

Temperature dependent properties of FePt: effective spin Hamiltonian model

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Abstract. – The model of magnetic interactions in the ordered ferromagnetic FePt is proposed on the basis of first-principles calculations of non-collinear magnetic configurations and shown to be capable of explaining recent measurements of magnetic anisotropy energy (MAE). The site (Fe,Pt) resolved contributions to the MAE have been distinguished with small easy-plane Fe and large Pt easy-axis terms. This model has been tested against available experimental data on temperature dependence of MAE and magnetization. It is shown that the observed unusual temperature dependence of MAE can be quantitatively explained within the proposed model. Thus we find that an effective anisotropic exchange mediated by the induced Pt moment is responsible for the MAE temperature dependence in this material. The latter is expected to be a common feature of 3d-5d(4d) alloys having 5d/4d elements with large spin-orbit coupling and exchange enhanced Stoner susceptibility.

Introduction. – Since the phenomenon of super-paramagnetism has been identified as one of the major limits for the conventional magnetic recording [1], significant research effort has been invested in the development of materials with large magnetic anisotropy energy (MAE). Recent progress in the fabrication and characterization of granular and nano-particulate FePt films [2] puts even more emphasis on the understanding of giant MAE of FePt and its temperature dependence. The latter property appears to be of critical importance for the development of future high density magnetic recording systems in particular for the heat-assisted magnetic recording [1].

A systematic understanding of the temperature dependence of the MAE in itinerant magnets remains a challenge and one of the long standing problems in the theory of magnetism. The proposed model deals with mixed localized and itinerant magnetic moments and thus bears general importance as large anisotropy is achieved by combining strongly magnetic elements with non-magnetic ones, where the latter have large spin orbit coupling.

The chemically ordered $L1_0$ phase of FePt has large uniaxial MAE with the first order anisotropy constant $K_1 \approx 10^8$ erg/cc [1] based on the simple angular variation of MAE $E^{anis} \sim K_1 \sin^2 \theta$. In the $L1_0$ phase the cubic symmetry is broken due to the stacking of alternate planes of the 3d element (Fe) and the 5d element (Pt) along the [001] direction. It

is well established that in this naturally layered ferromagnet the large MAE is mainly due to the contribution from the 5d element having large spin-orbit (s-o) coupling while the 3d element provides the exchange splitting of the 5d sub-lattice [3–5].

Theoretical framework for the temperature dependence of the MAE was mainly developed in the 50-60's. These efforts led to the formulation of a general quantum statistical perturbation theory (QSPT) summarized by Callen and Callen [6]. This theory provides a general approach for calculating the magnetic anisotropy free energy for the effective spin Hamiltonian $H = H^{iso} + H^{anis}$ with a large isotropic H^{iso} and a relatively small anisotropic part H^{anis} . The anisotropic part is taken in the form $H^{anis} = \sum_i k_2 L_2(\mathbf{S}_i)$, where $L_2(\mathbf{S}_i)$ is a normalized polynomial of 2nd order in the case of uniaxial symmetry with \mathbf{S}_i denoting a unit vector (in the classical case) at the atomic site i [6]. This form implicitly assumes well localized magnetic moments leading to a universal parametric relation between MAE ($K_1(T)$) and the magnetization $M(T)$ [6]. The two-sublattice modification of the QSPT proposed by Kuz'min [7] has been successfully applied to describe MAE of the localized 3d-4f magnets. Note, that all these theories predict that in the low temperature region MAE scales as $K_1(T) \sim M^3(T)$ except the very recent work by Skomski et. al. where a phenomenological mean-field two sub-lattice Hamiltonian for L1₀ CoPt led to a $K_1(T) \sim M^2(T)$ dependence [8].

