density at the nucleus. However, if the polarizability of *s* electrons does not vary substantially in these various materials, a correlation such as observed may be expected. One may also notice from Table I that the isomer shift is independent of concentration in a given alloy system. That fact, coupled with the correlation of Fig. 3, provides experimental evidence for the previous assumption that H_{op} is independent of concentration.

(iii) The hyperfine field of $(Eu^{2+})La^{3+}Al_2$ is -205 kOe. From Ref. 6 one finds a value of -140 kOe for $(Gd^{3+})Y^{3+}Al_2$. The difference may be easily understood as a repulsion of conduction electrons by the divalent Eu ion in the trivalent matrix. Thus H_{op} is decreased by 65 kOe.

(iv) The hyperfine field of $(Eu^{2+})Yb^{2+}Al_2$ is -235 kOe, as compared with the value[6] of -140 kOe for $(Gd^{3+})Y^{3+}Al_2$. Here the difference cannot be due to the charge screening effect just mentioned. Instead it is most likely due to an increase of 95 kOe in H_{op} due to the additional conduction electron available when one changes from the divalent to the trivalent matrix.

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PHYSICAL REVIEW B VOLUME 8, NUMBER 1 1 JULY 1973

Dynamic Spin Configuration for Hard Magnetic Bubbles in Translational Motion

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The detailed model of Vella-Coleiro *et al.* for steady translational motion of a hard bubble (i.e., a cylindrical magnetic domain with axial Bloch lines distributed nonuniformly around the domain wall) is shown to lead to essentially the same results obtained by Slonczewski and to exacly the same results obtained by Thiele in less specific calculations. These theoretical expressions are shown to give a good description of new experimental observations of hard-bubble dynamics.

Several theories have been reported[1-4] recently for the dynamic behavior of hard-magnetic-bubble domains (i.e., cylindrical domains whose walls contain axial Bloch lines). Steady translational motion is considered in Refs. 1, 3, and 4, while bubble-collapse dynamics is the subject of Ref. 2. Substantially different conclusions are presented in Ref. 1 than in Refs. 3 and 4. The purpose of the present paper is to show that the basic model used to develop the nonlinear theory of Ref. 1 can yield