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Effektív spin-modellek ab initio számítások alapján

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Effective spin models based on ab initio calculations

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1 Introduction

The first ab initio studies of the thermodynamical behavior of itinerant electron systems were carried out using the Stoner theory. It was successful to describe the ground state properties of magnetic systems, but it greatly overestimated the finite temperature magnetization. According to the Stoner model, the vanishing of the net magnetization can only happen if the local magnetization vanishes as well. To include transversal fluctuations one must introduce the concept of local spins into the theory of itinerant electron system.

Nowadays, most of the studies focusing on the finite temperature behavior of magnetic itinerant systems are based on mapping the energy of the system onto a Heisenberg Hamiltonian [1–5]. The exchange parameters are calculated by ab initio electron structure calculations. This mapping procedure is based on the adiabatic hypothesis. In this framework, the system can be divided into cells where the magnetization can be represented with a single ‘atomic momentum’. According to the hypothesis, the timescale of changes of atomic moments are much slower than the hopping time of the electrons.

In the present work we will investigate three different mapping techniques. The first one is useful in case of atomic moments the size of which is independent of the magnetic configuration [6]. Thereafter we will consider magnets that, beside of such strong moments, consist of induced moments as well. The size of these moments strongly depend on the magnetic configuration (order) of the strong moments. The FePt alloy is a prototypical example: the Fe moments are stable, while induced spins are formed on the Pt atoms. Ab initio calculations evidence that the local moment of Pt is about $0.32 \mu_B$ in the ferromagnetic (FM) state, but zero in the antiferromagnetic (AF) state. Since the model of Ref. [6] can not handle induced moments, special mapping techniques have been worked out to include longitudinal degrees of freedom of the spins [1, 2].

After describing these mapping models in details, for the case of the ordered FePt alloy, we will present our first ab initio results. We will test the resulting Heisenberg parameters with spin-dynamics simulations and we give estimates on the Curie temperature of FePt.

2 First principles electronic structure calculations

2.1 Density Functional Theory

Density Functional Theory (DFT) [7,8] allows us to perform first principles calculations of the ground state of interacting electrons. The basic concept of the theory is that the total energy of the ground state is a unique functional of the electron density, $n(\mathbf{r})$, and magnetization density (spin-density), $\mathbf{m}(\mathbf{r})$. By using the Kohn–Sham picture [9], the interacting many-electron system can be mapped onto a non-interacting system with the same ground state densities. Minimizing the energy functional of this non-interacting system yields the ground state densities that is necessary for calculating the physical properties of the system.

The Kohn–Sham equations of the non-interacting system can be obtained by varying the energy functional with respect to single-particle wave functions, $\psi_i(\mathbf{r})$,

$$\left(-\frac{\hbar^2}{2m}\Delta + V[n, \mathbf{m}](\mathbf{r}) + \mathbf{B}[n, \mathbf{m}](\mathbf{r})\boldsymbol{\sigma} \right) \psi_i(\mathbf{r}) = \varepsilon_i \psi_i(\mathbf{r}) , \quad (2.1)$$

where $\boldsymbol{\sigma}$ are the Pauli matrices and ε_i is the single-particle energy. The effective potential $V[n, \mathbf{m}](\mathbf{r})$ contains the external potential, $V_{\text{ext}}(\mathbf{r})$, the Coulomb potential of the nuclei, $W(\mathbf{r})$, the electrostatic (Hartree) potential of the electrons $V_{\text{el}}[n](\mathbf{r})$, and the so called exchange-correlation potential, $V_{\text{xc}}[n, \mathbf{m}](\mathbf{r})$ related to all the non Coulomb-like effects of the other electrons, whereas the effective field, $\mathbf{B}[n, \mathbf{m}](\mathbf{r})$, is the sum of the external magnetic field, $\mathbf{B}_{\text{ext}}(\mathbf{r})$, and the exchange field, $\mathbf{B}_{\text{xc}}[n, \mathbf{m}](\mathbf{r})$:

$$V[n, \mathbf{m}](\mathbf{r}) = V_{\text{ext}}(\mathbf{r}) + W(\mathbf{r}) + V_{\text{el}}[n, \mathbf{m}](\mathbf{r}) + V_{\text{xc}}[n, \mathbf{m}](\mathbf{r}) , \quad (2.2)$$

$$\mathbf{B}[n, \mathbf{m}](\mathbf{r}) = \mathbf{B}_{\text{ext}}(\mathbf{r}) + \mathbf{B}_{\text{xc}}[n, \mathbf{m}](\mathbf{r}) . \quad (2.3)$$

The exchange-correlation potential and field are determined by the functional derivative of the exchange-correlation functional, $E_{\text{xc}}[n, \mathbf{m}](\mathbf{r})$, with respect to $n(\mathbf{r})$ and $\mathbf{m}(\mathbf{r})$, respectively. However, this quantity is not known exactly, so different approximations are used. We will apply the local spin-density approximation (LSDA) where the density of the exchange-correlation energy of the interacting system is approximated by that of the free electrons.

The coupled Kohn–Sham equations are implicit, they can be solved by iteration. We start with well-guessed initial densities, then from the resulting wavefunctions we can generate new densities which will be different from the initial densities. Using the new densities as input we repeat the process until convergency, i.e. when the fix point is reached. Because of the highly non-linear nature of the Kohn–Sham equations, various

mixing schemes were introduced in order to avoid instabilities of the above self-consistent field (scf) process.

2.2 Constrained Local Spin-Density Approximation

Since, in a strict sense, DFT can only be used for ground state calculation, we must use some special treatment to calculate energies of excited states. A possible method is the constrained local spin-density approximation (CLSDA) [10], where we add Lagrange multiplier terms to the energy functional of the system, and we evaluate the variational process using the constrained energy formula.

In this work we deal with different magnetic configurations as excited states, so our aim is to fix the direction, $\mathbf{e}_i$, and the magnitude, m_i , of the atomic spin-moments,

$$\mathbf{m}_i = \int_{\Omega_i} d^3r \mathbf{m}(\mathbf{r}) = m_i \mathbf{e}_i, \quad (2.4)$$

where Ω_i denotes the spatial domain (cell) associated with site i . To this end, we need the Lagrange multipliers, $\mathbf{B}_{c,\perp}^i$ and $B_{c,\parallel}^i$ that set the amplitude of the i -th moment to $m_{i,c}$ and its direction to $\mathbf{e}_{i,c}$,

$$E_{\text{CLSDA}} = E_{\text{LSDA}} + \mathbf{B}_{c,\perp}^i \mathbf{e}_i + B_{c,\parallel}^i (m_i - m_{i,c}), \quad (2.5)$$

where $\mathbf{B}_{c,\perp}^i$ is perpendicular to $\mathbf{e}_{i,c}$. Minimizing this expression leads to an equation similar to Eq. (2.1) with the only difference that the effective field will also contain the constraining fields,

$$\mathbf{B}^i[n, \mathbf{m}](\mathbf{r}) = \mathbf{B}_{\text{ext}}^i(\mathbf{r}) + \mathbf{B}_{\text{xc}}^i[n, \mathbf{m}](\mathbf{r}) + \mathbf{B}_{c,\perp}^i + B_{c,\parallel}^i \mathbf{e}_i, \quad (2.6)$$

where $B_{c,\parallel}^i$ is parallel to $\mathbf{e}_i$.

We again solve the Kohn-Sham equations by iteration, however, a difficulty occurs since the constraining fields are not known *a priori*. Since in every iteration step we get new magnetization densities, we must update the constraining fields in each step. There are different schemes to update the constraints. These can be derived from the condition, that the constraining fields must be parallel/perpendicular to the magnetization direction and they should converge in the scf excited state [11]. Moreover, they should vanish in the unconstrained ground state.