Recent experimental results demonstrate that the uniaxial MAE of epitaxial FePt films can be very accurately fitted to a $K_1(T) \sim M^{2.1}(T)$ dependence in the low temperature range [9, 10]. This observation clearly demonstrates that the contribution of the single-ion anisotropy (leading to the M^3 dependence) is practically missing. Thus, the Hamiltonian used in the QSPT theory does not necessarily reflect all the essential features of the magnetic interactions in L1₀ FePt and possibly also of the other 3d-4d/5d ordered alloys.

In the following we present a model of magnetic interactions in FePt which is constructed and parameterized on the basis of first-principles calculations and is shown to be capable of explaining on the quantitative level recent measurements of the $K_1 \sim M^\gamma(T)$ dependence with non-integer exponent $\gamma = 2.1$ [9, 10]. Thus we propose a microscopic explanation of this unusual behavior and test our microscopic model of magnetic interactions.

The leading contribution to the anisotropic part of the spin Hamiltonian is described as anisotropic exchange mediated by the induced Pt atomic spin moments. The thermodynamic behavior of this Hamiltonian is investigated within the mean-field approximation (MFA) and in the classical limit using both Langevin dynamics and Monte Carlo simulations. We find that proper treatment of the magnetic interactions mediated by the induced Pt moment yield $K_1(M(T))$ and $M(T)$ dependences in a good quantitative agreement with experiment, including the value of T_c . Thus the proposed atomic scale model describes correctly the most important static magnetic properties and thus opens the way for modeling more complex dynamic switching properties [11].

First-principles calculations and model Hamiltonian. – Our analysis begins with an investigation of the isotropic part of the spin Hamiltonian. We start with the constrained local-spin-density-approximation (CLSDA) calculations [12] for a non-collinear arrangement of Fe and Pt atomic spin moments as summarized in Fig. 1. The CLSDA method [13] allows to reduce the many electron problem to a minimization of the Hohenberg-Kohn energy functional $E_{HK}(\rho(\vec{r}), \vec{\sigma}(\vec{r}))$ of charge, $\rho(\vec{r})$, and spin density, $\vec{\sigma}(\vec{r})$, with an additional constraint term which in the case of a non-collinear magnetic configuration leads to a CLSDA functional $E_{CLSDA}(\rho(\vec{r}), \vec{\sigma}(\vec{r}), \vec{h}_i^\perp)$ with an additional Lagrange multiplier \vec{h}_i^\perp having the meaning of an internal magnetic field [12]. This magnetic field is determined self-consistently according to the condition of the desired orientation of the atomic moment \vec{m}_i at the site i . The effect of thermal fluctuations on the electronic and spin sub-systems can be conveniently modeled with

the spin-spiral (SS) configurations representing various degrees of short range order which is found in 3d magnets for temperatures well above T_c [14]. The values of the Fe and Pt moments for these SS configurations are calculated in the local coordinate system, associated with the orientation of the local quantization axis at site i as $M_i = \int_{-\infty}^{e_F} [n_i^{up}(\varepsilon) - n_i^{dn}(\varepsilon)] d\varepsilon$, where the local density of electronic states is a diagonal matrix over the spin indices $n_i^{\sigma,\sigma}$ [15,16]. To summarize the most important results obtained for various SS configurations we introduce the convenient variable $h_\nu = H_\nu/H_\nu^0$ where H_ν is the exchange field at site ν of the Pt sublattice normalized by its value in the FM state H_ν^0 .

In Fig.1 we present the spin moments and total energies of the non-collinear magnetic configurations calculated self-consistently within the CLSDA, using a generalization of the electronic structure method to treat non-collinear magnetic order [12]. In Fig.1a we present normalized values of the Fe and Pt spin moments calculated as a function of h . The dependence shown in Fig. 1a clearly demonstrates a dramatic difference in the degree of localization for Fe and Pt spin moments, respectively. The Fe spin moment remains almost constant as a function of h (or angle θ) indicating its relatively localized nature in terms of the response to the thermal fluctuations. On the other hand, the Pt moment varies linearly with h . This result raises the important question how to develop an adequate model to describe a system with mixed localized and de-localized magnetic degrees of freedom.

