Local magnetic moment formation at ^{119}Sn Mössbauer impurity in RCo_2 (R=Gd,Tb,Dy,Ho,Er) Laves phase compounds

A. L. de Oliveira,[1,a)] N. A. de Oliveira,[2] and A. Troper[2,3]
[1]*Centro Federal de Educação Tecnológica de Química de Nilópolis/RJ, Rua Lúcio Tavares 1045, Nilópolis, Rio de Janeiro 26530-060, Brazil*
[2]*Instituto de Física, Universidade do Estado do Rio de Janeiro, Rua São Francisco Xavier 524, Rio de Janeiro 20550-013, Brazil*
[3]*Centro Brasileiro de Pesquisas Físicas, Rua Dr. Xavier Sigaud 150, Rio de Janeiro 22290-180, Brazil*

(Presented on 8 November 2007; received 16 October 2007; accepted 24 October 2007; published online 15 February 2008)

In this work, we theoretically study the local magnetic moment formation and the systematics of the magnetic hyperfine fields at a Mössbauer ^{119}Sn impurity diluted at the R site (R=Gd,Tb,Dy,Ho,Er) of the cubic Laves phase intermetallic compounds RCo_2. One considers that the magnetic hyperfine fields have two contributions, (i) the contribution from R ions, calculated via an extended Daniel-Friedel [J. Phys. Chem. Solids **24**, 1601 (1963)] model, and (ii) the contribution from the induced magnetic moments arising from the Co neighboring sites. Our calculated self-consistent total magnetic hyperfine fields are in a good agreement with recent experimental data. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2832668]

The description of the formation of the local magnetic moments and its connection to hyperfine interactions on impurities embedded in metallic systems have been challenging condensed matter theorists since decades. Initially, model approaches[1–9] have been proposed in which the electronic structures of the systems have been oversimplified in order to mimic the main features of the electronic metallic hosts. In these models, the scattering problem associated with by the impurity has been described using some empirical parameters.[1–9] On the other hand, the development of the density functional theory and its implementation to *ab initio* computational schemes yielded to an excellent understanding of the ground state properties of a large variety of systems.[10,11]

The purpose of this work is to theoretically study the formation of the local magnetic moments and the systematics of the magnetic hyperfine fields at a nonmagnetic *s-p* Sn impurity diluted on the R site of the Laves phase intermetallic compounds RCo_2. In order to calculate the local magnetic moment at a Sn impurity diluted in RCo_2, we use a Daniel-Friedel-like model,[12] from R ions, and an extra magnetic term, located at a distance $\mathbf{R}_0$ from the impurity site, to account for the contribution arising from the Co magnetic moments. We have in this case a two-center Blandin-Campbell-like problem,[13–15] where a magnetic 3*d* element located at a distance $\mathbf{R}_0$ from the Sn impurity gives an extra magnetization to a polarized electron gas which is strongly charge perturbed at the impurity site. We also include in the model, the nearest-neighbor perturbation due to the translational invariance breaking introduced by the impurity.[16–18]

We start the description of a Sn impurity diluted in RCo_2 intermetallic hosts by considering that the local magnetic moments and the total magnetic hyperfine fields have two important contributions, namely, (i) the contribution from R ions and (ii) the contribution from the coupling with the induced magnetic moments at the Co sites.[19] In order to account for both contributions to the formation of the local magnetic moments, we consider the RCo_2 intermetallic compounds as being an effective ferromagnetic rare-earth hosts,[15] with an extra term to account for the polarization arising from the magnetic moments of the transition elements surrounding the impurity site.[15] In the framework of our model, the Hamiltonian to describe the formation of the local *s-p* magnetic moment and hyperfine field at Sn impurity embedded in RCo_2 intermetallic hosts is given by

$$H = \sum_{i\neq 0,\sigma} \varepsilon_{i\sigma}^{c,h} c_{i\sigma}^{+} c_{i\sigma} + \sum_{i,j,\sigma} t_{ij}^{cc} c_{i\sigma}^{+} c_{j\sigma} + \varepsilon_{0\sigma}^{c,I} c_{0\sigma}^{+} c_{0\sigma} + \tau_c \sum_{i\neq 0,\sigma} t_{0l}^{cc} (c_{0\sigma}^{\dagger} c_{i\sigma} + c_{i\sigma}^{\dagger} c_{0\sigma}) + \sum_{l\neq 0,\sigma} V_{\mathrm{nn}\sigma}^{c} c_{l\sigma}^{\dagger} c_{l\sigma}. \quad (1)$$