2.3 The basics of the KKR method

To solve the Kohn-Sham equations we shall use the Korringa-Kohn-Rostoker (KKR) method or multiple scattering theory [12]. Here we just give a brief overview of this

method that is necessary to the understanding of the rest of this work. Let us consider a system with the Hamiltonian,

$$\hat{H} = \hat{H}_0 + \hat{V}, \quad (2.7)$$

where $\hat{H}_0$ denotes the free particle Hamiltonian and $\hat{V}$ the crystal potential. We define the resolvent operators of the corresponding systems as

$$\hat{G}_0(z) = (zI - \hat{H}_0)^{-1}, \quad \hat{G}(z) = (zI - \hat{H})^{-1},$$

where z is a complex energy value. Expanding $\hat{G}$ yields

$$\hat{G} = \hat{G}_0 + \hat{G}_0 \hat{V} \hat{G}_0 + \hat{G}_0 \hat{V} \hat{G}_0 \hat{V} \hat{G}_0 \hat{V} + \dots = \hat{G}_0 + \hat{G}_0 \hat{T} \hat{G}_0, \quad \hat{T} = \hat{V} + \hat{V} \hat{G}_0 \hat{V} + \dots$$

$\hat{T}$ is called the scattering operator. Note that for brevity we dropped the energy argument of the operators. For a potential confined to a single cell at site i (being zero out of the cell) it is termed as the single-site t operator, $\hat{t}_i$. For a crystal, i.e. a set of non-overlapping single-cell potentials, $\hat{V} = \sum_i \hat{V}_i$, the $\hat{T}$ operator can be expressed in terms of the single-site $\hat{t}$ operators,

$$\hat{T} = \sum_i \hat{t}_i + \sum_{i \neq j} \hat{t}_i \hat{G}_0 \hat{t}_j + \sum_{i \neq j \neq k} \hat{t}_i \hat{G}_0 \hat{t}_j \hat{G}_0 \hat{t}_k \dots \quad (2.8)$$

It is worth to introduce the scattering path operators (SPO), $\hat{\tau}_{ij}$,

$$\hat{T} = \sum_{ij} \hat{\tau}_{ij}, \quad (2.9)$$

which by using the convenient notations, $[\hat{\mathbf{t}}]_{ij} = \delta_{ij} \hat{t}_i$, $[\hat{\mathbf{G}}_0]_{ij} = \hat{G}_{0,ij}(1 - \delta_{ij})$ and $[\hat{\boldsymbol{\tau}}]_{ij} = \hat{\tau}_{ij}$, satisfies the following relationships,

$$\hat{\mathbf{G}} = \hat{\mathbf{G}}_0 + \hat{\mathbf{G}}_0 \hat{\boldsymbol{\tau}} \hat{\mathbf{G}}_0 \quad (2.10)$$

$$\begin{aligned} \hat{\boldsymbol{\tau}} &= \hat{\mathbf{t}} + \hat{\mathbf{t}} \hat{\mathbf{G}}_0 \hat{\mathbf{t}} + \hat{\mathbf{t}} \hat{\mathbf{G}}_0 \hat{\mathbf{t}} \hat{\mathbf{G}}_0 \hat{\mathbf{t}} + \dots \\ &= \hat{\mathbf{t}} + \hat{\mathbf{t}} \hat{\mathbf{G}}_0 \hat{\boldsymbol{\tau}} \end{aligned} \quad (2.11)$$

Representing the operators as matrices in a basis formed by the functions, $j_\ell(\sqrt{z}r_i)Y_\ell^m(\hat{\mathbf{r}}_i)\chi_s$, where $\mathbf{r}_i = \mathbf{r} - \mathbf{R}_i$, while j_ℓ , Y_ℓ^m and χ_s denote the spherical Bessel function, the spherical harmonics and spinor eigenfunctions, respectively, the above equations remain valid [12]. In what follows, we will label the corresponding matrices without 'hat'.

From equation (2.11) the SPO matrix can be obtained,

$$\boldsymbol{\tau} = (\mathbf{t}^{-1} - \mathbf{G}_0)^{-1}, \quad (2.12)$$

which, in turn, can be used to evaluate the real-space Green's function of the system [12, 13].

From the Green's function it is then straightforward to calculate physical observables, in particular, the charge and magnetization densities needed to solve the Kohn-Sham equations self-consistently. For the numerical evaluation we must limit the size of the matrices. The spherical harmonics will be used up to $\ell_{\max} = 2$. Furthermore we use the atomic sphere approximation (ASA) in which the effective potential and field in a cell are supposed to be spherical within a sphere of the same volume as the cell and out of the sphere the potential is constant.

2.4 The magnetic force theorem and the Lloyd formula

In order to calculate the exchange interactions the magnetic force theorem (MFT) is often used [6]. MFT relies on the assumption that the effective potentials and fields experience negligible change when changing the magnetic configuration from a state of reference (ground state), $\{\mathbf{e}_i\}$, to an excited one, $\{\mathbf{e}'_i\}$. From the stationary property of the LSDA ground state it can be shown that the change of the total energy can be approximated by the change of the single-particle energy, i.e. the band energy of the valence electrons,

$$\delta E = \delta \int_{-\infty}^{\varepsilon_F} d\varepsilon \varepsilon n(\varepsilon), \quad (2.13)$$

where ε_F is the Fermi energy and $n(\varepsilon)$ is the density of states (DOS). Since, however, the DOS for the magnetic state $\{\mathbf{e}'_i\}$ is not obtained from an scf LSDA calculation, charge neutrality is not strictly satisfied. Therefore, instead of the energy, the grand canonical potential should be used as the appropriate thermodynamic quantity ($T = 0$),

$$\Phi = E - \varepsilon_F N = \int_{-\infty}^{\varepsilon_F} d\varepsilon (\varepsilon - \varepsilon_F) n(\varepsilon) = - \int_{-\infty}^{\varepsilon_F} d\varepsilon N(\varepsilon). \quad (2.14)$$

where $N(\varepsilon)$ is the integrated density of states, $N(\varepsilon) = \int_{-\infty}^{\varepsilon} d\varepsilon' n(\varepsilon')$.

According to Lloyd's formula [14], the integrated DOS of a system described by the Hamiltonian (2.7) can be expressed as

$$N(\varepsilon) = N_0(\varepsilon) + \frac{1}{\pi} \text{Im Tr} \ln \boldsymbol{\tau}(\varepsilon)$$

where $N_0(\varepsilon)$ is the integrated DOS of the free electrons and Tr means the trace of a matrix in the composite site-, angular momentum, spin-space. Consequently, the change of grand canonical potential due to a magnetic excitation as mentioned above can be

calculated as

$$\delta\Phi = -\frac{1}{\pi} \int_{-\infty}^{\epsilon_F} d\epsilon \operatorname{Im} \operatorname{Tr} \ln [\boldsymbol{\tau}'(\epsilon) \boldsymbol{\tau}(\epsilon)^{-1}] , \quad (2.15)$$

where $\boldsymbol{\tau}(\epsilon)$ and $\boldsymbol{\tau}'(\epsilon)$ denote the SPO matrices related to the magnetic states $\{\mathbf{e}_i\}$ and $\{\mathbf{e}'_i\}$, respectively.

3 The isotropic classical Heisenberg model

The isotropic classical Heisenberg model,

$$H = -\frac{1}{2} \sum_{i \neq j} J_{ij} \mathbf{e}_i \mathbf{e}_j , \quad (3.1)$$

is often used to describe magnets characterized by stable atomic moments. It should be noted, that the only freedom of the moments are rotations and the exchange interaction parameters, J_{ij} 's, depend only on the lattice positions.

Let us consider a system with a ferromagnetic ground state. The parameters, J_{ij} , can be obtained by using the following recipe: compute the energy difference that appears between the ground state and some special excited states from both the model, Eq. (3.1), and from first principles calculations (the KKR Green's function method). Assuming that these energy differences are identical, we can sort out the interaction parameters.

