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^{160}Dy and ^{166}Er Mössbauer studies of concentrated and diluted rare-earth dihydrides: Single-line compounds and crystal-field effects

J. Stöhr* and J. D. Cashion†
Physik Department, Technische Universität München, D-8046 Garching, Germany

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Mössbauer-effect measurements on the cubic intermetallic rare-earth dihydrides are reported utilizing the 86.8-keV and the 80.6-keV $2^+ \rightarrow 0^+$ γ transitions of ^{160}Dy and ^{166}Er, respectively. All studies were carried out at liquid-helium temperatures on pure and diluted (with Y or Sc) paramagnetic samples. It is found that in the more concentrated alloys spin-spin (4*f*-4*f*) relaxation rates at 4.2 K are sufficiently fast to allow for the use of these systems as narrow-single-line Mössbauer sources and absorbers. Measurements on highly dilute ^{160}Dy and ^{166}Er impurities in YH_2 reveal the resolved paramagnetic hyperfine structure of a Γ_7 crystalline electric field (CEF) ground state in both cases. The results are discussed in terms of CEF theory.

I. INTRODUCTION

Because of their simple $2^+ \rightarrow 0^+$ $E2$ nuclear γ transition the Mössbauer resonances of ^{160}Dy (86.8 keV) and ^{166}Er (80.6 keV) are well suited for the study of the magnetic and quadrupole hyperfine (hf) interactions. Such investigations of rare-earth (RE) ions, especially in the paramagnetic regime, have recently received a lot of attention.[1] For the above isotopes the calculation of the Mössbauer-effect (ME) spectrum is always relatively easy, even for complicated electronic ground states or for the case of complex relaxation processes. Another advantage of the above ME resonances is the great ease of producing the source activities. ^{159}Tb and ^{165}Ho (both 100% abundant) metals are neutron irradiated to produce ^{160}Tb ($T_{1/2} = 72$ days) and ^{166}Ho ($T_{1/2} = 27$ h) which feed the ^{160}Dy and ^{166}Er Mössbauer resonances, respectively.[2] The question arises why the above ME resonances, in particular that of ^{160}Dy have not been applied as extensively in the past as most of the other RE resonances.[3] The main reason for this deficiency is the lack of good monochromatic sources and absorbers at liquid-helium temperature. The liquid-helium regime is of particular interest because Debye-Waller factors for the above isotopes are rather small at higher temperatures. However, unlike the similar ^{170}Yb (84.3 keV, $2^+ \rightarrow 0^+$) resonances, for which narrow-single-line sources and absorbers are available at 4.2 K[3] since Yb is often diamagnetic (Yb^{2+}: $4f^{14}$) in metallic enviroments, Dy and Er are always magnetic (Dy^{3+}: $4f^9$; Er^{3+} : $4f^{11}$) and nearly all intermetallic compounds exhibit magnetic order at 4.2 K.[4]

One aim of the present paper is to report the production of good single-line ME sources and absorbers for the ^{160}Dy and ^{166}Er ME resonances at 4.2 K. It will be shown that the RE dihydride (RH_2) intermetallic compounds are well suited for the above purpose[5] because of their low magnetic-transition temperatures[6] and sufficiently fast paramagnetic spin-spin relaxation times at 4.2 K. In addition they offer reasonable Debye-Waller factors and their preparation is extremely easy. We feel that the finding of such convenient single-line sources and absorbers will substantially improve the applicability of the above ME resonances. As a first application we then report the observation of paramagnetic *hf* structure of ^{160}Dy and ^{166}Er impurities in YH_2 and discuss the results in terms of crystalline electric field (CEF) theory.