In this Hamiltonian, $\varepsilon_{i\sigma}^{c,h}$ is the center of the *s-p* energy band, which depends on the 4*f* polarization of the effective rare-earth host. $\varepsilon_{0\sigma}^{c,I}$ is the *s-p* impurity state energy level. The term t_{ij}^{cc} is the electron hopping energy between i and j sites in the same *s-p* energy band. The parameter τ_c takes into account the change in the hopping energy associated with the presence of the impurity.[16–18,20] $V_{\mathrm{nn}\sigma}^{c}=-\sigma J^{\mathrm{sd}}\langle S^{\mathrm{Co}}\rangle$ is the change in band center due to the magnetic moments of the neighboring Co ions, where J^{sd} is an exchange parameter of the model.

Let us first solve the problem with $V_{\mathrm{nn}\sigma}^{c}=0$. We use the Hartree-Fock approximation to treat the electron-electron interaction. In this approach and using the Dyson equation, the perturbed Green's function $\tilde{g}_{ij\sigma}(z)$ due to the charge perturbation at the origin after some algebra is[21]

[a)]Electronic mail: alexandreo@cefeteq.br.

$$\tilde{g}^{cc}_{ij\sigma}(z) = g^{cc}_{ij\sigma}(z) + g^{cc}_{i0\sigma}(z)\frac{V^{c}_{\mathrm{eff}\ \sigma}(z)}{\alpha^2 - g_{00\sigma}(z)V^{c}_{\mathrm{eff}\ \sigma}(z)}g^{cc}_{0j\sigma}(z)$$
$$+ (\alpha_c - 1)^2\frac{g^{cc}_{00\sigma}(z)\delta_{i0}\delta_{0j}}{\alpha_c^2 - g^{cc}_{00\sigma}(z)V^{c}_{\mathrm{eff}\ \sigma}(z)}$$
$$- (\alpha_c - 1)\frac{\alpha_c[g^{cc}_{i0\sigma}(z)\delta_{0j} + \delta_{i0}g^{cc}_{0j\sigma}(z)]}{\alpha_c^2 - g^{cc}_{00\sigma}(z)V^{c}_{\mathrm{eff}\ \sigma}(z)}. \quad (2)$$

Here, $g^{cc}_{ij\sigma}(z)$ is the Green's function for the pure host with $z=\varepsilon+i0$. The nonlocal potential V^{c}_{σ} is given by $V^{c}_{\sigma}=V^{c}_{0\sigma}+\tau_c\Sigma_{i\neq 0\sigma}t^{cc}_{0l}(c^{\dagger}_{0\sigma}c_{i\sigma}+c^{\dagger}_{i\sigma}c_{0\sigma})$, where $V^{c}_{0\sigma}=(\varepsilon^{c,l}_{0\sigma}-\varepsilon^{c,h}_{0\sigma})$ is the local term, corresponding to the generalized Daniel-Friedel potential. The effective potential $V^{c}_{\mathrm{eff}\sigma}(z)$ is given by $V^{c}_{\mathrm{eff}\sigma}(z)=V^{c}_{0\sigma}+(\alpha_c^2-1)(z-\varepsilon^{c})$, with $\alpha_c=\tau_c+1$ and ε^{c} being the center of the *s-p* energy band.

Let us now solve the general problem defined by the Hamiltonian (1). In fact, one considers a magnetic 3*d*-element (Co) located at a distance $\mathbf{R}_0$ from a Sn impurity, which gives an extra magnetization to the strongly charge perturbed and polarized electron gas, defined by Eq. (2). The perturbed Green's function $G^{cc}_{ij\sigma}(z)$ is given by

$$G^{cc}_{ij\sigma}(z) = \tilde{g}^{cc}_{ij\sigma}(z) + \sum_{l\neq 0}\tilde{g}^{cc}_{il\sigma}(z)T^{c}_{l\sigma}\tilde{g}^{cc}_{lj\sigma}(z), \quad (3)$$

with $T^{c}_{l\sigma}=V^{c}_{\mathrm{nn}\sigma}[1-g^{cc}_{ll\sigma}(z)V^{c}_{\mathrm{nn}\sigma}]^{-1}$. Hence, the local Green's function $G^{cc}_{00\sigma}(z)$ at the impurity site can be written after some manipulation as[21]

