

Temperature-dependent magnetic properties of FePt: Effective spin Hamiltonian model

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Abstract. – A 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 Fe easy-plane and large Pt easy-axis terms. This model has been tested against available experimental data on the temperature dependence of MAE showing scaling of uniaxial MAE ($K_1(T)$) with magnetization ($M(T)$) $K_1(T) \sim M(T)^\gamma$ characterized by the unusual exponent of $\gamma = 2.1$. It is shown that this unusual behavior of the FePt can be quantitatively explained within the proposed model and originates from an effective anisotropic exchange mediated by the induced Pt moment. 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 the 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 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 $E^{\text{anis}} \sim K_1 \sin^2 \theta$. 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].

The theoretical framework for the temperature dependence of the MAE was mainly developed in the '50-'60s. 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 $H = H^{\text{iso}} + H^{\text{anis}}$ with a large isotropic H^{iso} and a relatively small anisotropic part H^{anis} . The anisotropic part is taken in the form $H^{\text{anis}} = \sum_i k_2 L_2(\mathbf{S}_i)$, 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 mean-field two-sub-lattice Hamiltonian for L1_0 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 L1_0 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 yields $K_1(M(T))$ 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 even more complex dynamic switching properties [11].

Effective spin Hamiltonian model based on first-principles calculations. – 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 $E_{\text{HK}}(\rho(\vec{r}), \vec{\sigma}(\vec{r}))$ 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 $E_{\text{CLSDA}}(\rho(\vec{r}), \vec{\sigma}(\vec{r}), \vec{h}_i^\perp)$ 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

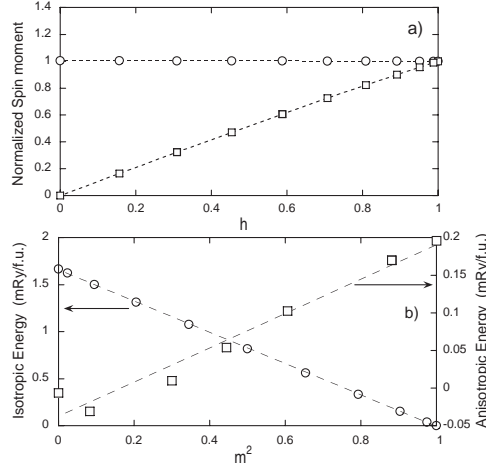


Fig. 1 – Results of the constrained self-consistent LSDA calculations for ordered $L1_0$ FePt: a) Fe (circles) and Pt (squares) spin moments normalized by their values in the FM state as a function of normalized exchange field h ; b) isotropic (squares) and anisotropic (circles) part of the total energy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

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 $M_i = \int_{-\infty}^{e_F} [n_i^{\text{up}}(\varepsilon) - n_i^{\text{dn}}(\varepsilon)] d\varepsilon$, 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

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

which relies on configuration-independent effective exchange interaction parameters J_{ij} and an effective single-ion anisotropy $k_{\text{Fe}}^{(0)}$. 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 $E_{\text{deloc}}^{\text{iso}}$ follows very closely the relation which can be derived from the Stoner-model expression for the total energy [17]:

$$E_{\text{deloc}}^{\text{iso}} = \int_{-\infty}^{\epsilon_{\text{F}}} d\varepsilon [n^{\text{up}}(\varepsilon) + n^{\text{dn}}(\varepsilon)]\varepsilon - \frac{1}{2}IM_{\text{Pt}}^2 \approx \tilde{I}[\mathbf{m}_{\nu}]^2, \quad (2)$$

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 $\tilde{I}_{\nu} = 1/2I_{\nu}[M_{\nu}^0]^2$ 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. As follows from the linear dependence in fig. 1(a,b), m_{ν} can be very accurately described by the relation

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

where the Pt sublattice χ_{ν} is found to have a weak magnetic configuration dependence; the $J_{i\nu}$ are the effective exchange parameters defined as the CLSDA total energy variation $\delta E_{\text{CLSDA}}/\delta \mathbf{S}_i \delta \mathbf{m}_{\nu}$ in the FM state, where \mathbf{S}_i is the Fe and \mathbf{m}_{ν} the Pt sub-lattice moment. The 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, $H_{\text{deloc}} = H_{\text{deloc}}^{\text{iso}} + H_{\text{deloc}}^{\text{anis}}$,

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

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 $k_{\text{Fe}}^{(0)}$ and the Pt single-ion $k_{\text{Pt}}^{(0)}$ contributions can be distinguished. In agreement with a previous study [4], we find that the Fe contribution is an easy-plane 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 $k_{\text{Pt}}^{(0)} = 1.427$ meV and $k_{\text{Fe}}^{(0)} = -0.097$ 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 $H =$

$H_{\text{loc}} + H_{\text{deloc}}$ 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 $\tilde{J}_{ij} = J_{ij} + \tilde{I}(\frac{\chi_\nu}{M_\nu^0})^2 \sum_\nu J_{i\nu} J_{j\nu}$ and an effective single-ion,

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

and a two-ion anisotropy contribution,

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

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 ferromagnetic order in the [001] direction [18]. Unlike the $J_{i\nu}$, the effective exchange interaction parameters between Fe moments J_{ij} appear to be sensitive to the chemical order and might be leading to the FM order instability [19].

