Magnetic properties of superparamagnetic particles by a Monte Carlo method

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We develop and carry out Monte Carlo simulations for an ensemble of superparamagnetic particles uniformly distributed in a nonmagnetic matrix. We find the magnetization below the blocking temperature T_B when it shows hysteresis and above T_B in the superparamagnetic region. We determine the blocking temperature for a set of anisotropy strengths from the magnetization and the susceptibility of the particles. A fixed number of Monte Carlo steps with a constrained acceptance rate is shown to be equivalent to an observation time in the simulations that is much shorter than experimental observation times. We show how the blocking temperature obtained in the simulations can be converted into the corresponding experimentally measurable blocking temperature by using this difference in the observation times. This provides a method to compare Monte Carlo simulation results with experiments, such as recent ones on fcc Co particles. [S0163-1829(96)02537-4]

A number of numerical simulations have been performed to study the magnetic properties of clusters or very small particles both at nonzero[1–8] and at zero temperatures.[9,10] The nonzero-temperature properties have been studied in Monte Carlo (MC) simulations[1,2] assuming that the anisotropy of the particles can be neglected. While this assumption is reasonable when the particles are studied on an atomic level or when the magnetization of the particles behaves essentially paramagnetically, it excludes the low-temperature properties when the magnetization shows hysteresis due to the anisotropy. Here we intend to study the combination of temperature and anisotropy effects on the magnetization of fine magnetic particles using the Metropolis Monte Carlo scheme.[11]

The description of the magnetic properties of single-domain particles in general for nonzero temperatures is based on the superparamagnetic theory.[12] The basic assumption of this theory is that the atomic magnetic moments within a particle are moving coherently and thus its magnetic moment can be represented by a single vector with a magnitude equal to $\mu=\mu_0 N$, where μ_0 is the atomic magnetic moment in Bohr magnetons and N is the number of atoms in the particle. The magnetic moment of the particle is considered coupled to a uniaxial anisotropy, for instance, due to crystal or stress anisotropy, and to the external magnetic field. In the absence of an external field, the uniaxial anisotropy leads to two equivalent equilibrium states of the moment. For an ensemble of identical particles which have been initially saturated in a given direction, the magnetization per particle will decrease from its initial value as $M=M_s e^{-t/\tau}$, where $M_s=\mu/V$ and V is the particle's volume, as equal populations of the two states are acquired due to thermal fluctuations.

The relaxation time τ is essentially the average time to reverse a particle's magnetization from one of the equilibrium states to the other and is determined by the Boltzmann factor $\exp(-D/k_B T)$ and a characteristic constant frequency f_0 (of the order of 10^{10} Hz) through the relation

$$\frac{1}{\tau}=f_0\exp(-D/k_B T), \tag{1}$$

where T is the temperature, k_B is Boltzmann's constant, and D is the energy barrier separating the two states.[13] D is determined by the anisotropy energy density K and the particle's volume, $D=KV$. For high T ($k_B T \gg D$) the time scale of the thermal relaxation τ in Eq. (1) is much shorter than any experimental observation time over which magnetization is measured, and so the system appears superparamagnetic. On the other hand, for low T ($k_B T \ll D$), the thermal reversal time scale τ becomes very large, much larger than any observation time, and the system appears ferromagnetic. The temperature determined from Eq. (1) with τ set equal to the experimental observation time t defines the blocking temperature T_B which separates the two regimes. However, the time t is determined by the experimental requirements, and so the definition of T_B is not unique, but can depend on the type of experiment. For $t=1000$ sec we have

$$T_B \approx D/30k_B, \tag{2}$$

where the numerical factor comes from $\ln(f_0 t)\approx 30$ for $f_0=10^{10}$ Hz. If we want to use Monte Carlo or even a spin dynamics simulation based on Landau-Lifshitz or Langevin equations to study the magnetic properties in the presence of anisotropy, it is impossible to make a long enough simulation that will correspond to 1000 sec. Therefore, part of our goal here is to describe how a laboratory value of T_B can be obtained from simulations that correspond to observation times much less than 1 sec.

