Henkel plots of single-domain ferromagnetic particles

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The use of Henkel plots as a tool to analyze the type and strength of interaction between particles in fine magnetic particle systems is wide spread. It is commonly accepted that noninteracting systems in general show linear Henkel plots, while interacting systems show curved plots. Using extensive Monte Carlo simulations Henkel plots for noninteracting and interacting systems of particles that show different anisotropies are studied. It is found that a direct relation between linearity and noninteraction exists only for systems of uniaxial particles at low temperatures, while particles with cubic anisotropy always show positive deviation of the Henkel plot in the whole range of temperatures. On the other hand, dipolar interaction always results in negative deviation. In the case of particles with cubic anisotropy and dipolar interaction, the deviation changes gradually from positive to negative with increasing strength of the interaction. © *2000 American Institute of Physics.* [S0021-8979(00)00910-5]

I. INTRODUCTION

Among the experimental techniques for determining the magnetic properties of systems composed by ultrafine magnetic particles, the use of remanence curves is becoming one of the most preferred.[1–5] The main reason is that the hysteresis loop is consequence of both reversible and irreversible processes, whereas remanence is a natural consequence of the irreversibility of the magnetization process. The measurement of the remanence curves is thus a very useful tool by which to separate the reversible and irreversible contributions to the magnetization process.

Remanence curves are related to the switching field distribution (SFD), but in addition they may give insight into the internal leading interaction between particles via Henkel plots[6] (also see below). Nevertheless there have been some uncertainties about the applicability of the results, since most of the derivations have been done using the Stoner-Wohlfarth (SW) model,[7] valid precisely only at a temperature 0 K and for noninteracting uniaxial particles. Except for a recently published article on Henkel plots of simple systems of uniaxial particles at higher temperatures,[8] up to the present no theoretical study of Henkel plots has included a wide range of temperatures, other anisotropies, and interactions. In this article, in order to understand the validity conditions of these results better, we have performed Monte Carlo simulations of the remanence curves in systems of single-domain particles showing uniaxial and cubic anisotropies, both with and without magnetic dipolar interactions.

The aim of our work is to try to answer the following questions.

(1) Is the model valid at temperatures different from 0 K?
(2) What happens when the particles show cubic instead of uniaxial anisotropy?
(3) What are the effects of dipolar interaction in both kinds of systems?

II. DESCRIPTION OF THE MODEL

In Sec. II we review the main results which link the different magnetization curves and give rise to the Henkel plots.

A. Main remanence curves

1. Isothermal remanent magnetization

For an isothermal remanent magnetization (IRM) experiment the starting point is a totally demagnetized sample after cooling it in zero magnetic field [zero field cooled (ZFC)]. When the experiment temperature is reached, a small external field H is applied and after a certain amount of time it is switched off and the remanence $M_R(H)$ is measured. The process is repeated, increasing the field H until the sample reaches saturation and the remanence takes the saturation value $M_R(\infty)$.

2. Direct current demagnetization

A dc demagnetization (DCD) experiment is basically similar to that of an IRM one, but initially the sample is in a saturated state. At a fixed temperature a small external field H in the direction opposite to magnetization is applied, and after some time it is switched off and the remanent magne-

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tization $M_D(H)$ is measured. This is repeated again, increasing the field H until saturation in the opposite direction is reached.

3. Alternating current demagnetization

Starting from the sample in saturation remanence, an ac demagnetizing (ACD) field of amplitude H is applied. This field will demagnetize particles with a critical field H_k smaller than this value. Then the magnetization $M'_D(H)$ is measured. The whole process is repeated, increasing the amplitude of the ac field.