3.1 The method of infinitesimal rotations

In this section we briefly sketch the mostly used method to calculate isotropic exchange interactions as introduced by Liechtenstein *et al.* [6]. Based on the Hamiltonian (3.1), in the FM state the energy of the system is given by

$$E_0 = -\frac{1}{2} \sum_{i \neq j} J_{ij} .$$

Let's consider an excitation when a single spin located at site i is rotated by an angle θ . In this case the energy difference with respect to the FM state is

$$\delta E(\mathbf{e}_i) = E(\mathbf{e}_i) - E_0 = - \sum_{j(\neq i)} J_{ji} (1 - \cos(\theta)) \approx - \left(1 - 1 + \frac{\theta^2}{2} \right) \sum_{j(\neq i)} J_{ji} = -\frac{\theta^2}{2} J_i .$$

where we took the limit of small (infinitesimal) θ and introduced the effective interactions $J_i = \sum_{j(\neq i)} J_{ji}$. Next we study the case when two spins at sites i and j are rotated with

respect to the uniform direction. The interaction energy between these two spins is defined as

$$\begin{aligned} E_{\text{int}}(\mathbf{e}_i, \mathbf{e}_j) &= [E(\mathbf{e}_i, \mathbf{e}_j) - E_0] - [E(\mathbf{e}_i) - E_0] - [E(\mathbf{e}_j) - E_0] \\ &= E(\mathbf{e}_i, \mathbf{e}_j) - E(\mathbf{e}_i) - E(\mathbf{e}_j) + E_0, \end{aligned} \quad (3.2)$$

where $E(\mathbf{e}_i, \mathbf{e}_j)$ is the energy of the system with two rotated spins. If the angles of rotations are $\theta/2$ at site i and $-\theta/2$ at site j ,

$$\begin{aligned} E_{\text{int},ij} \left(\frac{\theta}{2}, -\frac{\theta}{2} \right) &= E_{ij} \left(\frac{\theta}{2}, -\frac{\theta}{2} \right) - E_i \left(\frac{\theta}{2} \right) - E_j \left(-\frac{\theta}{2} \right) + E_0 \\ &= -J_{ij}(1 - \cos(\theta)) = -J_{ij} \frac{\theta^2}{2}. \end{aligned}$$

From here it follows that within the isotropic Heisenberg model

$$J_{ij} = -\frac{\delta^2 E}{\delta \mathbf{e}_i \delta \mathbf{e}_j} = -\frac{\partial^2}{\partial \theta^2} \left[E_{\text{int},ij} \left(\frac{\theta}{2}, -\frac{\theta}{2} \right) \right]. \quad (3.3)$$

According to the recipe mentioned above, by identifying the variation of the energy of the spin system with the variation of the electronic grand canonical potential, Eq. (2.15), a first principles expression for J_{ij} can be obtained within the KKR method.

A site-specific change in the t -matrix, $t'_i = t_i + \delta t_i$, implies

$$\mathbf{t}' = \begin{pmatrix} \ddots & & & & \\ & t_{i-1} & & & \\ & & t_i & & \\ & & & t_{i+1} & \\ & & & & \ddots \end{pmatrix} + \begin{pmatrix} \ddots & & & & \\ & 0 & & & \\ & & \delta t_i & & \\ & & & 0 & \\ & & & & \ddots \end{pmatrix} = \mathbf{t} + \delta \mathbf{t}, \quad (\mathbf{t}')^{-1} = \mathbf{t}^{-1} + \delta \mathbf{t}^{-1},$$

where the only non-vanishing block of $\delta \mathbf{t}^{-1}$ is $\delta t_i^{-1} = t_i'^{-1} - t_i^{-1}$.

If we use a coordinate system where the orientation of the ground state magnetization is parallel to z then a single-site t -matrix can be written in terms of spin-up and spin-down t -matrices,

$$t_i = \begin{pmatrix} t_{i\uparrow} & 0 \\ 0 & t_{i\downarrow} \end{pmatrix} = \frac{t_{i\uparrow} + t_{i\downarrow}}{2} I_2 + \frac{t_{i\uparrow} - t_{i\downarrow}}{2} \sigma_z, \quad (3.4)$$

where I_2 is the 2×2 unit matrix. Note that $t_{i\uparrow}$ and $t_{i\downarrow}$ are matrices with only angular-momentum indices. A rotation of the magnetization, $\mathbf{z} \rightarrow \mathbf{e}$, implies

$$\delta t_i = \frac{1}{2} (t_{i\uparrow} - t_{i\downarrow}) (\delta \mathbf{e} \boldsymbol{\sigma}), \quad \delta t_i^{-1} = \frac{1}{2} (t_{i\uparrow}^{-1} - t_{i\downarrow}^{-1}) (\delta \mathbf{e} \boldsymbol{\sigma}). \quad (3.5)$$

When evaluating the two-spin interaction energy, Eq. (3.2), based on Eq. (2.15), $\boldsymbol{\tau}'$ should be related to $\boldsymbol{\tau}$ using Eq. (2.12). Keep also in mind that if simultaneous rotations by $\theta/2$ and $-\theta/2$ are applied at sites i and j , $\delta\mathbf{t}^{-1} = \delta\mathbf{t}_i^{-1} + \delta\mathbf{t}_j^{-1}$. After a straightforward but lengthy derivation, where Eq. (3.5) is utilized and only the leading term, $\sim \theta^2$, is considered, we arrive at the famous formula [6],

$$\boxed{J_{ij} = -\frac{1}{4\pi} \int_{-\infty}^{\epsilon_F} d\varepsilon \operatorname{Im} \operatorname{Tr} \left(\Delta_i \tau_{ij,\uparrow} \Delta_j \tau_{ji,\downarrow} \right)}. \quad (3.6)$$

where $\Delta_i = t_{i\uparrow}^{-1} - t_{i\downarrow}^{-1}$ and $\tau_{ij,\uparrow}$ and $\tau_{ji,\downarrow}$ denoting the corresponding spin-diagonal and site off-diagonal blocks of the SPO matrix in the FM ground state.

4 Incorporating induced moments: Mryasov's renormalization

As we mentioned in the introduction, we would like to focus on magnets with induced moments. The isotropic Heisenberg model applies, however, only to strong moments. According to the thoughts of Oleg Mryasov [1], in this section we present a generalization of the Heisenberg model that includes the formation energy of the induced moments. The basic idea is to regain a renormalized spin-model for the strong moments.

Let us rewrite Eq. (3.1) by sorting out the strong and weak moments. From now on, Latin letters will denote strong moments (Fe) and Greek letters will denote the induced ones (Pt):

$$H = \underbrace{-\frac{1}{2} \sum_{i \neq j} J_{ij} \mathbf{e}_i \mathbf{e}_j}_{\text{Fe-Fe term}} - \underbrace{\sum_{i\nu} J_{i\nu} \mathbf{e}_i \mathbf{e}_\nu}_{\text{Fe-Pt term}} - \underbrace{\frac{1}{2} \sum_{\nu \neq \mu} J_{\nu\mu} \mathbf{e}_\nu \mathbf{e}_\mu}_{\text{Pt-Pt term}}. \quad (4.1)$$

The interaction between the induced moments is supposed to be negligible, we therefore neglect it in the following. Reformulating the second term,

$$H_{\text{Fe-Pt}} = - \sum_{i\nu} J_{i\nu} \mathbf{e}_i \mathbf{e}_\nu = - \sum_{\nu} \mathbf{h}_\nu \mathbf{e}_\nu, \quad \mathbf{h}_\nu = \sum_i J_{i\nu} \mathbf{e}_i,$$

the Weiss-field, $\mathbf{h}_\nu$, exerted by the strong moments onto a weak moment is introduced. Minimizing $H_{\text{Fe-Pt}}$ with respect to $\mathbf{e}_\nu$ yields $\mathbf{e}_\nu = \frac{\mathbf{h}_\nu}{h_\nu}$ and $H_{\text{Fe-Pt}} = - \sum_{\nu} h_\nu$. The obtained spin-model,

$$H = -\frac{1}{2} \sum_{i \neq j} J_{ij} \mathbf{e}_i \mathbf{e}_j - \sum_{\nu} \left| \sum_i J_{i\nu} \mathbf{e}_i \right|, \quad (4.2)$$

is, however, not a Heisenberg-like model and can hardly be used with usual methods of statistical physics. In addition, it still misses to consider the energy related to the formation of an induced local magnetic moment, μ_ν .

