Linear-response theory applied to the dynamics of submicronic magnetic particles

J-E. Wegrowe*
Laboratoire de magnétisme Louis Néel, CNRS, Boîte Postale 166, 38042 Grenoble Cedex 9, France

W. Wernsdorfer
Laboratoire de magnétisme Louis Néel, CNRS, Boîte Postale 166, 38042 Grenoble Cedex 9, France
and Centre de Recherche sur les Très Basse Température, CNRS, Boîte Postale 166, 38042 Grenoble, France

L. Thomas and B. Barbara
Laboratoire de magnétisme Louis Néel, CNRS, Boîte Postale 166, 38042 Grenoble Cedex 9, France

A. Sulpice, K. Hasselbach, and A. Benoit
Centre de Recherche sur les Très Basse Température, CNRS, Boîte Postale 166, 38042 Grenoble, France

D. Mailly
Laboratoire de Microstructure et Microéléctronique, CNRS, 196 Avenue H. Ravera, 92220 Bagneux, France
(Received 16 August 1995)

We first present magnetic aftereffect measurements of an array of submicronic amorphous noninteracting Co particles (300 nm $\times$ 200 nm $\times$30 nm). At low temperatures, the thermal dependence of the magnetic viscosity $S=dM/d(\ln(t))$ exhibits an unusual behavior. Comparison with measurements performed on a single particle allows the effects of the averaging over the statistical ensemble of the array of 2×10^7 Co particles to be studied experimentally. The link between both experiments is then performed by using the linear-response theory. This formalism allows the noise, $D(T)$, which accounts for the thermal dependence of the coupling of switching spins to their environment, to be defined. The consistency of this description is checked by comparing the noise measured in this way on the array of particle with the noise measured on the single particle through the Arrhenius law. In both cases, the thermal dependence of the noise $D(T)$ has a $\coth(T_0/T)$ form.

INTRODUCTION

In the last few years, the study of the dynamical properties of magnetic nanostructures at low temperature has been of considerable interest both from a fundamental and a technological point of view. Indeed, measurements on small systems showed some typical effects, which are relevant to the area of physics describing phenomena between the macroscopic and the microscopic world, i.e., relevant to the mesoscopic scale. An important and nontrivial question is to know how these typical effects can be observed by measuring an ensemble of noninteracting mesoscopic systems at the macroscopic scale.

Following Van Kampen's definition[1] mesoscopic effects occur when the fluctuations become predominant in the description of the system. Each realization of a single event, e.g., a magnetization jump due to the switch of a magnetic domain, accounts for the influence of the degree of freedom of the environment acting on the path taken by the system to realize this event. Indeed, the switching magnetic object is not isolated and interacts with its environment (the heat bath, impurities or surface spins). The effect of the environment on the system is modeled by a stochastic force whose time correlation defines the noise. This stochasticity manifests itself experimentally by a statistical distribution of the measured values. In the case of a macroscopic sample composed by a large set of noninteracting particles, the measure is directly averaged over the statistical ensemble of all particles and fluctuations around the mean value become negligible. However, fluctuations continue to play a fundamental role in dynamical properties and can be probed by relaxation measurements. In order to compare experimentally the fluctuations measured at both the nanoscopic and macroscopic scale, we need to master a second averaging process: the statistical ensemble of the set of all particles is not ideal and different distributions of probability exist. In other words, a distribution of mean values and fluctuations must be taken into account. The following problem arises: how can the averaging over the ensemble of particles be compared to the statistical averaging of a single particle?

Thanks to the progress in nanotechnology it is now possible to answer this question experimentally by using magnetic nanostructures. This article is divided in two parts. In an experimental report we present the relaxation measurements of an array of about 10^7 Co particles, followed by the report of the main results of dynamical measurements performed on one individual Co particle deposited onto a planar microbridge-dc-superconducting quantum interference device (SQUID). The detailed description of the measurements on the single particle is reported elsewhere.[2] In the theoretical interpretation, we present a model based on the linear-response theory, which allows to interpret the aftereffect measurements, performed on the macroscopic array, in terms of distribution of response functions. The consistency of this description is checked by comparing the magnetic noise deduced from aftereffect measurements on the array of particle