B. Relations between remanence curves for systems of noninteracting uniaxial particles

We start from the (SW) model,[7] which applies to a system of noninteracting single-domain particles. In this model every particle i consists of a single magnetic domain with all the atomic moments rotating coherently, resulting in a constant absolute value $|\vec{\mu}_i| = M_S V_i$ of the total magnetic moment $\vec{\mu}_i$. Here, V_i is the volume of particle i and M_S is the saturation magnetization, which we suppose to be independent of particle volume. The different particles present anisotropy energy, e.g., due to their shape and/or crystalline structure, and interact with a given external field $\vec{H}$, resulting in an energy function,

$$E^{(i)} = -KV_i \left(\frac{\vec{\mu}_i \cdot \vec{n}_i}{|\vec{\mu}_i|} \right)^2 - \vec{\mu}_i \cdot \vec{H}, \tag{1}$$

where K is the anisotropy constant and $\vec{n}_i$ with $|\vec{n}_i| = 1$ denotes the anisotropy axis.

When dealing with the SW model it is usual to employ reduced magnitudes. Considering the external field H along the z axis, (θ, ϕ) the spherical coordinates of the particle magnetization and ψ_i the angle between the easy axis and the field, the reduced energy function for a single particle can be written as

$$e^{(i)}(\theta, \phi) = -\tfrac{1}{2}(\sin\psi_i \sin\theta \cos\phi + \cos\psi_i \cos\theta)^2 - h\cos\theta, \tag{2}$$

where we divided Eq. (1) by $2KV_i$. Here $h = H/H_a$ with $H_a = 2K/M_s$ being the anisotropy field.

Within the framework of the SW model it is not difficult to obtain the relation between the different remanence measurements,[9] keeping in mind that these relations will be valid only for $T = 0$ K and for uniaxial systems. Starting with a system of aligned particles with their easy directions forming an arbitrary angle ψ with the applied field it is immediately seen that

$$M_{R_\psi}(H) = 0, \quad \text{if } H \leqslant H_k(\psi), \tag{3a}$$

$$M_{R_\psi}(H) = M_{R_\psi}(\infty), \quad \text{if } H > H_k(\psi), \tag{3b}$$

with M_{R_ψ} being the IRM of the aligned system and $H_k(\psi)$ its critical field, both quantities depending on orientation. Similarly we have

$$M_{D_\psi}(H) = M_{R_\psi}(\infty), \quad \text{if } |H| \leqslant H_k(\psi), \tag{4a}$$

$$M_{D_\psi}(H) = -M_{R_\psi}(\infty), \quad \text{if } |H| > H_k(\psi). \tag{4b}$$

The above expressions for every orientation ψ chosen can be summarized as

$$M_{D_\psi}(H) = M_{R_\psi}(\infty) - 2M_{R_\psi}(H). \tag{5}$$

For a system of noninteracting particles randomly oriented the total magnetization is the sum of the magnetizations for each orientation. Every loop is independent and hence the above expression is valid not only for aligned systems but for every kind of orientation distribution, i.e.,

$$M_D(H) = M_R(\infty) - 2M_R(H). \tag{6}$$

A similar relation exists between $M'_D(H)$ and $M_R(H)$. For a fixed orientation we have

$$M'_{D_\psi}(H) = M_{R_\psi}(\infty), \quad \text{if } H \leqslant H_k(\psi), \tag{7a}$$

$$M'_{D_\psi}(H) = 0, \quad \text{if } H > H_k(\psi), \tag{7b}$$

and using the same argument these expressions can be summarized for a random system as

$$M'_D(H) = M_R(\infty) - M_R(H). \tag{8}$$

C. Leading interaction between particles: Henkel plots

The relation between both remanence curves can be obtained directly with a simple plot. Dividing Eq. (6) by $M_R(\infty)$ we get

$$m_D(H) = 1 - 2m_R(H), \tag{9}$$

where we have used the lower-case letter ''m'' for $m_D(H)$ and $m_R(H)$ to denote the reduced magnitudes $M_D(H)/M_R(\infty)$ and $M_R(H)/M_R(\infty)$, respectively.