Based on ab initio calculations (see later), we shall suppose that the formation energy of an induced moment, H_ν , depends only on μ_ν , and μ_ν is a function of the magnitude of the Weiss field, h_ν ,

$$H_\nu = A\mu_\nu^2 + B\mu_\nu^4 + \dots \quad \mu_\nu = ah_\nu + bh_\nu^2 + \dots,$$

where we also made use of time-reversal symmetry. Combining these two equations, H_ν obviously can be expressed as a function of the Weiss field,

$$H_\nu = \frac{\chi_\nu^{(1)}}{h_{\nu 0}^2} h_\nu^2 + \frac{\chi_\nu^{(2)}}{h_{\nu 0}^4} h_\nu^4 + \dots \quad (4.3)$$

where we normalized the first and second order susceptibilities, $\chi_\nu^{(1)}$ and $\chi_\nu^{(2)}$ to the Weiss field in the FM state, $h_{\nu 0}$.

After substituting the expression, $h_\nu^2 = \sum_{i,j} J_{i\nu} J_{j\nu} \mathbf{e}_i \mathbf{e}_j$, and neglecting a constant and biquadratic terms we obtain an effective Heisenberg-like spin-model,

$$H = -\frac{1}{2} \sum_{i \neq j} \tilde{J}_{ij} \mathbf{e}_i \mathbf{e}_j, \quad (4.4)$$

$$\boxed{\tilde{J}_{ij} = J_{ij} - 2 \sum_\nu \frac{\chi_\nu^{(1)}}{h_{\nu 0}^2} J_{i\nu} J_{j\nu} - 4 \sum_\nu \frac{\chi_\nu^{(2)}}{h_{\nu 0}^4} \left(\sum_i J_{i\nu}^2 \right) J_{i\nu} J_{j\nu}}, \quad (4.5)$$

with

$$h_{\nu 0} = \sum_i J_{i\nu}.$$

Thus, by deriving from ab initio calculations the spin susceptibilities for the weak moments, $\chi_\nu^{(1)}$ and $\chi_\nu^{(2)}$, and calculating the bare interactions, J_{ij} and $J_{i\nu}$, as described in the previous sections, the renormalized interactions $\tilde{J}_{ij}$ can easily be obtained.

5 Renormalization based on the elimination of weak moments

Recently, a modified strategy has been published to obtain renormalized interactions between the strong moments [2]. As in the previous section, we will denote the strong moments by $\mathbf{m}_i$ and the induced ones by $\boldsymbol{\mu}_\nu$. The length of $\mathbf{m}_i$ is fixed, but $\boldsymbol{\mu}_\nu$ can change

it's amplitude as the magnetic configuration changes. In the Hamiltonian related to the weak moments we include a Weiss field term, since it is responsible for the precessional motion of the weak moments and we also will introduce a part which is responsible to the longitudinal variation of the weak moments,

$$H_\nu[\mathbf{h}_\nu, \boldsymbol{\mu}_\nu] = -\mathbf{h}_\nu \boldsymbol{\mu}_\nu + a\mu_\nu^2. \quad (5.1)$$

Note that higher-order terms in μ_ν are neglected. If $a \geq 0$, we deal with induced moments, otherwise we can have spontaneous magnetization. In absence of an external field, $\mathbf{h}_\nu$ is the Weiss-field exerted by the strong moments for which we assume

$$\mathbf{h}_\nu = \sum_i \kappa_{i\nu} \mathbf{m}_i. \quad (5.2)$$

where $\kappa_{i\nu}$ are phenomenological parameters (to be determined from ab initio calculations) related to the interactions between the induced and strong moments.

The Hamiltonian of the system then becomes

$$H[\{\mathbf{m}_i\}, \{\boldsymbol{\mu}_\nu\}] = -\frac{1}{2} \sum_{i \neq j} J_{ij} \mathbf{e}_i \mathbf{e}_j + \sum_\nu \left(a\mu_\nu^2 - \boldsymbol{\mu}_\nu \sum_i \kappa_{i\nu} \mathbf{m}_i \right), \quad (5.3)$$

where the second term can be manipulated as follows,

$$\sum_\nu \left(a\mu_\nu^2 - \boldsymbol{\mu}_\nu \sum_i \kappa_{i\nu} \mathbf{m}_i \right) = \sum_\nu \left[a \left(\boldsymbol{\mu}_\nu - \sum_i \frac{\kappa_{i\nu}}{2a} \mathbf{m}_i \right)^2 - \frac{1}{4a} \left(\sum_i \kappa_{i\nu} \mathbf{m}_i \right)^2 \right]. \quad (5.4)$$

We can further rewrite the last term as

$$\sum_\nu \sum_{i,j} \kappa_{i\nu} \kappa_{j\nu} \mathbf{m}_i \mathbf{m}_j = \sum_{i \neq j} K(i, j) \mathbf{m}_i \mathbf{m}_j + \sum_i K(i, i) m_i^2. \quad (5.5)$$

with

$$K(i, j) := \sum_\nu \kappa_{i\nu} \kappa_{j\nu}. \quad (5.6)$$

Since the magnitudes of the strong moments are fixed, the second term of Eq. (5.5) is constant, therefore, it can be neglected from the Hamiltonian. After all, we can cast the Hamiltonian of the system as

$$H[\{\mathbf{m}_i\}, \{\boldsymbol{\mu}_\nu\}] = H_R[\{\mathbf{m}_i\}] + H'[\{\mathbf{m}_i\}, \{\boldsymbol{\mu}_\nu\}], \quad (5.7)$$

where

$$H_R[\{\mathbf{m}_i\}] = -\frac{1}{2} \sum_{i \neq j} \tilde{J}_{ij} \mathbf{e}_i \mathbf{e}_j \quad \boxed{\tilde{J}_{ij} = J_{ij} + \frac{K(i, j) m_i m_j}{2a}} \quad (5.8)$$

manifests a renormalized Hamiltonian with the only variables $\{\mathbf{m}_i\}$, while

$$H'[\{\mathbf{m}_i\}, \{\boldsymbol{\mu}_\nu\}] = a \sum_\nu \left(\boldsymbol{\mu}_\nu - \sum_i \frac{\kappa_{i\nu}}{2a} \mathbf{m}_i \right)^2. \quad (5.9)$$

is a term that depends both on strong and weak moments.

At $T = 0$, the ground state of this model can be found easily. Since $a \geq 0$, $\boldsymbol{\mu}_\nu$ are determined by setting $H' = 0$,

$$\boldsymbol{\mu}_\nu = \sum_i \frac{\kappa_{i\nu}}{2a} \mathbf{m}_i, \quad (5.10)$$

and we are left with minimizing $H_R[\{\mathbf{m}_i\}]$.

In case of finite temperatures ($T > 0$), the equilibrium of the system is defined as the minimum of the free energy, thus we should investigate the canonical partition function,

$$\begin{aligned} Z &= \int d\{\mathbf{e}_i\} \int d\{\boldsymbol{\mu}_\nu\} e^{-\beta H[\{\mathbf{m}_i\}, \{\boldsymbol{\mu}_\nu\}]} \\ &= \int d\{\mathbf{e}_i\} e^{-\beta H_R[\{\mathbf{m}_i\}]} \int d\{\boldsymbol{\mu}_\nu\} e^{-\beta a \sum_\nu \left(\boldsymbol{\mu}_\nu - \sum_i \frac{\kappa_{i\nu}}{2a} \mathbf{m}_i \right)^2} \\ &= Z_R \left(\frac{\pi}{\beta a} \right)^{\frac{3N}{2}}, \end{aligned}$$

where $\beta = 1/k_B T$ and N is the number of weak moments in the system. We can see that, after integrating out the degrees of freedom of the weak moments, the partition function appears to be the partition function of the renormalized Hamiltonian of the strong moments multiplied by the partition function of $3N$ free oscillators. This is very convenient, because the thermodynamic quantities related to the strong moments can be described by using the renormalized Hamiltonian. As an example, we demonstrate this property for the expectation value of the magnetization of strong moments:

$$\begin{aligned} \langle \mathbf{m}_k \rangle &= \frac{1}{Z} \int d\{\mathbf{e}_i\} \int d\{\boldsymbol{\mu}_\nu\} \mathbf{m}_k e^{-\beta H[\{\mathbf{m}_i\}, \{\boldsymbol{\mu}_\nu\}]} \\ &= \frac{1}{Z} \left(\frac{\pi}{\beta a} \right)^{\frac{3N}{2}} \int d\{\mathbf{e}_i\} \mathbf{m}_k e^{-\beta H_R[\{\mathbf{m}_i\}]} \\ &= \frac{1}{Z_R} \int d\{\mathbf{e}_i\} \mathbf{m}_k e^{-\beta H_R[\{\mathbf{m}_i\}]} = \langle \mathbf{m}_k \rangle_R. \end{aligned}$$

This immediately implies that the Curie temperature of the renormalized system of strong moments and that of the original system of strong and weak moments are identical. After some derivation it can be proved that all correlation functions can be calculated by using the renormalized model only [2]. Also, the value of the weak moments can be expressed

as

$$\langle \boldsymbol{\mu}_\nu \rangle = \frac{1}{2a} \langle \mathbf{h}_\nu \rangle = \sum_i \frac{\kappa_{i\nu}}{2a} \langle \mathbf{m}_i \rangle = \sum_i \frac{\kappa_{i\nu}}{2a} \langle \mathbf{m}_i \rangle_{\text{R}} . \quad (5.11)$$

Summarizing, we can say that because of the Gaussian function form of $H'[\{\mathbf{m}_i\}, \{\boldsymbol{\mu}_\nu\}]$, Eq. (5.9), the weak moments could be eliminated from the spin-model.