II. SAMPLE PREPARATION

For the preparation of the RE dihydrides we used RE metals of 99.9% purity in the form of ingots. As diamagnetic diluents 99.99% purity Y and Sc metals were used. For the preparation of the mixed alloys (typical weight ~ 0.5 g) appropriate amounts of the respective metals were cleaned from surface oxide and then arc melted to obtain a homogeneous mixture of the components. For hydriding, the metals were wrapped in a Ti foil which acted as oxygen getter and then placed in a quartz tube on an out-gassed sheet of Ta. After evacuating the quartz tube to about 5×10^{-6} Torr it was heated in an Ohmic furnace to about 850° C. Purified hydrogen was then introduced to a pressure of about 200 Torr. Reaction of the RE alloys with hydrogen is almost immediate at 850 °C and cooking the samples for approximately 1–2 h was found to be sufficient. In order to avoid formation of the hexagonal trihydrides, the quartz tube was then cooled rapidly from 850 °C to room temperature. The RE and Y dihydrides had a dark blue color except $(Dy_{0.4}Sc_{0.6})H_2$ which was grey. It should be pointed out that the dihydrides just like other RE intermetallic compounds are completely stable against oxidation at room temperature. Debye-Scherrer x-ray photographs revealed that nearly all samples contained only the cubic phase. The lattice parameters were found to agree with the published values.[7,8] The stoichiometry of the so-called "di"-hydrides is usually not exactly RH_2. However, the compounds RH_x have a cubic CaF_2-type structure within the phase limits $1.94 \leq x \leq 2.08$.[8] The point symmetry of the RE (or Y, Sc) ions is $m3m$ (O_h).[9] Samples which showed traces of a second phase (e.g., hexagonal RH_3) were not considered for our studies.

For the ^{160}Dy resonance we used as absorbers the compounds DyH_2 ($T_N = 8$ K) and $(Dy_{0.4}Sc_{0.6})H_2$ ($T_N < 4.2$ K). The Dy metal, 80% enriched in ^{160}Dy, was obtained from Oak Ridge National Laboratories.[10] As sources we employed the alloys YH_2: Tb (2000 ppm), $(Tb_{0.08}Y_{0.92})H_2$, $(Tb_{0.05}Dy_{0.35}Y_{0.60})H_2$, and $(Tb_{0.1}Er_{0.9})H_2$, which are all nonmagnetic at 4.2 K (pure TbH_2 orders at 45 K). The samples were placed in an evacuated quartz tube during neutron irradiation which yielded an activity of about 10 mCi of ^{160}Tb. For our studies on ^{166}Er we used nonenriched ErH_2 ($T_N = 2.4K$) as absorber material. As sources we investigated YH_2:Ho (2000 ppm), $(Ho_{0.15}Y_{0.85})H_2$, $(Ho_{0.4}Y_{0.6})H_2$, and $(Ho_{0.1}Er_{0.9})H_2$ which all order below 4.2 K (pure HoH_2 orders at 8 K). Typical source activities were 20 mCi of ^{166}Ho.

III. EXPERIMENTAL DETAILS AND RESULTS

The reason for preparing these very alloys mentioned above is as follows. One aim was to prepare source and absorber materials which yielded a good monochromatic Mössbauer resonance line. Since the 4f-conduction electron coupling is usually too weak to cause a complete vanishing of the paramagnetic hf structure in the liquid-helium range,[1] a single ME line can only be obtained by using paramagnetic materials with sufficient spin-spin (4f-4f) interactions. Hence for single-line materials we only diluted (if at all) the pure RE dihyrides as much as was necessary to suppress the magnetic transition temperature below 4.2 K. For the second purpose of our paper, i.e., the study of CEF effects, we diluted the samples to an extent where spin-spin interactions were thought to be negligible (2000 ppm of RE in YH_2). In this case a paramagnetic hf structure characteristics of the electronic CEF ground state should be observable and the detailed shape of the hf structure depends on the electronic relaxation rate induced by the 4f-conduction electron coupling.[11]

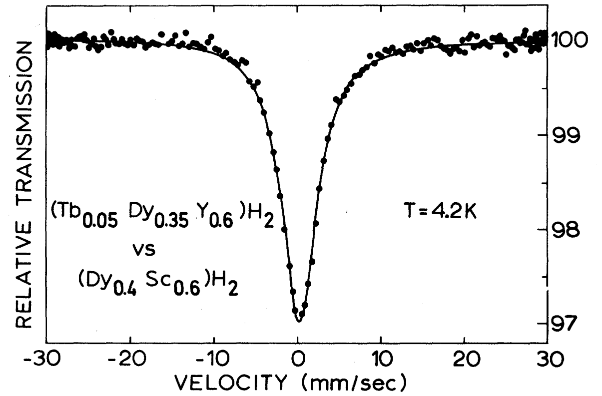

FIG. 1. Mössbauer spectrum of a $(Tb_{0.05}Dy_{0.35}Y_{0.6})H_2$ source vs a $(Dy_{0.4}Sc_{0.6})H_2$ absorber (30 mg ^{160}Dy per cm^2), both at 4.2 K.

