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Quantum Tunneling of Magnetization in Single Domain CoFe$_2$O$_4$ Particles

T. Oku, H. Takano, M. Ohwada, T. Sato and E. Ohta

Department of Materials Science, Keio University, 3-14-1, Hiyoshi, Kohoku-ku, Yokohama, Kanagawa, 223 Japan

Abstract. We systematically investigated the macroscopic quantum tunneling (MQT) phenomena of magnetization in CoFe$_2$O$_4$ particles with a diameter ranging from 30 to 92 Å in zero external magnetic field to remove the substantial influence of energy dissipation. For all of our samples, we observed the MQT phenomena in zero external magnetic field, in spite of the anticipation that a large magnetic field $H$ comparable with the coercive force $H_c$ would be necessary to lower the energy barrier so as to observe appreciable quantum tunneling of magnetization. The analysis shows that the crossover temperature $T^*$ from the thermal to the quantum regime increases with decreasing anisotropy field $H_a$. This relation between $T^*$ and $H_a$ is inconsistent with the theoretical prediction.

1. INTRODUCTION

In the classical model, magnetization reversal occurs by thermal activation and disappears at zero temperature. Recent theoretical studies predicted that magnetization could reverse by quantum process which was independent of temperature [1]. Moreover, the phenomena relevant to the macroscopic quantum tunneling (MQT) of magnetization have been observed experimentally in some systems consisting of single domain particles [2-4]. E.M. Chudnovsky and L. Gunther theoretically calculated the tunneling probability of magnetization and the crossover temperature $T^*$ from the thermal to the quantum regime under a large external field $H$ comparable with the coercive force $H_c$ [1]. Such a high magnetic field should cause sizable energy dissipation. The effect of dissipation was not taken into account in their calculation, in spite of the fact that it plays an important role in MQT. Most of the experimental works on MQT were performed under a large external field. So we wish to observe MQT phenomena irrelevant to energy dissipation in order to understand it exactly. In this paper, we systematically study the MQT phenomenon in CoFe$_2$O$_4$ particles with a median diameter ranging from 30 to 92 Å in zero external field to remove the substantial influence of energy dissipation.

2. EXPERIMENTAL

We prepared CoFe$_2$O$_4$ particles by the chemical coprecipitation method at some pH values as shown in Table 1, and the particles were coated with the nonmagnetic surfactant oleic acid to prevent the aggregation of particles. For magnetic measurements, the particles were packed in a Teflon tube. Magnetization curve was obtained at 6 and 10 K and the time dependence of thermoremanent magnetization (TRM) was measured after cooling the samples to a measuring temperature in a field of 1000 Oe and then turning it off.

3. RESULTS AND DISCUSSION

From TEM examination, the particles are approximately spherical and have a lognormal distribution with median diameters D$_p$ of 30, 63, 76 and 92 Å for samples A, B, C and D, respectively. Magnetization curves at 6 and 10 K are shown in Fig.1. In order to calculate the effective anisotropy constant $K_1$, we used the following expression for $H_c$ at temperature $T$ [5],

$H_c = \alpha (2K_1/\mu_b) \left[ 1 - (25 k_B T / E)^{(1/2)} \right], \quad (1)$

where $\mu_b$ is the saturation magnetization, the prefactor $\alpha=0.32$ [6] and the energy barrier $E = 1/4 K_1 V$ [7] for cubic anisotropy. Using the relations of $H_c$ measured at two temperatures, we deduced $K_1$ and the magnetic volume $V_m = 1/6 \pi <D_m>^3$ based on Eq.(1). In this calculation, we used the value of $\mu_b$ of bulk CoFe$_2$O$_4$, and neglected the influence of interparticle magnetic interaction on $H_c$, since the energy of magnetic interaction is much smaller than that of the anisotropy. The results are listed in Table 1. The increase in $K_1$ with decreasing particle size may be attributed to the surface effect [8].

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Table 1. Data characterizing all samples.

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<td>A</td>
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<tr>
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<td>10.5</td>
<td>75.8</td>
<td>67.6</td>
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<td>53,600</td>
<td>~6</td>
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<tr>
<td>D</td>
<td>13.0</td>
<td>92.1</td>
<td>76.5</td>
<td>910</td>
<td>38,100</td>
<td>6</td>
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</table>

Fig.1 Magnetization curves.

Fig.2 Temperature dependence of S.

The time dependence of TRM was described by the relation: M_{TRM} = M_0 - S \ln(t), and the magnetic viscosity S was plotted against T (Fig.2). In all of our samples, the magnetic viscosity S decreases with decreasing temperature and shows a tendency to be independent of temperature below a certain temperature T*. As shown in Fig.2. In the case of relaxation by thermal activation, S should be proportional to T at temperatures much lower than the blocking temperature [9]. Thus, the temperature independent behavior of S below T* must be mainly attributed to the quantum process of relaxation. The values of T* are listed in Table 1. T* increased with decreasing H_A and increasing particle size. According to the theoretical and experimental studies on MQT [10,11], T* should be proportional to the square root of the anisotropy field H_A (=2K1/Is) independently of particle volume. The opposite relation between T* and H_A suggests that there exists some contribution to T* except for H_A. Thus, we claim that the interparticle interaction is a candidate for mechanism bringing about such a size dependent change in T*, since the interaction energy should increase with increasing the particle size. A detailed study of the influence of the interparticle interaction on MQT will be presented elsewhere.

In conclusion, we observed the MQT phenomena in CoFe_2O_4 particles in zero external field in spite of the anticipation that a large magnetic field H = H_c would be necessary to observe appreciable quantum tunneling of magnetization. In the present samples, the crossover temperature T* increased with decreasing H_A and increasing the particle size, which is inconsistent with the previous theoretical prediction.

References