6 Calculations of the parameters for the spin models

In what follows we will denote the isotropic exchange parameters as calculated by means of the method of infinitesimal rotations, see Section 3.1, by *bare* exchange interactions, whereas the renormalized parameters in terms of Mryasov's model, Section 4, and by using the elimination method, Section 5, by *R1* and *R2* type of parameters, respectively. All the parameters were calculated in terms of the ab initio KKR method. The ordered FePt alloy has a face-centered tetragonal (fct) geometrical structure, see Fig. 1a. We used the experimental value of the lattice constants which means that the lattice is compressed along the z direction, $c/a = 0.964$, where a and c are the two lattice constants of the fct lattice.

6.1 Spin susceptibilities for spin model *R1*

Within the *R1* spin-model, the susceptibilities, $\chi^{(1)}$ and $\chi^{(2)}$ are defined in Eq. (4.3). Note that, since all the Pt atoms are identical, we omit the index ν . By manipulating the magnetic configuration of the Fe atoms, the total energy can be calculated as a function of the Weiss field, thus, $\chi^{(1)}$ and $\chi^{(2)}$ can be obtained from the data by fitting to the appropriate function.

As a suitable magnetic configuration we keep the Fe moments parallel to $\mathbf{z}$, and we will rotate all Pt moments by an angle of θ , see Fig. 1a. Performing constrained LSDA calculations with transversal constrains, Section 2.2, the constraining field compensates the perpendicular component of the Weiss field, see Fig. 1b. In Eq. (4.3) we then have to consider the component of the Weiss field parallel to the Pt moments, $h_\nu = h_{\nu 0} \cos \theta$. Our calculations show that the Fe moments are fairly insensitive to the variation of θ , $m_{\text{Fe}} = 2.82 \mu_{\text{B}}$, therefore, from the model we expect that the total energy of the system (normalized to a unit cell) varies as

$$E(\theta) = E_0 + \chi_\nu^{(1)} \cos^2 \theta + \chi_\nu^{(2)} \cos^4 \theta . \quad (6.1)$$

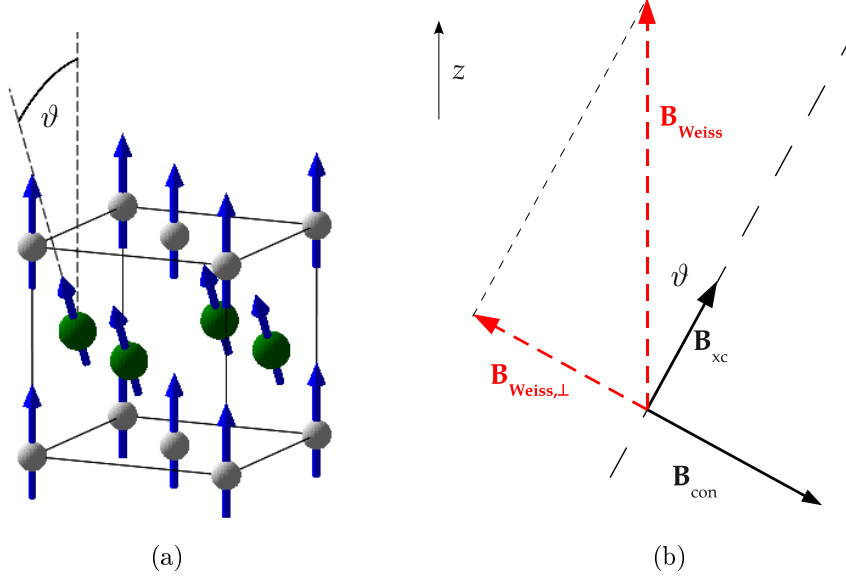


Figure 1: (a) The constrained magnetic configuration of the ordered FePt alloy as used for calculating the spin susceptibilities. Fe and Pt atoms are displayed by gray and green balls, respectively. The orientation of the spin moments are represented by arrows. (b) Different fields in the constrained LSDA calculations: the constraining field, $\mathbf{B}_{\text{con}}$, cancels the perpendicular component of the Weiss field, $\mathbf{B}_{\text{Weiss},\perp}$, thus, the Pt atoms experience a Weiss field and an exchange field, $\mathbf{B}_{\text{xc}}$, along the desired direction.

The results of the CLSDA calculations for the total energy are shown in Fig. 2. Reassuringly, we get an almost perfect dependence for $E(\theta)$ as predicted in Eq. (6.1) with the fitted parameters, $\chi^{(1)} = -10.89 \text{ mRyd}$ and $\chi^{(2)} = 1.72 \text{ mRyd}$. Apparently, the $\cos^2 \theta$ term applies just for small θ (near the ground state), while for larger θ values the fourth-order term can not be neglected.

6.2 Parameters for spin model $R2$

The phenomenological parameters, $\kappa_{i\nu}$ can, in principle, be calculated as follows: we set different configurations for the strong magnetic moments, $\{\mathbf{e}_i\}$, perform CLSDA calculations for these configurations and determine the values μ_ν and $\mathbf{m}_i$, while, finally, solve a set of linear equations, Eq. (5.10), for the unknown parameters, $\kappa_{i\nu}$. To simplify matter, in Ref. [2] it was supposed that $\kappa_{i\nu}$ differs from zero only for the nearest neighbor pairs of strong and weak moments and, quite obviously, all the NN pairs have the same parameter, κ . Adopting this approach, in the FM ground state we can write

$$\mu_0 = \frac{\kappa}{2a} N_c m_0 \quad \Rightarrow \quad \frac{\kappa}{a} = \frac{2\mu_0}{N_c m_0}, \quad (6.2)$$

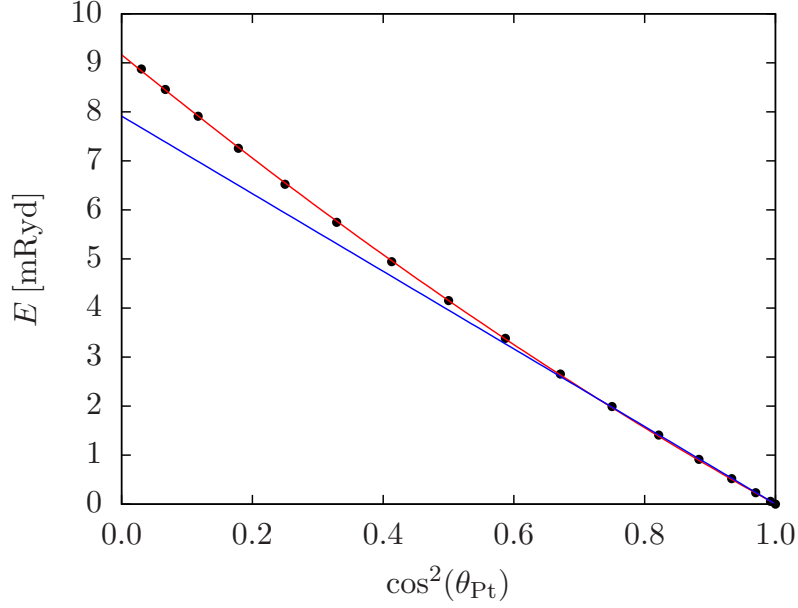


Figure 2: Ab initio total energies as calculated by using transversal constraint (black circles), see Fig. 1. The energy is measured from the FM state. The red line shows the fit as in Eq. (6.1). For comparison, the blue line shows the fit to $\cos^2 \theta$.

where N_c is the number of Fe nearest neighbors next to a Pt atom. In our case, the ground state values are $\mu_0 = 0.33 \mu_B$, $m_0 = 2.82 \mu_B$ and $N_c = 8$. Substituting these values into (6.2) we get

$$\frac{\kappa}{a} = 0.0292 .$$

On the other hand, a can be calculated from constrained calculations.