A. Single-line materials

Figure 1 shows the Mössbauer-effect spectrum of a $(Tb_{0.05}Dy_{0.35}Y_{0.6})H_2$ source versus a $(Dy_{0.4}Sc_{0.6})H_2$ absorber, both at 4.2 K. The absorber used for Fig. 1 contained 30-mg ^{160}Dy per cm^2. All ^{160}Dy spectra were recorded with a 2-mm-thick NaI scintillation counter (resolution ~10% at 87 keV). Such a system was found to be superior to conventional Ge(Li) detectors because of its higher counting capacity and because of its low detection probability for the various high-energy γ rays emitted in the decay of ^{160}Tb. The linewidth of the ME spectrum of Fig. 1 plus the results for some

TABLE I. Linewidths of various sources and absorbers for the ^{160}Dy resonance.

Source	Source temperature (K)	Absorber	Absorber thickness (mg ^{160}Dy cm^{-2})	Absorber temperature (K)	Γ_{exp}[a] (mm sec^{-1})
$(Tb_{0.05}Dy_{0.35}Y_{0.6})H_2$	4.2	$(Dy_{0.4}Sc_{0.6})H_2$	30	4.2	5.1 ± 0.1
$(Tb_{0.1}Er_{0.9})H_2$	4.2	$(Dy_{0.4}Sc_{0.6})H_2$	30	4.2	5.2 ± 0.1
$(Tb_{0.08}Y_{0.92})H_2$	30	DyH_2	25	20	8.1 ± 0.2
Tb metal[b]	4.2	$(Dy_{0.4}Sc_{0.6})H_2$	30	4.2	4.6[b]

[a] Γ_{exp} is the experimental full width at half-maximum (FWHM). Error bars represent accuracy of least-squares fit.

[b] For Tb metal, five resonance lines were observed (compare Fig. 2). The value $\Gamma_{exp} = 4.6$ mm sec^{-1} corresponds to an averaged value for the five linewidths.

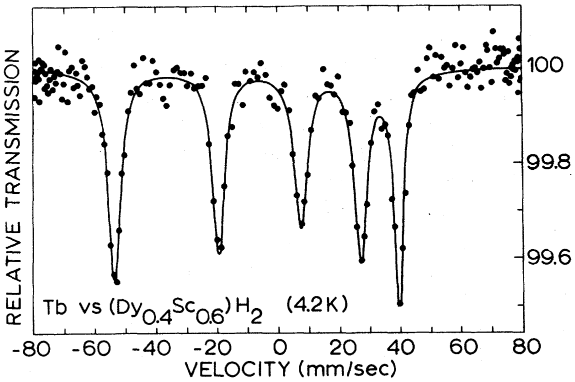

FIG. 2. Hyperfine structure of ^{160}Dy impurities in a Tb metal source analyzed with a $(Dy_{0.4}Sc_{0.6})H_2$ absorber, both at 4.2 K. Solid line through the data points represents a least-squares fit discussed in the text. The velocity convention is such that positive velocities correspond to positive hf splitting energies of the $I=2$ excited nuclear state.

TABLE II. Linewidths of several sources for the ^{166}Er resonance measured with an absorber of ErH_2 (25 mg ^{166}Er per cm^2) at 4.2 K.