$$G^{cc}_{00\sigma}(z) = \frac{g^{cc}_{00\sigma}(z)}{\alpha_c^2 - g^{cc}_{00\sigma}(z)V^{c}_{\mathrm{eff}\ \sigma}(z)} + \frac{\alpha_c^2 V^{c}_{\mathrm{nn}\sigma}}{[\alpha_c^2 - g^{cc}_{00\sigma}(z)V^{c}_{\mathrm{eff}\ \sigma}(z)]^2}$$
$$\times\left\{Z_{\mathrm{nn}}\frac{\partial g^{cc}_{00\sigma}(z)}{\partial z} + [g^{cc}_{00\sigma}(z)]^2\right\}. \quad (4)$$

Here, we assume that the spin potential $V^{c}_{\mathrm{nn}\sigma}$ is small as compared to the strong local charge and spin perturbation $V^{c}_{0\sigma}$; we take the Born approximation, i.e., $T^{c}_{l\sigma}\approx V^{c}_{\mathrm{nn}\sigma}$. Z_{nn} is the number of the first next neighbors of Co ions surrounding a Ta impurity (in the present case: $Z_{\mathrm{nn}}=8$). Note that in the local Green's function $G^{cc}_{00\sigma}(z)$, the first term is due to the rare-earth ions, whereas the second term comes from the polarization of Co ions. The local potential $V^{c}_{0\sigma}$ is self-consistently determined using the Friedel screening condition[2,20] for the total charge difference between impurity and host atoms. Here, we assume that the screening of the *s-p* charge difference is made by the *s-p* band, i.e., $\Delta Z^{c}=\Delta Z^{c}_{\uparrow}+\Delta Z^{c}_{\downarrow}$, where ΔZ^{c}_{σ}, $\sigma=\uparrow$ or $\downarrow$, is given by

$$\Delta Z^{c}_{\sigma} = -\frac{1}{\pi}\,\mathrm{Im}\ln[\alpha_c^2 - g^{cc}_{00\sigma}(z)V^{c}_{\mathrm{eff}\ \sigma}(z)]. \quad (5)$$

The local *s-p* density of states per spin direction at the impurity site is calculated by $\rho^{c}_{\sigma}(\varepsilon)=(-1/\pi)\mathrm{Im}\,G^{cc}_{00\sigma}(z)$. The local *s-p* electron occupation number, i.e., $n^{c}_{0\sigma}$, is obtained by integrating the corresponding local density of states up to the Fermi level ϵ_F. The total *s-p* magnetic moment $[\tilde{m}^{c}(0)]$ at a Sn impurity, given by $\tilde{m}^{c}(0)=n^{c}_{0\uparrow}-n^{c}_{0\downarrow}$ is

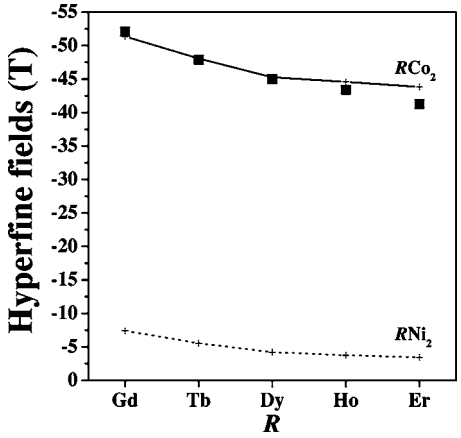

FIG. 2. Calculated total magnetic hyperfine field at Sn impurity in $R\mathrm{Co}_2$ intermetallic hosts (solid line). The dotted line stands for the total magnetic hyperfine field in the case of $R\mathrm{Ni}_2$. The squares represent experimental data collected from Ref. 23.

