On the local magnetic moments of transition-metal Mössbauer impurities in ferromagnetic rare earths

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Using isomer shift and hyperfine data of transition-metal Mössbauer impurities ^{193}Ir and ^{57}Fe diluted in the ferromagnetic rare-earth hosts Gd, Tb, and Dy, the formation of local moments at these impurities based on a two-band (s-d) model in the framework of an extended Ruderman-Kittel-Kasuya-Yosida picture is theoretically discussed. The experimental data of Mössbauer isomer shifts and hyperfine fields at the nobel probe ^{197}Au placed in Gd, Tb, and Dy are also briefly discussed, based on an extended Daniel–Friedel model. The results exhibit a good agreement with the available experimental measurements.

Experimental results[1,2] concerning the magnetic hyperfine fields and isomer shifts of the $5d$ transition-metal Mössbauer element ^{193}Ir diluted in the ferromagnetic heavy rare-earth metals Gd, Tb, and Dy are shown in Table I. From the Mössbauer isomer shifts reported in this table one concludes that the isomer shifts of ^{193}Ir in IrR (R = Gd,Tb, or Dy) are nearly constant when one goes from Gd to Dy, the value of 2 mm/s being in good agreement with the systematics of the isomer shift of ^{193}Ir in $5d$ transition-metal hosts.[3] The same feature of almost constant isomer shifts[4] of the $3d$ Mössbauer element ^{57}Fe placed in FeR (R = Gd,Tb, or Dy) is exhibited in Refs. 2 and 4.

Since isomer shifts of Mössbauer nuclei probes are directly related to the local Koster–Slater (KS) perturbed density of states $\tilde{\rho}_0(E)$, i.e., its electronic environment, this suggests that the host s-p- and d-band occupations are quite the same for these rare-earth metals. Band calculations[5-7] have shown indeed that Gd, Tb, and Dy metals exhibit d bands contributing a high density of states in the vicinity of the Fermi energy E_F, with Fermi surfaces unlike those expected for free electrons. In this sense these rare-earth metals can be regarded as being transitionlike metals in the beginning of a $5d$ series. One can estimate[8] from the above-mentioned band calculations the d- and s-p-band occupation numbers as being, respectively, $\langle n_d \rangle = 2.2$ electrons (per host atom) and $\langle n_c \rangle = 0.8$ electrons (per host atom) giving a 3^+ configuration for these metals: $\langle n_d \rangle + \langle n_c \rangle = 3.0$ electrons (per host atom).

The magnetic hyperfine fields of Mössbauer d impurities such as ^{193}Ir, ^{57}Fe, and the noble Mössbauer impurity ^{197}Au embedded in the ferromagnetic rare-earth hosts Gd, Tb, and Dy have been interpreted in the framework of the standard Ruderman-Kittel-Kasuya-Yosida (RKKY) theory which predicts a linear decrease when one passes from Gd to Dy[9]:

$$H_{hf} = C(g_J - 1)\langle J_z \rangle, \tag{1}$$

where H_{hf} is the total magnetic hyperfine field and C is, for each impurity, a constant across the lanthanide series.[10] For magnetic saturation and neglecting crystal field effects one has[9] $J_z = J$. The quantity $(g_J - 1)$, which is the projection of the spin S on the total angular momentum J of the rare-earth ions, decreases linearly with increasing atomic number of the rare earth. One has respectively $\frac{7}{2}, \frac{6}{2}$, and $\frac{5}{2}$ for Gd, Tb, and Dy. However, the experimental data deviate in all cases more or less strongly towards smaller hyperfine fields from the predicted linearity between H_{hf} and $(g_J - 1)J$ (see Figs. 1–3). Dunlap, Nowik, and Levy[11] pointed out that the breakdown of the standard RKKY theory is due to orbital and spin dipolar contributions because the nonzero angular momentum of the lanthanide ions, and by including further terms in Eq. (1) they can fit the experimental data.

In this work we want to suggest an alternative model to discuss the available hyperfine field data and therefore the local moments which can be obtained from these measurements. The model is based on an extended RKKY theory, which is beyond the usual single-band free-electron-like calculation. Our picture takes into account the transition-metal-like peculiarities of the heavy rare-earth band structure (s-d bands) and stresses the role of the effective $4f$ moments in deriving the ferromagnetic properties of these metals.[12-14] Moreover, all the parameters used in the present work are extracted from our previous calculations, where the systematics of d-transition impurities[12] and noble impurities[13] diluted in Gd metal as well as d-transition impurities[14] placed in Tb were theoretically discussed.

Some brief comments on these host metals are worthwhile.[15] Gd is the only rare earth which goes directly from the paramagnetic phase to the ferromagnetic one, with a Curie temperature T_C equal to 298 K. The saturation magnetic moment at very low temperature is 7.63 μ_B which is clearly larger than the 7 μ_B value corresponding to the trivalent

TABLE I. Magnetic hyperfine field (H_{hf}) and Mössbauer isomer shift (IS) of ^{193}Ir in the heavy rare-earth metals, Gd, Tb, and Dy, collected from Ref. 1.