Source	Source temperature (K)	Γ_{exp} [b] (mm sec^{-1})
$(Ho_{0.4}Y_{0.6})H_2$	4.2	7.7 ± 0.1
$(Ho_{0.15}Y_{0.85})H_2$	4.2	8.0 ± 0.1
	1.4	8.5 ± 0.1
$(Ho_{0.1}Er_{0.9})H_2$	4.2	8.5 ± 0.2
Ho metal[a]	4.2	5.9 ± 0.3

[a] For Ho metal, five resonance lines of equal width and intensity were observed. Fitting the line position according to $\mathcal{H}_{hf}=XI_z+Y[3I_z^2-I(I+1)]+S$ we obtain $X=55.5\pm0.5$ mm sec^{-1}, $Y=0.71\pm0.01$ mm sec^{-1} and $S=0$.

[b] Γ_{exp} is the experimental full width at half-maximum (FWHM). Error bars represent accuracy of least-squares fit.

other single-line sources and absorbers investigated are summarized in Table I. It should be noted that throughout this paper all experimental linewidths reported correspond to the full width at half-maximum (FWHM) of the ME resonance line. Least-squares fits with a Lorentzian absorption line slightly deviated from the observed line shapes. This is attributed to a small dispersion term in the ME absorption cross section due to interference between conversion and photoelectron absorption, and between Rayleigh and resonant scattering.[12] In our final analysis we have included such a term using the formula of Ref. 12 and as will be discussed below, this notably changes the derived isomer shifts.

In order to demonstrate the applicabilty of such good monochromatic sources or absorbers and to obtain further information on the minimum experimental linewidth possible in studies utilizing the ^{160}Dy Mössbauer resonance, we show in Fig. 2 the ME spectrum of ^{160}Dy in a magnetically ordered Tb metal ($T_N=228$ K) source at 4.2 K. As absorber we employed $(Dy_{0.4}Sc_{0.6})H_2$ (30 mg ^{160}Dy per cm^2) at 4.2 K. The experimental spectrum was least-squares fitted with five lines of free width and depth. The line positions were accounted for by the Hamiltonian $\mathcal{H}_{hf}=XI_z+Y[3I_z^2-I(I+1)]+S$, where $X=47.1\pm0.5$ mm sec^{-1}, $Y=-1.22\pm0.02$ mm sec^{-1} and $S=0$. For the linewidths we obtained the values (from left to right in Fig. 2) 4.7 ± 0.1, 4.3 ± 0.4, 5.3 ± 0.4, 4.9 ± 0.5, and 3.7 ± 0.3 (all in mm sec^{-1}). The slightly unequal intensities are probably due to the fact that our source sample was not completely polycrystalline.

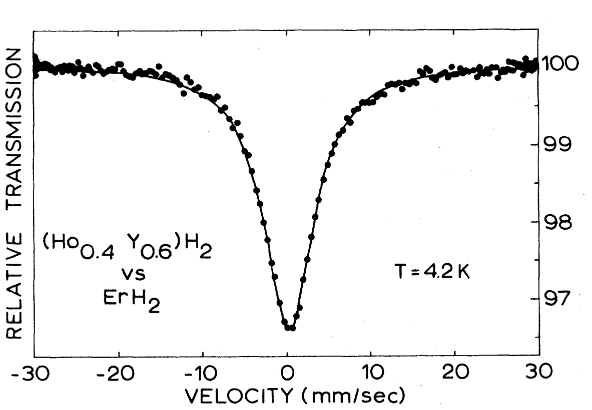

FIG. 3. Mössbauer spectrum of a $(Ho_{0.4}Y_{0.6})H_2$ source vs an absorber of ErH_2 (25 mg ^{166}Er per cm^2), both at 4.2 K.

In Fig. 3 we show the ME spectrum obtained with a $(Ho_{0.4}Y_{0.6})H_2$ source and an absorber of ErH_2, both at 4.2 K. The absorber contained 25 mg ^{166}Er per cm^2. For the ^{166}Er resonance ME spectra were always recorded with a Ge(Li) detector. The results on various single-line sources and absorbers are summarized in Table II. In the analysis of our single-line spectra we followed the approach used for ^{160}Dy.

