J. Braz. Chem. Soc., Vol. 19, No. 2, 268-272, 2008.
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0103 - 5053 $6.00+0.00

Article

Local Magnetic Moments and Hyperfine Fields at Ta Impurities Diluted in $X\text{Fe}_2$ (X = Y, Gd, Yb) Laves Phases Compounds

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Neste trabalho, estudamos, à temperatura finita, a sistemática na formação dos momentos locais de uma impureza de Ta diluída em compostos intermetálicos de fases de Laves $X\text{Fe}_2$ (X = Y, Gd, Yb). Utilizamos um hamiltoniano de Hubbard, extendido à duas sub-redes acopladas, para descrever as matrizes de fases de Laves. A interação eletrônica *d-d* é tratada via método da integral funcional, na aproximação quase estática. Efeitos da pressão dependentes da temperatura estão incluídos, considerando a interação elétron-fônon induzida que renormaliza a hibridização do elétron puro. Os campos hiperfinos magnéticos calculados, relacionados aos momentos magnéticos locais, estão em bom acordo com os dados experimentais avaliados.

In this work, we study the systematics, at finite temperature, of the formation of local magnetic moments at a Ta impurity diluted in intermetallic Laves phases compounds $X\text{Fe}_2$ (X = Y, Gd, Yb). We use an extended two-coupled sublattice Hubbard Hamiltonian, to describe the Laves phases host. The *d-d* electronic interaction is treated via a functional integral approach in the quasi-static saddle point approximation. Temperature dependent pressure effects are included considering induced electron-phonon interaction which renormalizes the pure electron hybridization. The calculated magnetic hyperfine fields related to the obtained local magnetic moments, are in a quite good agreement with available experimental data.

Keywords: intermetallic compounds, magnetic hyperfine fields, impurities

Introduction

The Laves phase intermetallic compounds $X\text{Fe}_2$ crystallize either in the cubic C15 or hexagonal C14 structure.[1] They exhibit an interesting variety of behaviors related to the changes in their magnetic, electronic, and lattice structures. For instance, in the intermetallic compound YFe_2 the magnetic order is sustained mainly by the itinerant electrons of the Fe sublattice and a small magnetic moment is induced at the Y sublattice. On the other hand the compounds of GdFe_2 and YbFe_2 exhibit magnetization associated both with the localized spins of the rare earth sublattice and the itinerant electrons of Fe sublattice. Although the rare earth Laves phase intermetallic compounds have been extensively studied, many properties of them still remain open. Therefore, the investigation of the magnetic hyperfine fields in the Laves phases intermetallic compounds may provide valuable information on the magnitude of the itinerant (*3d*) and also, in the case of rare earth intermetallic compounds, the localized (*4f*) magnetic moments. In this work we study, at finite temperature, the formation of the local magnetic moment and the systematics of the magnetic hyperfine fields at a Ta impurity diluted on the X site of the Laves phase intermetallic compounds $X\text{Fe}_2$ (X = Y, Gd, Yb). Experimental data show that Ta impurity diluted in the $X\text{Fe}_2$ intermetallic hosts enters in the X site.[2,3]

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Theoretical Model

In order to calculate the magnetic properties of the $X\mathrm{Fe}_2$ intermetallic hosts, we use a two sublattice Hamiltonian, describing a subsystem of itinerant *d*-electrons coupled with localized *f*-spins of rare earth ions.[4] One has:

$$H = \sum_{l\sigma} \varepsilon_0^X d_{l\sigma}^{\dagger} d_{l\sigma} + \sum_{lj\sigma} T_{lj}^{XX} d_{l\sigma}^{\dagger} d_{j\sigma} + U^X \sum_l n_{l\uparrow} n_{l\downarrow} + \sum_{j\sigma} \varepsilon_0^{\mathrm{Fe}} d_{j\sigma}^{\dagger} d_{j\sigma} + \sum_{jj'\sigma} T_{jj'0}^{\mathrm{FeFe}} d_{j\sigma}^{\dagger} d_{j'\sigma} + U^{\mathrm{Fe}} \sum_j n_{j\uparrow} n_{j\downarrow} + \sum_{lj\sigma} T^{X\mathrm{Fe}} \left(d_{l\sigma}^{\dagger} d_{j\sigma} + d_{j\sigma}^{\dagger} d_{l\sigma} \right) - J_{df} \sum_l S_l^d \cdot S_l^f , \quad (1)$$