We show in this paper that all of the results in the superparamagnetic theory can be obtained by Monte Carlo simulations where we use an observation time much smaller than the experimentally used values. Nevertheless, the data from the simulations can be compared with experimental results after we account for the shorter observation time of the Monte Carlo simulations.

We choose to do the simulations by using a Metropolis Monte Carlo scheme rather than using Landau-Lifshitz or Langevin equations because the implementation of the temperature is straightforward. However, it presents the usual problem with MC schemes that individual MC steps do not

correspond to real time, but are only sampling the phase space at some rate. If the MC acceptance rate can be set to some desired value (we use 30–40 %), however, this effectively sets the rate of motion in phase space. For the problem here, the magnetic moment can remain for many steps in one side of the double potential well due to the anisotropy term. Over some characteristic number of MC steps, it will typically pass near the barrier to go to the other side of the well. That is, there really will be a characteristic attempt frequency to jump to the other well, measured in attempts per MC step. This frequency should correspond approximately to f_0 in Eq. (1) and implies the conversion of MC steps, or ''MC time,'' into real time. We do not, however, try to measure the characteristic frequency directly in MC simulations. Instead, we use a fixed large number (1.2×10^6) of MC steps, keeping the acceptance rate from 30% to 40%, and determine T_B from magnetization and susceptibility results for different values of D. We find a linear relation between T_B and D, as in Eq. (2), but with the numerical factor of approximately 14.8 instead of 30. The smaller numerical factor is interpreted to mean that the 1.2×10^6 MC steps correspond to a much shorter ''observation time'' t_{MC} than the 1000 sec applied in Eq. (2), according to $\ln(f_0 t_{\mathrm{MC}})\approx14.8$.

By understanding this effective physical time scale over which the MC averages were made, it allows us to infer the corresponding laboratory measurements that would be made at the 1000 sec time scale. Although the model used here is simple, these ideas are important for the interpretation of any simulation of a more realistic model of fine magnetic particles with metastable states relaxing over a barrier at finite T.

We assume that our system consists of an assembly of spherical particles with identical sizes. The particles are embedded in a nonmagnetic matrix and can be considered approximately as noninteracting. Due to the spherical shape of the particles, we assume that the origin of the anisotropy is only from crystal anisotropy. Such a kind of a system has been realized experimentally (see, for example, Ref. 14).

Next, we assume that the magnetization is homogeneously distributed throughout the volume of the particle and thus can be represented by a single vector with a constant magnitude. The Hamiltonian of each particle is

$$\mathcal{H}=-\mu\vec{H}\cdot\hat{S}-D(\hat{n}\cdot\hat{S})^2, \tag{3}$$

where $\vec{H}$ is the external magnetic field, $\hat{S}$, the ''spin'' of the particle, is a unit vector along its magnetization, and $\hat{n}$ is a unit vector along the particle's anisotropy axis. The $\hat{n}$ vectors of the particles are assumed to be uniformly distributed in all directions. When we calculate the component of the magnetization of the system along the field, it is enough to calculate the average magnetization of the particles with anisotropy axes $\hat{n}$ in a single plane which includes the direction of the field $\vec{H}$. The magnetization of the system will then be obtained using the azimuthal symmetry about the direction of the field. We chose the z axis to be along $\vec{H}$ and take the xz plane as the plane containing $\vec{H}$ and $\hat{n}$. The Hamiltonian of each particle is invariant under the transformation $\hat{n}\rightarrow-\hat{n}$, and so we may restrict the angle θ between the field $\vec{H}$ and a given anisotropy axis $\hat{n}$ in the interval $0\leqslant\theta\leqslant\pi/2$. To obtain the magnetization per particle and the susceptibility of the system we perform two averages. First, we determine the thermal averages $\langle S_z\rangle_\theta$ and $\langle S_z^2\rangle_\theta$ for a given $\hat{n}(\theta)$ by Monte Carlo simulations, and second, we average these results over the uniform distribution of $\hat{n}$ on the unit sphere. Then the magnetization per particle will be