It should be noted that the effect of hydrogen content on the linewidths has not been investigated in detail. The results listed in Tables I and II represent the optimum linwidths achieved for a given RE concentration and within the cubic phase limits of hydrogen content.[8]

B. Highly diluted sources

The ME spectra of a 2000-ppm $YH_2:Tb$ source at 1.4 and 4.2 K versus an absorber of $(Dy_{0.4}$

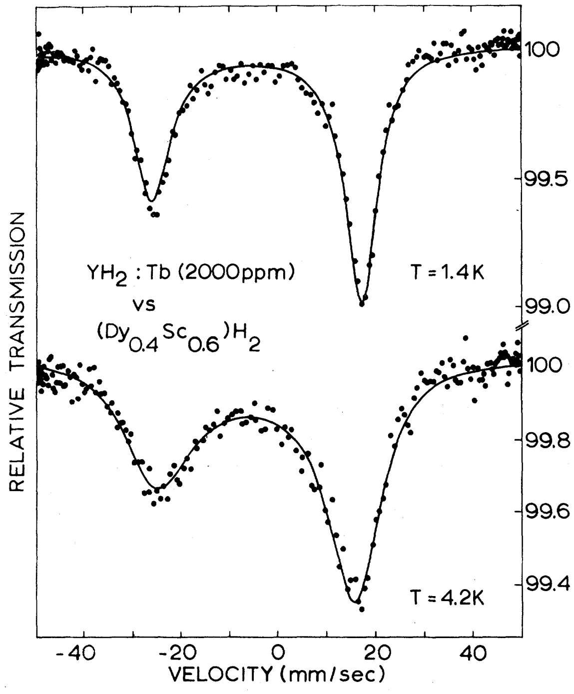

FIG. 4. Paramagnetic hyperfine structure of ^{160}Dy impurities in a 2000-ppm Tb in YH_2 source at 1.4 and 4.2 K. As absorber $(Dy_{0.4}Sc_{0.6})H_2$ (30 mg ^{160}Dy per cm^2) at 4.2 K was used. The least-squares fit with a microscopic relaxation theory is discussed in the text. Velocity convention as for Fig. 2.

$Sc_{0.6})H_2$ (30 mg ^{160}Dy per cm^2 kept at 4.2 K) is shown in Fig. 4. The observed paramagnetic hf spectrum of ^{160}Dy impurities in YH_2 is that of a Γ_7 CEF ground doublet.[13] The least-squares fit was carried out with a microscopic relaxation theory leaving the effective spin hf coupling constant A_s and the relaxation rate W as free parameters. We obtain $A_s = 17.6 \pm 0.2$ mm sec^{-1} for spectra both at 1.4 and 4.2 K. Using the value $g = 68/9$[14] for a Γ_7 doublet and $g_J = 4/3$ for the Landé factor of the $^6H_{15/2}$ free-ion ground state of Dy^{3+} we obtain the hf coupling constant $A = A_s g_J / g$ as $A = 3.10 \pm 0.04$ mm sec^{-1} (217 ± 3 MHz) for ^{160}Dy. For the relaxation rate we derive $W = 119 \pm 7$ MHz at 1.4 K and $W = 350 \pm 10$ MHz at 4.2 K. Thus the Korringa law[11] is exactly obeyed in this temperature region with $\Delta W / \Delta T = 84 \pm 5$ MHz/K. ME spectra taken with a source of $(Tb_{0.08}Y_{0.92})H_2$ at 1.4 and 4.2 K showed an increased relaxation rate relative to the 2000-ppm sample. However, even for this rather concentrated sample (8-at. % Tb) the spectrum still showed a paramagnetic hf splitting characteristic for a Γ_7 CEF ground state. It is because of this slow spin-spin relaxation in the paramagnetic regime of $(Tb_xY_{1-x})H_2$ that the single-line sources for the ^{160}Dy resonance had to be made by adding Dy or Er as has been discussed above.

