Magnetic aftereffect experiments at low temperature: Linear response and quantum noise

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An alternative approach to slow magnetic relaxation at low temperature is proposed in terms of a linear response theory involving collective variables characteristic of local metastable states. The model leads to the definition of two types of magnetic viscosity: S_0 due to the response to an excitation field and S_1 due to the response to an initial thermal perturbation. The thermal behavior of the magnetic viscosity is found to be $S_0 \propto K(T)/k_B T$ in the first case, and $S_1 \propto K(T)$ in the second case, where $K(T)$ is the thermal noise. Experimental results, interpreted as the manifestation of the magnetic viscosity S_0, show some evidence of a noise $K(T)$, which can be interpreted as a quantum form of the noise.

I. INTRODUCTION

Magnetic aftereffect measurements consist of measuring the time variation of the magnetization of a sample after a sudden change of the external magnetic field. The thermal dependence of this variation is usually expressed in terms of the magnetic viscosity $S(T)=dM/d[\ln(t)]$ which is the derivative of the magnetization with respect to the logarithm of the time.

Considerable interest in magnetic aftereffect measurements arose since deviations from the classical model of thermally-activated relaxation have been seen at low temperature.[1] These deviations are often interpreted as the manifestation of quantum tunneling of the magnetization. Indeed, the classical Kramers's stochastic law[2] which gives the expression of the temperature-dependent decay rate $\Gamma(T)$ of the activation process can be extended to very low temperatures where quantum tunneling prevails.[3] A description for all temperatures is then based on the assumption that the relaxation is entirely described by the same mechanism of hopping processes, i.e., is described by the single parameter $\Gamma(T)$ which gives the characteristic frequency of the transition from the side A to the other side B of each barrier potential of height E composing the sample. The definition of the elementary magnetic object which transits from A to B needs not to be explicitly defined at this observation scale because of the universality of the Kramers law $\Gamma(T)=\Gamma_0\exp(-E/k_B T)$. At the scale considered here, space and time have already been coarse grained and the microscopic degrees of freedom have already been reduced to the damping parameter contained in the attempt frequency $\Gamma_0=\kappa\omega_0$. κ stands for the correction due to the coupling with the heat bath and ω_0 is the frequency of small nondamped oscillation about the minimum of the potential well.

A basic limitation of the above description lies on the fact that the nature and the space distribution of the barriers inside the sample will obviously depend on the definition of the elementary magnetic object. Namely, the distribution of the height of the barriers cannot be defined independently. It has been shown[4] that in a system of small magnetic particles, the high-temperature dynamics is determined by the particle size distribution, whereas at low temperature, this is no longer the case. The micromagnetic structure plays a crucial role in the dynamics. Here we argue that thermal macroscopic characteristics of magnetic aftereffect brought about by the cooperation of many subsystems cannot be described without ambiguities using Kramers law as long as a microscopic study of the set of the underlying magnetic configurations is not performed. This problem is still poorly understood. Hence we will focus our attention on a more microscopic scale where dissipative effects are handled in terms of more precise statistical analysis.

A question arises: is it possible to describe the magnetic aftereffect at low temperature without the need to invoke barrier hopping processes? An answer to this question was given by Bray and Moore.[5] The authors derived the logarithmic time dependence and the linearity vs temperature of the classical correlation function for a set of coupled spins which stay in the vicinity of the bottom of the well. They assumed that, within the observation time scale, local metastable states can be taken as stable states, so that a thermal equilibrium can be defined. The measured relaxation can be then taken as the approach of collective variables to this equilibrium. In a previous work,[6] we have discussed the Brownian motion of these slow

variables in the quantum case using Mori-Zwanzig theory in order to study the thermal dependence of the magnetic viscosity in terms of linear response.

The aim of this paper is to show that linear response theory of magnetic systems provides an essential framework for a more comprehensive understanding of magnetic aftereffect measurements and allows access to the thermal dependence of the noise at low temperature. We start with the Langevin equation describing the magnetic system and show how it leads to the emergence of the slow variables under interest. Applying the linear response theory to these relevant slow variables, we define two different contributions to the magnetic viscosity, both depending on the thermal noise but in different ways. In Sec. III we summarize our experimental results on magnetic multilayers and on an array of etched magnetic microparticles where unusual behaviors were observed. Finally we discuss how linear response theory is helpful in interpreting experimental results, shedding light on magnetic aftereffect phenomena.