$$M=M_s\langle S_z\rangle=M_s\int_0^{\pi/2}\langle S_z\rangle_\theta\sin\theta\,d\theta \tag{4}$$

and the mass susceptibility is

$$\chi=\frac{\mu\sigma_s}{k_BT}\int_0^{\pi/2}(\langle S_z^2\rangle_\theta-\langle S_z\rangle_\theta^2)\sin\theta\,d\theta, \tag{5}$$

with thermal averages

$$\langle S_z^k\rangle_\theta=\int e^{-\beta\mathcal{H}\{\theta,\theta'\}}S_z^k\,d\Omega'\Big/\int e^{-\beta\mathcal{H}\{\theta,\theta'\}}d\Omega' \tag{6}$$

for $k=1,2$ and $\mathcal{H}\{\theta,\theta'\}=-\mu HS_z-D(n_xS_x+n_zS_z)^2$, $\hat{S}=(\sin\theta'\cos\phi',\sin\theta'\sin\phi',\cos\theta')$, $\hat{n}=(\sin\theta,0,\cos\theta)$, $\beta=1/k_BT$, and $d\Omega'=\sin\theta'\,d\theta'\,d\phi'$. The saturation magnetization $\sigma_s=M_sV/m$ is in emu/g, m is the particle's mass, and the magnetic moment μ of the particle is in Bohr magnetons.

The average over the distribution of anisotropy axes, i.e., taking the integrals in Eq. (4) and Eq. (5), is performed numerically using the extended midpoint rule, dividing the interval $0<\theta\leqslant\pi/2$ into 90 parts, and incrementing θ by $\Delta\theta=1°$. The thermal averages $\langle S_z\rangle_\theta$ and $\langle S_z^2\rangle_\theta$, defined by Eq. (6), are calculated in the Monte Carlo simulations for each θ using the Metropolis algorithm.[11] The most important part of our implementation of the Monte Carlo simulation is in the way of performing Monte Carlo steps (MCS). In a Monte Carlo step an attempt to change the spin from $\hat{S}_{\mathrm{old}}$ to $\hat{S}_{\mathrm{new}}$ is made such that the deviation of $\hat{S}_{\mathrm{new}}$ from $\hat{S}_{\mathrm{old}}$ is small but randomly chosen with a fixed limit δS_{max}. This approach allows the freedom to control the acceptance rate of MC moves by adjusting the limit δS_{max} and, more importantly, models the real system more accurately than other schemes used[2] where $\hat{S}_{\mathrm{new}}$ is completely randomly chosen independently from $\hat{S}_{\mathrm{old}}$. It is very important to understand that if we employed this latter type of move, the system would always be paramagnetic for any $T>0$ and no hysteresis would result. That is, for completely random and independent moves arbitrarily large fluctuations are allowed in a single Monte Carlo step and the system will escape very quickly from any metastable state which may be responsible for hysteresis. We actually perform the Monte Carlo in such a way that it samples the phase space only ''locally'' (near current position), allowing for confinement into metastable states responsible for hysteresis. It is important to use such a nonergodic scheme in order to obtain the blocking temperature.

The initial equilibration for each θ and given initial values of H and T uses the first 10 000–15 000 MCS, which are also used to adjust the acceptance rate to be approximately between 30% and 40%. These limits of the acceptance rate are chosen to optimize the simulations; however, it is essential that we use this same acceptance rate for different temperatures in order to produce a constant rate of sampling. In

this way, the mapping of MC ''time'' to experimental time will be considered to be the same for different temperatures. By using a constant phase space sampling rate, it is then possible to observe the obvious change from fast relaxation in the superparamagnetic regime to slower relaxation in the ferromagnetic regime.

