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The Curie temperature distribution of FePt granular magnetic recording media

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We present atomistic calculations of the magnetic phase transition behavior in an $L1_0$ FePt system to study the effect of grain size distribution on the Curie temperature (T_c) dispersion with relevance to heat assisted magnetic recording. Identifying the relation between the size and T_c of a grain by means of finite size scaling analysis of the differentiated magnetization versus T data allows to show that a lognormal size distribution transforms into a lognormal T_c distribution with moments dependent on the critical exponents. We also address the question of the universality class of FePt.
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Heat assisted magnetic recording (HAMR)[1–4] is the most likely candidate technology to achieve recording densities significantly beyond those accessible to conventional perpendicular magnetic recording. The recently announced demonstration of a density of 1 TBit/in^2 brings a transition to HAMR technology a step closer. An advantage of HAMR, noted in Ref. 1, is the fact that requirements of increasing head field gradients, which limit achievable areal densities in conventional perpendicular recording, are reduced by the large temperature gradients produced by the laser heating. However, in order to maximize the effect the rate of change of magnetization with temperature, dM/dT, must also be high, suggesting recording close to the Curie temperature T_c. This implies the necessity to understand static and dynamic magnetic behavior near T_c such as, for example, the recently predicted linear reversal mechanism[5,6] or the influence of grain size distribution $f_D(D)$ on the T_c dispersion $f_T(T_c)$—the understanding of which will be the main subject of the present work. Indeed, uncontrolled $f_T(T_c)$ may result in smeared $M(T)$ dependence and thus reduced dM/dT near T_c, and as such is a crucial factor with a potentially detrimental effect on HAMR.

The approach undertaken here to relate $f_D(D)$ and $f_T(T_c)$ relies on exploring the critical behavior in magnetic grains of variable size D. The task is to quantify the size dependence of the Curie temperature, $T_c(D)$, extracted from $M(T)$ data computed from an atomistic model of ordered ferromagnetic FePt (Fig. 1(a)), which is, with its low bulk Curie temperature T_c^∞ and large anisotropy constant K,[7,8] currently regarded as the best candidate material for HAMR media. The $T_c(D)$ dependence is expected to follow the finite size scaling law:

$$\epsilon_c(D) = \frac{T_c^\infty - T_c(D)}{T_c^\infty} = \left(\frac{d_0}{D}\right)^\lambda, \qquad (1)$$

where λ is the so-called phenomenological shift exponent and d_0 is the microscopic length scale close to the dimension of a single unit cell in the lattice structure of the material, see, e.g., Refs. 9–11. The exponent λ is related to the correlation length universal critical exponent ν to be defined below and it is expected that $\lambda \leq \nu^{-1}$ depending on the nature of the experiment;[12,13] $\lambda = \nu^{-1}$ in the absence of higher order system size effects, which will be shown to be the case also here.

Given only limited studies of size dependent critical behavior of FePt,[11,14] available critical parameters are insufficient for careful comparison with our study. Therefore, we consider two different models: an FePt effective Hamiltonian with a long-range exchange (LE) term derived from density functional theory (DFT) calculations,[8] and a "test" model with the nearest neighbor exchange (NE), which is expected to closely resemble the critical behavior of the well studied Heisenberg model[15] and thus allows validation of the present approach. Thereby we demonstrate that because of LE interaction, FePt may not belong to the universality class of the NE Heisenberg model, giving rise to a weaker variation of T_c with D according to Eq. (1). Equation (1) also allows relating $f_T(T_c)$ to $f_D(D)$, and we show analytically that the realistic choice of lognormal distribution $f_D(D)$ leads precisely to the lognormal form of $f_T(T_c^\infty - T_c)$.

The LE atomistic model used in the present study utilises an effective Hamiltonian of FePt parametrized based on DFT calculations,[8] which is of the form

$$\mathcal{H} = -\sum_{i \neq j}(J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j + K_{ij}S_i^z S_j^z) - \sum_i K_i (S_i^z)^2, \qquad (2)$$

where J_{ij} is effective exchange (considered long-ranged as its range extends beyond five lattice spacings), and K_i and K_{ij} are, respectively, the effective uniaxial single-ion and two-ion anisotropy contributions defined in more detail in Ref. 8. The NE Hamiltonian is obtained from Eq. (2) by specifying a nearest neighbor only exchange $J_{ij} = 3.0^{-21}$ J/link and by setting $K_{ij} = 0$. The anisotropy term K_i remains included. In both NE and LE Hamiltonians, the spin moments are expressed as unit vectors $\mathbf{S}_i = \mu_i/|\mu_i|$. The system is integrated using the Landau-Lifshitz-Gilbert equation with the Langevin Dynamics formalism.[16,17] The system is integrated using the Heun numerical scheme and a timestep