 © 1996 The American Physical Society

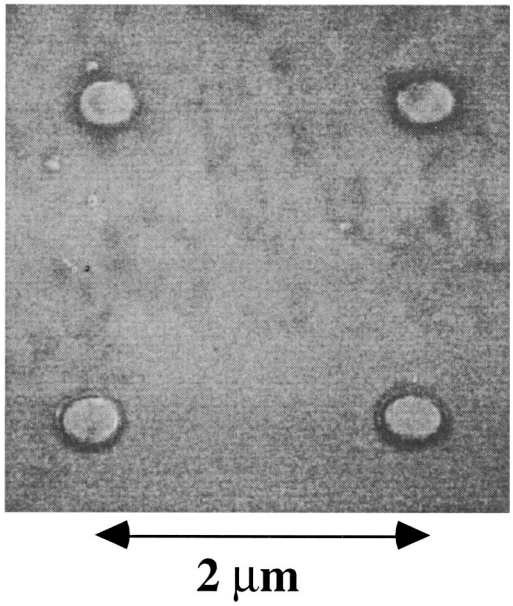


FIG. 1. Electron micrograph showing four Co particles (ellipticity 300 nm×200 nm, thickness 35 nm) belonging to an array of step 2000 nm.

with the noise measured on the single particle through an Arrhenius-like law.

I. DYNAMICAL MAGNETIZATION MEASUREMENTS ON SUBMICRONIC Co PARTICLES

A. Samples

We present measurements made on elliptic Co particles defined by liftoff techniques out of sputtered films (protected from oxidation by a 10 nm thin Si film). X-ray diffraction performed on the Co thin film bevor nanofabrication evidenced a nanocrystalline structure (5–10 nm). The particles have an elliptic contour with in-plane dimensions of 300 nm by 200 nm and a thickness of 30 nm. We controlled the particle's shape by scanning electron microscopy. The first sample fabricated is an array of 2×10^7 identical Co particles. The particles are placed on a Si substrate with a 2 μm spacing (see Fig. 1). Because of this large spacing, dipole interaction between particles are negligible. The second sample is an individual Co particle of the above dimensions which is deposited on a microbridge-dc SQUID. This detector allows the dynamical behavior of the magnetization reversal of the particle to be studied.

B. Hysteresis loops: Single particle and array

We studied the quasistatic magnetization reversal of the samples by measuring the hysteresis loops. Figure 2(a) shows the hysteresis loop of the array of 2×10^7 Co particles measured by a commercial SQUID magnetometer. The hysteresis loop of an individual Co particle of the same dimension is presented in Fig. 2(b). These hysteresis loops are both characterized by two magnetization jumps. Starting from saturation, the first jump can be associated to domain-wall nucleation and the second jump to domain-wall annihilation. The reversible central region describes the displacement of

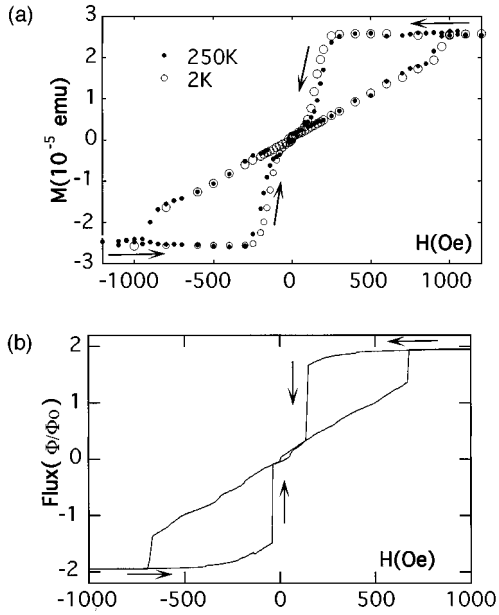


FIG. 2. (a) Hysteresis cycles of the array of about 2×10^7 Co particles (300 nm×200 nm×30 nm) measured at 2 K (○) and 250 K (●). The in-plane field is applied along the long axis of the particles. (b) Hysteresis loop of a single Co particle of same dimension.

the domain wall through the particle.[2] Figure 2(a) also compares the hysteresis loops of the array of particles at 2 K (open dots) and 250 K (black dots). We see a weak influence of the temperature on the hysteresis loops. The hysteresis loops plotted in Fig. 2(a) are an average of about 2×10^7 loops of individual particles, which a sample is plotted in Fig. 1. The jumps observed on the individual particle are events at a certain field changing the magnetization in a time interval smaller than 100 μs (our time resolution). Since particles have different mean switching fields, the two jumps in the hysteresis loop of the array are smoothed [in an interval of about 150 Oe as shown in Fig. 2(a)]. The slope dM/dH accounts for the distribution of the switching fields of each particle in the array. In the following we limit the studies on the domain-wall nucleation.

C. Relaxation measurements of the array of Co particles

The measurements on the array of Co particles were performed using a commercial Metronique Instruments SQUID magnetometer. The resolution of this magnetometer is better than 10^{-7} emu. Over the extraction length (42 nm) the field homogeneity has been found to be better than 10^{-3}. At low temperature, an accurate regulation, within an accuracy of 5 mK, is achieved over a long time interval.