Although Wohlfarth[9] had already obtained the relations between these different modes of acquisition of the remanent magnetization and pointed out the limitations of the model, it was Henkel[6] who first proposed to plot $m_D(H)$ vs $m_R(H)$, which is now called the Henkel plot. According to the Stoner-Wohlfarth model this should result in a straight line. Experimentally it is found that the IRM saturates at bigger fields than the DCD curve. This is attributed to the dipolar interaction which difficults the magnetization process and makes the Henkel plot deviate from linear behavior. The question is, Should the deviations from ideal behavior given by Eq. (9) be attributed mainly to the interaction between particles?

To try to quantify the deviations it has became customary[2] to define

$$\delta M = m_D(H) - [1 - 2m_R(H)], \tag{10}$$

which has a direct physical meaning in the framework of this model. The fraction of the global magnetic moment of the system switched at a particular field in IRM mode is $P_i = m_R(H)$ and the fraction switched in DCD mode is $P_d = [1 - m_D(H)]/2$ so

$$\delta M = 2(P_i - P_d), \tag{11}$$

and gives a direct measurement of the difference in the fraction of particles switched from IRM and DCD processes.

III. EXTENSION OF THE MODEL FOR PARTICLES WITH CUBIC ANISOTROPY AND FOR INTERACTING SYSTEMS

In the case of particles exhibiting cubic magnetocrystalline anisotropy the energy is expressed phenomenologically as a power series of the direction cosines (α,β,γ) of the magnetization in the orthogonal coordinate system formed by the lattice axes. It is customary to write the cubic anisotropy energy as

$$E_a = K_1 V_i(\alpha^2\beta^2+\alpha^2\gamma^2+\beta^2\gamma^2)+K_2 V_i\ \alpha^2\beta^2\gamma^2, \quad (12)$$

where K_1 and K_2 are the anisotropy constants.

If two of the anisotropy axes follow the directions given by $(\theta_1,\phi_1),(\theta_2,\phi_2)$ (the third one is then automatically determined) the direction cosines will be

$$\alpha = \sin\theta_1 \sin\theta\cos(\phi_1-\phi)+\cos\theta_1\cos\theta, \quad (13a)$$

$$\beta = \sin\theta_2 \sin\theta\cos(\phi_2-\phi)+\cos\theta_2\cos\theta. \quad (13b)$$

Dividing by $2|K_1|V_i$ to obtain a direct comparison to the Stoner-Wohlfarth results and taking into account that $\alpha^2+\beta^2+\gamma^2=1$, we get

$$e^{(i)}(\theta,\phi) = \pm\frac{1}{2}\left(\alpha^2+\beta^2-\alpha^4-\beta^4-\alpha^2\beta^2 + \frac{K_2}{K_1}\alpha^2\beta^2(1-\alpha^2-\beta^2)\right)-h\cos\theta, \quad (14)$$

where $h=H/H_a$, as before, and $H_a=2|K_1|/M_s$. The plus sign corresponds to the case where K_1 is positive, the minus sign to the case where K_1 is negative. In our simulations we have assumed for simplicity $K_2=0$.

The energy of the magnetic dipolar interaction between two particles i and j separated by $\vec{r}_{ij}$ is given by

$$E_D^{(i,j)} = \frac{\vec{\mu}_i\cdot\vec{\mu}_j}{r_{ij}^3}-\frac{3(\vec{\mu}_i\cdot\vec{r}_{ij})(\vec{\mu}_j\cdot\vec{r}_{ij})}{r_{ij}^5}. \quad (15)$$

The total reduced energy e_{tot} of our system is given by a sum over all particles, i.e.,

$$e_{\text{tot}} = \sum_i e^{(i)} + \frac{1}{4|K_1|V}\sum_i\sum_{j\neq i} E_D^{(i,j)}. \quad (16)$$

The long-range magnetic dipolar interaction between particles has been considered without any truncations or simplifications by applying periodic boundary conditions using the standard technique of Ewald's sum for an infinite sphere surrounded by vacuum.[10,11]