$$\tilde{m}^{c}(0) = \tilde{m}^{c}_{R}(0) + \tilde{m}^{c}_{\mathrm{ind}}(0), \quad (6)$$

where

$$\tilde{m}^{c}_{R}(0) = -\frac{1}{\pi}\sum_{\sigma}\int_{-\infty}^{\epsilon_F}\mathrm{Im}\,\frac{g^{cc}_{00\sigma}(z)}{\alpha_c^2 - g^{cc}_{00\sigma}(z)V^{c}_{\mathrm{eff}\ \sigma}(z)}dz \quad (7)$$

is the contribution from rare-earth ions and

$$\tilde{m}^{c}_{\mathrm{ind}}(0) = -\frac{1}{\pi}\sum_{\sigma}\int_{-\infty}^{\epsilon_F}\mathrm{Im}\,\frac{\sigma\alpha_c^2 J^{\mathrm{sd}}\langle S^{\mathrm{Co}}\rangle}{[\alpha_c^2 - g^{cc}_{00\sigma}(z)V^{c}_{\mathrm{eff}\ \sigma}(z)]^2}$$
$$\times\left\{Z_{\mathrm{nn}}\frac{\partial g^{cc}_{00\sigma}(z)}{\partial z} + [g^{cc}_{00\sigma}(z)]^2\right\}dz \quad (8)$$

is the contribution from the Co next neighbor ions. The total magnetic hyperfine field at a Sn impurity site is given by

$$B_{\mathrm{hf}} = A(Z_{\mathrm{imp}})\tilde{m}^{c}(0), \quad (9)$$

where $A(Z_{\mathrm{imp}})$ is the Fermi-Segrè contact coupling parameter.[7]

In order to calculate the local moments and the magnetic hyperfine fields at a Sn impurity diluted in $R\mathrm{Co}_2$ intermetallic hosts, we have to fix some model parameters. Here, we adopt a standard paramagnetic *s-p* density of states extracted from first-principles calculations.[22] The exchange splitting in the *s-p* energy bands induced by the local moments of the rare-earth ions was properly chosen to yield the *s-p* magnetic moment at the *R* sites of the host, which is assumed to be of the order of 0.1 of the *d* magnetization at the *R* sites. The parameter which renormalizes the hopping energy involving the impurity site was chosen by taking the ratio between the extension of the host and impurity *s-p* wave functions. In the case of $R\mathrm{Co}_2$ intermetallics, we adopted $J^{\mathrm{sd}}=0.4\times10^{-3}$ for heavy rare-earth ions in units of the *s-p* bandwidth of $R\mathrm{Co}_2$ hosts. The magnetic moments at the Co site in $R\mathrm{Co}_2$ intermetallic compounds $\langle S^{\mathrm{Co}}\rangle$ were estimated from Ref. 19. By keeping these parameters fixed, we self-consistently determined the local magnetic moment and the corresponding magnetic hyperfine field at the Sn impurity.

In Fig. 1, we plot the rare-earth contribution to the local magnetic moment $\tilde{m}_R(0)$ as well as the contribution to the

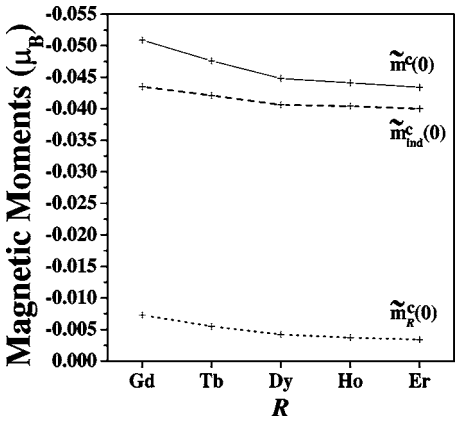

FIG. 1. Local magnetic moments at Sn impurity diluted in $R\mathrm{Co}_2$ intermetallic host. The dotted line corresponds to the contribution from the rare earth $\tilde{m}^c_R(0)$, whereas the dashed line corresponds to the contribution from the Co ions $\tilde{m}^c_{\mathrm{ind}}(0)$. The solid line represents the total magnetic moment $\tilde{m}(0)$.

local magnetic moment, originated from the Co neighboring ions, $\tilde{m}_{\mathrm{ind}}(0)$. The self-consistent calculations show that the Blandin-Campbell-like term[13,14] $\tilde{m}_{\mathrm{ind}}(0)$ dominates over $\tilde{m}_R(0)$, both for heavy and light rare earths. From Fig. 1, one can see that the contribution to the magnetic hyperfine field arising from the Co neighboring ions, $B^{\mathrm{ind}}_{\mathrm{hf}}=A(Z_{\mathrm{imp}})\tilde{m}_{\mathrm{ind}}(0)$, is larger than the contribution originated from the rare-earth ions, $B^R_{\mathrm{hf}}=A(Z_{\mathrm{ind}})\tilde{m}_R(0)$. Moreover, for heavy rare earths, $B^{\mathrm{ind}}_{\mathrm{hf}}$ decreases monotonically, being, however, always greater than B^R_{hf}; i.e., one has again a dominance arising from the contribution from the Co ions.