The theory of magnetic interactions due to localized magnetic moments is well established. Hence an effective spin Hamiltonian associated with the localized (Fe moments) degrees of freedom can be constructed in the form

$$H_{loc} = - \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i k_{Fe}^{(0)} [\mathbf{S}_i^z]^2, \quad (1)$$

which relies on configuration independent effective exchange interaction parameters J_{ij} and an effective single-ion anisotropy $k_{Fe}^{(0)}$. The \mathbf{S}_i are used to denote Fe sublattice spin moments which can be treated as unit vector in the classical limit. The form of the spin Hamiltonian Eq. 1 is well justified by our CLSDA results which clearly indicate that the Stoner excitations associated with Fe moments have much higher energy than those of the Pt.

On the other hand, the value of the induced Pt magnetic moment varies between 0 and a maximum value for the FM state. The CLSDA total energy calculations without s-o coupling (isotropic energy) presented in Fig. 1b allow to clarify how to deal with Pt magnetic degrees of freedom. Indeed, as the scalar-relativistic calculations show, the total energy associated with these delocalized degrees of freedom E_{deloc}^{iso} follows very closely the relation which can be derived from the Stoner-model expression for the total energy [17]

$$E_{deloc}^{iso} = \int_{-\infty}^{e_F} d\varepsilon [n^{up}(\varepsilon) + n^{dn}(\varepsilon)] \varepsilon - \frac{1}{2} I M_{Pt}^2 \approx \tilde{I} [m_\nu]^2 \quad (2)$$

where I is the intra-atomic exchange interaction parameter, M_ν^0 is the Pt magnetic moment in the FM state and n^{up}, n^{dn} are spin resolved densities of states forming delocalized Pt moments denoted as M_ν with $\tilde{I}_\nu = 1/2 I_\nu [M_\nu^0]^2$ and $m_\nu = M_\nu/M_\nu^0$. The dependence shown in Fig. 1a also demonstrates that both the value and orientation of the Pt moment are entirely due to the exchange field of the surrounding Fe moments, following very closely the relation $\mathbf{m}_\nu = \chi_\nu \mathbf{h}_\nu$, where χ_ν is the local Pt susceptibility constant which has very weak temperature dependence which is neglected in further statistical simulations. As follows from the linear dependence in Fig. 1a, m_ν can be very accurately described by the relation,

$$\mathbf{m}_\nu = \frac{1}{J_\nu^0} \sum_i J_{i\nu} \mathbf{S}_i, \quad (3)$$

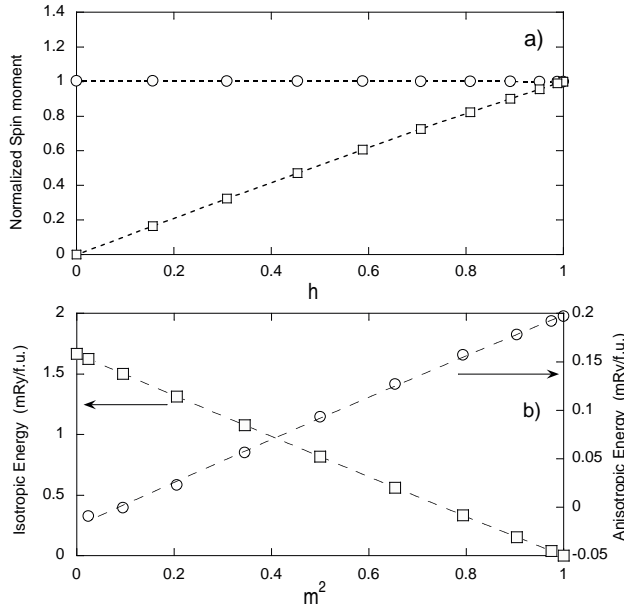


Fig. 1 – Results of the constrained self-consistent LSDA calculations for ordered $L1_0$ FePt: a) Fe (circles) and Pt (squares) spin moments normalized by their values in the FM state as a function of normalized exchange field h ; b) isotropic (squares) and anisotropic (circles) part of the total energy as a function $[m_\nu]^2$. Dashed lines used for the linear fit.