The next 1.2×10^6 MCS are used to collect data but the consecutive measurements are taken with a specific interval of MCS between them to minimize the correlation in the data. In the sense of our above discussion, the observation time of the simulation is determined by the number of MCS, $N_{\mathrm{MC}}=1.2\times10^6$, made, with a fixed acceptance rate, for each measurement. When changing either H or T a new fine equilibration is performed, adjusting also the acceptance rate before collecting data again. To consider how to use our simulation data to find T_B, we review how T_B is found in real experiments.

In experiments, the blocking temperature is determined by scanning $M(T)$ or $\chi(T)$, starting from a zero-field-cooled sample and then applying a very small field and taking measurements as a function of T. The magnetization of the system is very close to zero for very low temperature since approximately half of the particles with a particular anisotropy axis $\hat{n}$ have their moments along $\hat{n}$ and the other half along $-\hat{n}$; $\hat{n}$ is also uniformly distributed on the unit sphere. That is, the thermal fluctuations are very weak ($k_BT\ll D$), and they cannot move the moments from the metastable state (moment has component against H) to the global equilibrium state (moment has component along H) implied by the presence of the very small field, $\mu H\ll D$. Equivalently, the relaxation time τ in Eq. (1) will be much greater than any physical observation time. Increasing T will increase the thermal fluctuations (τ decreases) and thus the probability for a transition from the metastable state to the equilibrium state. This will increase $\langle S_z\rangle$. The increase will continue until the system reaches T_B. The assembly of particles will be in the superparamagnetic region for $T>T_B$ and $\langle S_z\rangle$ will decrease accordingly when we further continue to increase T. T_B can also be determined from the inverse susceptibility, which will have a minimum for $T=T_B$ and will increase linearly with T in the superparamagnetic region.

To determine T_B in the simulations we follow the experimental procedure. We start with two spins for each θ. Initially one of them is along $\hat{n}$ and the other along $-\hat{n}$ since in the zero-field-cooled sample one-half of the particles with this $\hat{n}$ will have on the average their magnetization along $\hat{n}$ and the other half along $-\hat{n}$. Then we apply very small constant field H and start to calculate $M(T)$. This is done for a number of values of the anisotropy coupling constant $100\leqslant D/k_B\leqslant700$ (K). These are typical values, for instance, for Co particles where $K_{\mathrm{bulk}}=2.7\times10^6$ erg/cm^3. Different values of D will correspond to different sizes of particles. The reduced magnetization M/M_s (M_s is the saturation magnetization) obtained by simulation (zero field cooled) is shown in Fig. 1 as function of T for $H=500$ Oe and different values of D/k_B. Selected error bars are shown in the superparamagnetic region; the error bars for $T<T_B$ are of the size of the symbols used or smaller. Changing the value of H to 100 Oe did not change the observed position of the peak of the magnetization but made the data more noisy, particularly

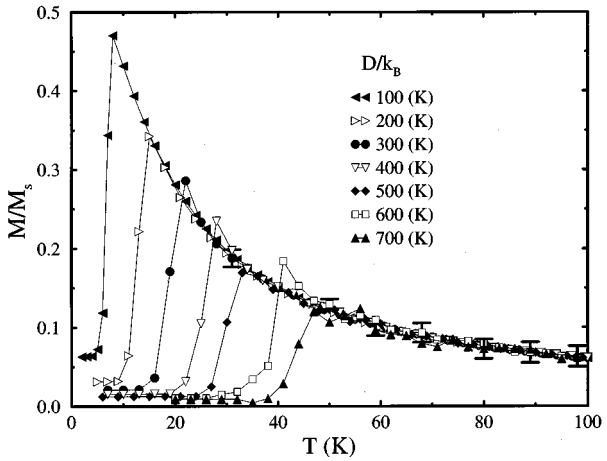

FIG. 1. Dependence of the reduced magnetization on the temperature for the zero-cooled system and different values of D/k_B. The external field is constant and equal to 500 Oe.

