Transition-metal impurities in Fe: Magnetic- and hyperfine-field properties

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A description of the local properties, hyperfine field, and local magnetic moment of dilute transition impurities in Fe, including the next-neighbor perturbation is presented. The Hartree-Fock approximation for the electron-electron interaction is adopted, as in the usual description of these alloys. The use of only a few parameters and a simple self-consistent procedure is enough to account for the observed experimental trends in the hyperfine fields of these systems.

INTRODUCTION

The formation of local moments and its connection to hyperfine interactions is one of the central problems in the description of the magnetic properties of metals.[1–5] The case of rare earths diluted in ferromagnetic hosts[6–8] has also been studied. Recently a formulation,[9] including the change in hopping between the impurity and next neighbors, applied to the case of Pd-based alloys, suggested a connection between first-principles calculations[10,11] and the model approaches for the impurity problem, as the classical Clogston-Wolff picture.[3]

In this work, the formulation of Ref. 9 is used to describe the magnetic and hyperfine properties of the general case of *nd* transition impurities dissolved in a ferromagnetic Fe host. A parametrized tight-binding approximation is adopted; this approach furnishes a simple theoretical framework to describe dilute impurities in a ferromagnetic host, providing a good description of the observed trends in the experimental data.

We include also the *sp* band to describe hyperfine interactions. The *sp* magnetization is taken proportional to the *d* one,[12] with proportionality constant, $\gamma(Z_{\rm imp})$. Here we use the qualitative description adopted in Refs. 3 and 14, where the hyperfine field is connected to the host magnetization, via the Fermi-Segré term $A(Z_{\rm imp})$, plus a core polarization contribution, proportional to the local *d* magnetization, multiplied by an effective coupling constant $A_{\rm cp}$. These quantities are both extracted from Ref. 13.

The plan of the paper is as follows. We first present the formulation, the involved approximations, and the obtained numerical results. Finally, we present a general discussion and the conclusions.

FORMULATION

We start from the *d* part of the transition impurity problem in the Fe ferromagnetic host, using the following single-band Hamiltonian:

$$H=\sum_{\sigma}\left[\sum_{jl\neq 0} t_{jl}a^{\dagger}_{j\sigma}a_{l\sigma}+\sum_{j\neq 0}U^{h}n_{j_{\uparrow}}n_{j_{\downarrow}}+\sum_{j\neq 0}\tilde{t}_{0j}(a^{\dagger}_{0\sigma}a_{j\sigma}+a^{\dagger}_{j\sigma}a_{0\sigma})\right]+(\varepsilon^{I}-\varepsilon^{c})a^{\dagger}_{0\sigma}a_{0\sigma}+U^{I}n_{0\uparrow}n_{0\downarrow}\ , \tag{1}$$

where $a^{\dagger}_{j\sigma}$ ($a_{j\sigma}$) is the creation (annihilation) operator, t_{jl} is the hopping between host sites, and $\tilde{t}_{0j}$ is the corresponding to processes involving the impurity site. The term ε^{I} is the energy level of impurity state, ε^{c} is the host-metal band center, and U^{h} (U^{I}) is the Coulomb interaction between electrons at the host (impurity) site.

Using the Hartree-Fock approximation to deal with the Coulomb interactions, one gets for the Hamiltonian

$$H=H_{0}+V_{\sigma}\ , \tag{2a}$$

$$H_{0}=\sum_{jl\sigma}t_{jl}a^{\dagger}_{j\sigma}a_{l\sigma}+\sum_{j\sigma}U^{h}\langle n_{j-\sigma}\rangle n_{j\sigma}\ , \tag{2b}$$

$$V_{\sigma}=(\Delta+\Delta U\langle n_{-\sigma}\rangle+U^{I}\delta\langle n_{0-\sigma}\rangle)a^{\dagger}_{0\sigma}a_{0\sigma}+\tau\sum_{j\neq 0}t_{0j}(a^{\dagger}_{0\sigma}a_{j\sigma}+a^{\dagger}_{j\sigma}a_{0\sigma})\ , \tag{2c}$$

where τ is an impurity-dependent parameter, describing the difference in hopping, between impurity-host and host-host sites, $\Delta=\varepsilon^{I}-\varepsilon^{h}$, $\Delta U=U^{I}-U^{h}$, and $\delta\langle n_{0\sigma}\rangle=\langle n_{\sigma}\rangle-\langle n_{0\sigma}\rangle$. H_{0} is the Hamiltonian of the unperturbed system, and V_{σ} is the potential due to the presence of the impurity. For $\tau=0$ this potential reduces to the case where the hopping is site independent.[5,14]