where the $J_{i\nu}$ are the effective exchange parameters defined as the CLSDA total energy variation $\delta E_{CLSDA}/\delta \mathbf{S}_i \delta \mathbf{m}_\nu$ in the FM state, where \mathbf{S}_i is the Fe and \mathbf{m}_ν the Pt sub-lattice moment. The $J_\nu^0 = \sum_i J_{i\nu}$ is the molecular field parameter defined in the FM state. Eq.3, which reflects the results of first-principles calculations, implies that χ_ν is practically independent of the magnetic configuration. The temperature dependence of χ_ν arising from the Fermi distribution smearing is weak and will be neglected in the following statistical simulations.

The anisotropic part of the effective spin Hamiltonian is calculated within the CLSDA, with s-o interactions included self-consistently, and is presented in Fig. 1b. We find that it can be very accurately approximated by a quadratic dependence on the m_ν parameter. We should emphasize that owing to the delocalized nature of 5d/4d elements, in the general case this dependence cannot be guessed prior to the rigorous calculations. However, our result for FePt allows us to identify the form of the spin Hamiltonian associated with delocalized magnetic degrees of freedom, $H_{deloc} = H_{deloc}^{iso} + H_{deloc}^{anis}$.

$$H_{deloc} = - \sum_{\nu} \tilde{I} \mathbf{m}_\nu^2 - \sum_{\nu} k_{Pt}^{(0)} (\mathbf{m}_\nu^z)^2. \quad (4)$$

The magnetic energy is partitioned into localized and delocalized contributions using the CLSDA approach allowing for a unified description of the electronic degrees of freedom within the one-electron approximation. In particular, the Fe $k_{Fe}^{(0)}$ and the Pt single-ion $k_{Pt}^{(0)}$ contributions can be distinguished. In agreement with a previous study [3] we find that the Fe contribution is negative while Pt gives rise to a large easy axis contribution. The Fe and Pt contributions to MAE have been calculated within the LSDA and then corrected according to the previous LSDA+U calculations [5]. We find $k_{Pt}^{(0)} = 1.427$ meV and $k_{Fe}^{(0)} = -0.097$

meV which corresponds to the macroscopic uniaxial anisotropy constant $K_1(T=0) = 7.7 \cdot 10^7$ erg/cc.

Finally, with Eqs. (1,3,4) we can introduce an effective spin Hamiltonian reflecting all the above features revealed by our first-principles calculations. It is constructed as $H = H_{loc.} + H_{deloc}$ and can be reduced to the convenient form

$$H = - \sum_{i \neq j} \tilde{J}_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i d_i^{(0)} (\mathbf{S}_i^z)^2 - \sum_{i \neq j} d_{ij}^{(2)} \mathbf{S}_i^z \mathbf{S}_j^z. \quad (5)$$

We note that the spin Hamiltonian is now expressed in terms of the Fe degrees of freedom, with effective exchange interaction parameters $\tilde{J}_{ij} = J_{ij} + \tilde{I}/(J_\nu^0)^2 \sum_\nu J_{i\nu} J_{j\nu}$ and an effective single-ion anisotropy,

$$d_i^{(0)} = k_{Fe}^{(0)} + \frac{k_{Pt}^{(0)}}{(J_\nu^0)^2} \sum_\nu J_{i\nu}^2, \quad (6)$$

and a two-ion anisotropy contributions

$$d_{ij}^{(2)} = \frac{k_{Pt}^{(0)}}{(J_\nu^0)^2} \sum_\nu J_{i\nu} J_{j\nu}. \quad (7)$$

As can be seen from these expressions, the Pt induced spin moments result in additional isotropic and anisotropic contributions, both depending on the effective exchange interaction parameters $J_{i\nu}$ defined in the ferromagnetic ground state. We find that $J_{i\nu}$ are relatively strong and positive, resulting in isotropic and anisotropic exchange interactions both stabilizing the ferromagnetic order in the [001] direction (see [18]). Unlike the $J_{i\nu}$, the effective exchange interaction parameters between Fe moments J_{ij} appear to be sensitive to the lattice spacing and may change sign from positive to negative as a function of the chemical ordering [19]. Importantly, the additional anisotropic contribution (Eqs.7) which controls $K_1(M(T))$ does not depend on J_{ij} .

