Individual-domain-wall motion in $Ni_{0.77}Mn_{0.23}$ observed via resistance fluctuations

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(Received 18 November 1994)

Both equilibrium magnetic noise and Barkhausen noise in $Ni_{0.77}Mn_{0.23}$ films were observed via electrical resistance. The duty cycle of one two-state switcher could be adjusted by changing the magnetic field, as is expected for thermally activated single-domain-wall motion. Two switchers showed non-Arrhenius behavior near the Curie point. Temperature and frequency dependence of the noise indicated the presence of smaller fluctuating domains throughout the ferromagnetic regime. At some temperatures when a large oscillating magnetic field was applied, individual Barkhausen jumps were seen at reproducible points in each field cycle.

I. INTRODUCTION

Electrical transport noise in ferromagnetic materials is acquiring technical importance as magnetoresistive devices are adopted for magnetic memory applications, and especially as the size of such devices is reduced.[1] The detection of individual domain fluctuations via transport measurements is also becoming of interest as a technique which should be able to test theories of macroscopic magnetic quantum tunneling when extended to low enough temperatures.[2]

There are two broad categories of domain fluctuations. The most familiar is Barkhausen noise,[3,4] which results when domains rotate or domain walls move in discrete steps as the magnetic field is swept. The relative importance of domain-wall pinning by defects[5] and of the collective self-organization of domain walls in the Barkhausen effect has become a matter of dispute.[6]

Although spontaneous thermal fluctuations have not been studied as extensively as the Barkhausen effect, fluctuations of this second category can be observed in quasiequilibrium when the magnetic field is not inducing a change in magnetization on experimental time scales.[7–12] In this paper we show both quasiequilibrium and Barkhausen fluctuations of individual domains in partially short-range-ordered *Ni*Mn thin films.[10] An interesting experiment, complementary to this one,[13] has shown reproducible magnetization reversals of single-domain ferromagnetic particles, partly due to thermal energy, after large changes in the applied field.

II. BACKGROUND

Resistance fluctuations are often quantified in terms of a convenient, dimensionless parameter:

$$\alpha(f,T)=\frac{N_a f S_R(f,T)}{R^2}\ , \qquad (1)$$

where f is the frequency, R is resistance, T is temperature, $S_R(f,T)$ is the power spectral density of resistance fluctuations, and N_a is the number of atoms in the sample. We also define spectral slope $\gamma\equiv-\partial \ln S_R(f,T)/\partial \ln f$. α can depend not only on f and T but also on magnetic field H with the form of these dependences determined by the detailed mechanism generating the noise in a particular material.[14]

In many ferromagnets, the main factor that couples magnetization fluctuations to resistance fluctuations is the spontaneous resistive anisotropy[10] (SRA), which is the fractional difference between the resistivity parallel and perpendicular to the magnetic moment within a domain of constant magnetization. (Other factors, such as giant magneto-resistive effects at domain walls, can be more important in some materials.[11]) When the magnetization in some region rotates by any angle other than 180° (e.g., when a domain wall moves), the local resistivity tensor changes. At equilibrium, in samples sufficiently small that only a few FM domain walls fluctuate in the experimental frequency range, the resistance of the sample will have observable discontinuous jumps as a function of time. These are similar to Barkhausen jumps except that they occur reversibly, under constant conditions, such as a constant magnetic field. In the temperature range explored here, thermal energy activates the domain walls over some energy barrier. Arrhenius fluctuations obey the relation $f_p=f_0 e^{-E_a/kT}$, where f_p is α's peak frequency, f_0 is the attempt rate, E_a is the activation energy, and k is Boltzmann's constant.