for large D when μH is more than two orders of magnitude smaller than D. The susceptibility is shown in Fig. 2 for the same set of parameters. The blocking temperatures determined from the peak of the magnetization or from the minimum of the susceptibility do not differ by more than 1 (K). The dependence of T_B on D and a linear fit are shown in Fig. 3. We have $D/k_BT_B^{\mathrm{MC}}=14.81\pm0.25$ from the slope of the linear fit. Therefore, if we assume that the characteristic frequency f_0 is the same as in the experiment (10^{10} Hz), we can use Eq. (1) to define an effective observation time t_{MC} of the Monte Carlo simulations. Substituting $D/k_BT_B^{\mathrm{MC}}=14.81$ in Eq. (1), we obtain $t_{\mathrm{MC}}=2.70\times10^{-4}$ sec for a measurement consisting of $N_{\mathrm{MC}}=1.2\times10^6$ MCS. Increasing the number of MCS decreases T_B^{MC} and increases t_{MC}, but to use a relaxation time equal to 1000 sec in the simulations, we would have to use approximately $1.2\times10^6\times\exp(15.21)$ MCS per data point.

Nevertheless, we can compare the magnetization curves from our simulations with the experimental ones after we take into account the much smaller observation time in the simulations compared to the observation time of 1000 sec used in the experiments. To account for this difference we

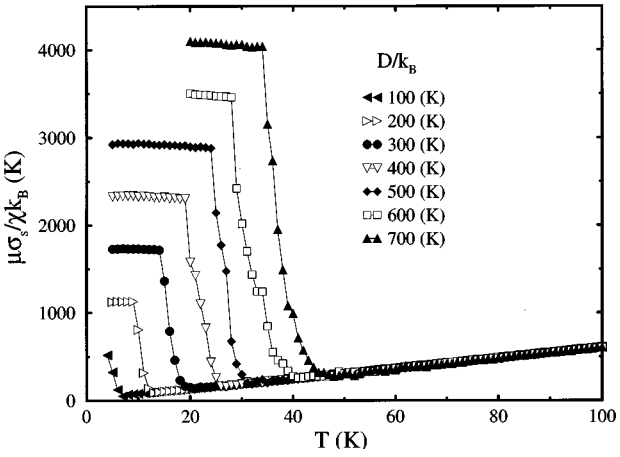

FIG. 2. Reduced inverse susceptibility of the system vs T for the same values of D/k_B as in Fig. 1.

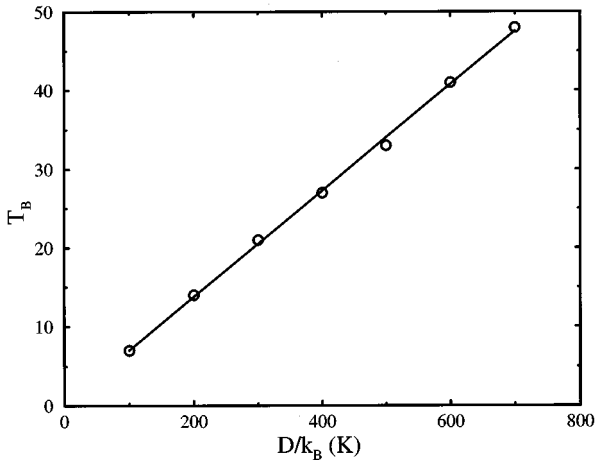

FIG. 3. Dependence of the blocking temperature on the anisotropy energy and a linear fit to it.

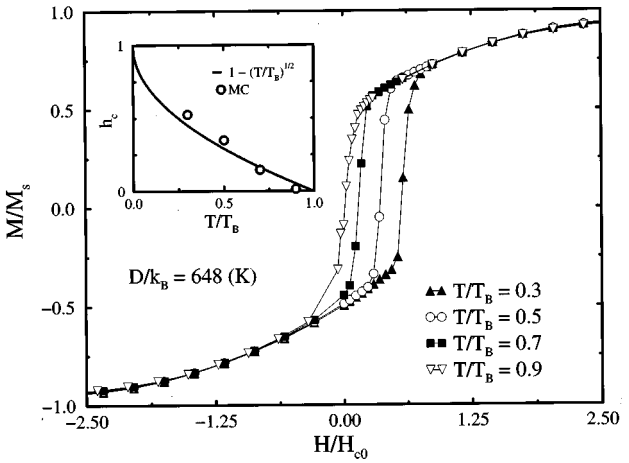

FIG. 4. One-half of the hysteresis loops for a set of temperatures below T_B, where $T_B = 44$ K. The inset shows the comparison of the reduced coercivity h_c (see the text) with the superparamagnetic theory.

