Measurement of the Dynamics of the Magnetization Reversal in Individual Single-Domain Ferromagnetic Particles

M. Lederman and S. Schultz
Department of Physics and Center for Magnetic Recording Research, University of California, San Diego, La Jolla, California 92093-0319

M. Ozaki
Department of Chemistry, Yokohama City University, 22-2 Seto, Kanazawa-Ku, Yokohama 236, Japan
(Received 28 February 1994)

We have measured the spontaneous thermal switching of the magnetization of individual single-domain ellipsoidal ferromagnetic γ-Fe_2O_3 particles. At room temperature, and in applied fields close to coercivity, the statistics of the reversal cannot be described by activation over a single-energy barrier as originally proposed by Néel. We suggest the dynamics of reversal occurs via a complex path in configuration space, and a complex theoretical approach is required to provide a correct description of thermally activated magnetization reversal even in single-domain ferromagnetic particles.

PACS numbers: 75.60.−d, 75.10.Hk

The mechanism and dynamics of the thermally activated switching of the magnetization of a single-domain ferromagnetic particle has been of considerable interest to theorists and experimentalists since the pioneering work of Néel [1–3]. An understanding of this problem is important from both a fundamental science and a technological point of view. It is of intrinsic interest because the process may be complex even for this simple ferromagnetic system, currently viewed as a two-level system. The insights gained may be relevant to current problems, such as the interpretation of data proposed as evidence for magnetic quantum tunneling (MQT) and magnetic quantum coherence (MQC) [4] and the fundamentals of magnetic interacting systems [5], such as spin glasses [6] and collections of ferromagnetic particles [3,7] where complex time-dependent functional forms are observed. As recording densities increase, these ferromagnetic units will approach sizes where a detailed understanding of their thermal-magnetization switching behavior is necessary.

The determination of the statistics of the thermally activated magnetization reversal as a function of the orientation, magnitude, and duration of the applied magnetic field requires thousands of measurements of the remanent magnetization. In combination with the transmission electron microscopy (TEM) techniques of Salling *et al.* [8] for sample preparation and morphological characterization, we have used magnetic force microscopy (MFM) as an ideal tool to very efficiently determine the sign of the remanent magnetization.

The study of the magnetization reversal of individual single-domain particles is difficult because of their small size (here 0.30 μm by 0.065 μm) and their small moment (typically 10^{-13} emu). Prior attempts at characterizing isolated-individual single-domain particles have been reported [9,10], but they lacked the ability to provide quantitative information and verification of the particle morphology. As an alternative, there have been many experiments on collections of particles, where measurements are made of thermal viscosity [7,11,12], the frequency dependence of the coercivity [13,14], and of discrete hysteresis jumps in CoCr [15]. But they generally lack the ability to provide a critical test of the underlying dynamical processes of an individual model particle, because of the complications due to the spread of individual particle parameters and the consequences of their interactions [16].

Even for permalloy particles prepared by nanolithography, and hence with well separated nearly identical particle sizes [17,18], we have found a distribution of switching fields of 180 Oe between 370 and 550 Oe [19]. Measurements of the probability of thermally activated magnetization reversal $P(t)$ in such particles indicated that the process could not be described by an Arrhenius process for a single-energy barrier. However, since the angular dependence of the switching field $H_s(\theta)$ was very different from the Stoner-Wohlfart model of uniform rotation (SW), one might naturally attribute the deviations from an Arrhenius process to the complexity of the mode of reversal. Here we report on two γ-Fe_2O_3 prolate ellipsoids, where one has $H_s(\theta)$ in agreement with the predictions of SW but where $P(t)$ is also not described by a simple Arrhenius process.

Recently, Salling *et al.* [20] have reported measurements of the angular dependence of the switching field for individual γ-Fe_2O_3 particles by Lorentz TEM, which also allows for *in situ* characterization of the particle size, crystal structure, and morphology. We have made analogous measurements utilizing MFM [21,22] and have analyzed the data by comparison to numerical calculations of the full micromagnetic equations used to determine the angular dependence of the nucleation field $H_s(\theta)$ [23]. This analysis indicates that when the field is applied very near the long (easy) axis of the particle, the mode of reversal is curling [24], but for larger angles ($\theta > 30°$), the mode of reversal is consistent with uniform rotation [25]. In this Letter we address the more difficult problem of determin-

 0031-9007/94/73(14)/1986(4)$06.00 © 1994 The American Physical Society

ing the probability of spontaneous thermal-magnetization reversal as a function of the system parameters.