The potential V_{σ} must be determined consistently with Friedel's sum rule. In this case the problem is more complicated than the usual one. To solve it, we use Dyson's equation, obtaining

$$G^{\sigma}_{jl}=g^{\sigma}_{jl}+g^{\sigma}_{j0}V^{\sigma}_{0}G^{\sigma}_{0l}$$
$$+\tau g^{\sigma}_{j0}\sum_{m\neq 0}t_{0m}G^{\sigma}_{ml}+\tau\left[\sum_{m\neq 0}g^{\sigma}_{jm}t_{m0}\right]G^{\sigma}_{0l} \quad (3a)$$

with

$$V^{\sigma}_{0}=\Delta+\Delta U\langle n_{-\sigma}\rangle+U^{I}\delta\langle n_{0-\sigma}\rangle\ . \quad (3b)$$

Following the same procedure used in Ref. 9, one has for the change in the density of states

$$\Delta\rho_{\sigma}=\frac{1}{\pi}\mathrm{Im}\frac{-(\alpha^2-1)g^{\sigma}_{00}-[(z-\varepsilon_c)(\alpha^2-1)+V^{\sigma}_{0}]\partial g^{\sigma}_{00}/\partial z}{\alpha^2-g^{\sigma}_{00}[(z-\varepsilon_c)(\alpha^2-1)+V^{\sigma}_{0}]}\ , \quad (4)$$

where $z=\varepsilon+i\delta$ and $\alpha=\tau+1$. Integrating this expression up to the Fermi level (ε_F), one gets for the change in the occupation of σ spin states:

$$\Delta Z_{\sigma}=(-1/\pi)\mathrm{Im}\ln\{\alpha^2-g^{\sigma}_{00}[(\varepsilon_F-\varepsilon_c)(\alpha^2-1)+V^{\sigma}_{0}]\}\ . \quad (5)$$

The potentials V^{σ}_{0} are self-consistently determined using Friedel's screening condition for the total charge difference $\Delta Z=\Delta Z_{\uparrow}+\Delta Z_{\downarrow}$. Once these potentials are obtained, the magnetic moment at the impurity site is calculated using $\tilde{m}_d=n_{0_{\uparrow}}-n_{0_{\downarrow}}$. The occupation numbers at the impurity site are given by

$$n_{0\sigma}=-\frac{1}{\pi}\int_{-\infty}^{\varepsilon_F}\mathrm{Im}\frac{g^{\sigma}_{00}}{\alpha^2-g^{\sigma}_{00}[(z-\varepsilon_c)(\alpha^2-1)+V^{\sigma}_{0}]}dz\ . \quad (6)$$

Within the simplifying approximations used in Refs. 5 and 14 to describe the *d*-induced *sp* magnetization and with the notation defined above, one has for the hyperfine fields

$$H_{\mathrm{hf}}=-A_{\mathrm{cp}}\tilde{m}_d+A(Z_{\mathrm{imp}})m_s$$
$$=-A_{\mathrm{cp}}\tilde{m}_d-A(Z_{\mathrm{imp}})\gamma(Z_{\mathrm{imp}})m_d\ , \quad (7)$$

where $m_s=-\gamma(Z_{\mathrm{imp}})m_d$ and m_d is the *d*-host magnetization.

NUMERICAL RESULTS

The numerical calculations were performed starting from a density of states previously used.[14] The parameters of the present model are the host and impurity Coulomb interaction strengths and also the α and γ quantities; cf. Eqs. (4) and (7). Assuming suitable values for the Coulomb interaction parameters and pure host spin-dependent occupation numbers combined with Friedel's sum rule, we self-consistently determine the impurity potentials via an interactive numerical procedure.

