Magnetic coupling induced increase in the blocking temperature of $\gamma$-Fe$_2$O$_3$ nanoparticles

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In this article, we report the magnetic properties of surfactant coated $\gamma$-Fe$_2$O$_3$ nanoparticles which are pressed under different pressures. With increasing pressure, the sample volume decreases, density increases, and a 55% density change has been achieved. The blocking temperature is increased from 50 to 80 K. Analyzing the data of blocking temperature versus densities, which exhibits linear relationship, and comparing the magnetic properties, the increase in blocking temperature is understood in terms of increased magnetic interactions between neighboring nanoparticles, which is due to the reduced average interparticle distance by the applied pressure. © 2000 American Institute of Physics. [S0021-8979(00)06410-0]

INTRODUCTION

Systems consisting of magnetic nanoparticles have been widely studied in recent years, and their superparamagnetic properties have attracted much attention. The susceptibility as a function of temperature reveals some of the main features of a superparamagnetic system. It is well known that the thermal stability of the magnetic particles depends on the anisotropy of the particles, and it is also affected by the interparticle interactions. The blocking temperature $T_B$, below which the particle moments are blocked, is usually considered an important parameter when studying a magnetic nanoparticle system. In general, $T_B$ can be obtained by analyzing the zero field cooled and field cooled (ZFC/FC) susceptibility versus temperature curve. The magnetic behavior of these particle systems has been explained by theoretical models based on the work of Néel, Brown, and Bean and Livingston. In these models, one can use the treatment that the atomic magnetic moments within the particles are acting coherently and their magnetic moments can be represented by a single vector with a magnitude $\mu = N \mu_0$, where $N$ is number of atoms in the particle and $\mu_0$ is the average magnetic moment of an atom. Considering that the relaxation time $\tau$ is a function of temperature $T$ and anisotropy barrier $E_a$,

$$\tau^{-1} = f_0 \exp(-E_a/k_B T),$$

where $k_B$ is Boltzmann’s constant, $f_0$ is a frequency factor on the order of $10^9$ s$^{-1}$, $E_a$ is the anisotropy barrier that can be determined by $E_a = KV$ in which $K$ is the anisotropy energy density constant, and $V$ is the volume of particle. The definition of blocking temperature of an ideal superparamagnetic particle system is given as follows:

$$T_B = E_a/k_B \ln(tf_0),$$

where $t$ is the experimental measuring time. $k_B \ln(tf_0)$ can be treated as a constant. Herein only the anisotropy energy barrier $E_a$ has been considered. If we include the interaction between particles, the energy barrier and blocking temperature will be modified

$$T_B = (E_a + E_{int})/k_B \ln(tf_0),$$

where $E_{int}$ is introduced to indicate the interaction energy. Obviously higher interaction may cause higher $T_B$. The theoretical model of the interaction energy $E_{int}$ has been introduced by Dormann et al., in which the interparticle interactions are treated as magnetic dipole–dipole interaction. Samples containing magnetic nanoparticles (powder or bulk) can be made by several different ways: either chemical synthesis or physical methods like ball milling and film deposition. In an experiment, the blocking temperature $T_B$ is normally determined by measuring the peak position of ZFC susceptibility versus temperature curve. Often, the interactions between magnetic particles are ignored and the susceptibility data are analyzed without considering the interparticle distance or the density of particles. In reality these parameters influence strongly the interparticle magnetic interaction in the nanoparticle systems. We have designed an experiment to study how applied pressure and sample density affect the magnetic properties of a nanoparticle system, specifically how the blocking temperature is changed by compressing it.

EXPERIMENTS AND DISCUSSION

$\gamma$-Fe$_2$O$_3$ particles of spherical shape ranging from 6 to 7 nm in diameter were prepared using the method for the synthesis of magnetoliposomes. In this method, an aqueous ferrous solution was first trapped inside the phospholipid vesicles consist of dimyristoylphosphatidylcholine (DMPC) by direct injection. Ammonia solution was subsequently added to this system and its diffusion into the ferrous-containing vesicles causes the formation of nanosized par-
particles of $\gamma$-Fe$_2$O$_3$ since those vesicles as nanoreactors restrict the growth of as-precipitated particles within nanoscaled dimensions. Since there was no further purification, after drying, the $\gamma$-Fe$_2$O$_3$ particles were surrounded and isolated by the residual of DMPC surfactant. The thickness of the surfactant layer was estimated to be about 3 nm. Such a sample in which magnetic particles are embedded in a porous nonmagnetic matrix exhibits superparamagnetic behaviors; Figure 1 is the schematic view of this system. Particles are isolated from each other by the surrounding DMPC, and the system is compressible.