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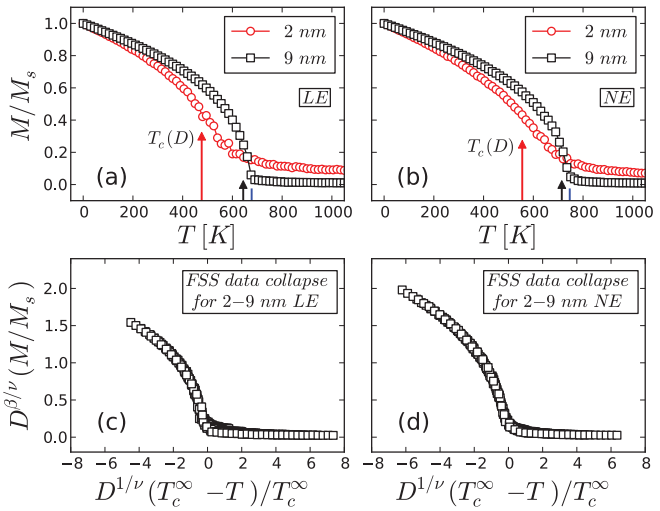

FIG. 1. (a) Computed M vs T data for LE FePt model and (b) nearest NE model, for 2 nm and 9 nm grains. Vertical arrows indicate finite size Curie temperatures $T_c(D)$ determined as minima of $\Delta M/\Delta T$ vs T for every grain size D (also in Fig. 2). The vertical line corresponds to the bulk T_c^∞ listed in Table I. (c) A scaling collapse according to Eq. (3) for LE and (d) NE models, including grain sizes $2, 3, \ldots, 9$ nm and critical parameters T_c^∞, ν, and β summarised in Table I.

of 1.0×10^{-15} s. Since we are interested only in equilibrium properties we set the precessional damping constant $\alpha = 1.0$ for computational efficiency. Simulated samples were cut from a bulk crystal in the shape of a cylinder with diameter D and height D varying in the range 2 nm to 9 nm in 1 nm increments. The system for every D was equilibrated at each temperature for 20 000 time steps and then a thermodynamic average magnetization calculated over further 20 000 time steps. Examples of $M(T)$ data computed in this way for $D = 2$, 9 nm for LE FePt and NE models are shown Figs. 1(a) and 1(b), respectively.

In general terms, the behavior of a bulk system near its Curie temperature T_c^∞ is described by divergent tendencies of some of its thermodynamic variables.[15] For example, the correlation length ξ which quantifies the extent of magnetic ordering in a system is expected to follow a power law divergence $\xi \sim d_0|\epsilon|^{-\nu}$ where the reduced temperature is defined as $\epsilon = (T_c^\infty - T)/T_c^\infty$ similarly to the critical ϵ_c introduced in Eq. (1), and ν is the correlation length universal critical exponent. Other characteristics approach a finite value as $T \to T_c^\infty$. For example, the magnetization M approaches zero following a power law with the "magnetization" universal critical exponent β : $M \sim |\epsilon|^\beta$. Strictly speaking, such non-analytic behaviors exist only in the thermodynamic limit. Thermodynamic quantities for systems with some characteristic dimensions D finite are always rounded, shifted, and smeared out, and as a result, the critical region becomes hard to access. These difficulties appear as soon as the correlation length ξ becomes comparable with the smallest characteristic dimension D, i.e., $\xi \sim D$ or equivalently $d_0|\epsilon|^{-\nu} \sim D$, which directly implies a shift of the size-dependent $T_c(D)$ away from the bulk value T_c^∞, thus motivating the phenomenological Eq. (1).

