# HYPERFINE FIELDS ON ACTINIDE IMPURITIES IN FERROMAGNETIC Fe AND Ni HOSTS*

A.L. de Oliveira[a], N.A. de Oliveira[b] and A. Troper[a,b]

[a]Centro Brasileiro de Pesquisas Físicas
Rua Dr. Xavier Sigaud, 150, Rio de Janeiro 22290-180, Brazil
[b]Instituto de Física, Universidade do Estado do Rio de Janeiro
Rua São Francisco Xavier, 524, Rio de Janeiro 20550-013, Brazil

*(Received July 10, 2002)*

We discuss the local magnetic moments and magnetic hyperfine fields on actinide impurities diluted in Fe and Ni hosts. One adopts a Anderson–Moriya model in which a localized $5f$ level is hybridized with a spin polarized and charge perturbed $d$-conduction band. Our self-consistent numerical calculations for the hyperfine fields on the impurity sites are in good agreement with the available experimental data.

PACS numbers: 75.10.Lp, 75.20.En, 75.30.Hx

The most part of actinide and rare earth impurities, develop large electronic magnetic moments, composed of both spin and orbital contributions. Therefore, it is very important to consider both spin and orbital magnetic moments in the theoretical calculations on hyperfine fields of these impurities, similarly to Coqblin and Blandin treatment used to discuss the stability of magnetic moments in pure metals [1].

In this work, we want to study the local magnetic moment formation on actinide impurities diluted in Fe and Ni hosts and to bring out the fundamental role played by the orbital contribution. We use an Anderson model Hamiltonian [2, 3] in which the $5f$ impurity level is coupled to a spin polarized $d$-band. In what follows we adopt the same approach used to discuss magnetic hyperfine fields and local moments of the series of lanthanide impurities diluted in Fe, Co and Ni hosts [4], where we can find more details about the model and the theoretical calculation. We start with the following

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

Hamiltonian [1]

$$H = H_0 + V^d_{0\sigma} + \tau_d \sum_{j\neq 0,\sigma} t^d_{0j}\left(d^\dagger_{0\sigma} d_{j\sigma} + d^\dagger_{j\sigma} d_{0\sigma}\right) + V^f + V^{df}_0 \sum_{m,\sigma}\left(d^\dagger_{0\sigma} f_{0m\sigma} + f^\dagger_{0m\sigma} d_{0\sigma}\right), \tag{1}$$

where

$$V^f = \sum_{m,\sigma} \varepsilon^f_0 f^\dagger_{0m\sigma} f_{0m\sigma} + \sum_{m,m',\sigma} \frac{U^f_{mm'}}{2} n^f_{m\sigma} n^f_{m'-\sigma} + \sum_{\substack{m,m',\sigma \\ (m\neq m')}} \frac{\widetilde{U}^f_{mm'}}{2} n^f_{m\sigma} n^f_{m'\sigma}. \tag{2}$$

$H_0$ is the Hamiltonian for the $d$-band of the pure ferromagnetic host; $V^d_{0\sigma}$ is a local spin dependent impurity potential which is self-consistently determined using the Friedel screening condition for the total charge difference [4,5]. $\tau_d$ is an impurity-dependent parameter which renormalizes the energy hopping between impurity and host sites with respect to the hopping energy between one host site and another [4–6]. $V^f$ describes the degenerate impurity actinide $5f$ level where $m$ and $m'$ denote $f$ orbital labels. $V^{df}_0$ describes the coupling between the degenerate $f$-level and the $d$-band. $d^\dagger_{j\sigma}$ ($d_{j\sigma}$) is the creation (annihilation) operator at site $j$ with spin $\sigma$, $t^d_{jl\sigma}$ is the $d$-electron energy hopping. $U^f_{mm'}$ is the usual Coulomb interaction, which can be intra and inter-orbital. In principle $U^f_{mm}$ may be different from $U^f_{mm'}$ ($m'\neq m$); $\widetilde{U}^f_{mm'} = (\bar{U}^f_{mm'} - J^f_{mm'})$, $m'\neq m$. $\bar{U}^f_{mm'}$ and $J^f_{mm'}$ are the Coulomb and exchange correlations between *different* $5f$ orbitals. In Eq. (2), for simplicity, we neglect spin-flip terms between actinide ions.