II. MODEL

As experiments were performed on disordered magnetic materials (amorphous), the simplest way to describe a macroscopic spin system is to start with the following Hamiltonian:

$$H=-\frac{1}{2}\sum_{ij}J_{ij}\mathbf{S}_i\mathbf{S}_j \ , \tag{1}$$

where J_{ij} has a given distribution with respect to the sites i and j of the spin.[5]

We assume that the spins are frozen in different potential minima with local mean values $\mathbf{S}_i^0$ and deviations $\delta\mathbf{S}_i$:

$$\mathbf{S}_i=\mathbf{S}_i^0+\delta\mathbf{S}_i \ . \tag{2}$$

Writing $\delta\mathbf{S}_i=\sum_\alpha \delta S_i^\alpha \mathbf{e}_\alpha(i)$, the linearized Langevin equation corresponding to the Hamiltonian (1) writes

$$\frac{d\delta S_i^\alpha}{dt}=-\sum_{j,\beta}A_{ij}^{\alpha\beta}\delta S_j^\beta+f_i^\alpha \ , \tag{3}$$

where

$$A_{ij}^{\alpha\beta}=\frac{\partial^2 H}{(\partial\delta S_i^\alpha)(\partial\delta S_j^\beta)} \tag{4}$$

and f_i^α is the Langevin stochastic force, defined by

$$\langle f_i^\alpha\rangle=0, \quad \langle f_i^\alpha(t+\tau),f_j^\beta(t)\rangle=2K(T)\delta_{ij}\delta^{\alpha\beta}\delta(\tau) \ , \tag{5}$$

where $K(T)$ is the noise.

The description of independent particles moving in a viscous environment can then be obtained by diagonalizing the matrix $[A]$. If γ is an eigenvalue of $[A]$, Eq. (3) can be written as a set of uncoupled equations of the type

$$\frac{d\delta S\gamma}{dt}=-\gamma\delta S_\gamma+f_\gamma \ . \tag{6}$$

In other words, each normal mode defines a magnetic Brownian particle with a damping coefficient γ. All the topological complexity of the sample is then contained in the spectrum of the eigenvalues γ.

We have still[5]

$$\langle f_\gamma\rangle=0, \quad \langle f_\gamma(t+\tau),f_{\gamma'}(t)\rangle=2K(T)\delta_{\gamma\gamma'}\delta(\tau) \ . \tag{7}$$

The autocorrelation function is then well known:

$$\langle\delta S_\gamma,\delta S_\gamma(t)\rangle=\frac{K(T)}{\gamma}e^{-\gamma t} \ . \tag{8}$$

In order to describe the collective behavior for the sample we perform an average over all possible modes γ. Assuming that the noise is identical for all modes and taking a wide uniform distribution of damping coefficients $g(\overline{\gamma})$ from $\gamma_{\min}$ to a cutoff frequency $\gamma_{\max}$, we obtain a logarithmic time dependence of the autocorrelation function:[5,7]

$$\langle\delta S,\delta S(t)\rangle\approx K(T)g(\overline{\gamma})\ln(\gamma_{\min}t) \ . \tag{9}$$

The application of the Kubo equation for the linear response to a relaxation field h, constant in time, and switched on at time $t=0$, leads to the expression

$$\langle\delta S\rangle(t)=-\frac{h}{k_BT}\langle\delta S,\delta S(t)\rangle \ . \tag{10}$$

In the quantum case, the correlation function is the Kubo-Mori scalar product.[6]

We define $S_0=-d\langle\delta S\rangle/d[\ln(t)]$ as the magnetic viscosity due solely to the response to a local field. From Eq. (10), it takes the simple form

$$S_0=h\frac{K(T)}{k_BT}g(\overline{\gamma}) \ . \tag{11}$$

On the other hand, the linear response to a thermal excitation can be expressed by[6]

$$\langle\delta S\rangle(t)=-\lambda\langle\delta S,\delta S(t)\rangle \ . \tag{12}$$

λ is then the Lagrange multiplier, corresponding to the "local statistical equilibrium" described by the distribution function $\exp(H/kT+\lambda\delta S)/Z$.

We then deduce the magnetic viscosity $S_1=-d\langle\delta S\rangle/d[\ln(t)]$ due solely to the response to thermal excitations at initial time. From Eq. (12) it takes the simple form

$$S_1=\lambda K(T)g(\overline{\gamma}) \ . \tag{13}$$

It is important to note that the state at $t=0$ is not accessible experimentally. This is the state which would be reached by the subsystem, after the external field is set equal to the relaxation value, and just before the subsystem begins to see its driving force h.