where ε_0^X (ε_0^{Fe}) is the energy of the center of *X* (Fe) sublattice, $d_{j\sigma}^{\dagger}$ ($d_{j\sigma}$) is the creation (annihilation) operator, $n_{j\sigma} = d_{j\sigma}^{\dagger} d_{j\sigma}$ is the number operator. T_{lj}^{XX} (T_{lj}^{FeFe}) is the hopping integral between only *X* (Fe) sub-lattice atoms, and $T^{X\mathrm{Fe}}$ is the corresponding to the processes involving *X* and Fe sublattices atoms. J_{df} is an exchange interaction parameter and U^{Fe} (U^X) is the Coulomb interaction parameter for the Fe (*X*). Notice that in the case of the YFe_2 host the last term of the equation (1) does not appear, since there is no magnetic moment at the Y sublattice.

We use the functional integral technique to treat the *d-d* electron-electron interaction.[4-7] In this framework of the functional integral method, the initial Hamiltonian with Coulomb correlations is mapped into an effective one-body Hamiltonian in which the electrons are under the action of fluctuating charge (ν) and spin (ξ) fields. These fluctuating fields which are randomly distributed all over the sites, define a "disorder problem" in both sublattices. We treat this intrinsic disorder in the Coherent Potential Approximation (CPA) and so one defines effective media $\Sigma_\sigma^{\mathrm{Fe}}$ and $\Sigma_\sigma^{\mathrm{X}}$ to restore the translational invariance of the pure Laves phase host. The self-energies are self consistently determined by a CPA equation,[7] and thereby the *X* sublattice host is completely described at any finite temperature.

We consider that the Ta impurity diluted in the *X* sublattice, defines a Wolf-Clogston problem in the effective medium at the *X* sublattice. The local *d*-density of states per spin direction at the impurity site is given by

$$\rho_{0\sigma}(\varepsilon) = -\frac{1}{\pi} \mathrm{Im}\, G_{00\sigma}^{XX}(z), \quad (2)$$

where $G_{00\sigma}^{XX}(z)$ is the local perturbed Green function given by:

$$G_{00\sigma}^{XX}(z) = \frac{g_{00\sigma}^{XX}(z)}{1 - g_{00\sigma}^{XX}(z)\left(\varepsilon_\sigma^{\mathrm{Ta}} - \Sigma_\sigma^X(z)\right)} \quad (3)$$

and $g_{00\sigma}^{XX}(z)$ is the host Green function written in terms of the self-energy. The energy ε_0^{Ta} should be self consistently determined by the Friedel screening condition

$$\Delta Z = \Delta N_\uparrow^X + \Delta N_\downarrow^X , \quad (4)$$

where ΔN_σ^X is the change in the occupation number at the *X* sublattice calculated by:[8]

$$\Delta N_\sigma^X = -\frac{1}{\pi} \mathrm{Im} \ln\left[1 - g_{00\sigma}^{XX}(\varepsilon_F)\left(\varepsilon_\sigma^{\mathrm{Ta}} - \Sigma_\sigma^X(z)\right)\right]. \quad (5)$$

The electron occupation number at the impurity site ($n_{0\sigma}$) is obtained by integrating the local density of states up to the Fermi level ε_F, i.e.,

$$n_{0\sigma} = \int_{-\infty}^{\varepsilon_F} \rho_{0\sigma}(\varepsilon) f(\varepsilon) \mathrm{d}\varepsilon \quad (6)$$