IV. SIMULATION TECHNIQUE

We have performed a Monte Carlo (MC) simulation[12] to investigate the remanence curves of single-domain particle systems. The particles can show uniaxial or cubic anisotropy and they interact via dipolar interaction. A MC simulation always consists of two parts: thermalization and experiment. During the first part, the system is led adiabatically to its thermodynamical equilibrium. This first part is very important, since one starts normally with a nonequilibrium state. It is particularly important in the simulation of the IRM curves of interacting systems since these are very sensitive to the initial demagnetized state, since it is necessary to start with the system in a state of minimum energy.[4,8] After the system is fully thermalized, we can study its evolution and obtain the properties of interest under the influence of external parameters.

The simulations were performed with a set of 250 randomly orientated particles in the case of noninteracting systems and a set of 64 for systems interacting dipolarly. The particles with equal volume $V_i=V_0$ are placed randomly in the simulation box, with their positions determined by an inverse MC simulation.[13] The angles defining the orientation of the easy axes of the particles are chosen randomly and are kept constant during the simulation. The two remaining angles (θ,ϕ) per particle defining the direction of its magnetization will be variable throughout the MC simulation. Due to thermalization, their initial values can be chosen arbitrarily.

The MC simulation consists of many elementary steps. In every elementary step a particle i is chosen at random and an attempted orientation $\vec{\mu}_{\text{att}}^{(i)}$ of the magnetization is generated. The attempted direction is chosen in a spherical segment around the present orientation $\vec{\mu}^{(i)}$, which is used as an azimuthal axis, with $\overline{\phi}\in[0,2\pi]$ and $\overline{\theta}\in[0,\delta\theta]$. Then the difference in energy Δe between the attempted and the present orientation is calculated. If $\Delta e\leqslant 0$, the magnetization is always changed to $\vec{\mu}_{\text{att}}^{(i)}$. If $\Delta e>0$, the magnetization is changed with probability $\exp(-\Delta e/t)$ and remains unchanged with probability $1-\exp(-\Delta e/t)$ (Metropolis rates, the random number generator used is the Kirkpatrick-Stoll R250). Here, $t=k_BT/(2|K_1|V)$ denotes the reduced temperature and k_B is the Boltzmann constant. In any case variable counting of the elementary steps is increased and the process is continues with the next elementary step. A complete Monte Carlo step consists of N elementary steps, N being the number of particles in the system, so that in every MC step on each average particle is considered once. Varying the aperture angle $\delta\theta$, i.e., the maximal jump angle, it is possible to modify the range of acceptance to optimize the simulation. Using this kind of local dynamic permits us to detect confinement in metastable states responsible for the hysteresis[14] and has been successfully used by the authors to study hysteresis loops of noninteracting particles, showing cubic crystalline anisotropy[15] and ZFC and field cooling (FC) of interacting systems.[16] As a compromise between simulations at low and high temperatures we choose $\delta\theta=0.075$. Usually about 10 000 MC steps are used for thermalization, which is one to two orders of magnitude larger than the algorithm's autocorrelation time at zero field. Once the desired temperature is reached, we start the corresponding remanence curve by slowly varying the reduced applied field in steps of 0.025 in the following way: After changing the field, 2000 MC steps are done, the field is cut, another 2000 MC steps are done and the magnetization is measured. The field is changed again, and the routine is repeated until reaching

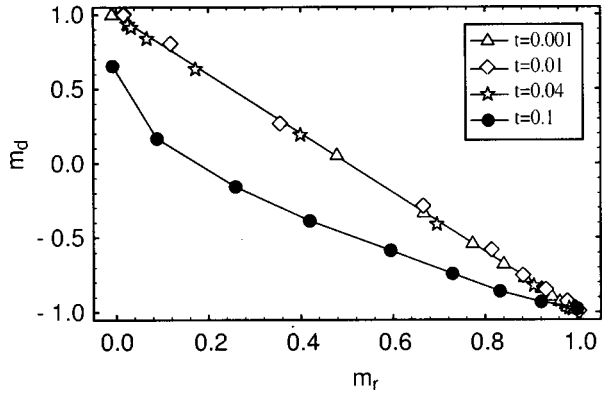

FIG. 1. Henkel plots for systems of uniaxial noninteracting particles. For lower temperatures $t=0.001$ (open triangles), $t=0.01$ (open diamonds) and $t=0.04$ (open stars) the plot is a straight line but it curves at higher temperature $t=0.1$ (solid circles). The error bars are smaller than the size of the symbols. The solid lines are guides to the eye. Only a subset of all points obtained is represented.