The results for ^{166}Er impurities in YH_2 are similar to those of ^{160}Dy. The ME spectrum of a 2000-ppm YH_2 : Ho source at 4.2 K is shown in Fig. 5. The absorber used was ErH_2 (25 mg ^{166}Er per cm^2) at 4.2 K. Again the observed paramagnetic hf structure is that of a Γ_7 electronic ground state.[13] The parameters deduced from the spectrum are $A_s = 21.7 \pm 0.3$ mm sec^{-1} and $W = 260 \pm 20$ MHz. With [14]$g = 34/5$ and $g_J = 6/5$ for the $^4I_{15/2}$ Hund's ground state of Er^{3+} we obtain $A = 3.83 \pm 0.04$ mm sec^{-1} (249 ± 3 MHz) for ^{166}Er. Opposite to the case of Tb in YH_2, the ME spectrum of Ho in YH_2 rapidly collapses with increasing Ho concentration. Ho concentrations of a few percent resulted in a complete vanishing of the hf structure leaving a broadened single line.

IV. DISCUSSION OF RESULTS

A. Single-line materials

At first let us compare our results on single-line materials to those published previously. For ^{160}Dy either[15] $(Tb_{0.2}Y_{0.8})Al_2$ or [16]$(Tb_{0.05}La_{0.95})Al_2$ have been used. When kept above 20 K such sources were found to emit an unsplit resonance line which under optimal absorber conditions yielded a FWHM of about 13 mm sec^{-1},[3] or about eight times the minimum experimental linewidth $2\Gamma_n = 1.59$ mm sec^{-1}.[2] As an absorber DyF_3 has been previously used.[15] This absorber yields an essentially unsplit line at 20 K, but there are broad shoulders in addition to the central line probably indicating an unresolved paramagnetic *hf* structure. From the spectrum published by Cohen[15] the FWHM of such an absorber measured with a $(Tb_{0.2}Y_{0.8})Al_2$ source at 25 K is estimated to be about 20 mm sec^{-1}. When these results are compared to the linewidths of our sources and absorbers (see Table I) the improvement is striking. Our sources and

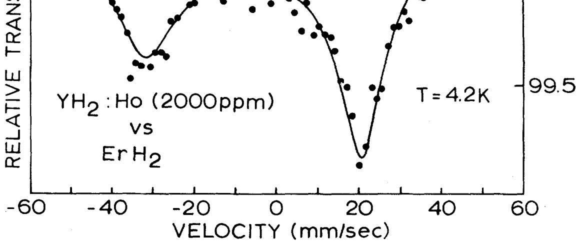

FIG. 5. Paramagnetic hyperfine splitting of ^{166}Er in a 2000-ppm Ho in YH_2 source analyzed with ErH_2 (25 mg) ^{166}Er per cm^2) absorber, both at 4.2 K. The least-squares fit is discussed in the text. Velocity convention as for Figs. 2 and 4.

absorbers not only yield linewidths which are at least a factor 3 better but in addition they can be employed at 4.2 K. It is also apparent from Figs. 1 and 3 that the resonance effect in the hydrides is quite high which ensures reasonable measuring times.

Unlike the resonance of ^{160}Dy which has almost completely been neglected in the past in favor of that of ^{161}Dy ($5/2^- \rightarrow 5/2^+$, 25.6 keV) the ME resonance of ^{166}Er has been commonly applied before.[3] Here good monochromatic sources and absorbers have been used. The only disadvantage of the previously used single-line materials is that they have to be employed at temperatures above 4.2 K. $HoAl_2$ ($T_c = 27$ K)[3] or Ho in Al (~10 wt.%)[12] have usually been taken as single-line sources. As monochromatic absorbers $ErAl_2$ ($T_c = 14.5$ K)[17] or cubic (β) $ErAl_3$ ($T_N = 5.1$ K)[13,18] have been used. When kept above their respective magnetic transition temperatures the best line widths measured with such single-line sources or absorbers is about 2–3 times[3,17,18] the minimum theoretical line width $2\Gamma_n = 1.87$ mm sec^{-1}.[2]