Figure 2 exhibits the self-consistently calculated magnetic hyperfine fields at a Sn impurity site, $R\mathrm{Co}_2$ (solid line). This figure shows that our theoretical model predicts that as we go from Gd to Er (heavy rare earths), $|B_{\mathrm{hf}}|$ decreases. The total self-consistently calculated magnetic hyperfine fields for $R\mathrm{Co}_2$ hosts plotted in full line exhibit an excellent agreement with available experimental data.[23]

Now, comments on a Sn impurity diluted in $R\mathrm{Ni}_2$ compounds are worthwhile. As previously pointed out,[15,21] $R\mathrm{Ni}_2$ behaves as an effective rare-earth metal, since no magnetization is present in the Ni sites.[19,24] So one assumes that the formation of the local magnetic moment at Sn diluted in $R\mathrm{Ni}_2$ can be described by the Hamiltonian (1), with $V^c_{\mathrm{nn}\sigma}=0$. Systematic measurements of Sn in $R\mathrm{Ni}_2$ shoud be made in order to test further our model.

[1] J. Friedel, Nuovo Cimento, Suppl. **7**, 287 (1958).
[2] E. Daniel and J. Friedel, J. Phys. Chem. Solids **24**, 1601 (1963).
[3] P. A. Wolff, Phys. Rev. **124**, 1030 (1961).
[4] A. M. Clogston, B. T. Matthias, N. Peter, H. J. Williams, E. Orenzwit, and R. J. Sherwood, Phys. Rev. **125**, 541 (1962).
[5] J. Kanamori, J. Appl. Phys. **16**, 929 (1965).
[6] I. A. Campbell and A. A. Gomes, Proc. Phys. Soc. London **91**, 319 (1967).
[7] I. A. Campbell, J. Phys. C **2**, 1338 (1969).
[8] I. A. Campbell, J. Phys. F: Met. Phys. **2**, L47 (1972).
[9] I. A. Campbell, W. D. Brewer, J. Flouquet, A. Benoit, B. W. Mardsen, and N. J. Stone, Solid State Commun. **15**, 711 (1974).
[10] H. Akai, M. Akai, S. Blugel, and B. Dederich, Prog. Theor. Phys. Suppl. **101**, 261 (1990).
[11] P. H. Dederichs, R. Zeller, H. Akai, and H. Herbert, J. Magn. Magn. Mater. **100**, 261 (1991).
[12] A. L. de Oliveira, M. V. Tovar Costa, N. A. de Oliveira, and A. Troper, J. Appl. Phys. **81**, 4215 (1997).
[13] A. Blandin and I. A. Campbell, Phys. Rev. Lett. **31**, 51 (1973).
[14] I. A. Campbell and A. Blandin, J. Magn. Magn. Mater. **1**, 1 (1975).
[15] A. L. de Oliveira, M. V. Tovar Costa, N. A. de Oliveira, and A. Troper, J. Magn. Magn. Mater. **177–181**, 1091 (1998).
[16] W. Speier, J. F. van Acker, and R. Zeller, Phys. Rev. B **41**, 2753 (1990).
[17] J. F. van Acker, W. Speier, and R. Zeller, Phys. Rev. B **43**, 9558 (1991).
[18] J. F. van Acker, W. Speier, J. C. Fuggle, and R. Zeller, Phys. Rev. B **43**, 13916 (1991).
[19] P. de la Presa, S. Müller, A. F. Pasquevich, and M. Forker, J. Phys.: Condens. Matter **12**, 3423 (2000).
[20] N. A. de Oliveira, A. A. Gomes, and A. Troper, Phys. Rev. B **52**, 9137 (1995).
[21] A. L. de Oliveira, N. A. de Oliveira, and A. Troper, Phys. Rev. B **67**, 012411 (2003).
[22] H. Yamada, J. Inoue, K. Terao, S. Kanda, and M. Shimizu, J. Phys. F: Met. Phys. **14**, 1943 (1984).
[23] N. N. Delyagin and V. I. Krylov, J. Phys.: Condens. Matter **19**, 086205 (2007).
[24] A. Troper and A. A. Gomes, Phys. Rev. B **34**, 6487 (1986).