This behavior is demonstrated in Figs. 1(a) and 1(b) where in both cases of LE FePt and NE models, the reduction of the finite size $T_c(D)$ from the bulk T_c^∞ becomes more pronounced for small 2 nm grains. Figure 2 further confirms this systematic trend for grains of diameters in the range $2, \ldots, 9$ nm. As conventional in experiments, e.g., Refs. 10 and 11, we determined the finite size $T_c(D)$ as corresponding to the minimum of derivative $\Delta M(T)/\Delta T$ as illustrated in the inset for a 3 nm grain. Figure 2 shows that the variation of $T_c(D)$ is slightly weaker for the LE FePt model than for the NE model, which suggests a difference between the shift exponents λ. This issue will be addressed below. To apply Eq. (1) to the data in Fig. 2, it is first necessary to identify the bulk T_c^∞ which, although often readily accessible in experiments, is hard to access in computations.

To extrapolate the finite size computational data to the thermodynamic limit to identify the bulk T_c^∞, we utilize the finite system size scaling (FSS) analysis.[18,19] The FSS method allows, in addition to T_c^∞, a direct extraction of critical exponents ν and β. It is based on the concept of universality,[19] which in the context of the present study implies (1) independence of universal critical exponents on D and (2) data similarity near the critical point, i.e., that after rescaling the $M(T)$ data for all different D by the same appropriate combinations of critical exponents and non-universal parameters, they will all collapse onto a universal curve which we denote as $y = \tilde{\mathcal{M}}(x)$. Specifically, the FSS scaling theory for the one-dimensional scaling relevant to the present study gives the following FSS scaling ansatz when expressed in terms of the reduced temperature ϵ:[19]

$$M(\epsilon) \sim D^{-\beta/\nu}\tilde{\mathcal{M}}(D^{1/\nu}|\epsilon|). \tag{3}$$

Thus, it is expected that for the optimum set of parameters T_c^∞, β, and ν, the $M(T)$ data for all different particle sizes $D = 2, \ldots, 9$ will collapse onto $\tilde{\mathcal{M}}(x)$ after rescaling the M-axis by the factor $D^{\beta/\nu}$ and T-axis by $D^{1/\nu}|\epsilon|$. This is indeed confirmed in Figs. 1(c) and 1(d), which show excellent quality of data scaling for both LE FePt and NE models assuming scaling parameters summarized in Table I. The optimization

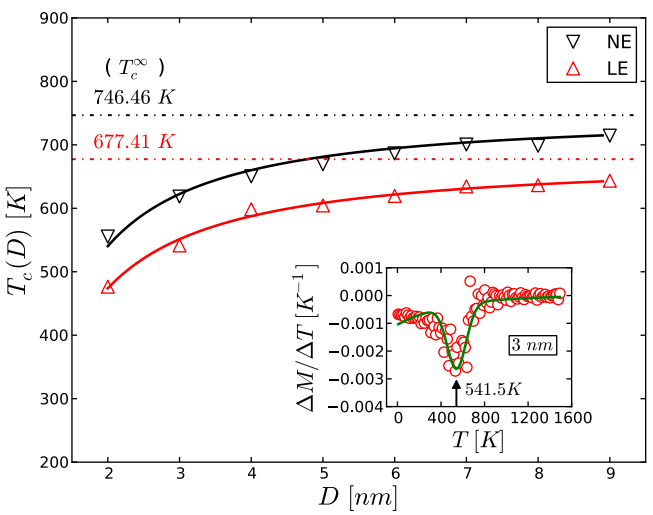

FIG. 2. Dependence of the Curie temperature T_c on the particle diameter D for NE and LE FePt systems. T_c for every D corresponds to a minimum of the differentiated M vs T dependence, as illustrated in the inset for 3 nm LE FePt grain. Solid lines through the triangular points are fits of Eq. (1) with d_0 being the only fit parameter and the bulk T_c^∞ (dashed lines) and ν obtained from the FSS analysis and listed in Table I.

procedure for finding the parameters T_c^∞, β, ν in Table I leading to the best scaling shown in Figs. 1(c) and 1(d) was based on minimizing, in the least-squares sense, the differences between the vertical coordinates of all data points for every D using the interpolation approach reviewed in Ref. 20.