It is interesting to note that the best linewidths for all RE Mössbauer transitions are obtained when either the ME atom occurs in a nonmagnetic electronic state (which is not possible for Er^{3+} and Dy^{3+}) or when the ME spectrum of one or both of the source and absorber is magnetically split. When measuring a single-line source against a single-line absorber considerably larger linewidths are obtained. For example, with a source of 10-wt.% Ho in Al and an absorber of $ErAl_3$ (10 mg ^{166}Er per cm^2) one obtains[12] $\Gamma_{exp} = 6.85 \pm 0.07$ mm sec^{-1} when both are kept at 45 K. The above effect is also revealed by our experimental results in both Table I and II where the best linewidths were obtained when using the magnetically ordered Tb ($T_N = 228$ K) or Ho ($T_N = 133$ K) metal sources. This indicates that the broadening observed for the single-line materials must be due primarily to an incompletely vanishing paramagnetic *hf* structure.

The fact that all linewidths for the ^{166}Er resonance are larger than those of ^{160}Dy can also be at least partly explained by the above effect. Because of the smaller total electronic spin of Er^{3+} ($S = \frac{3}{2}$) than of Dy^{3+} ($S = \frac{5}{2}$), relaxation rates within the Γ_7 CEF ground state of Er^{3+} are expected to be smaller than those within the Γ_7 ground doublet of Dy^{3+} (compare the results of Sec. III B for W for Er^{3+} and Dy^{3+} in YH_2 at 4.2 K). Second, the hf splitting (i.e., the nuclear correlation frequency) is larger for ^{166}Er than for ^{160}Dy (compare the above results for A for Er^{3+} and Dy^{3+} in YH_2). Thus a paramagnetic hf splitting or broadening is more easily observable for the ^{166}Er resonance. It is thought that it is this mechanism rather than the fact that the minimum possible linewidth is slightly smaller for ^{160}Dy (1.59 mm sec^{-1}) than for Er (1.87 mm sec^{-1}) which causes the relatively broader lines observed for the ^{166}Er resonance.

As has been pointed out previously, the single-line ME spectra were analyzed by allowing for a dispersion term. Spectra which were least-squares fitted with a mere Lorentzian line shape showed an apparent isomer shift between +0.20 and +0.30 mm sec^{-1}. Because of the small $\delta\langle r^2\rangle$ values for ^{160}Dy and ^{166}Er, and because the employed sources and absorbers ought to have similar charge densities, the above value is thought to be too large. As was pointed out previously by Wagner *et al*,[12] such apparent isomer shifts may arise if the interference effects of outgoing photons and electrons are neglected. Indeed, when including a dispersion term the values derived for the insomer shifts were considerably reduced. In general isomer shifts < 0.10 mm sec^{-1} resulted. The values derived for the dispersion parameter ξ agreed well with those found by Wagner *et al.* for ^{166}Er.

B. Highly diluted sources

Following the notation of Lea, Leask, and Wolf (LLW),[19] the CEF effects in cubic symmetry depend on the CEF parameters $A_4 = B_4/\beta\langle r^4\rangle$ and $A_6 = B_6/\gamma\langle r^6\rangle$. The lattice sums A_4 and A_6 are expected to be of comparable magnitude and of the same sign for the various RE ions in a given CEF. The *signs* of A_4 and A_6 are of particular interest since they may under favorable circumstances be determined from the knowledge of the CEF ground state alone and may then be compared to the sign which results from a simple point-charge model.