have to consider the measurement of the magnetization in two intervals of time with different lengths. If the temperature and the magnetic field are fixed, the magnetization measured in intervals of time t_1 and t_2 ($t_1 < t_2$) will be greater for the shorter interval of time t_1 since the magnetization will fluctuate less on a shorter time scale. Thus, if we are to measure the same value of the magnetization in two intervals of time $t_1 \neq t_2$ at the same external field, we have to perform the measurements at different temperatures. A particular rate of the fluctuations in a given interval of time can be seen in a shorter interval of time if the temperature is higher since the rate of fluctuations of the magnetization will increase with temperature. Therefore, we expect that a value of the magnetization measured in an interval of time t_1 and at temperature T_1 should be measured also in a time interval t_2 ($t_1 \neq t_2$) but at temperature T_2 such that $T_1/T_{B1} = T_2/T_{B2}$, where T_{B1} and T_{B2} are the blocking temperatures determined from Eq. (1) with relaxation time τ set to t_1 and t_2, respectively.

We can also apply Eq. (1) to obtain a relation between the blocking temperature in MC simulations and the blocking temperature in experiments. If an MC observation time t_{MC} leads to a blocking temperature T_B^{MC} and the longer laboratory observation time t_L leads to the lower blocking temperature T_B^L then these are related by

$$\frac{D}{k_B T_B^L} = \frac{D}{k_B T_B^{\mathrm{MC}}} + \ln\left(\frac{t_L}{t_{\mathrm{MC}}}\right). \tag{7}$$

If the absolute scale of time for the MC simulation were known, then this would specify how to obtain the laboratory blocking temperature from the simulation. However, t_{MC} is not known in an absolute sense, since the ''time'' in the MC simulation is measured only in MC steps, with no physical dimensions. Alternatively, we can adopt a different interpretation of Eq. (7). In the MC simulation we found $D/k_B T_B^{\mathrm{MC}} = 14.81$, and we can use Eq. (7) to determine the value of t_{MC} that recovers the usual experimental result $D/k_B T_B^L \approx 30$. Using $t_L = 10^3$ sec, this leads to $t_{\mathrm{MC}} = t_L \exp(-15.12) = 2.70 \times 10^{-4}$ sec, the same value as stated earlier. We should also note that Eq. (7) can be generalized to the case where the anisotropy constant D takes a different value in the simulation than in the laboratory [i.e., change $D \rightarrow D'$ on the right-hand side (RHS)]. The formula really only specifies how the ratio D/T_B changes with observation time.

With these ideas in mind, we can compare our results from the simulations with the data for fcc Co particles 18 (Å) in diameter.[14] The experimental values for the anisotropy energy and the magnetic moment per particle are $D/k_B \approx 648$ K and $\mu = 550\mu_B$ with $T_B = 22 \pm 2$ K. The blocking temperature determined from the Monte Carlo simulations (Fig. 1) for $D/k_B = 648$ K is $T_B^{\mathrm{MC}} = 44 \pm 1$ K. By Eq. (7), this maps into $T_B^L = 21.7$ K, in agreement with the experimental value, as must be the case, because the experimental value of the anisotropy constant is determined from the experimental blocking temperature via Eq. (2).

The hysteresis loops (magnetization for H increasing only) are shown in Fig. 4 for the same value of D/k_B and a set of temperatures $T < T_B$. The observed coercivity agrees well with the theoretical value given by

$$H_c = 0.48 \times \frac{2K}{M_s}(1 - \sqrt{T/T_B}) \tag{8}$$

for an assembly of particles with randomly oriented anisotropy axes.[15] The inset of Fig. 4 shows $h_c = H_c/H_{c0}$, where $H_{c0} = 0.96K/M_s$, from the Monte Carlo data and from the theory. The reduced remanent magnetization M_r/M_s also agrees well with the theoretical value of 0.5 and starts to deviate from it when T approaches T_B.