In order to start from an equilibrium state a magnetic field of 1.5 kOe was applied. After 15 min, the field was changed within 1 to 2 min to the final desired constant relaxation field, comprised in the interval 40–250 Oe. The time variation of the magnetization $M(t)$ was then measured during 3 h. This variation is about logarithmic as shown in Fig. 3. The rate of relaxation is given by the magnetic viscosity $S=dM(t)/d(\ln(t))$ which can be defined over about two decades.

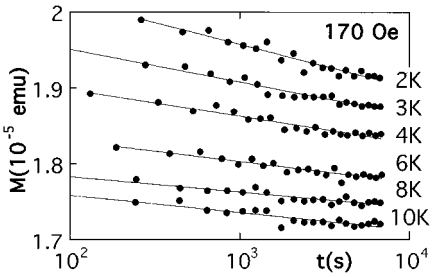
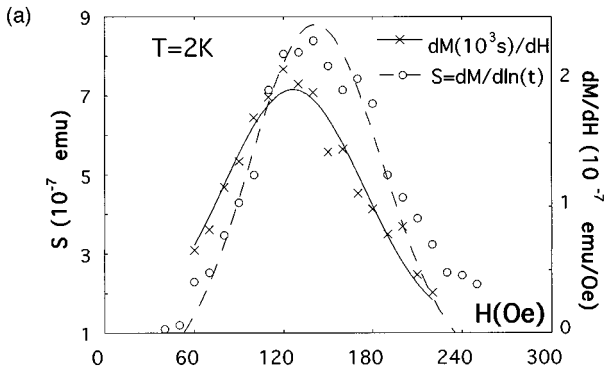


FIG. 3. Relaxation of magnetization of the array of Co particles at $H=170$ Oe. The slope $S=dM/d(\ln(t))$ defines the magnetic viscosity.

Special attention has been given to the field dependence of the magnetic viscosity $S(H)$, which is compared to the switching field distribution in the array, given by the slope $dM(t)/dH$. In Fig. 4(a), $dM(t)/dH$ (crosses) and $S(H)$ (open dots) vs applied field H are plotted on different scale at $T=2$ K. The magnetization $M(t)$ was measured during the relaxation at $t=1000$ s. The distribution of mean switching



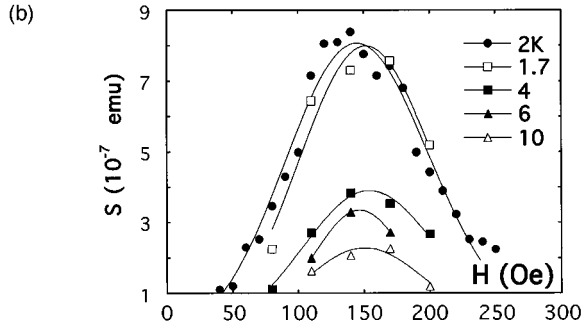


FIG. 4. (a) Field dependence of the slope dM/dH of the magnetization (crosses) plotted with the field dependence of the magnetic viscosity $S(H)=dM/d(\ln(t))$ (open dots). M is taken at time $t=10^3$ s during the relaxation. The slope dM/dH is fitted by a Gaussian $G_{Hc,\sigma}(H)$ and the dashed line is the function $S(H)=aHG_{Hc,\sigma}(H)$, where a is the unique fitting parameter. (b) Field dependence of the magnetic viscosity $S(H)$ for several temperatures, fitted by $HG_{\langle H\rangle,\Sigma}(H)$ functions.

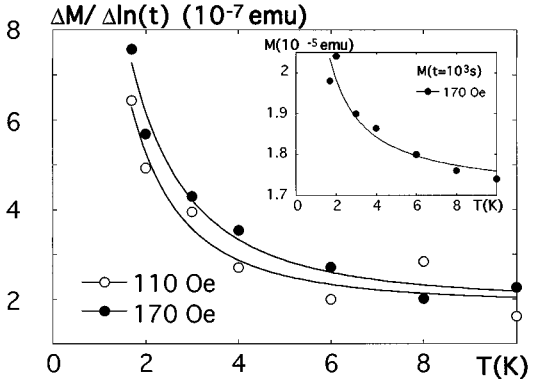


FIG. 5. Temperature dependence of the magnetic viscosity $S=dM/d(\ln(t))$ at 110 and 170 Oe. The data are fitted by a $\coth(T_0/T)T$ dependence ($T_0\approx 0.6$). (Inset) Temperature dependence of the magnetization M at $t=10^3$ s.

fields $\langle H_{sw}\rangle$, deduced from the slope $dM(t)/dH$, is fitted by a Gaussian function $G_{Hc,\sigma}(H)$, where the mean value $H_c=126$ Oe (± 3 Oe) defined the coercive field of the array and σ is the variance. The magnetic viscosity is fitted by the same Gaussian function multiplied by the applied field, $S(H)=aHG_{Hc,\sigma}(H)$, where a is the unique fitting parameter.