The γ-Fe_2O_3 particles (with a bulk saturation magnetization $M_s = 350$ emu/cm^3 and a coercivity of 350 Oe[20]) have been made by the process described by Ozaki and Matijevic [26] and are deposited on a TEM grid [8]. Particles are examined in the TEM, and those selected for measurement are isolated and a good approximation to a prolate ellipsoid 3000 Å long and 650 Å wide. Their small size and large aspect ratio ensure that the particle is in a single-domain state, and that the remanent magnetization is parallel to the long axis of the particle. Two examples of the angular dependence of the switching field $H_s(\theta)$ for typical individual particles are presented in Fig. 1. H_s is bounded by the highest field that does not reverse the magnetization and the lowest field that always does, when the field is applied for a time of 1 s. The solid line is the prediction of the SW model [25] for a prolate ellipsoid with an aspect ratio of 4.6 and a saturation magnetization $M_s = 350$ emu/cm^3. The interpretation of these data based on the studies presented in Ref. [23] is that near 0° the switching is best represented by a curling mode, but for large angles (>30°) the switching field is in agreement with that predicted by the SW model [25,27]. In the data that follow, we present measurements in both regimes.

The Néel [1] and Brown [2] model of the thermal switching of the magnetization of a single-domain particle is appealing in its simplicity. For $H = 0$, a single-domain particle has two equivalent ground states of opposite magnetization separated by an energy barrier. By applying an external field, the barrier decreases in height and, if the field is large enough, $H \leq H_s$, thermal fluctuations are sufficient enough to drive the system to overcome the barrier, and the magnetization is reversed. To date, analytical expressions for the barrier height exist only for the case of uniform rotation, i.e., when the magnetization acts as a rigid spin during the reversal process [25]. Expressions for other modes of reversal have recently been proposed for particular geometrics and anisotropy axes [28], but in all cases, the dynamics of the reversal is assumed to be described by a thermal activation process in which the probability after a time t that the magnetization has *not* switched is given by

$$P(t) = e^{-t/\tau}, \tag{1}$$

where τ is a characteristic time, depending on the ratio of the height Δ of the energy barrier to the temperature T, and is given by

$$\tau = \tau_0 e^{\Delta/k_B T}. \tag{2}$$

The numerical value of the prefactor τ_0 is specified by the particular theory. For the size and parameters of the particles under discussion, τ_0 is typically between 10^{-10} and 10^{-8} s.

As mentioned, prior to this Letter only the time dependence of the magnetization $M(t)$ of collections of particles has been studied [3,11,12,15] because of the extremely small moment of the individual particles. In all cases, $M(t)$ has failed to follow Eq. (1). Instead, in the range of times accessible to experimentalists, typically 3 to 5 decades in time [29], $M(t)$ approximates the logarithmic dependence [7]:

$$M(t) = M_0 S(T) \ln(t), \tag{3}$$

where $S(T)$ is the magnetic viscosity. The reason why Eq. (3) is observed instead of Eq. (1) is postulated to be due to a distribution of characteristic switching times $P'(\tau)$ because, in a collection of noninteracting particles, each one has its own τ. Therefore $M(t)$ may be written as

$$M(t) = \int_0^\infty P'(\tau) e^{-t/\tau} \, d\tau, \tag{4}$$

with an appropriate distribution $P'(\tau)$ in a restricted time interval of 2 to 3 decades.

Our experiments determine $P(t)$ for individual-isolated single-domain particles. The measurements were made using a MFM with an *in situ* electromagnet [19], as follows: (a) The magnetic sensing tip is moved away from the sample, and a large negative field (1.5 kOe) is applied, which sets the remanent magnetization in a known direction. This set field is followed by a positive test field **H** (antiparallel to the 1.5 kOe) applied for a time interval t and for which overshoot is carefully prevented. After **H** = 0, the tip is brought back close to the sample, and a scan is made to determine whether the magnetization has switched or not. (b) Procedure (a) is repeated N times so as to collect statistics. The ratio of the number of cases where the magnetization did *not* switch, divided by N, is taken to be $P(t)$, the probability of not switching, which is to be compared with Eq. (1) [30]. [In our experiments, N was ~30 when $P(t)$ was close to 50% and ~100 when $P(t)$ was close to either 0% or 100%.] (c) The time t during which **H** is applied is changed, and we repeat procedures (a) and (b). For