$f(\varepsilon)$ being the Fermi function. The local *d*-magnetic moment at the Ta impurity is then given by

$$\tilde{m}_d(0) = \sum_\sigma \sigma n_{0\sigma}. \quad (7)$$

We also consider the *s-p* sublattice band to describe hyperfine interactions.[9] The local *s-p* magnetic moment is obtained by

$$\tilde{m}_c(0) = -\gamma m_d^X , \quad (8)$$

where m_d^X is the *X* sublattice *d*-magnetization and the parameter γ is of order of 0.1.[9] The total magnetic hyperfine field at the Ta impurity site, is made up by a conduction electron polarization (CEP) contribution due to the *s-p* conduction electrons and by a core polarization (CP) due to the *d* conduction electrons. One has

$$B_{hf} = A(Z_{\mathrm{imp}})\tilde{m}_c(0) + A_{cp}^d \tilde{m}_d(0), \quad (9)$$

where $A(Z_{\mathrm{imp}})$ is the Fermi-Segrè contact coupling parameter and A_{cp}^d is *d*-core polarization parameter.[10]

Results and Discussions

In order to obtain the numerical results for a Ta impurity diluted at $X\mathrm{Fe}_2$ metallic host, we need to follow some self-consistent calculation steps.

Firstly, we describe the intermetallic hosts in the absence of the impurity. Hence we solve two set of equation: *(i)* for the itinerant system (*d*-electrons), and *(ii)* for the localized system due to the *4f* rare earth electrons, in the cases of GdFe_2 and YbFe_2 hosts. So, the electronic and magnetic properties of the system are described properly. Notice

that, as mentioned before the *s* electrons are not considered explicitly and they contribute to the appearance of the local moment through equation (8). Moreover, since the *s-p* band is large and flat, the local *s*-moment is constant with temperature, and is not considerately affected by the presence of the Ta impurity since the charge screening is made by the *d*-band.

Secondly, once the self-consistent calculations for the pure host were made at 0 K temperature, for a fixed set of the model parameters, we determine the self-energies Σ_σ^{Fe} and Σ_σ^{X} for at a given temperature. After that, we determine the impurity energy ε_0^{Ta} *via* the Friedel screening condition and calculate the local magnetic moment and the related magnetic hyperfine fields at a Ta impurity.

In Figures 1(b), 2(b), and 3(b) we plot as a function of temperature, the calculated local magnetic moments at a Ta impurity dilute in YFe_2, $GdFe_2$, and $YbFe_2$.

In Figures 1(a), 2(a), and 3(a) we plot as a function of temperature, the calculated magnetic hyperfine field at a Ta impurity dilute in YFe_2, $GdFe_2$, and $YbFe_2$. Our theoretical calculations exhibit a very good agreement with experiments,[2,3] which show that the magnetic hyperfine