Figure 4(b) shows $S(H)$ for different temperatures. The maximum of the magnetic viscosity $S(H)$ versus applied field is about 150 Oe with about 5 Oe variation in the temperature interval of 1.7 to 10 K. This temperature dependence is negligible compared with the width of the curves. The temperature dependence of the magnetic viscosity S at fixed field is shown in Fig. 5. The magnetic viscosity $S(T)$ decreases as temperature increases. The relaxation above 10 K is negligibly small. Furthermore the thermal dependence of the magnetization at a given time follows also a similar variation (see inset of Fig. 5). It is to notice that in common magnetic relaxation measurements, the magnetic viscosity is about proportional to the temperature.[3]

D. Relaxation measurements of an individual Co particle

In order to understand the relaxation of the array of Co particles, we have also studied the dynamical magnetization reversal of a single particle. We use two independent techniques to access to the dynamical nucleation properties. We will call the first approach ''switching field'' measurements[2] and the second ''switching time'' measurements. In the case of the switching field measurements, the applied field is decreased at a given rate and fixed temperature and the field value is stored as soon as the sample magnetization switches, i.e., when the domain wall nucleates. After about 100 cycles, a switching field histogram is established, allowing to define a mean switching field $\langle H_{sw}\rangle$ and its widths.[2] We focus here on switching time measurements. In these experiments, we decrease at a given temperature the magnetic field until the set point is reached (near the nucleation of the domain wall). Then we measure the time it takes until the domain wall nucleates. This process is repeated again about 100 times and

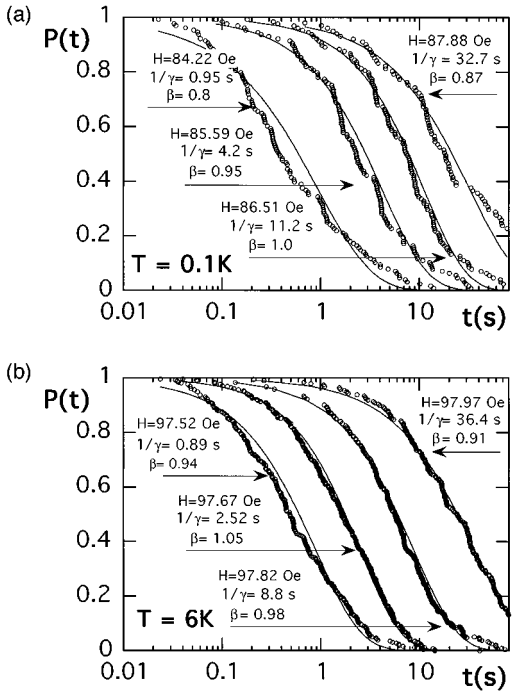


FIG. 6. Probability of not switching of magnetization as a function of the time at different applied fields at (b) 6 K and (a) 0.1 K. Full lines are fits to the data with a stretched exponential as given by formula (1). The fitting parameters β and $1/\gamma$ are indicated in the graphs.

we obtain a switching time histogram of the nucleation process. The integral of this histogram gives the probability of not switching.

Figure 6 shows examples of the measured probability of not switching in the temperature range between 0.1 and 6 K. A good and simple fit of the data is given by a stretched exponential.

$$P(t)=e^{-(\gamma t)^{\beta}}. \tag{1}$$

This fit enables the mean switching time $\langle\tau\rangle=1/\gamma c(\beta)$, with $c(\beta)=\Gamma(2/\beta)/\Gamma(1/\beta)$ expressed in terms of the Γ function, to be deduced. The measurements show that β and γ are temperature and field dependent. The fluctuations around the mean value of the switching time are of the order of $1/(\gamma\beta)$. Note that these fluctuations extend over three decades. Figure 7 shows that β is close to 1 (exponential relaxation) at all temperature so that the factor $c(\beta)$ is also about 1 and $\langle\tau\rangle \approx 1/\gamma$. The variations of γ is fitted by $\ln[\gamma(T,H)]=\ln[a_{(T,0)}]+b_{(T)}H$ in Fig. 8. At fixed temperature, a field variation of 5 Oe covers more than 3 decades of the mean switching time. For a variation of the external field of 15 Oe, the mean switching time variation covers more than three decades for all temperatures.

II. STATISTICAL DESCRIPTION OF THE RELAXATION

In the following, we give a statistical description of the aftereffect during a nucleation process of the array of particles by taking into account the dynamical characteristics of the magnetization reversal of a single particle.

The studied processes are nonequilibrium and dissipative.