What remains for us to calculate is the noise $K(T)$ in the quantum (or quasiclassical) limit. According to the classical equipartition theorem, k_BT expresses the average energy per unit bandwidth so that the autocorrelation (8) at initial time is $\langle(\delta S_\gamma)^2\rangle/M=k_BT$. This defines the magnetic "mass" M of our system. The noise $K(T)$ can then be written as [see Eq. (8)] $K(t)=M\gamma k_BT=\eta k_BT$, where η is the friction coefficient which depends only on the nature of the coupling to the heat bath (i.e., on the experimental setup) and not on the parameters describing

the spin system. However, as pointed out in Ref. 6, the equipartition theorem is no longer valid in the general case of a quantum spin system. A crossover is then to be expected between the classical regime and the quantum regime. According to Dyson, Lieb, and Simon,[8] we might have the following inequality for $K(T)$:

$$K(T) \leq C \coth\left[\frac{C}{\eta k_B T}\right], \tag{14}$$

where C has no explicit temperature dependence.

Taking both the low-temperature limit and the high-temperature limit and inserting in (11), we get the viscosity due to the response to a local excitation field:

$$S_0 \leq \begin{cases} hg(\bar{\gamma})\eta & \text{if } k_B T \gg \dfrac{C}{\eta}, \\ hg(\bar{\gamma})\dfrac{C^2}{\eta}\dfrac{1}{k_B T} & \text{if } k_B T \ll \dfrac{C}{\eta}. \end{cases} \tag{15}$$

Using Eq. (13) yields the response to a thermal perturbation

$$S_1 \leq \begin{cases} \lambda g(\bar{\gamma})\eta k_B T & \text{if } k_B T \gg \dfrac{C}{\eta}, \\ \lambda g(\bar{\gamma})\dfrac{C^2}{\eta} & \text{if } k_B T \ll \dfrac{C}{\eta}. \end{cases} \tag{16}$$

We shall now briefly describe some unusual results of magnetic aftereffect measurements performed on three different amorphous magnetic samples which can be interpreted in the framework of the above model. We emphasize here that the linear response theory applies to dynamical processes not far from the equilibrium. Experimentally, we will therefore not consider the dynamics at short times where nonlinearities arise due to big avalanche effects. The full details of the measurements will be reported elsewhere.

III. EXPERIMENTS

We first present results obtained on a multilayer system made of an amorphous film of $SmCo_2/GdCo_2/SmCo_2$ layers where the central $GdCo_2$ (1700 Å) layer is separated in two equal parts by an ultrathin $SmCo_2$ (50 Å) layer [see scheme of Fig. 1(b)]. The sample was prepared by sputtering the constituent elements of the alloy onto a glass substrate and protected by a 1200 Å Ag layer. A high uniaxial anisotropy was induced in the $SmCo_2$ layers by annealing the sample at 300 °C in an in-plane magnetic field of 50 kOe, in order to keep the boundary conditions constant: the magnetization of the two extremes $SmCo_2$ (750 and 1200 Å) layers did not switch during the magnetic aftereffect measurements. Consequently, the system under interest is actually the $GdCo_2$ (850 Å)/$SmCo_2$ (50 Å) multilayer, with the central layer playing the role of a planar defect for a domain wall propagating from one $GdCo_2$ layer to the next one.

(a) The hysteresis loop measured with a vibrating sample magnetometer is shown in Fig. 1. A negative field $H = -70$ kOe was applied before sweeping the field from