The value of the parameter α decreases from the beginning to the end of the same series and up a column in the Periodic Table. This is a consequence of the decreasing extent of the impurity wave function. Its value for each impurity was determined (all other parameters were fixed) by the best fit between the calculated local magnet-

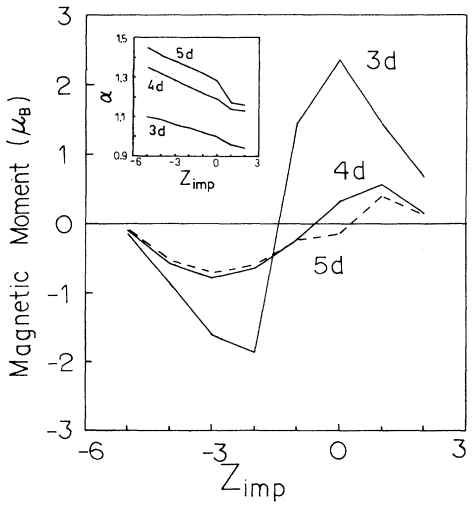

FIG. 1. Calculated magnetic moments for *nd* series impurities in Fe. Inset: Hopping matrix elements α as a function of the position in the Periodic Table for the *nd* series.

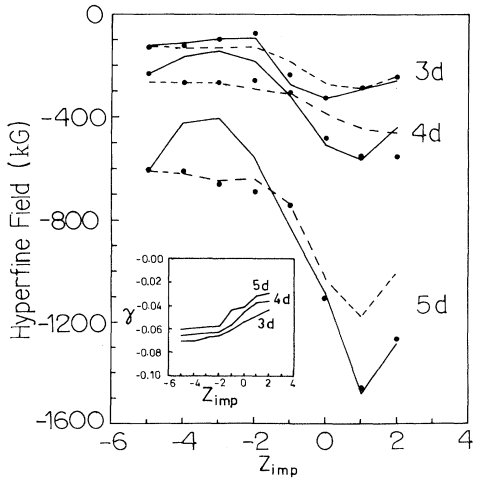

FIG. 2. Hyperfine fields for the *nd* series impurities in Fe. The full line corresponds to the present calculation, the dashed line corresponds to the results of Refs. 9 and 10, and the circles are the experimental data. Inset: variation of the γ parameter as a function of the position in the Periodic Table for the *nd* series.

ic moment by the present method and the first-principles calculations;[9,10] the adjusted values are shown in the inset of Fig. 1. The final values for the d-magnetic moments at the impurity site, are shown in Fig. 1.

The parameter γ depends also on the impurity atom in Fe. This is to be associated to the origin of the antiparallel alignment of the host sp polarization with respect to the d one,[14] driven by the sp-d hybridization. The extent of the impurity d-wave function modifies the hybridization matrix element, introducing the impurity dependence indicated by $\gamma(Z_{\text{imp}})$. The existing hyperfine field data enabled us to fix the values of γ as a function of Z_{imp}. The results of this fit are presented in the inset of Fig. 2. These changes are directly connected to the hybridization strength of the impurity nd states with the *conduction* $4sp$ *states*. The obtained hyperfine fields for impurities of the $3d$, $4d$, and $5d$ series are shown in Fig. 2.

We observe a good agreement with experimental data for the $3d$ series. In the other cases there is a deviation in the beginning of the series, except for the first element used to fix γ. This deviation may be a consequence of the absence of a local impurity-induced sp-d hybridization in the present model Hamiltonian.

For comparison, we also show the hyperfine field calculations extracted from Refs. 9 and 10. Note that first-principles calculations deviate from experiment at the end of the series, a region where our model is in fair agreement with experiment. Further study within our model, including the additional impurity-induced hybridization term, is required.

FINAL DISCUSSIONS AND CONCLUSIONS

In this work we have used an extension of the classical parametrized tight-binding approach[5] to include finer details of the impurity host hopping and a simple picture of the hybridization-induced sp host polarization essential[13] for hyperfine field calculations. The motivation and interest of this calculation is to exhibit in detail the intrinsic merits and limitations of our parametrized and simple approach, as compared to first-principles calculations, in what concerns local magnetic moments and hyperfine fields. We conclude, in agreement with Refs. 13 and 5, that a series of improvements to the Wolff-Clogston formulation[3] are enough to provide a reasonable description of experimental data for $3d$ impurities. Also the period dependence of the deviations from experiment and first-principle calculations indicates that our method is adequate not only for pure $3d$ ferromagnetic hosts but also for other $3d$ intermetallic compounds.

Finally we want to stress that sizeable discrepancies, with respect to experimental data, still occur in first-principles approaches for $4d$ and $5d$ impurities in Fe. This encourages us to proceed with working along the above-suggested lines of including impurity-induced hybridization effects in our formulation.

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