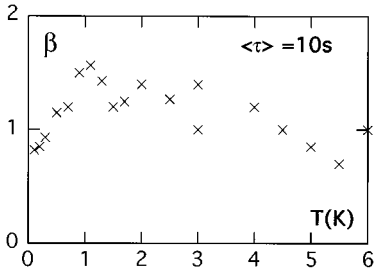


FIG. 7. Temperature dependence of the stretching components β (for $1/\gamma=10$ s) as used for fitting the probabilities of not switching [Eq. (1)].

The small size of the magnetic particles of the array suggests that the slow dynamics of the array are dominated by single-particle fluctuations. Indeed, the time scale of the aftereffect experiments on the array is of the same order than the time fluctuations around the mean value $\langle\tau\rangle$ shown in Fig. 6 for a single particle (few decades). A simple way to deal with fluctuations in nonequilibrium systems is linear-response theory. Using this formalism, we are able to make a link between the time-dependent statistical averaging of the magnetization $\langle m\rangle(t)$ with the temperature-dependent noise $D(T)$ responsible for the relaxation. We can then study how this parameter emerges from the aftereffect measurements after averaging over the ensemble of particles in the array. The consistency of this description with the more usual description in terms of activated process[3] is checked by comparing the noise *D(T)* measured on the array with the noise measured on the single particle through an Arrhenius-like law for the switching time.

A. The linear-response theory applied to an ensemble of identical particles

In the following, the mean features of the aftereffect protocol are described in terms of linear-response theory applied to an ideal array of identical particles, i.e., defined by a fixed parameter γ (since β is close to 1, we suppose in the following that it does not play an important role). This ensemble is ergodic: fluctuations measured of this ideal ensemble are identical to the fluctuations measured in time on a single particle. Before the initial time $t=0$, the magnetization of the statistical ensemble of particles is saturated by a high mag-

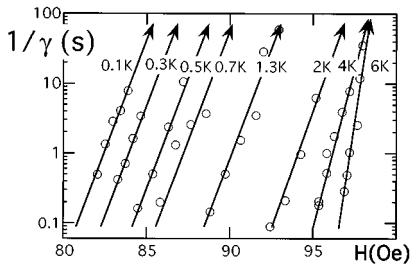


FIG. 8. Field dependence of the inverse of the damping coefficient $1/\gamma$ at different temperatures. Arrows indicate the variation of $1/\gamma$ for applied field *H*.

netic field: the magnetization of the ideal array is in equilibrium and can be described by a stationary Hamiltonian. Then, the applied field is reduced to the measuring field H. During this process the Hamiltonian of the system is modified by a time-dependent perturbative quantity $E_p = mH_p(t)$, where m is the magnetization of the ideal array. If we call m_0 the magnetization discontinuity due to one domain-wall nucleation in the hysteresis loop [Fig. 2(b)], the magnetization of the ideal array can for instance be written $m(t)=N(t)m_0$, where $N(t)$ is the number of particles which contribute to the relaxation process at time t. The field H_p accounts for the energy dependence between the state corresponding to a high positive field and the state corresponding to the field H of unstable states, close to the switching field H_{sw}. The energy E_p is proportional to the external field H (see, e.g., the Stoner-Wolfarth model[4]). We suppose that the applied field is set instantaneously with respect to the time scale of the experiment so that at the time $t=0$, particles are suddenly submitted to the perturbation field. The perturbation field can then be written: $H_p(t)=H_p\Theta(t)$ where $\Theta(t)$ is the step function at time $t=0$. At very long times ($t\rightarrow\infty$), the magnetization reaches a second equilibrium value, which corresponds to the state where all particles of the ideal array are in the reversible part after the nucleation jump in the hysteresis loop shown in Fig. 2(b). The dynamics of the number of switched particles can be described by the response to the perturbation field H_p:

$$\langle \delta m\rangle(t)=\langle m\rangle(t\rightarrow\infty)-\langle m\rangle(t). \tag{2}$$

In the linear-response approximation, the response to a step function is given by the following Kubo formula:[5]

$$\langle \delta m\rangle(t)=\frac{H_p}{kT}\langle mm(t)\rangle \tag{3}$$

where $\langle mm(t)\rangle$ is the time autocorrelation function at equilibrium (in the quantum regime this correlation function is the Kubo-Mori product). The empirical formula (1) gives the time dependence of the process (the distribution of probability of the switching times obeys the same Fokker-Planck equation as the distribution of probability of the variable, see, e.g., Ref. 6). The magnetization (3) of the ideal array can hence be written as follows:

$$\langle \delta m\rangle(t)=\frac{H_p}{kT}\langle m^2\rangle e^{-(\gamma t)^\beta}, \tag{4}$$