Ferromagnetic $Ni_{1-x}Mn_x$ has a large SRA,[15,16] up to 10% in bulk disordered material at 4 K and low Mn concentrations. When a domain switches between two different orientations, the fractional step size in the resistance $\Delta R/R$ due to the SRA is[10]

$$\frac{\Delta R}{R}=\frac{\Delta\rho}{\rho}\frac{V_D}{V_s}(\cos^2\theta_1-\cos^2\theta_2)\ , \qquad (2)$$

where $\Delta\rho/\rho$ is the SRA, θ_1 and θ_2 are the two angles of the magnetic moment of the domain with respect to the current density, and V_D and V_s are the volumes of the domain and the sample, respectively.

Besides V_D, it is helpful to find μ_D, the magnetic moment of the fluctuating region. μ_D can be inferred from the effect of H on the energy asymmetry between the two states. This energy difference appears in the logarithm of

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the Boltzmann factor between the two states, an equilibrium property which does not depend on the details of the kinetic pathway between the states, which affect only E_a. The Boltzmann factor is simply the ratio t_u/t_d, of the average times spent in the two states. If we apply H along the direction defined as $\hat{z}$, we then find

$$\Delta \ln(t_u/t_d)=(\Delta\mu_D)_z H/kT , \quad (3)$$

where $(\Delta\mu_D)_z$ is the difference between the magnetic moment's z components in the two states, and k is Boltzmann's constant. We define H_s to be the field that causes t_u/t_d to change by a factor of e. Then

$$\mu_D \geq \frac{kT}{2H_s} . \quad (4)$$

For a single FM domain,

$$V_D=\frac{\mu_D}{\mu_a n} \geq \frac{kT}{2H_s\mu_a n} , \quad (5)$$

where μ_a is the average magnetic moment per atom within the domain and n is the atomic density. If μ_a and the SRA are known at the temperature where the dependence of t_u/t_d on H and $\Delta R/R$ are measured, Eqs. (2) and (5) provide independent estimates of the domain volume.

*Ni*Mn is a material known to have a ferromagnetic regime and a reentrant-spin-glass regime, although the exact nature of these regimes, including whether or not they are true phases, is disputed.[17–19] Previous electrical noise measurements in relatively large samples $[(6\times1280\times0.11)\ \mu\mathrm{m}^3]$ found large, discrete switching objects which were identified as clusters of ferromagnetic domains, since the sizes obtained from the resistance switches greatly exceeded the net moments consistent with the relatively weak field dependences.[10] In this paper we examine the noise in smaller samples, in which individual domains should be detectable.

III. SAMPLES AND TECHNIQUES

A $Ni_{0.77}Mn_{0.23}$ film was deposited onto a sapphire substrate by dual electron beam evaporation at 3×10^{-8} Torr base pressure, at a rate near 3 Å/sec.

The Mn concentration was determined by the inductively coupled argon-ion plasma (ICP) technique to be 23 at. %. The two samples were fabricated from the same film at different locations on the substrate. A sample for dc magnetization (M) measurements, 1_m, was grown on a glass substrate. M vs T, shown in Fig. 1, and hysteresis loops were measured. The Curie temperature T_C of sample 1_m was 285±10 K, in agreement with the T_C obtained from ac susceptibility. In samples 1 and 2, the T_C may have been higher due to annealing at 90 °C during fabrication, which could have increased the amount of atomic short-range ordering. From ac-susceptibility measurements, the crossover into the ferro-spin-glass regime occurred at about 65 K, below the temperature range of the FM domain noise presented here.

Although the SRA was not measured in these samples, similar films[10] had a low-temperature SRA of 0.5% below

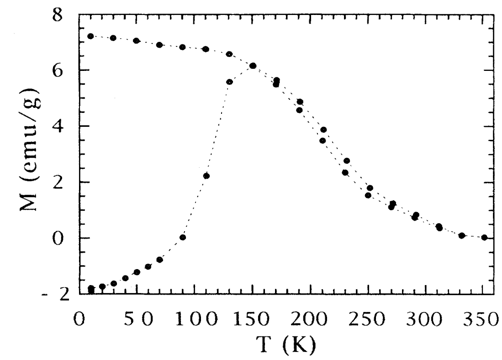

FIG. 1. M vs T of sample 1_m. The onset of strong coercivity occurred at about 130 K. Below 150 K, the upper curve shows data obtained while cooling in 20 Oe. The lower curve corresponds to cooling in near zero field, although there was a slight negative offset from zero.