According to the summary given in Table I, the $T_c^\infty \sim 677.4 \pm 10.7$ K obtained from the FSS analysis of the LE FePt model data is somewhat smaller than the expected value of 750 K for bulk FePt.[11] However, given that there are no adjustable parameters (all parameters for the FePt Hamiltonian are determined from DFT calculations[8]), the agreement is reasonable. Moreover, according to Ref. 7, the size dependent structural ordering effect not included in the present modeling can result in the increase of T_c^∞ of the bulk FePt, which might account for the difference. For the NE model, critical exponents agree within the confidence interval with $\beta = 0.36$ and $\nu = 0.71$ for the isotropic Heisenberg model[15] which validates the present approach. This also suggests that in the NE case the value of the anisotropy constant K_i in Eq. (2) is insufficient to induce a crossover to the universality class of the 3D Ising model for which $\beta = 0.325$ and $\nu = 0.630$.[15] For the LE FePt model, the value of the exponent ν is slightly higher than in the NE case suggesting that the FePt system might belong to a universality class different from that of the isotropic Heisenberg model. However, more extensive analysis which would allow extraction of a complete spectrum of critical exponents is required to quantify the university class of FePt system. Such analysis is beyond the scope of the present work.

Having obtained the bulk T_c^∞, we now mimic the experimental procedure and fit Eq. (1) to the $T_c(D)$ data in Fig. 2 with d_0 being the only fit parameter. We fix the phenomenological shift exponent to $\lambda = \nu^{-1}$ with the values of ν as obtained from the FSS analysis in Table I. Fig. 2 shows an excellent agreement between the one-parameter fits of Eq. (1) and the data, with the values of fitted d_0 listed in Table I. The parameter d_0 obtained for LE FePt agrees closely with the experimentally reported value,[11] validating the approach further. We note that in experiments the exponent λ is often considered as a fit parameter as well because ν is often unknown and, moreover, λ does not always agree with ν^{-1} due to the importance of higher order corrections to finite size scaling which are not included in Eq. (1).[9,12,13] We found that such two-parameters fits of Eq. (1) to our data yield $\nu = 0.94 \pm 0.03$ and $d_0 = 0.56 \pm 0.03$ for the NE model, and $\nu = 0.85 \pm 0.04$ and $d_0 = 0.72 \pm 0.04$ for the LE FePt model. While the values for the NE model are unrealistically off the expected range for the Heisenberg model probably due to high sensitivity to parameter correlation during the fitting, for the LE FePt model the agreement is remarkable. Hence, the approach based on fixing λ to the bulk value of ν^{-1} obtained from the FSS is preferred here.

TABLE I. Critical exponents β and ν, and the bulk T_c^∞ obtained from the FSS analysis according to Eq. (3) for the NE and the LE FePt Hamiltonians. The microscopic length d_0 was obtained by fitting Eq. (1) to the T_c vs D dependence obtained by differentiating the M vs T data for different sizes D (Fig. 2). For comparison the experimental data of Ref. 11 and the values for the Heisenberg Hamiltonian[15] are also given.

	NE $\mathcal{H}$	LE-FePt $\mathcal{H}$	Experimental[11]	Heisenberg Hamiltonian[15]
β	0.38±0.03	0.33±0.10	—	0.36
ν	0.79±0.11	0.85±0.10	0.91±0.10	0.71
T_c^∞ (K)	746.5±5.45	677.4±10.66	775±10	—
d_0 (nm)	0.71±0.02	0.72±0.02	0.84±0.05	—

Now that Eq. (1) has been fully quantified in terms of the parameters T_c^∞, λ, and d_0, we proceed to analyze the relationship between the distribution functions $f_T(T_c)$ and $f_D(D)$, assuming the realistic scenario of a lognormal distribution of diameter $f_D(D) = (D\tilde{\sigma}_D\sqrt{2\pi})^{-1}\exp(-(\ln D - \tilde{D})^2/2\tilde{\sigma}_D^2)$ with $\tilde{D}$ and $\tilde{\sigma}_D^2$ being, respectively, the mean and variance of the random variable $\ln(D)$.[21] Given that the dependence $T_c(D)$ as defined by Eq. (1) is monotonic and that distribution functions f_T and f_D are univariate, the transformation law reads $f_T(T_c) = |D'(T_c)|f_D(D(T_c))$,[22] where $D(T_c)$ is the inverse of Eq. (1) and the prime denotes the derivative d/dT_c. Thus, rearranging Eq. (1) gives $D(T_c) = d_0\epsilon_c^{-1/\lambda}$ and $D'(T_c) = \lambda^{-1}d_0/(T_c^\infty\epsilon_c^{1+1/\lambda})$, which after algebraic manipulations and using the substitution $\epsilon_c = (T_c^\infty - T_c)/T_c^\infty$ leads to the expression