where the damping coefficient γ is close to the inverse of the mean switching time $\langle\tau\rangle$. The application of the fluctuation-dissipation theorem allows us to link the magnetic fluctuations $\langle m^2\rangle$ with the noise $D(T)$ through the damping constant γ. Assuming that the process can be described by a white noise, we have[5]

$$\gamma=\frac{\eta D(T)}{\langle m^2\rangle}, \tag{5}$$

where η is the temperature-independent friction coefficient, defined in order to have a noise $D(T)$ in energy units [in the case of Brownian motion $D(T)=kT$]. Therefore for a fixed field, the response (4) of the ideal array of particles becomes

$$\langle \delta m\rangle(t)=\eta H_p\frac{D(T)}{kT}\frac{e^{-(\gamma t)^\beta}}{\gamma}. \tag{6}$$

B. Magnetization of the array of particles

The magnetization decay of the real array of particles $M(t)$ accounts for the contribution of all particles switching during the aftereffect measurement: $M(t)$ is then given by the sum of those particles having switching times compatible with the experimental time scale. In order to compare the measurements performed on one single particle to those performed on the array of particles, we first have to define in the array, the number of different ideal arrays of particles of identical switching time distribution. An ideal array is now defined as the set of particles with a damping coefficient γ contained in the interval $[\gamma,\gamma+d\gamma]$. If $f_\gamma d\gamma$ is the number of such ideal arrays in the sample, $f_\gamma d\gamma\langle m\rangle(t)$ is the time-dependent magnetization of this subsystem. The magnetization $M(t)$ is then given by the summation over all ideal arrays weighted by f_γ.

The question arises as to how the distribution f_γ of damping constants in the array of particles can be deduced. The variation of the mean switching time with the applied field H, for a single particle is shown in Fig. 8. Three decades of the mean switching time are covered by a field variation ΔH of about 15 Oe for all temperatures. In the macroscopic array of particles, the distribution of γ is due to the distribution of mean switching field $\langle H_{\mathrm{sw}}\rangle$, given in Fig. 4. A variation of 15 Oe is negligible with respect to the width of the distribution in the vicinity of H_c. Then, the distribution of switching fields can be taken as uniform, and the distribution can be approximated by a uniform distribution $\bar{f}$ over the frequency interval $\Delta\gamma$. The summation writes

$$\langle M\rangle(t)=H_p\eta\frac{D(T)}{kT}\bar{f}\int_{\Delta\gamma}\frac{e^{-(\gamma t)^\beta}}{\gamma}d\gamma. \tag{7}$$

The noise $D(T)$ has been taken to be independent of γ [in Fig. 8 a field-independent noise can indeed be extracted also through the thermal dependence of the slope of the arrows in Fig. 8 (Ref. 2)].

Setting $U=(\gamma t)^\beta$ in the integral, the limits of integration become $\Delta U=t(\gamma_{\max}-\gamma_{\min})^\beta$. The upper integration limit $\gamma_{\max}$ is of the order of the inverse of the second and t is in hours, the contribution to the integral from $t\gamma_{\max}$ to infinity is negligible so that (7) can be written

$$\langle M\rangle(t)=H_p\eta\frac{D(T)}{kT}\bar{f}\frac{1}{\beta}\int_{U_{\min}}^{+\infty}\frac{e^{-U}}{U}dU. \tag{8}$$

The last integral can be rewritten using an expansion in logarithm[7]

$$-\int_{U_{\min}}^{+\infty}\frac{e^{-U}}{U}dU=\ln(U_{\min})+\varepsilon(t)+\mathrm{Eu}. \tag{9}$$

Eu is Euler's constant and $\varepsilon(t)$ tends to zero when $U_{\min}$ tends to zero.

$U_{\min}=(t\gamma_{\min})^{\beta}$ where t is the time of the observation. The difference $2(1/\gamma_{\min}-t)$ accounts for the fluctuations and is more than a decade (see Fig. 6) so that the magnetization $M(t)$ rewrites

$$\langle M\rangle(t)\approx -H_p\eta\frac{D(T)}{kT}\bar{f}\left[\ln(t\gamma_{\min})+\frac{\mathrm{Eu}}{\beta}\right]. \tag{10}$$

The magnetic viscosity $S=|d\langle M\rangle/d(\ln(t))|$ can be written

$$S(T)\approx H_p\eta\frac{D(T)}{kT}\bar{f}. \tag{11}$$

As long as $\gamma_{\min}$ is not temperature dependent, $\langle M\rangle(t)$ and $S(T)$ should have a similar thermal dependence. Experimentally, it is observed that the magnetization $M(t=10^3$ sec) (inset of Fig. 5) measured during the relaxation has the same temperature dependence as the magnetic viscosity $S(T)$ (Fig. 5), which suggests that $\gamma_{\min}$ is indeed temperature independent.