~100 K. $\Delta\rho/\rho$ decreased gradually to near zero as the T_C was approached. μ_a, the average magnetic moment per atom within a FM domain, was estimated from the saturation magnetization of sample 1_m to be $0.2\mu_B$ at 5 K. From earlier results, the temperature dependence of μ_a and $\Delta\rho/\rho$ can be estimated,[20] yielding the domain volume at higher temperatures to within a factor of 3.

Because the T_C was higher than it would be in atomically disordered bulk samples,[10] we conclude that the samples were partially short-range ordered (SRO). It should be noted that such SRO films are truly ferromagnetic below T_C and above the reentrant spin-glass temperature. In Ref. 10, as well as in the data of this paper, magnetoresistive jumps corresponded to magnetically ordered volumes that were too large to be consistent with superparamagnetism in this temperature regime. Also, hysteresis loops and noise results of similar samples[10] suggested a long-range antiferromagnetic interaction among ferromagnetic domains at low T, implying a domain structure similar to that of bulk disordered samples.[21] Finally, in a different sample, with T_C well above 300 K, domains were directly observed by scanning electron microscopy.[10]

Samples were fabricated into five-point resistance bridges by standard optical lithography. The film was etched by ion milling in an Ar atmosphere of 3×10^{-4} Torr. To minimize contact resistance, Ag was evaporated onto the contact areas, and then 36 gauge Cu wire was connected by using pressed In pads. The samples were 32 μm long and 55 nm thick. Sample 1 was 1.8 μm wide, and sample 2 was 2.0 μm wide. These had 300 times smaller volume than the samples of Ref. 10.

Most of the noise measurements were made with five-probe balanced-bridge samples using a 2.41-kHz ac probe current and a homemade lock-in amplifier, limiting the upper measurement frequency to about 1 kHz. The applied voltages were close to 100-mV rms. The actual voltage fluctuation time traces shown were filtered at about 5 Hz. All spectral data used a standard antialias filter before the analog-to-digital conversion. The Barkhausen experiments were made with a four-probe arrangement and dc current giving about 200 mV. The Barkhausen voltages were sampled at 10 kHz, after standard antialias filtering.

IV. RESULTS

A. Equilibrium noise

Several easily resolved two-state switchers were found in the time-dependent resistance. Typical time traces are shown in Figs. 2(a), 2(b), and 3(a). For one of these, we measured the magnetic-field dependence of t_u/t_d, shown in Fig. 2(c). The strong field dependence clearly illustrates that the switcher is magnetic in nature. Figure 2(c) shows some weak hysteresis, possibly from the environment of the particular magnetic fluctuator, but it is not large enough to prevent an accurate fit of t_u/t_d to an exponential dependence on H. Equation (3) gives $(\Delta\mu_D)_z = (1.0\pm0.1)\times10^7\mu_B$.

Between 300 and 311 K, the magnitude of the switcher's step size, $\Delta R/R$, varied by less than about 12%. Because the SRA must vanish as T approaches T_C, the lack of a decrease in $\Delta R/R$ means that T_C for this particular region was at least about 400 K.[20] Then the SRA would be at least ~0.2 of its low-temperature (5 K) value, and μ_a was at least $\sim0.4\times\mu_a$ (5 K). Assuming that this fluctuator is all or part of a FM domain which switches between two states, Eq. (2) then gives $V_D\approx1.5\times10^{-15}$ cm^3, and Eq. (5) gives $V_D\approx3\times10^{-15}$ cm^3. Given that our estimates of μ_a and the SRA are uncertain to about a factor of 2, the two estimates are in satisfactory agreement. Thus, in all probability this fluctuator consisted of a single domain or a portion of a domain near a domain wall. This result contrasts sharply with the larger fluctuators seen in bigger samples, which required a picture of clusters of antiferromagnetically aligned domains.[20]