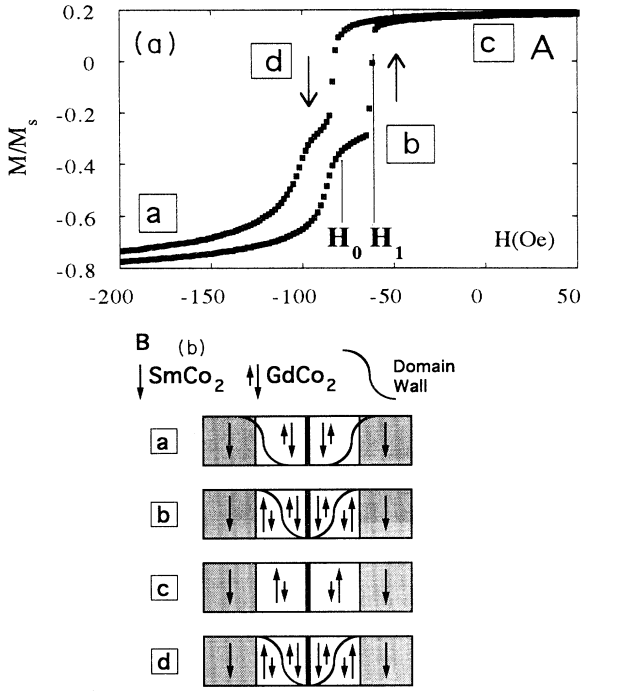

FIG. 1. (a) Hysteresis loop of $\{SmCo_2[/GdCo_2/SmCo_2/GdCo_2/]\ SmCo_2\}$ multilayer sample where the magnetization of both external $SmCo_2$ layers are fixed in the negative direction with respect to the applied field. The configuration *a* is obtained by saturating the sample at −70 kOe. The loop is then measured by sweeping the field from +0.2 to −0.2 kOe. The magnetic aftereffect measurements have been performed by suddenly changing the field from −2 kOe up to H_0, i.e., in the configuration *b*, or up to H_1, i.e., in the configuration close to *c*. (b) Schematic representation of the magnetic configuration in *a*–*d*.

−0.2 kOe (point *a*) to 0.2 kOe (point *c*) and back to −0.2 kOe. The resulting hysteresis loop can easily be understood in terms of the sequence of magnetic layer configurations shown in Fig. 1(b). The relaxation measurements were performed using a commercial superconducting quantum interference device (SQUID) magnetometer from Quantum Design Inc. The resolution was better than 10^{-7} emu and the temperature was regulated with a precision of about 5 mK. In order to start from the same initial magnetic state, the temperature T was first fixed, then the field was swept from 2 to −2 kOe, before being set at the relaxation field. The time needed to sweep the magnetic field from −2 kOe to the relaxation field was 1 kOe/min. The spontaneous magnetization M_s at −2 kOe did not vary, to within experimental accuracy, in the temperature range of the experiments.

Comparative plots of the relaxation are shown in Fig. 2 for the two different values of the applied field and starting from the same initial state. The first field H_0 corresponds to the maximum of the magnetic viscosity vs magnetic field in the vicinity of the configuration *b* of Fig. 1(b), where the 180° domain wall of the Co spins is compressed on the central $SmCo_2$ layer. The second field H_1 corresponds to the maximum of the magnetic viscosi-

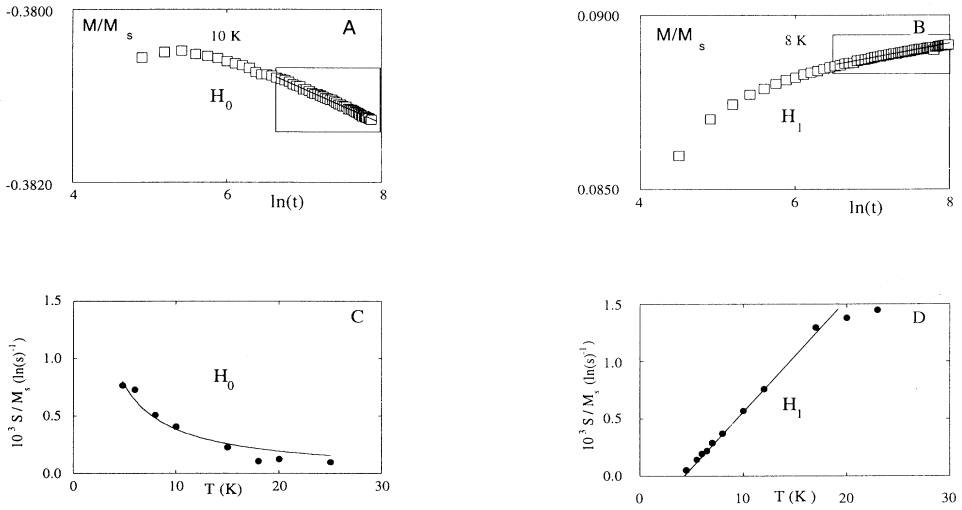

FIG. 2. Time variation (t in seconds) of the relative magnetization M/M_S and thermal variation of the magnetic viscosity S/M_S measured on the $\{SmCo_2[/GdCo_2/SmCo_2/GdCo_2/]SmCo_2\}$ in the field H_0 [graphs (a) and (c)] and H_1 [graphs (b) and (d)]. The continuous line in graph (c) is a fit $S/M_S \propto 1/T$.