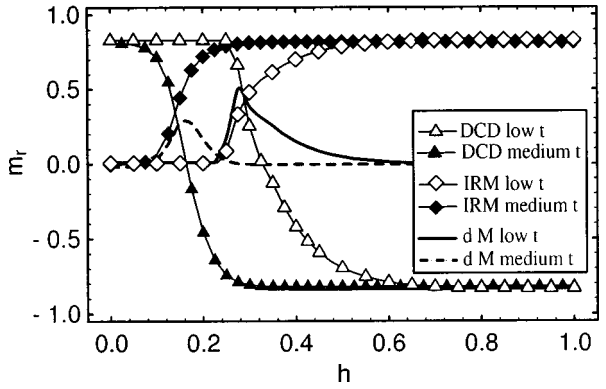

FIG. 2. Reduced IRM (diamonds) and DCD (triangles) curves for cubic noninteracting systems at $t=0.0001$ (open symbols) and $t=0.01$ (closed symbols). The lines represent the δM calculated from both curves. The error bars are smaller than the size of the symbols. The solid lines are guides to the eye. Only a subset of all points obtained is represented.

saturation. The whole remanence curve is repeated for a large number of independent configurations to perform an ensemble average.

V. RESULTS

A. Noninteracting systems

Since dynamical aspects are very important in ferromagnetic nanoparticle systems we first analyze the effect of temperature on the remanence curves of noninteracting uniaxial systems. We have performed simulations in a range of temperatures from $t=0.001$ to 0.02 which are considered to be very low and intermediate temperatures, respectively. To have a reference, ZFC/FC simulations of these systems with the same parameters show a blocking temperature around $t=0.15$. Part of the results shown in Fig. 1 correspond to IRM and DCD curves calculated for $t=0.001$ and 0.04. In this range of temperatures the relation given by Eq. (6) is completely fulfilled. Consequently, the corresponding Henkel plot is a straight line.

When the temperature becomes of the same order as the blocking temperature there is appreciable relaxation of the magnetization value during the time of measurement. The value of saturation of the remanence is around 50% of the value given by the SW model. In this range of temperatures it cannot be expected that the above relation still holds; this is confirmed by the simulation results for $t=0.1$. The consequence for the Henkel plot (see Fig. 1) is a negative δM.

The model is valid for uniaxial particles at low temperatures, but in this article we were mainly interested in particles with cubic anisotropy. There is a basic difference between the two kinds of anisotropy since in the case of uniaxial anisotropy there are only two possible directions in which the energy reaches a minimum value. This allows analytical calculation of the hysteresis loops and remanence curves. On the contrary, the case of cubic anisotropy has many possible minima and the analysis done before cannot be extended to this kind of anisotropy. In fact it is not possible to follow the Stoner-Wohlfarth method to obtain the magnetization direction. Here MC simulation shows its strenghts, weighting the different minima accordingly with the correct Boltzmann statistics. Similar to the uniaxial case, several studies have been performed in a wide range of temperatures. Shown in Fig. 2 are the results of the simulations for $t=0.0001$ and 0.01, which again correspond to low and intermediate temperatures. (The energy barrier in the cubic case with $K_1>0$ and $K_2=0$ is 1/4 of the uniaxial case and consequently the blocking temperature is four times smaller.) The continuous and dashed lines show that there is appreciable positive deviation of δM. This deviation decreases with increasing temperature. The corresponding Henkel plots are shown in Fig. 3.