$$f_T(\Delta T_c) = \frac{1}{\sqrt{2\pi}\Delta T_c\tilde{\sigma}_T}\exp\left(-\frac{(\ln\Delta T_c - \tilde{T})^2}{2\tilde{\sigma}_T^2}\right), \quad (4)$$

with $\Delta T_c = T_c^\infty - T_c$. Equation (4) is a lognormal distribution function with logarithmic mean $\tilde{T} = \lambda(\ln(d_0(T_c^\infty)^{1/\lambda}) - \tilde{D})$ and variance $\tilde{\sigma}_T^2 = \lambda^2\tilde{\sigma}_D^2$. Examples of the distribution functions for both NE and LE FePt models are shown in Fig. 3. It can be seen that the characteristic tail of the lognormal distribution is to low values of T_c, essentially because of the cut-off imposed by the parameter T_c^∞.

In addition to $\tilde{T}$, $\tilde{\sigma}_T^2$, and $\tilde{D}$, $\tilde{\sigma}_D^2$ from the practical point of view, it is also useful to identify the relationship between the arithmetic mean values and variances: $\langle T_c\rangle$, σ_T^2, and $\langle D\rangle$, σ_D^2. Straightforward manipulation of standard expressions relevant to the lognormal distribution[21] gives

$$\frac{\langle T_c\rangle}{T_c^\infty} = 1 - \left(\frac{d_0}{\langle D\rangle}\right)^\lambda\left(1 + \frac{\sigma_D^2}{\langle D\rangle^2}\right)^{(\lambda^2+\lambda)/2}, \quad (5a)$$

$$\frac{\sigma_T^2}{\langle\Delta T_c\rangle^2} = -1 + \left(1 + \frac{\sigma_D^2}{\langle D\rangle^2}\right)^{\lambda^2}. \quad (5b)$$

Equation (5a) reduces to Eq. (1) in the absence of randomness $\sigma_D \to 0$, in which case also $\sigma_T \to 0$ in Eq. (5b), as expected. Moreover, given the λ values found here, increasing σ_D results in increase of $\langle T_c\rangle$ and σ_T.

In conclusion, we have performed a finite size scaling analysis based on the calculated $M(T)$ data sets for grains of different size D and extracted the bulk value of Curie temperature and critical exponents. This allows the quantification of the $T_c(D)$ dependence based on the scaling relation Eq. (1) and identification of the relationship between the T_c distribution $f_T(T_c)$ and grain size distribution $f_D(D)$ as Eq. (4). Equation (4) demonstrates that a lognormal form of the distribution function is preserved under the critical scaling

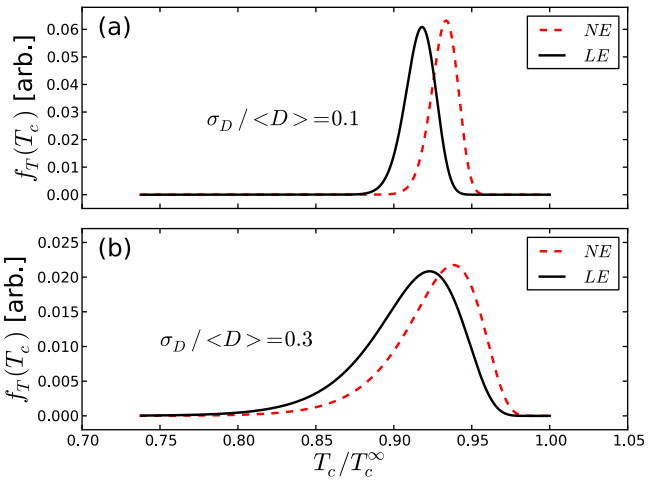

FIG. 3. Distribution function $f_T(T_c)$ calculated from Eq. (4) for $\langle D\rangle = 6$ nm and $\sigma_D/\langle D\rangle = 0.1$ (a) and 0.3 (b), for NE and LE FePT models.

Eq. (1). Importantly, we show analytically that standard deviations of $f_T(T_c)$ and $f_D(D)$ are related through the shift exponent λ as $\tilde{\sigma}_T = \lambda\tilde{\sigma}_D$ or, equivalently, σ_T and σ_D through Eq. (5). Consequently, λ is potentially an important physical quantity in relation to HAMR, and one which is expected to be material dependent through the physical concept of universality, as demonstrated here by comparing the long range exchange FePt model with a Heisenberg-like system with short range interaction.

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