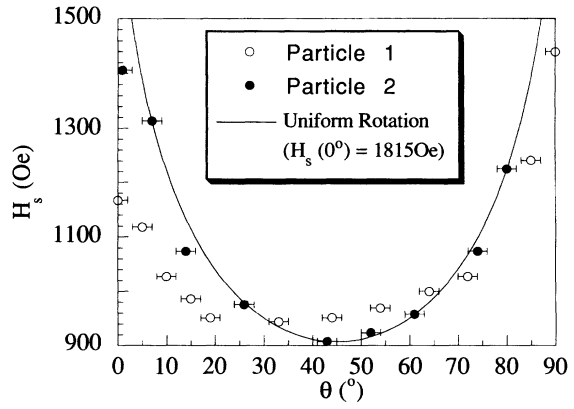

FIG. 1. Angular dependence of the switching field $H_s(\theta)$ of two isolated single-domain γ-Fe_2O_3 particles. The solid curve represents $H_s(\theta)$ predicted by Stoner and Wohlfart (uniform rotation) with the parameters given in the text.

our experiments, t varied from 100 ms to 10 s. (d) The strength of **H** is changed, and we repeat procedures (a), (b), and (c). Data were taken for **H** intervals of 3.3 Oe, with the range determined by the shortest time interval that could be reliably measured (100 ms) and the longest time chosen for applying the field (10 s).

We present the results of these measurements in Fig. 2, where the probability of not switching is plotted versus log$t$. We note that this "y-log$x$" semilog format is just the opposite of that normally used to test for a presumed exponential decay process but has the advantage that $P(t) = e^{-t/\tau}$ is represented by a universal curve, whereby, changing τ simply corresponds to sliding the curve along the x axis. Our main observations are as follows: (1) The time that corresponds to $P(t = t_{50\%}) = 50\%$ varies from $\approx$10 s when $H = 1067.3$ Oe to $\approx$0.25 s when $H = 1077.1$ Oe. Therefore, a change in the field of only $\sim$1% corresponds to a change in the characteristic time of 4000%. The very dramatic effect means that when $H = 0$ the magnetization is extremely stable and unlikely to spontaneously switch directions during any time scale of our measurements. (2) In the range $20\% \leq P(t) \leq 80\%$, $P(t)$ appears to be logarithmic [reminiscent of Eq. (1)], but as $P(t)$ approaches 0% and 100%, it deviates from a log$t$ curve. In Fig. 3, we replot the data of Fig. 2 in the range $20\% \leq P(t) \leq 80\%$, with a solid line as a guide to the eye. (3) We note that the slope of $P(t)$ in Fig. 3 increases as H increases, which is reminiscent of measurements on a collection of particles where the viscosity [see Eq. (3)] increases as the coercive field H_c is approached [31].

In Fig. 3, we also present (in dashed lines) the function e^{-t/τ_0}, where τ_0 is adjusted for a best fit to the data. It is observed that for the two lowest fields, while a simple exponential can fit the data within the error bars, for the two largest fields, the data are not well described by

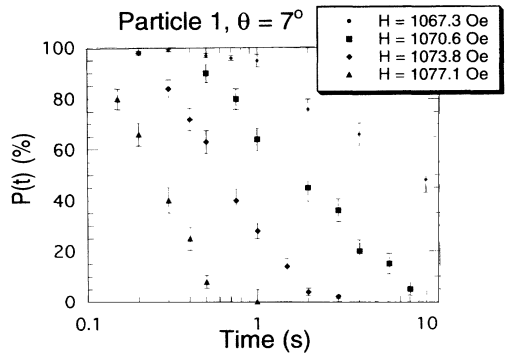

FIG. 2. For particle 1 in Fig. 1, we plot the probability $P(t)$ that the magnetization direction of the particle does *not* switch when a field H is applied for a time t, plotted as a function of log$t$. The field was applied at an angle $\theta \sim 7°$ with respect to the long axis of the particle. $P(t)$ was determined using the protocol described in the text. The four fields $H = 1067.3$, 1070.6, 1073.8, and 1077.1 Oe correspond to an appreciable change in $P(t)$ in the experimental time window $0.1 \leq t \leq 10$ s. The measurements were made at $T = 23.0 \pm 0.3$ °C.