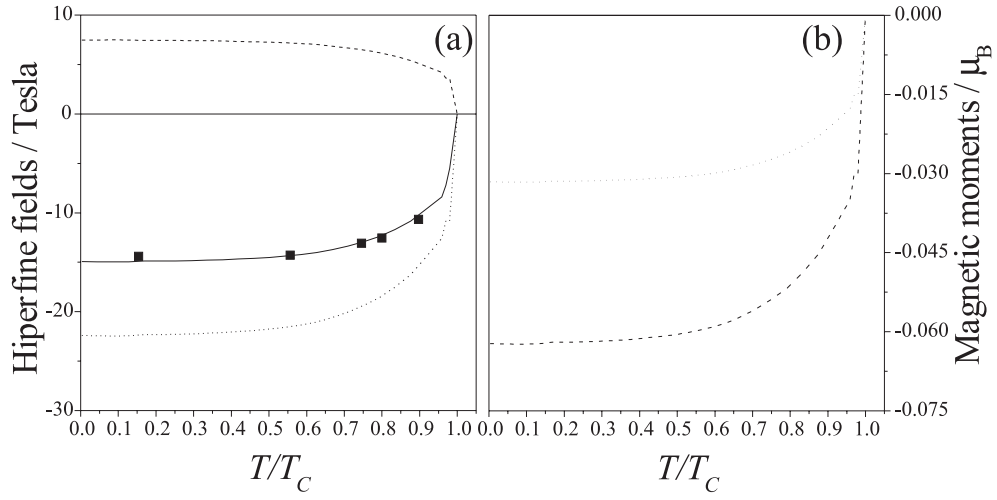

Figure 1. (a) Magnetic hyperfine field at Ta impurity diluted in YFe_2 intermetallic host. The dotted line corresponds to the CEP contribution and the dashed line corresponds to the CP contribution. The solid line represents the total hyperfine field. The squares represent the experimental data collected from reference 2. (b) Local magnetic moments at Ta impurity diluted in YFe_2 intermetallic host. The dotted line corresponds to the $\tilde{m}_c(0)$ contribution and the dashed line corresponds to the $\tilde{m}_d(0)$ contribution.

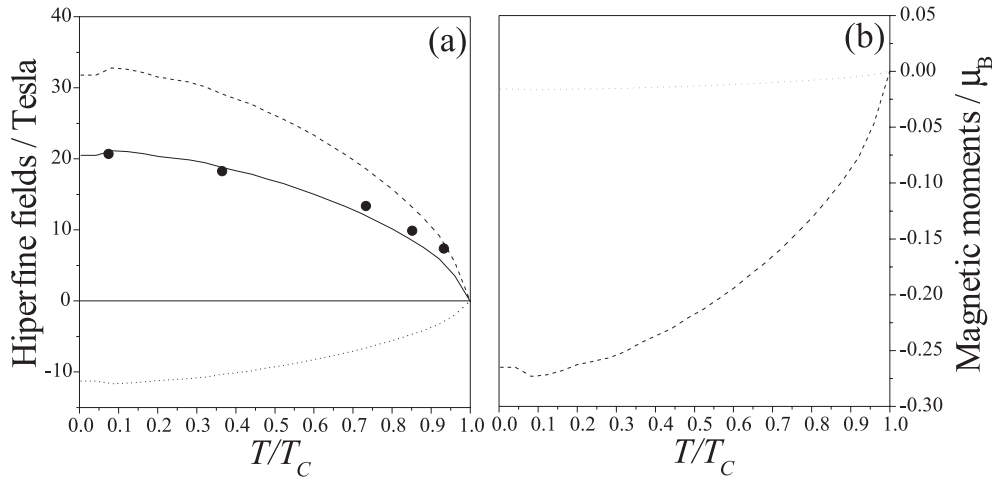

Figure 2. (a) Magnetic hyperfine field at Ta impurity diluted in $GdFe_2$ intermetallic host. The dotted line corresponds to the CEP contribution and the dashed line corresponds to the CP contribution. The solid line represents the total hyperfine field. The circles represent the experimental data collected from reference 3. (b) Local magnetic moments at Ta impurity diluted in $GdFe_2$ intermetallic host. The dotted line corresponds to the $\tilde{m}_c(0)$ contribution and the dashed line corresponds to the $\tilde{m}_d(0)$ contribution.