Host	H_{hf} (kG)	IS (mm/s)
Gd	− 624(6)	− 1.99(2)
Tb	− 318(5)	− 2.01(3)
Dy	− 155(5)	− 2.03(3)

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TABLE II. Magnetic hyperfine field (H_{hf}) and Mössbauer isomer (IS) of ^{197}Au in the heavy rare-earth metals Gd, Tb, and Dy, collected from Ref. 10.

Host	H_{hf} (kG)	IS (mm/s)
Gd	−809(8)	−8.09(5)
Tb	−426(13)	−8.20(5)
Dy	−258(6)	−8.09(5)

Gd^{3+}. In contrast, Tb and Dy have two magnetic transitions characterized by the Néel temperature T_N and the Curie temperature T_C and between the two temperatures a periodic arrangement of the magnetic moments is present. In Tb metal the transition temperatures are $T_N = 229$ K and $T_C = 221$ K, and at very low temperatures the saturation magnetic moment is $9.34\,\mu_B$ whereas a theoretical value of $9\,\mu_B$ is expected for the trivalent ion Tb^{3+}. In the case of Dy one has $T_N = 178$ K and $T_C = 85$ K, and at very low temperatures the saturation magnetic moment is $10.2\,\mu_B$ which has to be compared to the $10\,\mu_B$ expected for the trivalent ion Dy^{3+}.

In spite of the peculiarities of these hosts, which are also reflected in the somewhat different "accidents" appearing in their densities of states, the ferromagnetism of these metals is described within a simple Stoner-type picture by the two d-spin subbands split by an average exchange energy ϵ_d.[12–14]

One assumes that the polarized d electrons contribute to create an effective magnetic splitting ϵ_c in the s-p electrons through s-d hybridization. This mechanism provides a negatively polarized s-p conducting band,[16,17] m_c, given by

$$m_c = -\alpha_{cd} m_d, \tag{2}$$

where m_d is the d magnetization of the host. α_{cd} being a phenomenological parameter, estimated to be of the order of 0.1.[16]

When the d-transition-metal Mössbauer probe is placed in a rare-earth substitutional site, it introduces a localized charge and spin dependent potential. One assumes here that such perturbation can be described by effective intraband potentials $V_{c\sigma}$ and $V_{d\sigma}$ acting respectively on the s-p and on the d conduction bands. The localized potential $V_{c\sigma}$ is of a Daniel–Friedel[18] type,

$$V_{c\sigma} = V_{0c} - \sigma\lambda_c\epsilon_c. \tag{3}$$

λ_c is a phenomenological parameter which takes into account the RKKY transferred magnetization at the impurity site.[17]

The localized potential $V_{d\sigma}$ is written in a Hartree–Fock approximation as

$$V_{d\sigma} = V_{0d} - \sigma\lambda_d\epsilon_d + \Delta U\tilde{n}_{d-\sigma}. \tag{4}$$

$V_{0\alpha}$ ($\alpha = s$-p or d) is a purely electrostatic potential arising from the charge difference between impurity d electrons and host d conducting electrons. ΔU is the difference of Coulomb-type correlations at the d-transition site between impurity and host and $\tilde{n}_{\alpha\sigma}$ is the number of α electrons with spin σ at the impurity site.

One gets the local moments $\tilde{m}_\alpha$:

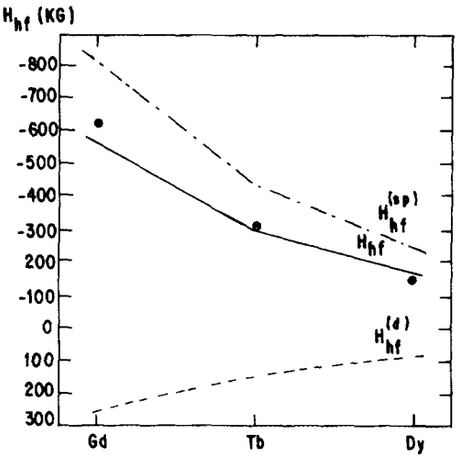

FIG. 1. Self-consistent numerical results for the CEP field and CP hyperfine field at ^{193}Ir impurity placed in Gd, Tb, and Dy (dotted lines). The full line is the total hyperfine field. We adopted $\lambda_c = 0.2$, $\lambda_d = 1.0$, $\Delta U_{sd} = 0.14$, and $A_{CP}^{(d)} = -1200$ K/Oe/μ_B (see Ref. 12). The cross circles represent the experimental data for the total hyperfine field collected from Ref. 1.

$$\tilde{m}_\alpha = \sum_\sigma \sigma\tilde{n}_{\alpha\sigma}, \tag{5}$$

and hence, the total local moment at the d Mössbauer probe is

$$\tilde{m}_0 = \tilde{m}_d + \tilde{m}_c. \tag{6}$$

The localized magnetic moments of dilute d impurities in these ferromagnetic metals can be estimated from their contributions to the magnetic hyperfine field H_{hf} at the impurity site.