ty vs magnetic field close to the configuration c of Fig. 1(b), i.e., during the annihilation process of the 360° domain wall of the Co spins. Both fields H_0 and H_1 are constant with temperature. There was no evidence for a logarithmic time dependence of the magnetization at short times. A logarithmic time dependence takes place after the time $\ln(t)=7$ (about 18 min). We have defined the magnetic viscosity as $S \equiv |M[\ln(t)=8]-M[\ln(t)=7]$ (see Fig. 2). The main unusual feature of the measurements is that the relaxation for H_0 is anomalous with respect to the one expected for metastable states. One observes a variation of the magnetization opposite to that which would converge to the anhysteresis cycle as always observed at high temperature. In this case, the thermal variation of the magnetic viscosity follows a $1/T$ behavior at low temperature. In contrast, the relaxation for H_1 is consistent with that observed usually with respect to the hysteresis loop and the thermal variation of the magnetic viscosity follows the usual linear behavior associated with thermal activation.

Magnetic aftereffect measurements were also performed on a sputtered $SmCo_2$ (600 Å)/Mo (200 Å)/$SmCo_2$ (600 Å) multilayer system. In this sample, the initial state was that obtained by setting the magnetic field at 30 kOe after having set the temperature at the relaxation value (within the range of 1.8–50 K). The relaxation measurements were performed with the SQUID magnetometer, and with the same protocol as above. The relaxation field was fixed to 0.6 kOe at all temperatures. The time variation of the magnetization is logarithmic and is consistent with that observed usually with respect to the hysteresis loop. However, the magnetic viscosity, measured at a fixed relaxation field, follows a $1/T$ variation at low temperature (Fig. 3).

Our third sample was a macroscopic array of ellipses with main axes of 1 and 0.8 μm of size, separated from each other by a distance of 1 μm. The sample was prepared by ion beam etching of a sputtered amorphous $Co_{81}Zr_9Mo_8Ni_2$ thin film of 50 nm of thickness. Various magnetic measurements on different samples showed that the ellipses were magnetically independent from each other.[9]

Thorough relaxation measurements were performed using a commercial SQUID magnetometer from Metronique Instruments. The resolution of this magnetometer is better than 10^{-7} emu. Over the extraction length ($\approx$42 mm) the field homogeneity has been found to be better than 10^{-3}. At low temperature, an accurate regulation, within a precision of 5 mK, is achieved over a long time interval. In order to start from the same initial magnetic state, the sample was first heated at 50 K, then a magnetic field of 0.6 kOe was applied. Fifteen minutes later, the sample was slowly cooled in this field down to the desired temperature. After 15 more minutes, the field was changed within 1 to 2 minutes to the final desired

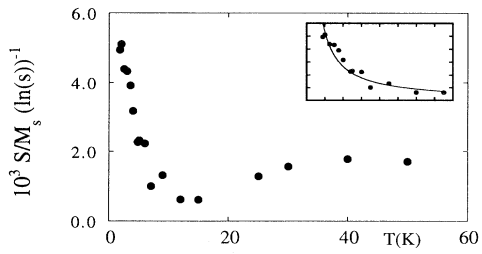

FIG. 3. Thermal variation of the magnetic viscosity of the $SmCo_2/Mo/SmCo_2$ multilayer. Inset: fit with $S/M_S \propto 1/T$ at low temperatures.

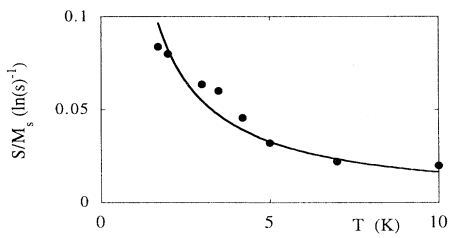

FIG. 4. Thermal variation of the magnetic viscosity S/M_S of the array of $Co_{81}Zr_9Mo_8Ni_2$ ellipses. The continuous line is a fit $S/M_S \propto 1/T$.

value H_c. H_c is the coercive field defined as that corresponding to the maximum of viscosity vs applied field at fixed temperature. In contrast to the above samples, it shows a temperature dependence. The time variation of the magnetization was then measured during 4 h. It is logarithmic and consistent with that observed usually with respect to the hysteresis loop, but the magnetic viscosity follows a $1/T$ variation at low temperature (see Fig. 4). No relaxation was detected at temperatures higher than 10 K, within the time window of the experiment.