From the structure of the Eq. 5 one can draw initial conclusions. One of them is that the anisotropic part of the H_{deloc} leads to an effective two-ion contribution mediated by $J_{i\nu}$ exchange interactions. Within the MFA, the two-ion term contributes to the temperature dependence as $k_{Pt}^{(0)}/(J_\nu^0)^2 \sum_\nu J_{i\nu} J_{j\nu} \langle \mathbf{S}_{Fe}^z \rangle^2 \sim M^2(T)$. In order to assess the relative magnitudes of the single- and two-ion terms, consider for clarity nearest neighbor interaction only. From Eqs. 6 and 7, given the small magnitude of $k_{Fe}^{(0)}$, $d_i^{(0)}$ and $d_{ij}^{(2)}$ are of the same magnitude. However, the total two-ion contribution at site i involves a sum over nearest neighbors, resulting in an increase of the total two-ion contribution over the single-ion contribution equal to the coordination number. Clearly, the exact increase will depend on the distance dependence of the J_{ij} meaning that the exact ratio will be alloy dependent. However, it is reasonable to expect that in general the two-ion contribution will be an order of magnitude larger than the single-ion term. In the following we present calculations beyond MFA to provide a quantitative analysis of the proposed model in terms of its ability to explain the non-integer exponent of the $K_1 \sim M^{2.1}$ dependence.

Thermodynamic properties. – In the following, we use Langevin dynamics simulations described in [20] to investigate properties of the effective spin Hamiltonian Eq. 5 within the classical approximation. The exchange interactions are long-ranged and are taken into account for a distance of up to 5 atomic unit cells via fast-Fourier-transformation, within the same