$$H_{hf} = H_{0p} + H_{CEP} + H_{CP}. \tag{7}$$

H_{0p} is due to overlap polarization and can be disregarded here, since the metallic radius of Gd, Tb, or Dy is much larger than that of any d impurity. H_{CEP} arises from conduction electron polarization (CEP), which is due to the magnetization of s-p conduction electrons. It can be written as

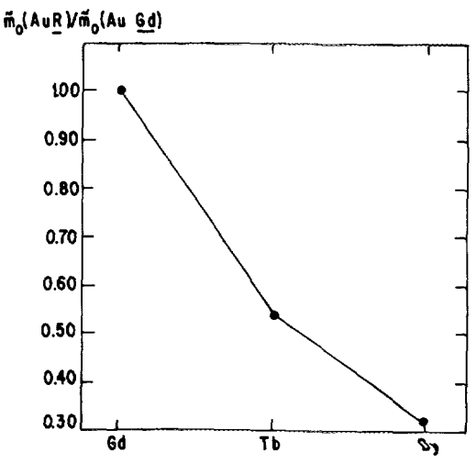

FIG. 2. Self-consistent numerical results for the local magnetic moments at ^{197}Au impurity in Gd, Tb, and Dy normalized to the local moment at ^{197}Au in Au*Ge* [see Eq. (12) in the text]. The cross circles represent the experimental data taken from Ref. 10.

$$H_{\mathrm{CEP}} = H_{hf}^{(sp)} = A(Z)\tilde{m}_c, \tag{8}$$

where $A(Z)$ is the Fermi–Segrè contact parameter.[19]

H_{CP} is the core polarization (CP) field, originated from the localized d magnetic moment of the Mössbauer probe which polarizes the impurity electron core. One has

$$H_{\mathrm{CP}} = H_{hf}^{(d)} = -A_{\mathrm{CP}}^{(d)}\tilde{m}_d, \tag{9}$$

where $A_{\mathrm{CP}}^{(d)}$ is a positive core polarization coupling parameter, which is constant for a given n-d ($n = 3,4,5$) series.[19]

The total magnetic hyperfine field is given by

$$H_{hf} = H_{hf}^{(sp)} + H_{hf}^{(d)}. \tag{10}$$

In the present numerical calculation, following the same procedure adopted in previous works[12,14,20] we considered Moriya-type bands.

Here only integrations over the densities of states are relevant to calculate the local magnetic moments. So, the "accidents" in the densities of states, which are peculiar to each rare-earth host considered in this work, are almost wiped out.

The self-consistent numerical results for the CEP hyperfine field and for the CP hyperfine field at the 5d-Mössbauer impurity ^{193}Ir is shown in Fig. 1. We have adopted the same parameters used previously in the discussion of the systematics of the hyperfine fields of 5d impurities in Gd.[12] One sees that our two-band RKKY calculations reproduce the measured hyperfine fields quite well. The theoretically calculated local magnetic moments turn out to be

$$\tilde{m}_0(\mathrm{Ir}Gd) = -0.26\ \mu_B, \quad \tilde{m}_0(\mathrm{Ir}Tb) = -0.15\ \mu_B,$$

and

$$\tilde{m}_0(\mathrm{IrDy}) = -0.08\ \mu_B,$$

which are always antiparallel to the host magnetization. Moreover, the theoretically calculated local magnetic moments, normalized to $\tilde{m}_0$ (IrGd) are almost proportional to the ratio of the experimentally measured excess of magnetization of the hosts with respect to the observed excess of magnetization in Gd.

In the same way, our self-consistent theoretical results for ^{57}Fe diluted in Gd, Tb, and Dy obey the same tendency observed in the case of IrR alloys.

It should be emphasized that our self-consistent calculations, although describing quite adequately the observed hyperfine fields main features, are based on a semiphenomenological tight-binding approach. So, it would be desirable to perform *ab initio* calculations, similar to those obtained in the case of Fe (Ref. 21) and Ni (Ref. 22) hosts.

Finally, in Fig. 3 we show the behavior of the relative local magnetic moments for the Mössbauer noble impurity ^{157}Au, which also follows the same tendency exhibited by ^{193}Ir and ^{57}Fe impurities. The Au impurity has the electronic configuration $(5d)^{10}(6s)^1$, therefore a CP hyperfine contribution is not expected. So, the total hyperfine field is only due to CEP contribution[20]:

$$H_{hf} = H_{hf}^{(sp)} = A(Z)\tilde{m}_c = A(Z)\tilde{m}_0, \tag{11}$$

and, hence,

$$\frac{H_{hf}(\mathrm{Au}R)}{H_{\mathrm{hf}}(\mathrm{Au}Gd)} = \frac{\tilde{m}_0(\mathrm{Au}R)}{\tilde{m}_0(\mathrm{Au}Gd)}. \tag{12}$$

A more complete theoretical description of the behavior of the magnetic hyperfine fields of s-p and noble Mössbauer impurities placed in these heavy rare-earth ferromagnetic metals, based on an extended Daniel–Friedel model[17,20] will be presented elsewhere.

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