B. Interacting systems

In order to study the influence of the dipolar magnetic interaction on the Henkel plots we have used different concentrations ranging from $c=0$ (no dipolar interaction) to $c=0.32c_0$. Here $c\equiv\Sigma_i V_i/V_{\text{tot}}$ is the ratio between the volume

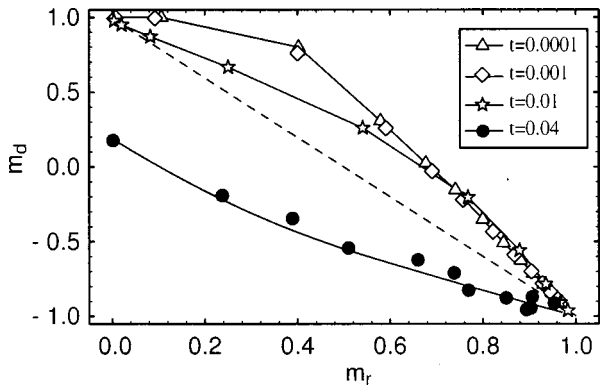

FIG. 3. Henkel plot for systems of noninteracting particles showing cubic anisotropy with $K_1>0$. Even at very low temperatures $t=0.0001$ (open triangles) there is clear deviation from a straight line. The rest of the symbols correspond to $t=0.001$ (open diamonds), $t=0.01$ (open stars) and to a high temperature $t=0.04$ (closed circles). The error bars are smaller than the size of the symbols. The solid lines are guides to the eye. Only a subset of all points obtained is represented.

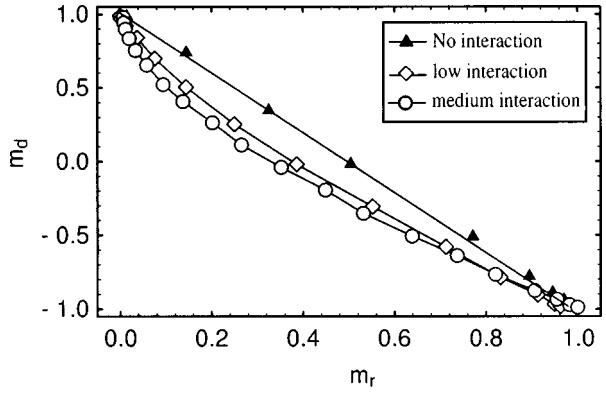

FIG. 4. Henkel plots for systems of interacting uniaxial particles with $c/c_0=0.064$ (open diamonds) and $c/c_0=0.128$ (open circles) at very low temperature $t=0.0001$. Closed triangles are for the noninteracting system. Even at very low temperatures there is clear deviation from a straight line. The error bars are smaller than the size of the symbols. The solid lines are guides to the eye. Only a subset of all points obtained is represented.

$\Sigma_i V_i$ occupied by the particles and the total volume of the sample V_{tot} and $c_0 \equiv M_S/H_A$ is an unitless material constant of order one.[17]

The simulations on dipolarly interacting systems of uniaxial particles confirm the behavior expected. At low temperatures the result is that δM is negative in the whole range of the curves. This results in negative deviation in the Henkel plots. Deviation from a straight line increases with increasing interaction.The Henkel plots shown in Fig. 4 correspond to $c/c_0=0.064$ and 0.128 done at very low temperature. From the shape of the IRM and DCD curves (not shown) it can be seen that the SFD broadens considerably when the particle concentration increases.

In the case of interacting systems of particles with cubic anisotropy there is a gradual change from the positive deviation of the noninteracting case to the negative deviation of the strongly interacting systems. In Fig. 5 we present Henkel plots of five different systems at low temperature. The concentrations are $c/c_0=0$, 0.016, 0.032, 0.064 and 0.128. It is interesting to note that the intermediate concentration $c/c_0=0.032$ shows an almost straight plot.