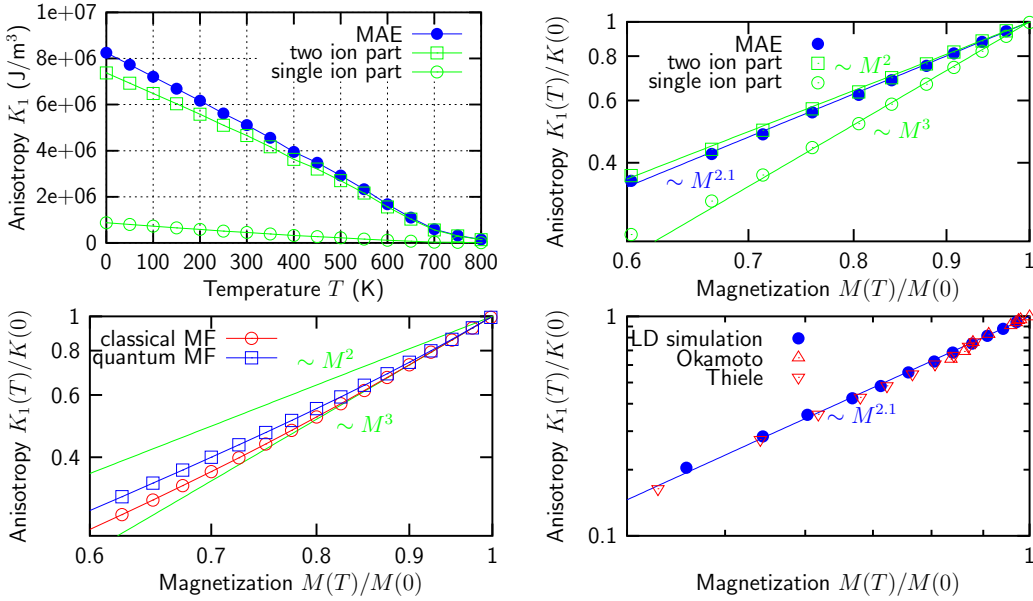


Fig. 2 – (a) $K_1(T)$ dependence using LD simulations with the effective spin Hamiltonian and its single and two-ion contributions; (b-d) log-log plots for $K_1(T)/K_1(0)$ vs. reduced magnetization $M(T)$: (b) using LD simulation within the classical approximation; (c) calculated within the MFA QSPT for quantum $j = 3/2$ and classical $j = \infty$, $M^2(T)$ and $M^3(T)$ dependence are presented for comparison; (d) comparison of $K_1(T)$ calculated with LD and experimental data of Thiele et al. [9] and Okamoto et al. [10]. The solid lines gives a power law with exponent 2.1.

calculation as the dipolar interaction. We simulate spherical nanoparticles with open boundary conditions and sizes up to 14464 moments, corresponding to diameters up to 9.2nm. The anisotropy constant defined as the free energy difference between magnetization oriented parallel or perpendicular to the easy axis is calculated according to the first order thermodynamic perturbation theory [6] as the internal energy difference

$$K_1(T) = E_a(T, \vec{B} = B\vec{e}_\perp) - E_a(T, \vec{B} = B\vec{e}_\parallel). \quad (8)$$

Then, according to Eq. 5 single-ion and two-ion contributions can be distinguished as summarized in Fig. 2a. One can see that the two-ion term is the dominant contribution which is nearly nine times larger. Furthermore the data indicate a Curie temperature close to the experimental value of 750K [9, 10]. In Fig. 2b we present the calculated $K(M)$ dependence along with its single and two-ion contributions. Within these calculation, which we stress go beyond MF classical calculations, the two-ion term scales as $M^2(T)$ in a wide T range, while the single-ion term follows $M^3(T)$ scaling only at low temperatures. Since $K_1(T)$ has both contributions, the expression for the low T expansion

$$K_1(T)/K_1(0) \approx \alpha M^2 + (1 - \alpha) M^3 \sim M^{3-\alpha}, \quad (9)$$

contains the α coefficient originating from the normalized two-ion contribution and the second term originating from the single-ion contribution. The parameters $d_i^{(0)}$ and $d_{ij}^{(2)}$ following from our first-principles calculations allow us to evaluate finally the exponent of $3 - \alpha = 2.09$.

Before proceeding to a comparison with experiments we examine the range of validity of our classical statistical approximation. In Fig. 2c we present results of the MFA QSPT calculations of the $K_1(M)$ dependence and corresponding two and single-ion contributions. Both, the classical and the quantum $K_1(M)$ dependence are identical for the two-ion term in the whole range of temperatures and for the single-ion term in the low temperature range. Considering that the single-ion contribution is dominant, we can compare our Langevin dynamics calculations with available experiment as shown in Fig. 2d. As one can see, our spin Hamiltonian with ab-initio parameterization agrees very well in a wide range of temperatures, especially given that the low temperature measurements by Okamoto, et al. also yielded an exponent of 2.1 [10].

Summary and Conclusions. – To summarize, we propose an atomic-scale model of magnetic interactions in ordered L1₀ FePt with an effective spin Hamiltonian constructed and parameterized on the basis of first-principles calculations. The proposed model is investigated analytically and using statistical simulations. We find that the model describes on the quantitative level the experimentally observed anomaly in the temperature dependence of the magnetic anisotropy energy. We demonstrate that this observed, anomalous temperature dependence ($K_1 \sim M^{2.1}(T)$) is due to the delocalized induced Pt moments, leading to an exchange mediated two-ion anisotropy which dominates the usually expected M^3 contribution of the single-ion anisotropy. We believe that this mechanism is common for various 3d-5d/4d ordered alloys having 5d/4d nominally non-magnetic elements with large s-o coupling and exchange enhanced susceptibility.

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