Reexamination of Macroscopic Quantum Tunneling in Ferritin - Temperature dependence of Magnetic Relaxation -

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Abstract

The existence of pure quantum tunneling of macroscopic magnetic moment in the Kelvin regime is reexamined for antiferromagnetic nanoparticles in ferritin. In the present study, we discuss effects of temporary temperature change on magnetic relaxation. The results show that the relaxation slows down when the temperature is decreased, as expected for thermal activation process.

Key words: macroscopic quantum tunneling, ferritin, magnetic relaxation

Recently, pure and thermally assisted quantum tunneling have been intensively investigated for macroscopic magnetic moment ($\approx 300\mu_B$) of antiferromagnetic nanoparticles in ferritin [1-4]. For thermally assisted quantum tunneling, field dependence of magnetic relaxation has been discussed in the temperature range higher than about 2 K [2]. Recently, a strict approach was proposed [3]. Hence, thermally assisted quantum tunneling will be clarified in the near future. On the other hand, temperature-independent magnetic viscosity S was observed at the temperatures lower than about 2 K [1,4]. Although this result suggests the existence of pure quantum tunneling, it is not a conclusive proof. Hence, another approach is required in order to clarify the existence of pure quantum tunneling in the Kelvin regime. In this study, the approach proposed by Sappey et al. [5] is applied to the temperature dependence of magnetic relaxation.

The sample was a commercial ferritin from horse spleen (Wako Pure Chem. Ind. Ltd., 100 mg/cm^3). The solution was concentrated by drying in a vacuum desiccator. The measurements were performed after cooling from 35 K to the desired temperature T_m in zero field (ZFC), or in static field H_{FC} (FC). The magnetization curve was measured below 80 kOe after ZFC. The thermoremanent magnetization $M_r(t, T_m)$ was recorded as a function of time t after cutting off H_{FC}.

We found that the magnetization curve is not saturated at the highest field. There is an additional linear component in the high field range. This behavior is consistent with the results of the previous study which interpreted it as the susceptibility of the antiferromagnetically ordered core [6]. It was found that the decay of M_r for $H_{FC} = 30$ kOe is almost proportional to ln t between 1 ks and 10 ks. Therefore, we can estimate the magnetic viscosity S that is the slope of M_r in the ln t plot. The result shows that S decreases with decreasing temperature in the range between 2.3 K and 8 K, and then it flattens out below about 2.3 K. This behavior is consistent with the results of the previous study [1], and it may suggest the dominance of pure quantum tunneling below 2.3 K. However, it is not sufficient evidence[5,7]. In order to verify such dominance, a scaling relationship is usually examined for the relaxation curves at different temperatures in the $T_m\cdot$ ln(t/τ_0) plot, where $1/\tau_0$ is the attempt frequency [7]. If thermally activated process dominates the relaxation, the relaxation curves on the same route collapse onto an unique master curve. However, we can not use this approach, because it is difficult to prepare the same

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initial state at different temperatures. We should recall that the magnetization curve can not be saturated at 80 kOe. For this reason, the existence of pure quantum tunneling must be examined by using another approach.

The approach has been given by Sappey et al. [5]. They gave attention to that temperature change during magnetic relaxation does not affect the relaxation by non- thermal processes. We should notice that this approach is independent of the quality of the initial state. In this study, we measured the decay of M_r for $H_\mathrm{FC} = 30$ kOe at $T_\mathrm{m} = 2.0$ K. At t_1 the temperature was temporary shifted to $T_\mathrm{m} + \Delta T$, and then the temperature was returned to T_m at t_2. During it, the measurement was continued. Figure 1 shows the relaxation curves for $\Delta T = \pm 0.05$K. It can be found that the relaxation slows down during the temporary cooling, while it accelerates during the temporary heating. The relaxation curve after the temporary temperature change can be onto the isothermal relaxation curve at T_m, if we regard $t - (t_2 - t_1) + t_\mathrm{eff}$ as an effective time. This result indicates that the relaxation at $T_\mathrm{m} + \Delta T$ for $t_2 - t_1$ is equivalent to the relaxation at T_m for t_eff. In other words, both the relaxations surmount the same barrier at different rates. Figure 2 shows that the ratio of t_eff to $t_2 - t_1$ exponentially varies with ΔT. The estimated height $E_\mathrm{a}/k_\mathrm{B}$ of the barrier is about 60 K, and it is comparable to the maximum barrier height that can be surmounted in 10 ks at 2 K. Here, τ_0 is assumed at 10^{-9} s. The same results can be obtained for $H_\mathrm{FC} = 4.8$ Oe. These results clearly show that some thermally activated process dominates the magnetic relaxations

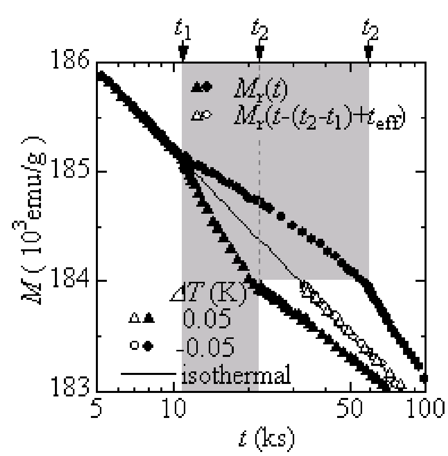

Fig. 1. Relaxation curves of M_r with temporary temperature change for $\Delta T = \pm 0.05$K. The solid line shows the isothermal relaxation curve at $T_\mathrm{m} = 2.0$ K. The open symbols represent M_r as a function of effective time $t - (t_2 - t_1) + t_\mathrm{eff}$, while the closed symbols show M_r as a function of the total time t. The shaded region represents the period of the temporary temperature change.

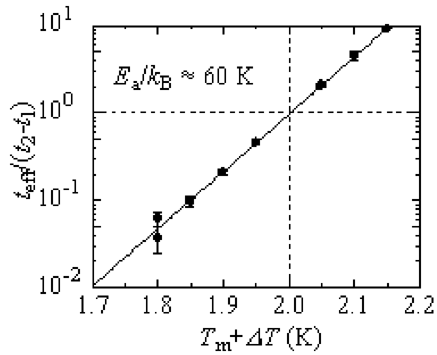

Fig. 2. Ratio of t_eff to $t_2 - t_1$ for various ΔT.

in the vicinity of 2 K, in contrast with the suggestion by the analysis of the magnetic viscosity. Needless to say, the existence of pure quantum tunneling is not denied at the other temperatures. When the remained possibility is examined, it will be useful to keep in mind that the approach using temperature change is suitable as shown in this study.

Acknowledgements

We acknowledge helpful discussions and technical assistance by Dr. T. J. Sato.

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