IV. DISCUSSION

We have defined a slow variable $\delta S\gamma$ in order to describe, at the macroscopic scale, the collective behavior of microscopic spins during aftereffect measurements at low temperature and we reinterpret the magnetic viscosity in terms of a linear response process. Experimentally, we observe unusual behaviors. For the first multilayer sample, a variation of the magnetization depending on the internal (local) field for a fixed initial state is observed. In this sample, the relaxation due to the action of a local field (described by the viscosity S_0 in the model) can be easily separated from the relaxation due to thermal perturbations (described by the viscosity S_1) because they correspond to opposing variations of the magnetization with respect to time. The thermal behavior displayed in Fig. 2 shows that S_0 remains very small and constant at high temperature [2(c)] while S_1 decrease linearly [2(d)]. In the case of the amorphous $Co_{81}Zr_9Mo_8Ni_2$ sample, we have not been able to measure the relaxation above 10 K. In this sample, the small ellipses are coupled with the heat bath at the mesoscopic scale so that the relaxation due to thermal perturbations (described by the viscosity S_1) is negligible. All the results at high temperature are in agreement with the classical parts of Eqs. (15) and (16) where the noise is given by the equipartition theorem $[K(T)=\eta kT]$. On the other hand, the dramatic increase of the magnetic viscosity S_0 at low temperature must be interpreted as a correction to the equipartition theorem. The low temperature and the mesoscopic size of the sample suggest that this correction may be due to quantum effects. Indeed, if the upper bound is taken in the inequality (15) in order to obtain the quantum correction to the classical noise, a $1/T$ law is obtained, which is in agreement with our experimental results.

As a final remark, we want to stress that a formal equivalence exists between the Langevin equation (6) describing the magnetic variable δS_γ with an additional potential well and the Langevin equation describing the voltage difference V across a resistively shunted Josephson Junction. The analogy between both magnetic and superconducting systems corroborates the relevance of the apparition of the quantum form of the noise $K(T)$, which can be measured at the same size and temperature scale on a shunted Josephson junction (see, e.g., Koch, Van Harlingen, and Clarke[10]). The link between the stochastic fluctuations used in the approach described here and the approach in terms of quantum tunneling through a parabolic barrier at zero temperature has been discussed by Ford, Lewis, and O'Connell,[11] starting from the same Langevin equation. An extension to disordered magnetic systems is straightforward, using Eq. (6).

ACKNOWLEDGMENTS

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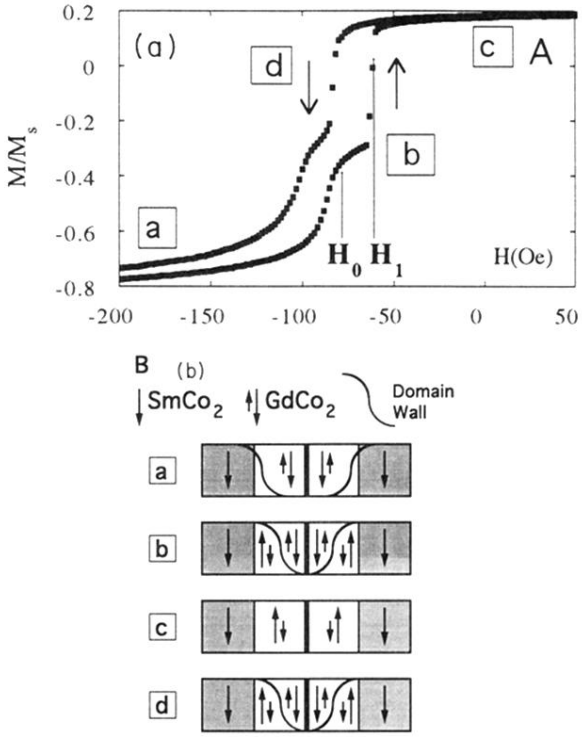

FIG. 1. (a) Hysteresis loop of $\{SmCo_2[/GdCo_2/SmCo_2/GdCo_2/]\ SmCo_2\}$ multilayer sample where the magnetization of both external $SmCo_2$ layers are fixed in the negative direction with respect to the applied field. The configuration *a* is obtained by saturating the sample at -70 kOe. The loop is then measured by sweeping the field from $+0.2$ to -0.2 kOe. The magnetic aftereffect measurements have been performed by suddenly changing the field from -2 kOe up to H_0, i.e., in the configuration *b*, or up to H_1, i.e., in the configuration close to *c*. (b) Schematic representation of the magnetic configuration in *a*–*d*.