Our experimental results of obtaining a Γ_7 CEF ground state for Dy^{3+} and Er^{3+} in YH_2 impose a negative A_4 and positive A_6 or two positive parameters A_4 and A_6.[19] In order to distinguish between the two possibilities let us consider the results of our measurements on more concentrated samples. As was mentioned before, a source of 8-at.% Tb in YH_2 yielded similar results to the very dilute 2000-ppm source. This can only be understood in terms of a nonmagnetic CEF ground state for TB^{3+} in YH_2 since for a Tb concentration of 8% and for a magnetic ground state of Tb^{3+} the ME spectrum is expected to change (collapse) significantly owing to the presence of spin-spin relaxation. A nonmagnetic CEF ground state for Tb^{3+} in YH_2 is completely confirmed by the two possibilities of sign for A_4 and A_6 mentioned above. For both cases a nonmagnetic CEF state (Γ_1, Γ_2, or Γ_3) is found to be lowest for Tb^{3+}. For Ho in YH_2, the rapid increase of the relaxation rate with concentration can only be explained if Ho^{3+} possesses a magnetic ground state. From the graphs of LLW it is seen that for negative A_4 and positive A_6 the CEF ground state of Ho^{3+} is indeed magnetic. In this case a magnetic Γ_5^2 triplet or a nonmagnetic Γ_3^2 doublet nearly degenerate with a magnetic Γ_4^2 triplet are found to be lowest. For positive A_4 and A_6, a Γ_3^2 nonmagnetic doublet or a Γ_1 nonmagnetic singlet forms the ground state. Although in this case a Γ_4^2 triplet as the first excited state may be close enough to the ground state to have nonvanishing thermal population, the fast relaxation times observed for the more concentrated $(Ho_xY_{1-x})H_2$ alloys seem to point to a *negative* sign of A_4 and a *positive* sign of A_6.

We conclude the discussion of CEF effects by comparing the above result to those obtained by a simple point-charge calculation. In the RH_2 compounds there are eight hydrogen nearest neighbors in an eightfold cubic coordination.[9] If we assume that the CEF is only determined by these closest charges a point-charge calculation yields the following results:[19,20] (a) for H^+ charges one gets a positive A_4 and negative A_6, and (b) for H^- charges we obtain a *negative* A_4 and a *positive* A_6. Hence we find complete agreement between our experimental CEF results and those predicted by a simple *hydric*-hydrogen model. Although we do not want to press our results in terms of the query whether the hydric (H^-) or protonic (H^+) model is correct for the RH_2 systems,[7] we mention, however, that point-charge calculations have proven to explain the CEF effects in a variety of RE intermetallic compounds.[21]

Finally, we comment briefly on relaxation effects and the measured hyperfine coupling constants of ^{160}Dy and ^{166}Er impurities in YH_2. Since the Korringa law is well obeyed below 4.2 K for Dy impurities in YH_2, we may use the value $\Delta W/\Delta T = 84 \pm 5$ MHz/K to extract information on the $4f$–conduction-electron coupling.[11] Assuming this coupling to be of vector type[11,23] we obtain $|J_{sf}N(E_F)| = 1.34\times10^{-2}$.[22] With the value $N(E_F) = 0.3$ (eV spin atom)$^{-1}$ for the conduction-electron density of states in YH_2 estimated from the value $A = 249$ MHz for ^{166}Er impurities in YH_2 and from Fig. 1 of Ref. 23, we obtain $J_{sf} = 4.5\times10^{-2}$ eV for the exchange coupling constant of Dy impurities in YH_2.

Both values obtained for the hyperfine coupling constants are slightly larger than those observed in insulator hosts $A(^{160}\mathrm{Dy}) = 209$ MHz and $A(^{166}\mathrm{Er}) = 245$ MHz.[23] For ^{160}Dy in YH_2 we observe an increase of 8 MHz or about 4%, for ^{166}Er in YH_2 the value of A is 4 MHz or about 1.5% larger. Similar effects have also been observed for ^{166}Er impurities in various host metals[23] and have been attributed to a contribution to the hf coupling by conduction electrons polarized by the $4f$ impurity spin.[17,23-25] In this picture the increase in the hf coupling constant is expected to be larger for Dy^{3+} than for Er^{3+} since the Hund's ground state of Dy^{3+} ($^6H_{15/2}$) has a larger spin ($S=\frac{5}{2}$) than that of Er^{3+} ($^4I_{15/2}$, $S=\frac{3}{2}$).[24] Our results are well within expectations based on this simple theoretical model.[24]

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[*] Present address: Lawrence Berkeley Laboratory, University of California, Berkeley, Calif. 94720.

[†] Permanent address: Dept. of Physics, Monash University, Clayton, Victoria 3168, Australia.

[1] For recent Mössbauer-effect studies of rare-earth ions in nonmagnetic metals and insulator hosts see J. Phys. (Paris) 35, 21 (1974); 35, 421 (1974); 35, 425 (1974); 35, 437 (1974), and references therein.

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