Transversal constraints We might try to apply the previous CLSDA calculations with transversal constraint. At $T = 0$, the energy is stationary at any constrained state, so $\frac{\partial H_\nu}{\partial \mu_\nu} = 0$. Using $h_\nu = h_{\nu 0} \cos \theta$,

$$0 = \frac{\partial H_\nu}{\partial \mu_\nu} = \frac{\partial}{\partial \mu_\nu} [-h_\nu \mu_\nu(\vartheta) \cos(\vartheta) + a \mu_\nu^2(\vartheta)] = -h_\nu \cos(\vartheta) + 2a \mu_\nu(\vartheta) \quad (6.3)$$

$$\mu_\nu(\vartheta) = \frac{h_\nu}{2a} \cos(\vartheta) = \mu_{\nu,0} \cos(\vartheta) , \quad (6.4)$$

then substituting this result into the energy formula, yields

$$H_\nu(\vartheta) = a \mu_{\nu,0}^2 \cos^2(\vartheta) . \quad (6.5)$$

Comparing this result with Eq. (6.1) one can notice the relationship $a = -\chi^{(1)}/\mu_0^2$. In this way, we get a value of $a = 100.1 \text{ mRyd}/\mu_B^2$. Note, however, that the ab initio

CLSDA calculations give credit also to a non-negligible fourth order term in the total energy, see Fig. 2, therefore, the transversal constraint doesn't seem to be suitable to derive the parameter a . It should, however, be mentioned that the variation of the Pt moment, Eq. (6.4), is verified by the ab initio calculations.

Longitudinal constraints Applying longitudinal constraint, i.e. fixing the value of the weak moment μ_ν in the ab initio calculations, provides a direct tool to calculate the parameter a , since $H_\nu(\mu_\nu)$ in Eq. (5.1) can be rewritten as

$$H_\nu(\mu_\nu) = a(\mu_\nu - \mu_{\nu,0})^2 - a\mu_{\nu,0}^2, \quad (6.6)$$

with $\mu_{\nu,0} = \frac{\hbar\nu}{2a}$. This is consistent with the requirement that $H_\nu(\mu_\nu)$ should be stationary at $\mu_\nu = \mu_{\nu,0}$. The curvature of the parabola is directly related to the parameter a .

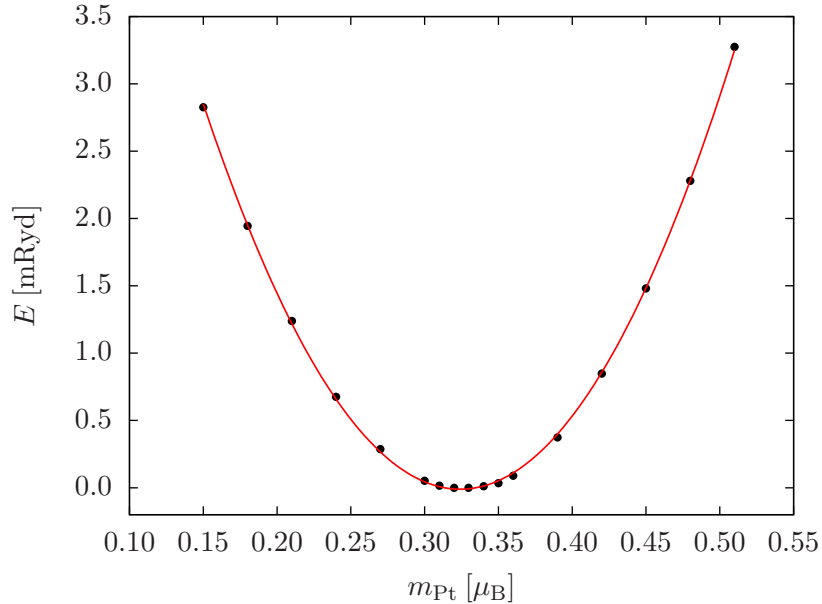


Figure 3: Ab initio total energy as calculated by using longitudinal constraint on the Pt moments. The red solid line shows a parabolic fit.

We performed CLSDA calculations with all moments parallel to $\mathbf{z}$, and applying a longitudinal constraining field only on the Pt moments. The results are depicted in Fig. 3. Obviously, the quadratic behavior of $E(\mu)$ applies not only in the vicinity of $\mu_0 = 0.32 \mu_{\text{B}}$, but in a wide range of μ . This makes the longitudinal constraint more reliable to fit the parameter a as compared to the transversal constraint. The parameter of the fitted parabola is $a = (94.4 \pm 0.4) \text{ mRyd}/\mu_{\text{B}}^2$. Reassuringly, however, this value is very close to that obtained by using transversal constraint. We summarized the resulting parameters in Table 1.

Calculation type	a [mRyd/ μ_B^2]	κ [mRyd/ μ_B^2]
Transversal constraint on Pt	100.1	2.923
Longitudinal constraint on Pt	94.4	2.756

Table 1: The a and κ parameters obtained from CLSDA calculations by using different constraints.

neighbor shell	bare [mRyd]	$R1$ [mRyd]	$R2$ [mRyd]
1	1.143	1.644	1.787
2	-0.184	0.224	0.460
3	0.576	0.890	0.898
4	0.304	0.564	0.626
5	-0.368	-0.206	-0.207
6	-0.261	-0.144	-0.261
7	0.108	0.175	0.108
8	0.029	0.089	0.029
9	-0.018	0.017	-0.018

Table 2: Isotropic exchange interactions calculated by the method of infinitesimal rotations (bare), and as renormalized within the $R1$ and the $R2$ models.

6.3 Exchange interactions from different models

First, by using Eq. (3.2) we calculated the bare Fe-Fe and Fe-Pt interactions. The results are listed in Table 2 and also plotted in Fig. 4a. Note that the first, third, sixth and seventh nearest neighbors (NN) are within the same (001) layer, while the second, fourth, and fifth NN pairs connect subsequent (001) layers. Positive and negative signs of the interactions indicate ferromagnetic (FM) and antiferromagnetic (AF) couplings, respectively. Summing up the interactions between layers, we get effective couplings between layers. These layer-layer couplings are plotted in Fig. 4b. Apparently, the first and second NN interlayer couplings are positive and negative, respectively, indicating that taking into account just bare Fe-Fe interactions the Fe atoms would order parallel within each layer, but antiparallel between adjacent layers (layered AF magnetic structure). However, as clear from the inset of Fig. 4a, the dominantly ferromagnetic Fe-Pt interactions seem to stabilize an FM state for the system.

Next we calculated renormalized Fe-Fe interactions within the spin models $R1$ and $R2$ by using Eqs. (4.5) and (5.8), respectively. In particular, for model $R2$ we used a

simplified expression (see previous section),

$$\tilde{J}_{ij} = J_{ij} + \frac{\kappa^2 m_{\text{Fe}}^2}{2a} c(i, j) , \quad (6.7)$$

where $c(i, j)$ is the number of common first NN Pt atoms of the Fe atoms at sites i and j . For these calculations we employed the spin susceptibilities, $\chi^{(1)}$ and $\chi^{(2)}$, as well as the parameters κ and a as derived from CLSDA calculations. In case of spin model *R2* we choose the longitudinal CLSDA results, because the parabolic fit was quite accurate. We summarized the results in Table 2 and plotted them also in Fig. 4a.