Following the same approach as in Ref. [4], one obtains the $d$- and $f$-occupation numbers at the impurity site, with spin $\sigma$:

$$\widetilde{n}^d_{0m\sigma} = -\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{\varepsilon_{\mathrm{F}}} \frac{\left(z - \varepsilon^f_{m\sigma}\right)\widetilde{g}^{dd}_{00}(z)}{z - \varepsilon^f_{m\sigma} - \left|V^{df}_0\right|^2 \widetilde{g}^{dd}_{00}(z)}\,\mathrm{d}z\,, \tag{3}$$

$$\widetilde{n}^f_{0m\sigma} = -\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{\varepsilon_{\mathrm{F}}} \frac{1}{z - \varepsilon^f_{m\sigma} - \left|V^{df}_0\right|^2 \widetilde{g}^{dd}_{00}(z)}\,\mathrm{d}z\,, \tag{4}$$

where $\varepsilon_{\mathrm{F}}$ is the Fermi level. The local Green function $\widetilde{g}^{dd}_{00}(z)$ is calculated disregarding the $5f$ level and considering only the charge and spin perturbation due to the $7s6d$ states of the impurity [4,5]. $\varepsilon^f_{m\sigma}$ is the renormalized energy level in the mean field approximation, and is given by

$$\varepsilon^f_{m\sigma} = \varepsilon^f_0 + \sum_{m'} U^f_{mm'} \left\langle n^f_{m'-\sigma} \right\rangle + \sum_{m' \neq m} \widetilde{U}^f_{mm'} \left\langle n^f_{m'\sigma} \right\rangle . \qquad (5)$$

The $6d$ contribution to the magnetic moment at the impurity site is given by $\widetilde{m}_d(0) = \sum_m \left(\widetilde{n}^d_{0\uparrow} - \widetilde{n}^d_{0\downarrow}\right)$. The total $f$ magnetic moment at the impurity site is given by $\widetilde{m}_f(0) = \widetilde{m}^{\rm spin}_f(0) + \widetilde{m}^{\rm orb}_f(0)$, where $\widetilde{m}^{\rm spin}_f(0) = \sum_m \left(\widetilde{n}^f_{0m\uparrow} - \widetilde{n}^f_{0m\downarrow}\right)$ and $\widetilde{m}^{\rm orb}_f(0) = \sum_m m \left(\widetilde{n}^f_{0m\uparrow} + \widetilde{n}^f_{0m\downarrow}\right)$ are, respectively, the spin and orbital contributions. So, the total local magnetization at the impurity site is given by $\widetilde{m}(0) = \widetilde{m}_d(0) + \widetilde{m}_f(0) + \widetilde{m}_c(0)$. We assume that the $s$–$p$ local moment $\widetilde{m}_c(0)$ is antiparallel and proportional to the $d$-host magnetization [5], *i.e.*, $\widetilde{m}_c(0) = -\gamma m_d = -\gamma \sum_\sigma \sigma\, n^d_\sigma$, with the proportionality constant $\gamma$ being of the order of 0.1 [5]. Once the magnetic moments are determined, the total hyperfine field $B^{\rm tot}_{\rm hf}$ is given by:

$$B^{\rm tot}_{\rm hf} = B^{\rm c}_{\rm hf} + B^{\rm d}_{\rm hf} + B^{\rm f}_{\rm hf} = A(Z_{\rm imp})\widetilde{m}_c(0) + A^{6d}_{\rm cp}\widetilde{m}_d(0) + A^{5f}_{\rm cp}\widetilde{m}_f(0), \qquad (6)$$

where $A(Z_{\rm imp})$ is the impurity dependent Fermi–Segrè coupling, $A^{6d}_{\rm cp}$ and $A^{5f}_{\rm cp}$ are the core polarization coupling parameters for $6d$ and $5f$ electrons, respectively [7].

In order to obtain numerical results, we considered the parameter $U^f_{mm'}$ to be independent of the $5f$-level, *i.e.*, $U^f_{mm'} = U^f_{mm} = U^f$ and we adopted $U^f = 0.9$ (in half bandwidth units). Moreover, we adopted for $\widetilde{U}_m = \widetilde{U}_0 = 0.9U^f$ (case of Ac impurity, where orbital contribution is absent). For the other actinide impurities, we considered $\widetilde{U}_m$ ($|m| = 0, 1, 2, 3$) assuming values with small variations about $\widetilde{U}_0$.

In Fig. 1 we plot the calculated magnetic hyperfine fields on actinide impurities in Fe and Ni hosts. One obtains a good agreement with the available experimental data. Moreover, in the case of U$\underline{\rm Fe}$, Np$\underline{\rm Fe}$ and Pu$\underline{\rm Fe}$ our calculations are in excellent agreement with first principles calculations [10]. Some discrepancies with experiments are also observed, *e.g.*, in the cases of Th$\underline{\rm Fe}$, Pu$\underline{\rm Fe}$. This is because the experiments were performed at $T \approx 300$K, whereas our calculations (as well the ab initio calculation for U$\underline{\rm Fe}$, Np$\underline{\rm Fe}$ and Pu$\underline{\rm Fe}$) were performed at $T = 0$K [10]. The core polarization contributions, which reflect at $T = 0$K, the "accidents" of the $d$-band as well as the Moriya $f$-hump are strongly affected by temperature and a decrease in the core polarization is expected, whereas the $s$-$p$ conduction electron contribution remains mainly unaffected by temperature. Therefore, the total hyperfine field would decrease, and so being in accordance with the available