In order to assess the relative magnitudes of the single- and two-ion terms (eq. (5)), consider for clarity the nearest-neighbor (NN) interaction only with N being the number of NNs. Then the magnetic-anisotropy free energy ($F_{\text{anis}}(T)$) within the first-order thermodynamic theory [6] (justified by $\langle H_{\text{anis}} \rangle / \langle H_{\text{iso}} \rangle \approx 0.1$, see fig. 1b) has an effective single- and two-ion contributions with the latter involving a sum over nearest neighbors:

$$F_{\text{anis}}(T) \approx \langle H_{\text{anis}} \rangle_T = d_i^{(0)} f^1(T) + (N-1) d_{ij}^{(2)} f^2(T), \quad (8)$$

where $f^1(T) = \langle S_i S_i \rangle_T$ and $f^2(T) = \langle S_i S_j \rangle_T$ are the single-site and pair correlation functions, $d_i^{(0)} \approx k_{\text{Fe}}^{(0)} + k_{\text{Pt}}^{(0)} / N$ and $d_{ij}^{(2)} \approx k_{\text{Pt}}^{(0)} / N$ within the NN approximation. Given the small magnitude of $k_{\text{Fe}}^{(0)}$, the ratio between the single- and two-ion contributions $(N-1) d_{ij}^{(2)} / d_i^{(0)} \approx (N-1)$. Then, given the dominance of the two-ion contribution, and since within the MFA-type approximation $f^2(T) \sim M^2(T)$, one can arrive at a qualitative explanation of the observed MAE temperature dependence. Clearly, for more accurate evaluation of the ratio between two- and single-ion contributions the distance dependence of the $J_{i\nu}$ has to be taken into account. In the following we present calculations beyond MFA and NN approximations 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 [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 as well as dipolar interactions. We simulate spherical nano-particles with open boundary conditions and sizes up to 14464 moments, corresponding to diameters up to 9.2 nm. The anisotropy constant is defined as the free-energy difference between magnetization oriented parallel or perpendicular to the easy axis which, according to the first-order perturbation theory [6], is given by the internal

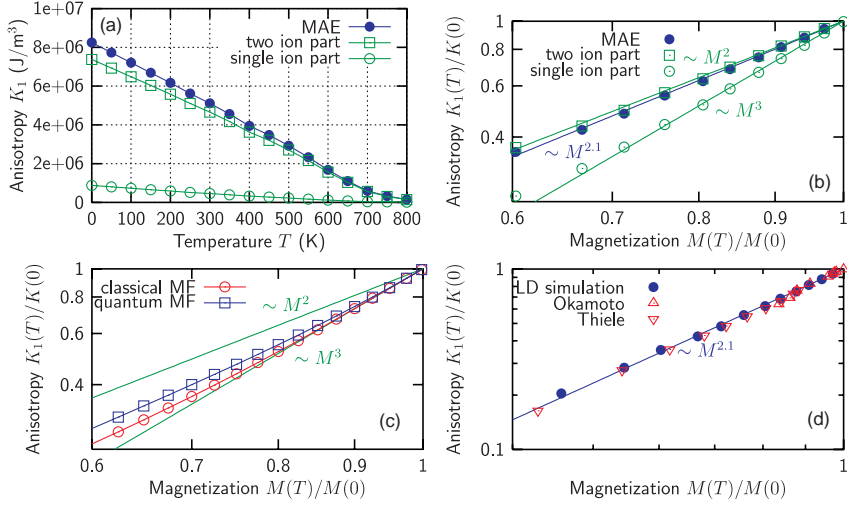


Fig. 2 – (a) $K_1(T)$ -dependence using LD simulations with the effective spin Hamiltonian and its single- and two-ion contributions; (b-d) log-log plots for $K_1(T)/K_1(0)$ vs. reduced magnetization $M(T)$: (b) using LD simulation within the classical approximation; c) calculated within the MFA QSPT 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 give a power law with exponent 2.1.

anisotropic energy difference $K_1(T) = E_a(T, \vec{B} = B\vec{e}_\perp) - E_a(T, \vec{B} = B\vec{e}_\parallel)$ for the external field \vec{B} . 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 750 K [9,10]. In fig. 2b we present the calculated $K(M)$ -dependence along with its single- and two-ion contributions. Within these calculations, which we stress go beyond MF classical approximation, 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. This 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 Stoner enhanced susceptibility.

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