As can be seen, the coupling constants are increased due to the renormalization. The spin model *R2* changes only interactions between the first five Fe pairs since it uses only the nearest Pt neighbors. Quite importantly, the second NN Fe-Fe interaction connecting adjacent Fe layers becomes positive in the renormalized spin models. As a consequence, the first Fe-Fe interlayer coupling becomes positive, see Fig. 4b, implying a FM ground state for the Fe sublattice (and for the whole system, as well). Quite clearly, the two types of renormalization gives rise to very similar Fe-Fe exchange interactions.

6.4 Discussion of the spin models for induced moments

A common feature of the renormalized spin models is that the induced Pt moments mediate an additional interaction between the Fe atoms. Concerning spin model *R1*, in the sums of Eq. (4.5) every possible connections over Pt sites are taken into account between the i -th and the j -th Fe spins. A drawback of the model is that, in order to produce the Weiss field exerted by the Fe atoms on the Pt moments, the bare $J_{\text{Fe-Pt}}$ interactions are used as calculated within Liechtenstein's method. This approach is very questionable, since it treats the Pt moments as if they were strong moments. We stress that even for very small deviations from the ground state the energy change of the system consists of a contribution due to the change of the magnitude of the Pt moments that is not included within the method of infinitesimal rotations.

The above drawback of the spin model *R1* is seemed to be cured in spin model *R2*, since the couplings between the Fe and Pt atoms, $\kappa_{i\nu}$, are obtained from ab initio total energy calculations, where the change of the induced Pt moments due to the magnetic configuration of the Fe atoms is naturally included. Although in this work we just considered nearest neighbor Fe-Pt interactions, the model can be generalized for higher order Fe-Pt interactions as discussed in Section 5. From the expression of the Weiss field,

$$\mathbf{h}_\nu = \sum_j \kappa_{i\nu} m_{\text{Fe}} \mathbf{e}_i,$$

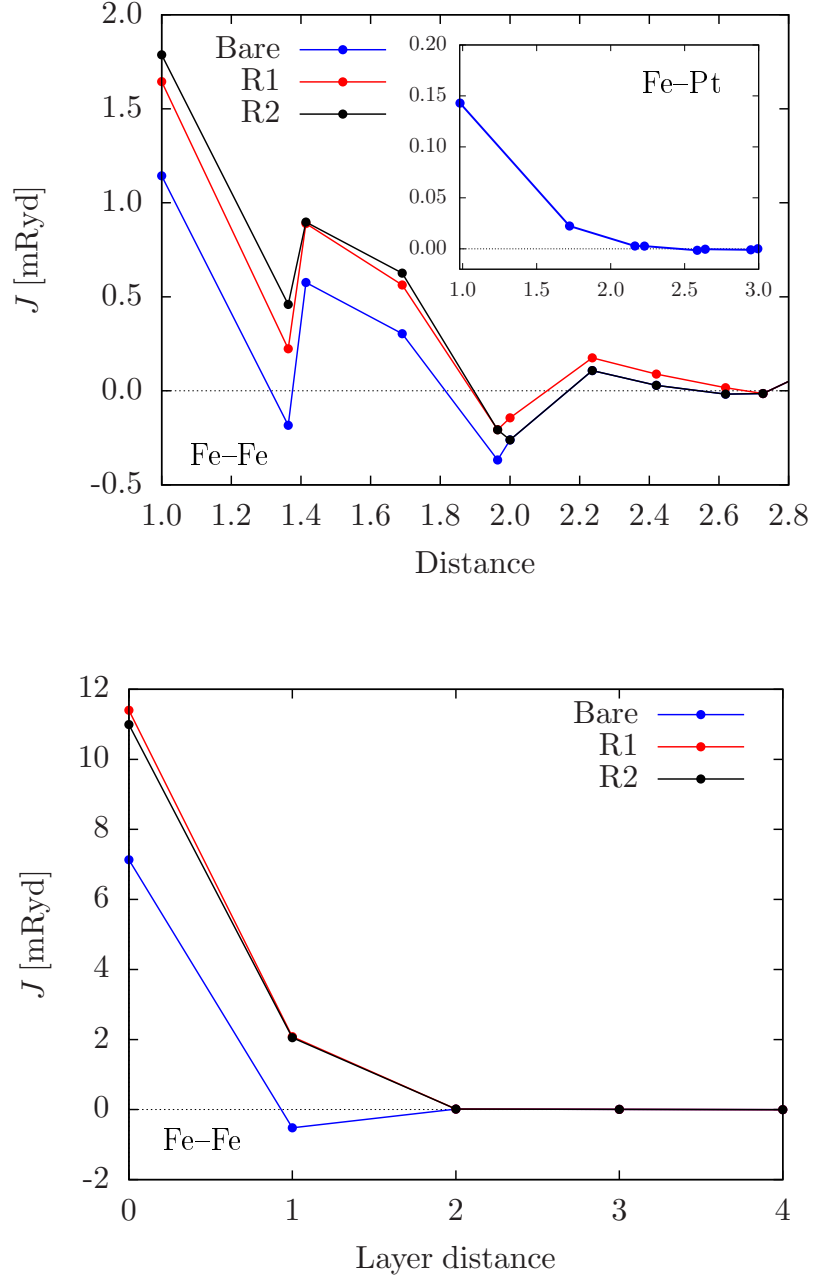


Figure 4: Isotropic exchange interactions calculated within the different models, see Table 2. In the upper panel, the Fe-Fe exchange parameters are plotted as a function of the distance of the Fe sites. The bare Fe-Pt couplings are shown in the inset. In the bottom panel, the exchange couplings between the Fe layers are displayed.

we anticipate the relationship, $\kappa_{i\nu} = J_{i\nu}/m_{\text{Fe}}$. This relationship remains, however, just formal, since $\kappa_{i\nu}$ includes the formation energy of the induced moments, while $J_{i\nu}$ stems from a calculation treating all the moments as strong moments.

Interestingly, there is a formal similarity between the first corrections to J_{ij} ,

$$\frac{m_{\text{Fe}}^2}{4a} \sum_{\nu} \kappa_{i\nu} \kappa_{j\nu} \rightarrow \frac{1}{4a} \sum_{\nu} J_{i\nu} J_{j\nu} \rightarrow -\frac{2\chi^{(1)}}{h_0^2} \sum_{\nu} J_{i\nu} J_{j\nu},$$

where we made use of the relationships $\chi^{(1)} = -a\mu_0^2$ and $\mu_0 = \frac{h_0}{2a}$. The spin model *R2* can, however, provide neither a term similar to the second correction to J_{ij} of spin model *R1*, nor biquadratic couplings [1]. This can be corroborated by the limitation of the spin model *R2* that it can only be used when the induced moment appears up to second order in the energy, Eq. (5.1).

7 Spin-dynamics calculations

After obtaining all the coupling parameters, we can set up finite temperature spin-dynamics calculation. We would like to test the different types of parameters by calculating the magnetization as a function of temperature and also the Curie temperature.

For the simulations we will use the phenomenological Landau–Lifshitz–Gilbert (LLG) equations,

$$\frac{\partial \mathbf{m}_i}{\partial t} = -\gamma \mathbf{m}_i \times \mathbf{h}_i + \frac{\alpha}{m} \mathbf{m}_i \times \frac{\partial \mathbf{m}_i}{\partial t}, \quad (7.1)$$

where γ is the gyromagnetic ratio and α is a damping parameter. The first term describes the precession of the moments. The second term is related to the damping. $\mathbf{h}_i$ is the effective field which contains the Weiss field and a possible external magnetic field. The Weiss field can be deduced from the coupling constants and the magnetic configuration. To investigate finite temperature phenomena we must include the effects of the thermal excitations into the LLG equation. This can be done by adding a stochastic magnetic field to the effective field. By knowing the magnetization at all sites as a function of time, we can obtain the averaged magnetization. For further description of the spin-dynamics calculations see Ref. [15].