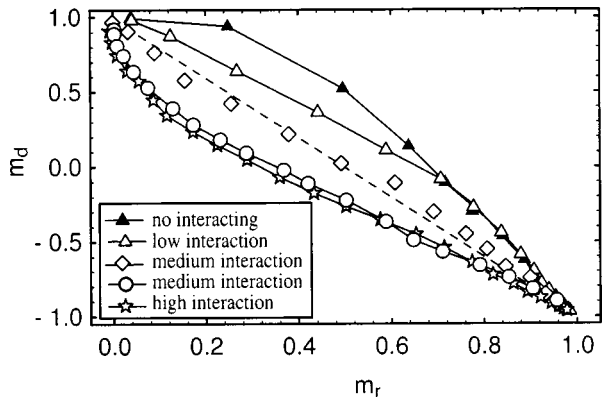

FIG. 5. Henkel plots for systems of interacting particles showing cubic anisotropy at very low temperature $t=0.0001$. The closed triangles are the result for the noninteracting system while $c/c_0=0.016$ (open triangles) and $c/c_0=0.032$ (open diamonds) correspond to weakly interacting systems, and $c/c_0=0.064$ (open circles) and $c/c_0=0.128$ (open stars) are relatively highly interacting. Even at very low temperatures there is clear deviation from a straight line. The error bars are smaller than the size of the symbols. The solid lines are guides to the eye. Only a subset of all points obtained is represented.

VI. CONCLUSIONS

The IRM and DCD curves of random systems of single domain particles exhibiting different anisotropy energies and different concentrations have been studied. The Henkel plot which is based on an analytical relation between these two curves was obtained and the validity of the model was verified. Systems composed of uniaxial particles at low and intermediate temperatures show a linear Henkel plot, as the model predicts. At temperatures of the order of the blocking temperature the model is no longer valid and dynamic effects start to play an important role. In this case negative deviation in the Henkel plot is observed. For systems of uniaxial particles interacting via magnetic dipolar interaction, the experimentally observed negative deviation is reproduced in the simulation. The stronger the interaction the stronger the deviation.

For systems of noninteracting particles with cubic anisotropy positive deviation is observed, which must not be attributed to some kind of exchange interaction. The effect is counterarrested by dipolar interaction, resulting in negative deviation at a relatively high interacting system. At intermediate concentrations the Henkel plot may resemble a straight line, which might lead to the incorrect conclusion that it is a noninteracting system.

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[13] During this inverse MC simulation, the particles can move freely and interact only by a standard Lennard-Jones pair potential $v_{\text{LJ}}(r)$

$=\varepsilon[(r_0/r)^{12}-2(r_0/r)^6]$ with periodic boundary conditions (Ref. 11). We choose $\varrho^\star=0.85r_0^{-3}$ as density, start with a very high temperature and decrease it slowly until its final value $T^\star=0.2\varepsilon/k_B$. The values chosen $(\varrho^\star,T^\star)$ belong to the liquid phase (Ref. 11). Of course, for systems without dipolar interaction (i.e., concentration $c=0$) the total energy is independent of the particles' positions.

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[17] To give an estimate for the concentrations in real units: Iron-nitride nanoparticles reported by H. Mamiya, I. Nakatani, and T. Furubayashi, Phys. Rev. Lett. **80**, 177 (1998) show $M_S=1182$ emu/cm^3 and $K=10^6$ erg/cm^3, which yields $c_0=1.43$. Maghemite particles with mean diameters of around 7.5 nm reported by T. Jonsson, J. Mattsson, C. D. Djurberg, F. A. Khan, P. Nordblad, and P. Svedlindh, Phys. Rev. Lett. **75**, 4138 (1995) show $M_S=420$ emu/cm^3 and and $K=1.9\times10^5$ erg/cm^3, which gives $c_0=2.15$. Thus, the largest concentration considered in this work corresponds to $c=0.46$ for ion-nitride nanoparticles and $c=0.69$ for maghemite particles.