While the values of h_c are in good agreement with the superparamagnetic theory, the experimental values for the same ratio of T/T_B are approximately one order of magnitude smaller. This could be caused by existence of imperfections in the crystal structure of the particles though it does not seem to be the case.[14] There are also considerable deviations from the bulk properties when the size of the particles is decreased. For instance, the 18 Å diameter particles show an anisotropy energy density about an order of magnitude larger than the bulk value and the atomic magnetic moment also increases. These are attributed to surface effects which

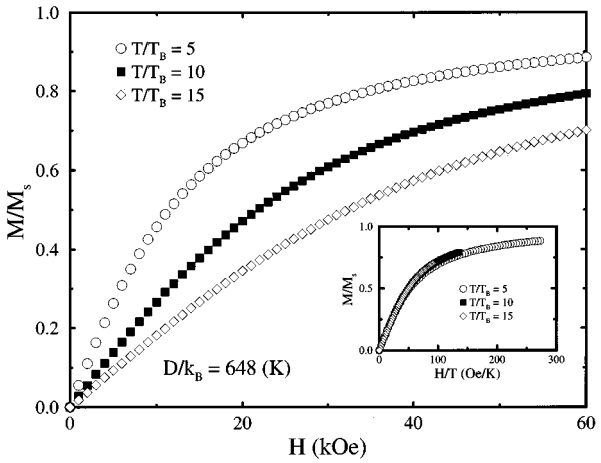

FIG. 5. Reduced magnetization of the system vs H for $T>T_B=44$ K. The inset shows the overlap of the magnetization curves when plotted vs H/T. The error bars are of the order of the symbols or smaller.

should lead to deviations from coherent rotation of the atomic magnetic moments, particularly of the surface spins which have less nearest neighbors and could fluctuate more. The number of surface sites is considerable in such small particles, and so the smaller coercivity may be due to surface effects too.

Finally, the magnetization curves for three values of the temperature above T_B are shown in Fig. 5 again for $D/k_B=648$ K. These results show that the MC simulations are consistent with the superparamagnetic behavior which is illustrated by the overlap of the magnetization curves when plotted vs H/T, the inset in this figure. The experiment also shows superparamagnetic behavior but with approximately half as large saturation magnetization at large external magnetic fields for the same values of T/T_B which is attributed to a core-shell structure of the particles[14] with a saturation magnetization of the shell of the particle much smaller than the net magnetic moment of the atoms in the shell. Since the small saturation magnetization of the particles at large fields cannot be obtained assuming coherent rotation of the atomic spins, it is of importance to consider the magnetic properties of the particles with their internal structure taken into account when the surface starts to dominate their properties.

In conclusion, we carried out Monte Carlo simulations for an assembly of particles with randomly oriented uniaxial anisotropy. The simulation recovers all of the results in the superparamagnetic theory. The observation time in the Monte Carlo simulations is much smaller than the corresponding time in the experiments. The effect of the smaller observation time leads to a higher blocking temperature in the simulations and generally the superparamagnetic properties which are observed in the experiments for a given temperature should be observed in the simulations at a higher temperature. A comparison of the simulations with the experiments on fcc Co particles 18 Å in diameter for the same ratio of T/T_B shows that while the properties of these particles are generally superparamagnetic there are two deviations from the theory and from the simulations. The coercivity in the experiment is about an order of magnitude smaller than the values of the coercivity in the superparamagnetic theory and in the simulations, and the saturation magnetization for $T>T_B$ is about half as large as in our simulations. These deviations are assumed to be caused by the surface of these particles which requires a study of such particles on an atomic level. The method we have used suggests a possible extension for simulations on a single particle with its atomic structure considered and anisotropy included.

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