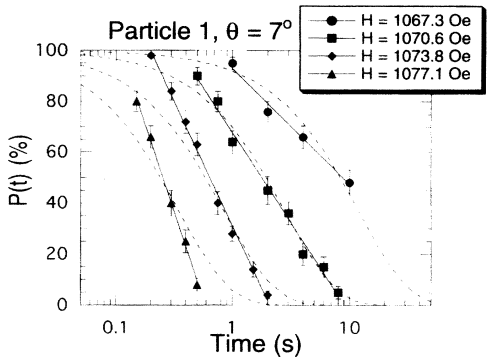

FIG. 3. The data of Fig. 2 are replotted in the range $20\% \leq P(t) \leq 80\%$. In this range, $P(t)$ is nearly linear in log$t$. The solid lines are guides to the eye for the four fields used. The dashed lines correspond to $f(t) = e^{-t/\tau_0}$, where the values of τ_0 are chosen for a best fit to the data. The implications of the deviations of the data from e^{-t/τ_0} are discussed in the text.

Eq. (1). Moreover, our measured $P(t)$ is such that (i) at short times ($t \ll \tau_0$), $P(t) > e^{-t/\tau_0}$, and (ii) at long times ($t \gg \tau_0$), $P(t) < e^{-t/\tau_0}$.

As shown in Fig. 1, particle 2 has an $H_s(\theta)$ very close to the SW prediction [25]. In Fig. 4 we present data for $P(t)$ taken at $\theta = 45°$, which is clearly in the SW regime and where $H_s(\theta)$ is independent of angle. It can again be seen that $P(t)$ obeys the above inequalities at both short and long times for all test fields. Since these inequalities are observed at both $\sim$0° (Fig. 3) and $\sim$45° (Fig. 4), *it suggests that they apply independent of the mode of reversal.*

In Ref. [27] we show that the integral in Eq. (4) has the following property. If $P'(\tau)$ is a very general function of τ with a mean τ_0, then (i) at short times ($t \ll \tau_0$),

$$\int_0^\infty P'(\tau) e^{-t/\tau}\, d\tau < e^{-t/\tau_0},$$

and (ii) at long times ($t \gg \tau_0$),

$$\int_0^\infty P'(\tau) e^{-t/\tau}\, d\tau > e^{-t/\tau_0},$$

which is just the opposite of our data. This indicates that the magnetization reversal of one isolated single-domain ferromagnetic particle is not only not well described by thermal activation over a single barrier but also not well represented by a series of independent thermally activated processes occurring in parallel [which is the meaning of Eq. (4)]. Instead, the reversal likely occurs via many complex paths in configuration space. For example, the paths between the initial and final state may correspond physically to the reversal nucleating in different locations of the particle and subsequently propagating via many statistically possible paths. Other reversal scenarios are also possible.

Our data show that the model of overcoming a single-energy barrier via thermal fluctuations is too simplistic

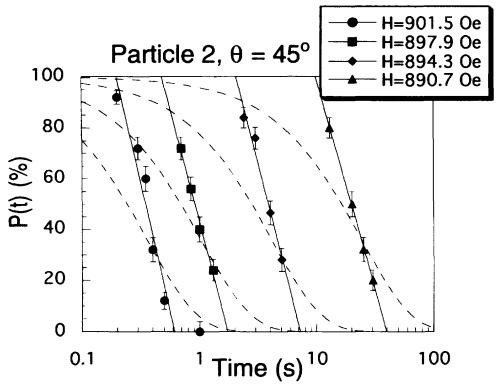

FIG. 4. $P(t)$ for particle 2 in Fig. 1 with $\theta \sim 45°$, an angle where H_s is consistent with the uniform rotation prediction. The implications of the very large deviations of the data from e^{-t/τ_0} are discussed in the text.

to describe the magnetization reversal in "real" single-domain ferromagnetic particles. We believe these results suggest that interpretations of the temperature dependence of magnetic viscosity, as evidence for MQT or MQC, should be viewed with caution, as the dynamics of even a single particle may be more complex than tunneling through a single barrier.

We have initiated measurements on pairs of particles similar to the ones described here. The results will provide information regarding the influence of interactions on both the magnetostatics and the dynamics of magnetization reversal. While we expect that numerical simulations in progress based on either magnetostatics [32] or damped Landau-Lifshitz-Gilbert equations [33,34] can model these systems (including particles having rough surfaces, holes, crystal anisotropy, etc.), to provide nucleation fields and equilibrium magnetization distributions, they have not yet provided a description of the thermally assisted dynamics. Thus, we suggest that an extended formulation of the dynamical reversal process for single-domain ferromagnetic particles is warranted.

We would like to thank Dr. W. Chen and Prof. D. Fredkin for very stimulating conversations, Dr. G. A. Gibson for building the MFM, R. O'Barr for helping us in the sample preparation, and G. Kassabian for building the excellent electronics necessary to do this study. This work was sponsored by the Center for Magnetic Recording Research, and by NSF Grant No. DMR-90-10908.