To keep the calculations tractable, we must limit the number of interactions. This is fairly possible, since the interaction strength decays strongly with distance (see Fig. 4a). To guess the cutoff distance, we used mean field approximation for the Curie temperature, T_C . In case of a simple lattice, $T_C^{\text{MF}} = \frac{1}{3k_B} \sum_i J_{ij}$, while for a system with two sublattices, the formula becomes [16]

$$T_C^{\text{MF}} = \frac{1}{3k_B} \cdot \max. \text{ eigenvalue} \begin{pmatrix} \sum_i J_{ij} & \sum_i J_{i\nu} \\ \sum_{\nu} J_{i\nu} & \sum_{\nu} J_{\nu\mu} \end{pmatrix}.$$

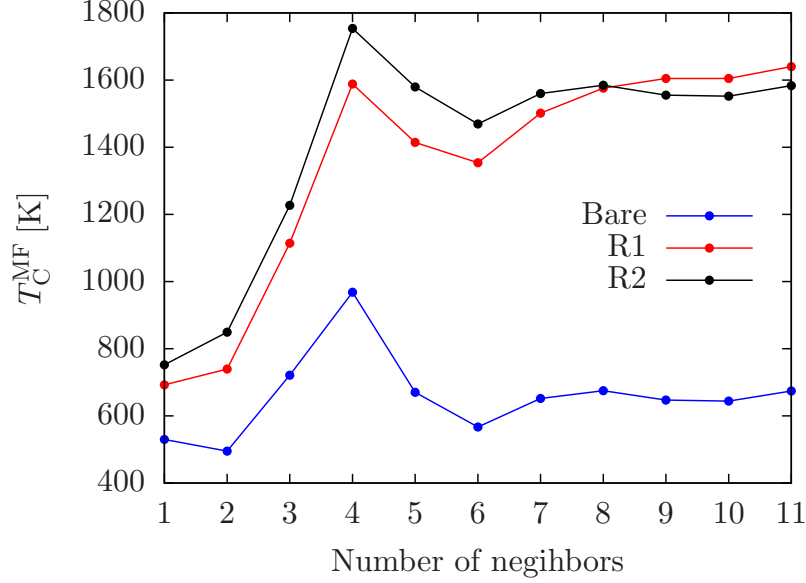


Figure 5: Mean field Curie temperatures of FePt within the different spin models as calculated by considering different numbers of neighbor shells for the Fe atoms.

According to Fig. 5, it is sufficient to consider only 9 neighbor shells, because the interactions between the more distant neighbors don't modify the Curie temperature significantly.

The calculated temperature dependent magnetizations are shown on Fig. 6. As we see, the magnetization curves calculated with the bare couplings and with the renormalized parameters are quite different. The renormalized models results to quite similar Curie temperatures. The critical exponent β was fitted to about 0.33 in both cases. As a consequence of the different lattice structure used in the calculations, the critical exponent from the simulations for the two-sublattice case (bare interactions) is slightly different. Note that the β critical exponent for classical Heisenberg model is $\beta = 0.36$ [17].

The experimental value of the Curie temperature is about 760 K [1]. The calculation using the bare interactions results to a much smaller Curie temperature, $T_C \simeq 192$ K. This seems to be reasonable, since in this case there are only transversal excitations, all the thermal fluctuations try to eliminate the FM order.

On the other hand, since the renormalized models include longitudinal excitations of the Pt moments, these models result in much higher ordering temperatures. However, the simulated Curie temperatures appear to be too high, $T_C \simeq 1300$ K, as compared to the experiment. The spin model *R2* provides a T_C by about 100 K smaller than the spin model *R1*. This means that the exchange interactions for distant pairs are smaller in the

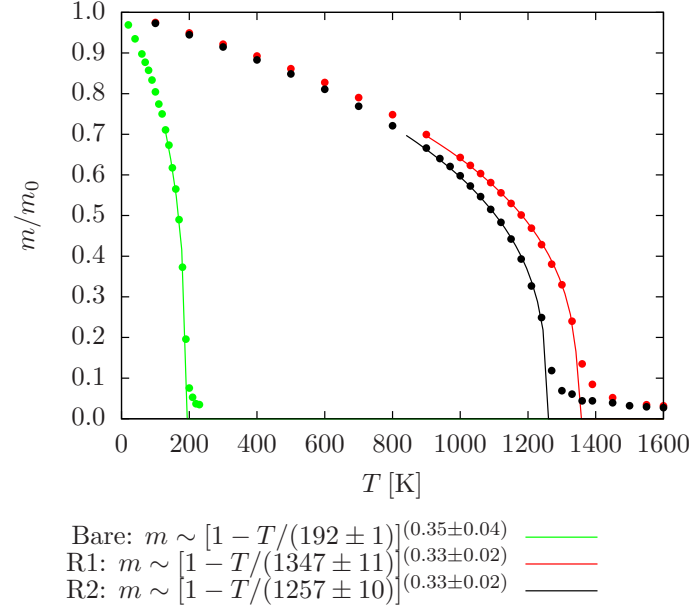


Figure 6: Average magnetization as a function of the temperature. In case of the renormalized spin models the simulation included only the Fe moments. In case of the 'bare model', both the Fe and Pt sublattices were taken into account. The details of the computations: system size $20 \times 20 \times 20$, damping parameter $\alpha = 0.1$, number of Fe neighbors 9, number of Pt neighbors 3.

spin model *R2* than in the spin model *R1*, see Fig. 4a.

8 Summary

The main objective of this work was to compare different strategies to construct spin models for magnetic materials containing induced moments from ab initio calculations. The problem is substantial, since advanced methods of statistical physics applied to magnetic systems, Monte Carlo or Langevin spin-dynamics simulations, can only deal with the orientational degree of freedom of spins, i.e. with spins of fixed length. Lacking new types of spin models that can account for the formation energy of the induced moments, would exclude a large family of magnetic materials, most of them being of primary importance for future technologies, from the previously mentioned theoretical investigations.

First, we briefly reviewed the ab initio basis of our work, namely, the Constrained Local Density Functional Theory together with the Korringa-Kohn-Rostoker method. Beside the widely used method of infinitesimal rotations [6], suitable to construct an isotropic Heisenberg model for stable moments, we described two methods that have been published recently in the literature with the purpose of treating induced moments. These methods share the idea that the induced moments are completely coupled to the magnetic configuration of the strong moments, while the corresponding formation energy is a unique function of the locally induced moment. This enables to design renormalized spin models for the strong moments. The method of Mryasov [1] (model *R1*) allows for quite a general functional form of the formation energy, however, it makes use of the exchange interactions between the strong and the weak moments calculated from the method suitable only for strong moments. The effective spin model proposed by Lezaic et al. [2] (model *R2*) relies on a more sound theoretical basis and the coupling strengths between the strong and weak moments are related to ab initio total energies. In contrast to model *R1*, it is limited to a quadratic form of the formation energy, thus, it can provide with renormalized spin-interactions of less hierarchy than model *R1*.

For the case of ordered FePt alloy, we presented ab initio calculations of these spin models. Our results for the total energy in different constrained magnetic states were consistent with the functional forms assumed in the models, therefore, we could safely deduce the necessary microscopic parameters. The exchange interactions between the Fe atoms turned out to enhance due to renormalization. Moreover, the mostly antiferromagnetic interlayer Fe-Fe interactions become ferromagnetic, stabilizing thus a FM ground state for the system. We also performed spin-dynamics simulations with the renormalized models. Whereas the Curie temperature obtained for model *R2* is by about 100 K less than that obtained from model *R1*, the value of $T_C = 1257$ K is too high as compared to

the experiment, $T_C = 760$ K.

Based on our present results, we cannot decide which method is more accurate, since both models provided with very similar exchange couplings and largely overestimated the experimental Curie temperature. A possible reason for this inadequacy is that all exchange couplings were evaluated from the electronic structure at $T = 0$. It is well-known that transversal spin-fluctuations can modify the electronic structure quite a lot, consequently, the spin-interactions will be modified as well. The electronic theory which accounts for this effect is called the method of Disordered Local Moments (DLM) [18]. In the next future, we plan to use the DLM scheme to calculate the exchange interactions. We also plan further calculations to other magnetic systems, for example to FeRh, where Rh exhibits an induced moment as large as $1 \mu_B$ in the FM state. In addition, FeRh shows a temperature induced AF-FM transition. The nature of this transition is theoretically not yet fully explored [19] and we hope that applying the renormalized spin models will add to the understanding of this transition.

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