The validity of the linear-response hypothesis can be checked by using our knowledge of the switching field distribution. Due to the ''local uniformity'' of the distribution $\bar{f}$, the field dependence of the slope $\Delta\langle M\rangle/\Delta H$ in the vicinity of the applied field H_0 is proportional to the distribution

$$\left.\frac{\Delta\langle M\rangle(t)}{\Delta H}\right|_{H=H_0}\propto\bar{f}(H_0), \tag{12}$$

the assumption of the linear-response hypothesis ''H_p proportional to the applied field H_0'' leads to Eq. (11). Inserting Eq. (12), we then have

$$S(H_0)\propto H_0\left.\frac{\Delta\langle M\rangle(t)}{\Delta H}\right|_{H=H_0}, \tag{13}$$

in agreement with the fit of Fig. 4(a).

C. Consistency of the model with the experiments on the single particle

The noise $D(T)$ can be measured independently of the above model on a single particle and by using the usual formalism of activated process which describes the thermal dependence of the relaxation. If we suppose that for each temperature and at fixed applied field H, the same magnetic object relaxes over an energy barrier E, the thermal dependence of the activation process is given by the mean switching time. The Kramers formula gives[6]

$$\langle\tau\rangle(T)=\langle\tau\rangle_H^0\exp\left(\frac{E}{D(T)}\right), \tag{14}$$

where the prefactor $\langle\tau\rangle_H^0$ is the switching time in the limit where $E/D(T)$ tends to zero. The diffusion coefficient $D(T)$ has been identified with the noise defined in (5) through the fluctuation-dissipation theorem.[8] The measured temperature dependence of $\ln(1/\gamma)=\ln(1/\gamma^0)+E/D(T)$ has been plotted in Fig. 9 where some points have been extrapolated from the arrows of Fig. 8 at fixed field H. At each external field, experimental data are fitted by $\tanh(T_0/T)$ curve. The low-temperature deviation from the usual white noise $D_{cl}(T)=k_BT$ at low temperature has already been measured in various magnetic nanostructures.[9,10]

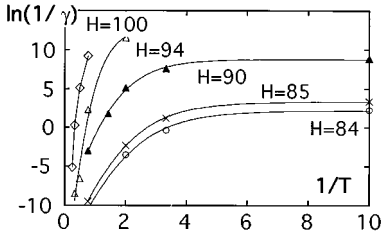


FIG. 9. Arrhenius plot of the system for various values of the external fields. The logarithm of the mean switching time is plotted versus the inverse of the temperature in order to evidence the shift from the noise $D(T)=k_BT$ occurring at low temperature. Some of the points are obtained from extrapolations of the arrows of Fig. 8. All the curves are fitted by a $\tanh(T_0/T)$ function where T_0 is varying from 0.4 to 2.

Note that in the usual approach in terms of activation processes,[3] the relaxation function (1) with $1/\gamma$ given by (14) is integrated over a distribution of energy barriers E. This procedure leads to a result formally identical to the integral (8),

$$\langle M\rangle(t)\approx M(0)D(T)\bar{f}_E\ln(t\gamma_{\min}), \tag{15}$$

with the exception of the thermal dependence contained in the prefactor $M(0)D(T)$ which cannot be described in this approach without more statistical arguments about the initial state.

Equation (11) predicts a thermal dependence of the viscosity $S(T)$ in the form $D(T)/T$. We have plotted in Fig. 10 the thermal dependence $D(T)/T$ deduced from the fits of Fig. 9 (curve at $H=100$ Oe). In the left scale, data of the magnetic viscosity taken at $H=110$ Oe has been plotted. Both data points are fitted by $1/T\coth(T_0/T)$ curves: both thermal variations are qualitatively in agreement. The temperature T_0 is of the order of 1 K.

CONCLUSION

Relaxation measurements on magnetic Co particles at low temperature have been performed at two different scales

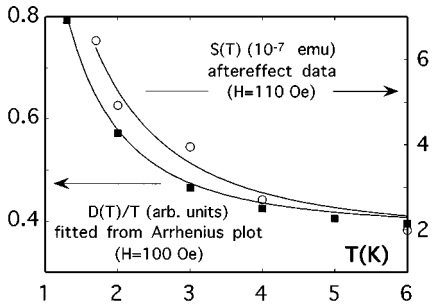


FIG. 10. The thermal dependence of $D(T)/T$, deduced from the fit of Fig. 9 (curve at $H=100$ Oe), is plotted (left scale) together with the data of the magnetic viscosity S taken at $H=110$ Oe and $H=170$ Oe (right scale). Both sets of data points are fitted to $\coth(T_0/T)/T$ where T_0 is varying from 1 to 3.