We used a steel die to apply the pressure on the bulk samples. Some nonmagnetic buffer powders have been used, which are separated from the samples by sample holders, to allow uniform pressure distribution. Both scotch tape and Cu foil have been used as the sample holders, and we found that there was no significant difference in experimental results when we used different sample holders. For each compression, we kept the pressure for 15 min to stabilize the volume change. By measuring the thickness of the compressed sample, the volume change of sample and its density were obtained. The susceptibility and magnetization measurements were made with a Quantum Design superconducting quantum interference device with temperatures varying from 5 to 300 K.

Figure 2 illustrates that the susceptibility–temperature behavior of the system has been significantly changed after compressing under a pressure $P = 5.0 \times 10^8$ N/m$^2$. The data were taken under both ZFC and FC conditions with an applied field of 50 Oe. Although no big change in the maximum value of the susceptibility after compression was found, the blocking temperature $T_B$ has been increased significantly. It was enhanced by more than 50% after the sample was compressed under a pressure of $6.3 \times 10^8$ N/m$^2$.

Figure 3(a) shows the pressure dependence of the blocking temperature $T_B$, and Fig. 3(b) shows the blocking temperature as a function of sample density. The blocking temperature $T_B$ varies linearly with the density of the samples in the data range of our experiment.

To determine the causes of the increase in $T_B$, one may consider the changes in sample density, thus the interparticle distance and interaction, or the change in particle size, or the particle shape. As will be seen later, the change of interparticle distance is the main reason for the increase in $T_B$ observed in our experiments.

We have compared the particle size before and after compression, which were measured by x-ray diffraction. The average particle size is about 6–7 nm in diameter and it was found that there was no significant change of the particle size after the samples had been compressed. The effect of pressure is essentially the densification of the porous DMPC matrix. With regard to the possible change of particle shape due to the pressure, the susceptibility and magnetization of the

![FIG. 1. The schematic view of $\gamma$–Fe$_2$O$_3$ nanoparticles surrounded by DMPC surfactant. The $\gamma$-Fe$_2$O$_3$ nanoparticles are well separated by the surfactant.](image1)

![FIG. 2. ZFC and FC $\chi$–$T$ curves of $\gamma$–Fe$_2$O$_3$ samples before and after compressing. The blocking temperature $T_B$ has been increased by ~ 28 K after the sample was compressed under pressure $P = 5.0 \times 10^8$ N/m$^2$.](image2)

![FIG. 3. (a) The blocking temperature $T_B$ as a function of applied pressure. (b) $T_B$ as a function of the sample density; a linear relationship between the $T_B$ and density $D$ exists.](image3)
compressed sample were measured in two different orientations: the applied field is parallel to the direction in which pressure was applied ($\mathbf{H} \parallel \mathbf{P}$), and perpendicular to it ($\mathbf{H} \perp \mathbf{P}$). As shown in Fig. 4, there is no difference in the susceptibilities and magnetizations between the two orientations. The coercivity measured at 10 K is also the same for the two orientations. This implies the $\gamma$-Fe$_2$O$_3$ particles do not change significantly from the spherical shape due to the applied pressure. So we are inclined to interpret the variation of $T_B$ as being due to the change of sample density, that is, the change of particle distance, which affects the interparticle interaction. It should be pointed out that the $\gamma$-Fe$_2$O$_3$ nanoparticles are well separated by DMPC, and it is unlikely that the pressure can cause direct contact between neighboring particles and form a larger particle.

We consider the dipole–dipole interaction as the main coupling mechanism between magnetic particles. For two identical particles with magnetic moment $M$, interaction can be denoted as follows:

$$E_{\text{int}} = \frac{M^2}{r} (3 \cos \psi_1 \cos \psi_2 - \cos \alpha),$$

where $r$ is the distance between the particles, $\psi_1$ and $\psi_2$ are the angles between $r$ and the two moments, respectively, and $\alpha$ is the angle between the two moments. Compressing changes the sample density and decreases the average distance $r$, which causes an increased interaction. According to Eq. (3), an enhanced $T_B$ is expected. Considering that the density $D$ is a function of particle distance $r$, $D \propto r^{-3}$, from Eqs. (2), (3), and (4) one can easily see the blocking temperature $T_B$ is a linear function of the sample density $D$; that is $T_B \propto D$. This result agrees well with our experimental data shown in Fig. 3(b).

**SUMMARY**

In summary, we have studied a highly compressible system in which nanoparticles of $\gamma$-Fe$_2$O$_3$ are covered with a DMPC surfactant layer and well isolated from each other. The average interparticle distance can be controlled by applying a given pressure. The pressure reduces the interparticle distance, which leads to an increased magnetostatic interaction. With increasing coupling, the effective volume of the particles increases, and the blocking temperature $T_B$ is greatly enhanced. The linear relationship found between $T_B$ and sample density supports this explanation. Our results demonstrate the interactions between magnetic particles, which play an important role in the magnetic properties of superparamagnetic systems, and can be controlled by adjusting average particle distance through sample density in a properly chosen system.

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