[1] L. Néel, Ann. Geophys. **5**, 99 (1949).
[2] W. F. Brown, Phys. Rev. **130**, 1677 (1963).
[3] *Studies of Magnetic Properties of Fine Particles and their Relevance to Materials*, edited by J. L. Dormann and D. Fiorani, Proceedings of the International Workshop on Studies of Magnetic Properties of Fine Particles and their Relevance to Materials Science; Rome 1991 (Elsevier, New York, 1992).
[4] A recent review of MQT/MQC can be found in P. C. E. Stamp, E. M. Chudnovsky, and B. Barbara, Int. J. Mod. Phys. B **6**, 1355 (1992).
[5] P. Hänggi, P. Talkner, and M. Borkovec, Rev. Mod. Phys. **62**, 251 (1990).
[6] The latest results are reviewed in *Recent Progress in Random Magnets*, edited by D. H. Ryan (World Scientific, Singapore, 1992).
[7] R. Street and J. C. Wooley, Proc. Phys. Soc. London, Sect. A **62**, 562 (1949).
[8] C. Salling, S. Schultz, I. McFadyen, and M. Ozaki, IEEE Trans. Magn. **27**, 5184 (1991).
[9] A. H. Morrish and S. P. Yu, Phys. Rev. **102**, 670 (1956).
[10] J. E. Knowles, IEEE Trans. Magn. **17**, 3008 (1981).
[11] R. W. Chantrell, J. Magn. Magn. Mater **95**, 365 (1991).
[12] A. Aharoni, in Ref. [3].
[13] P. Flanders and M. Sharrock, J. Appl. Phys. **62**, 2918 (1987).
[14] S. B. Oseroff, D. Clark, S. Schultz, and S. Shtrikman, IEEE Trans. Magn. **21**, 1495 (1985).
[15] C. Bucknell Webb and S. Schultz, J. Phys. (Paris), Colloq., Suppl. 12 **49**, C8-1975 (1988).
[16] C. Salling, Ph.D. thesis, University of California at San Diego, 1992 (unpublished).
[17] J. F. Smyth, S. Schultz, D. R. Fredkin, T. R. Koehler, I. R. McFayden, D. P. Kern, and S. A. Rishton, J. Appl. Phys. **69**, 5262 (1991).
[18] G. A. Gibson, S. Schultz, J. F. Smyth, and D. P. Kern, IEEE Trans. Magn. **27**, 5000 (1991).
[19] M. Lederman, G. A. Gibson, and Schultz, J. Appl. Phys. **73**, 6961 (1993).
[20] C. Salling, R. O'Barr, S. Schultz, I. R. McFayden, and M. Ozaki, J. Appl. Phys. **75**, 7989 (1994).
[21] Y. Martin and H. K. Wickramasinghe, Appl. Phys. Lett. **50**, 1455 (1987).
[22] G. A. Gibson and S. Schultz, J. Appl. Phys. **69**, 5877 (1991).
[23] D. Fredkin, M. Lederman, and S. Schultz (unpublished).
[24] E. H. Frei, S. Shtrikman, and D. Treves, Phys. Rev. **106**, 446 (1957).
[25] E. C. Stoner and E. P. Wohlfart, Philos. Trans. R. Soc. London A **240**, 599 (1948).
[26] M. Ozaki and E. Matijevic, J. Colloid Interface Sci. **107**, 199 (1985).
[27] M. Lederman, D. Fredkin, R. O'Barr, M. Ozaki, and S. Schultz, J. Appl. Phys. **75**, 6217 (1994).
[28] H. B. Braun, Phys. Rev. Lett. **71**, 3557 (1993).
[29] C. Bucknell Webb, S. Schultz, and S. B. Oseroff, J. Appl. Phys. **63**, 2923 (1988).
[30] This problem is the same as the simple random walk in one dimension. The error in $P(t)$ here is the same as in that problem. See F. Reif, *Fundamentals of Statistical and Thermal Physics* (McGraw-Hill, New York, 1965).
[31] S. B. Oseroff, D. Franks, V. M. Tobin, and S. Schultz, IEEE Trans. Magn. **23**, 2871 (1987).
[32] D. R. Fredkin and T. R. Koehler, J. Appl. Phys. **67**, 5544 (1990).
[33] Y. Nakatani, Y. Uesaka, and N. Hayashi, Jpn. J. Appl. Phys. **28**, 2485 (1989).
[34] S. Yuan, H. N. Bertram, J. Smyth, and S. Schultz, IEEE Trans. Magn. **28**, 3171 (1992).