(single nanoparticle and array of independent particles). The comparison of the experimental results show that two levels of statistic distributions must be taken into account. The switching process of the magnetization measured on a single particle is stochastic and accounts for the intrinsic fluctuations. The time dependence of the relaxation process, measured statistically, shows that the mean switching time follows an Arrhenius-like law with an effective noise $D(T)$ of the form $\coth(T_0/T)$. The measurements performed on the array of the noninteracting particles confirm the existence of a distribution of mean values (mean switching time, mean switching field) of the individual particles. The magnetization of the array has been described by applying the linear-response theory, and using our knowledge of the statistical parameters of the sample. This model predicts a $D(T)/T$ thermal dependence of the array's magnetic viscosity $S = dM/d(\ln(t))$, which has been observed experimentally. In a previous work[9] two types of responses of magnetic systems have been described; the response to an external excitation field, which is relevant here, and the response to a thermal excitation. It seems that this last situation cannot be observed in the present nanostructured sample, because the coupling to the heat bath is performed at a nanoscopic scale. However, if the sample were thermally excited at time $t=0$, the linear-response formalism would lead to an equation similar to (11) where the prefactor H_p/kT is replaced by a Lagrange multiplier λ, which does not usually depend on the temperature. The thermal dependence of the viscosity may then be identical to the thermal dependence of the noise $D(T)$, and may lead to a $\coth(T_0/T)$ profile (proportional to T when $T \gg T_0$). It seems that this last situation has been observed by aftereffect measurements in various systems and interpreted in the framework of the theory of the macroscopic quantum tunneling of the magnetization.[11,12]

Although the physical interpretation of the noise $D(T) \propto \coth(T_0/T)$ has not been completely clarified in the present work, especially the quantum nature of $D(T)$,[9] our results show that the magnetic aftereffect process at low-temperature reduces essentially to dissipative phenomena. These phenomena, related to the investigation field of quantum stochastic and quantum noise, have to be better understood before concluding about macroscopic quantum tunneling of the magnetization.[13,14]

ACKNOWLEDGMENTS

We would like to thank J. M. Rubí, J. L. Porteseil, and O. McGrath for their interest and useful suggestions.

*Electronic address: wegrowe@labs.polycnrs-gre.fr

[1] N. G. Van Kampen, Adv. Chem. Phys. **34**, 245 (1976).

[2] W. Wernsdorfer, K. Hasselbach, A. Sulpice, A. Benoit, J-E. Wegrowe, L. Thomas, B. Barbara, and D. Mailly, Phys. Rev. B **53**, 3341 (1996).

[3] R. Street and J. C. Wooley, Proc. Phys. Soc. London Ser. A **62**, 562 (1949).

[4] E. C. Stoner and E. P. Wohlfarth, Philos. Trans. R. Soc. London Ser. A **240**, 599 (1948).

[5] E. Fick and G. Sauermann, in *The Quantum Statistics of Dynamic Processes*, edited by Peter Fulde, Springer Series in Solid-State Sciences, Vol. 86 (Springer-Verlag, Berlin, 1990), p. 116.

[6] K. Matsuo, K. Lindenberg, and K. E. Shuler, J. Stat. Phys. **19**, 65 (1978).

[7] M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1972), p. 229.

[8] Equations (5) and (14) are not in contradiction since $\langle m^2 \rangle$ in (5) is defined in the framework of the linear-response approximation. A demonstration of the Arrhenius law (14) using projection operator formalism can be found in W. Weidlich and H. Grabert, Z. Phys. B **36**, 283 (1980), and it can be shown that the linear approximation of the stochastic equation obtained by this last formalism is equivalent to the Mori equation from which Eq. (5) is derived (see Ref. 5).

[9] J-E. Wegrowe, R. Ballou, B. Barbara, V. Amaral, A. Sulpice, and D. Mailly, Phys. Rev. B **52**, 3466 (1995).

[10] W. Wernsdorfer, K. Hasselbach, D. Mailly, B. Barbara, A. Benoit, L. Thomas, and S. Suran, J. Magn. Magn. Mater. **145**, 33 (1995).

[11] See, e.g., the Symposium of the Pittsburgh MMM Conference on Macroscopic Quantum Tunneling [J. Appl. Phys. **73**, 6697 (1993)], and references therein.

[12] M. Stamp, E. Chudnovsky, and B. Barbara, Int. J. Mod. Phys. B **6**, 1355 (1992)

[13] E. G. Harris, Phys. Rev. A **49**, 629 (1994), and references therein.

[14] P. Politi, A. Rettori, F. Hartmann-Boutron, and J. Villain, Phys. Rev. Lett. **75**, 537 (1995).