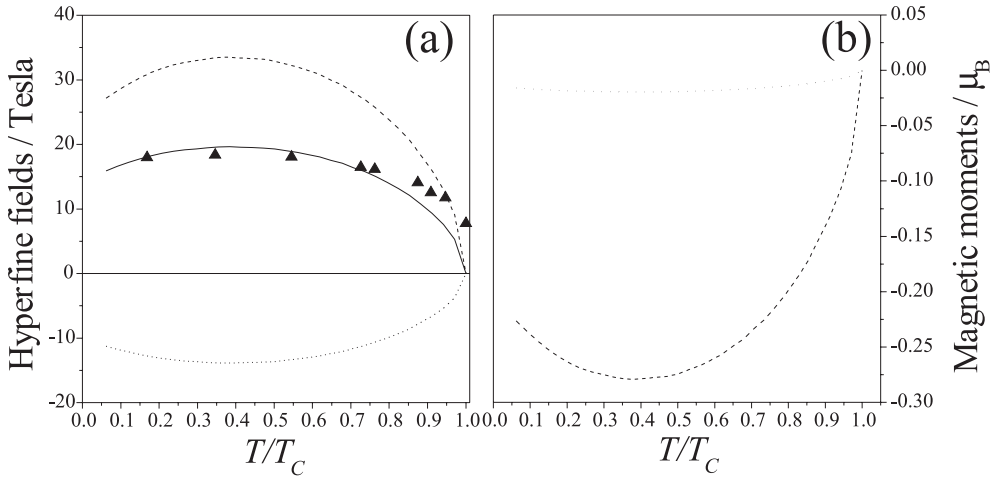

Figure 3. (a) Magnetic hyperfine field at Ta impurity diluted in $YbFe_2$ intermetallic host. The dotted line corresponds to the CEP contribution and the dashed line corresponds to the CP contribution. The solid line represents the total hyperfine field. The triangles represent the experimental data collected from reference 3. (b) Local magnetic moments at Ta impurity diluted in $YbFe_2$ intermetallic host. The dotted line corresponds to the $\tilde{m}_c(0)$ contribution and the dashed line corresponds to the $\tilde{m}_d(0)$ contribution.

field at the Ta impurity in $GdFe_2$ and $YbFe_2$ is antiparallel to the magnetization of the Fe sublattice being positive in both cases.

In Figure 4, we show a semi phenomenological calculation to include pressure effects.[11] In this case, we assume that external pressure excites the elastic degrees of freedom of the host. Therefore one has an electron-phonon interaction which modifies the pure electronic hybridization-like term,[13,14] $V_\sigma^X = \varepsilon_\sigma^{Ta} - \Sigma_\sigma^X(\varepsilon_F)$ (see equation (5)).

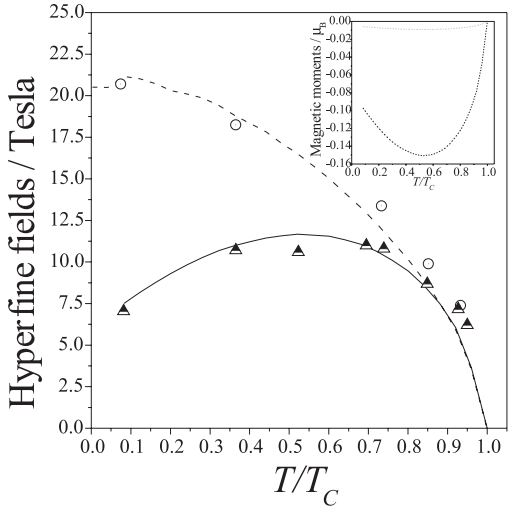

Figure 4. Magnetic hyperfine field at Ta impurity diluted in $GdFe_2$ intermetallic host. The solid line corresponds to the high pressure calculated total magnetic hyperfine field and the dashed line to the total calculated hyperfine field at normal pressure. Triangles and circles represent experimental data collected from reference 3. Inset: Local magnetic moments at Ta impurity in $GdFe_2$ intermetallic host with pressure effect. Dotted line and dashed line represent the $\tilde{m}_c(0)$ and the $\tilde{m}_d(0)$ contributions, respectively.

When the electron-phonon interaction is included in equation (5), one has a temperature dependence of V_σ. Figure 4 illustrates such pressure effects. The experimental data were obtained for a high pressure of 7.7 GPa and our theoretical results are again in a good agreement with the experimental data reported in reference 3. So, we have shown, that a functional integral approach, using a mean field approximation can describe the magnetic hyperfine field behavior of Laves phase intermetallics. The theory should be enlarged to account also for light rare earth, in the beginning of the *4f* series such as Ce, Pr and Nd, following the calculation made at $T = 0$ K made in reference 15.

Acknowledgment

This article was written for a special volume in honor of Professor Ricardo Ferreira. Professor Ricardo Ferreira is a true "natural philosopher" and his contribution to the development of natural sciences (physics, chemistry and biology) in Brazil has been extremely important. In his scientific work, originality and imagination are always present, thus being a source of inspiration to a whole generation of scientists.

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Received: August 21, 2007
Published on the web: February 29, 2008