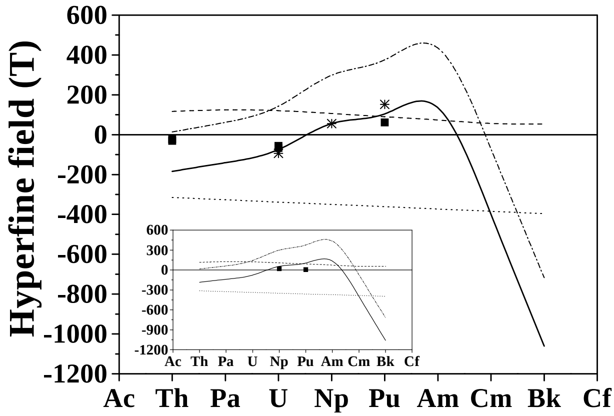


Fig. 1. Systematics of the magnetic hyperfine fields on the actinide impurities in Fe host. The solid line corresponds to the total hyperfine field, the dotted line, dashed line and dash-dotted line correspond to $B_{\rm hf}^{\rm c}$, $B_{\rm hf}^{\rm d}$ and $B_{\rm hf}^{\rm f}$, respectively. The experimental data for Fe (squares) host have been collected from Refs. [8, 9] and the stars correspond to first principles calculations results [10]. Inset: The same for Ni host.

experimental data [8,9]. In Fig. 2 we plot the contributions to the magnetic moments as well as the total local magnetic moments. One sees that the behaviour is almost the same as in the case of rare earth impurities, a change in sign of the total moments occurring around the middle of the series [4].

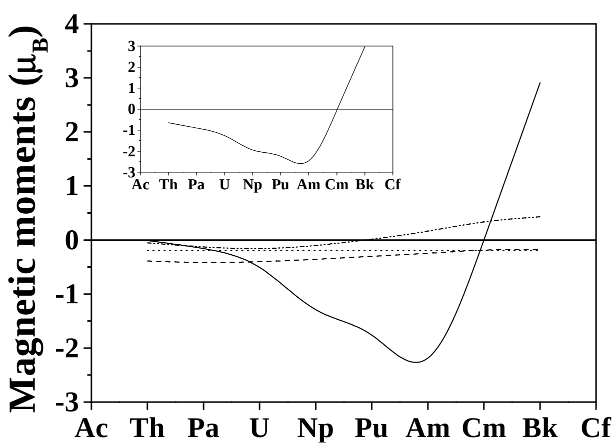


Fig. 2. Systematics of the magnetic moments on the actinide impurities in Fe host. The solid line corresponds to $\widetilde{m}_f^{\rm orb}(0)$, the dash-dot-dotted line to $\widetilde{m}_f^{\rm spin}(0)$, the dashed line to $\widetilde{m}_d(0)$ and the dotted line to $\widetilde{m}_c(0)$. Inset: The total magnetic moments, $\widetilde{m}(0)$, on the actinide impurities in Fe.

We would like to thank Conselho Nacional de Desenvolvimento Científico e Tecnológico — CNPq for financial support. This work was partially performed under the frame of PRONEX, project number 66201998-9.

## REFERENCES

[1] B. Coqblin, A. Blandin, *Adv. Phys.* **17**, 281 (1968).

[2] P.W. Anderson, *Phys. Rev.* **124**, 41 (1961).

[3] T. Moriya, *Prog. Theor. Phys.* **34**, 329 (1965).

[4] A.L. de Oliveira, N.A. de Oliveira, A. Troper, *J. Phys.: Condens. Matter* **14**, 1949 (2002).

[5] N.A. de Oliveira, A.A. Gomes, A. Troper, *Phys. Rev.* **B52**, 9137 (1995).

[6] J.F. van Acker, W. Speier, J.C. Fuggle, R. Zeller, *Phys. Rev.* **B43**, 13916 (1991) and references therein.

[7] R. Jullien, A.A. Gomes, B. Coqblin, *Phys. Rev. Lett.* **29**, 482 (1972).

[8] G.N. Rao, *Hyperfine Interact.* **26**, 1119 (1985).

[9] B.D. Dunlap, G.M. Kalvius, in *Handbook on the Physics and Chemistry of the Actinides*, North-Holland, Amsterdam 1995, p. 392.

[10] A.B. Shick, V.A. Gubanov, *Phys. Rev.* **B49**, 12860 (1994).