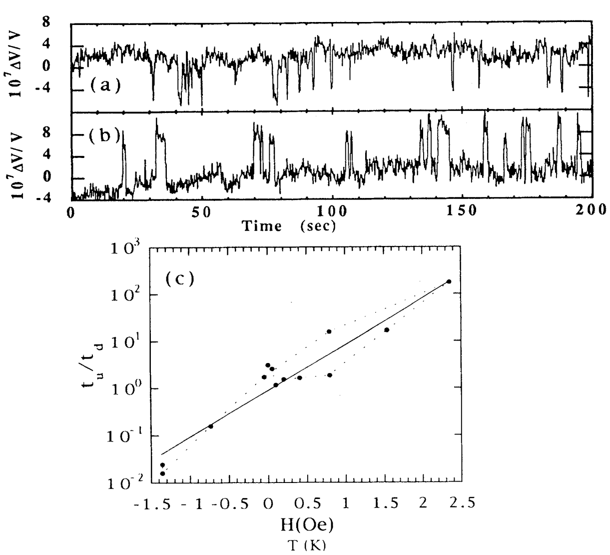

FIG. 2. (a) Time series of sample 1 at $H=1.5$ Oe and $T=307.8$ K. (b) $H=-0.7$ Oe. (c) Ratio of the times spent in the "up" and "down" states, t_u/t_d vs H. H was first increased from 0 to 2.4 Oe and then decreased to -1.4 Oe. (a) and (b) are portions of time series from which two points on (c) are calculated.

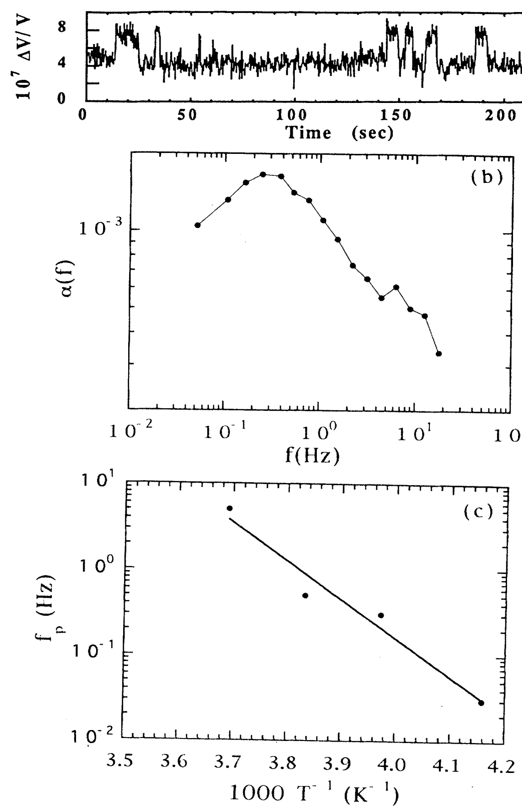

FIG. 3. (a) Time series of sample 1 at 240.5 K. (b) Spectrum α vs f at 252 K. (c) Arrhenius plot of the peak frequency vs $1/T$.

The temperature dependence of this fluctuator's characteristic rate could be tracked only over a narrow range of T, over which significant history-dependent effects were found. If the temperature dependence were to be fit with an Arrhenius form, it would have an activation energy of about (1.7 ± 0.3) eV and an attempt rate f_0 of roughly $10^{27\pm5}$ Hz, but these numbers may be distorted by the dependence of the rate on thermal history. Clearly, since the attempt rate is not physically plausible, the activation energy is somewhat temperature dependent if not also dependent on thermal history.

Another switcher was seen in sample 1, somewhat smaller than the one described above. This one was observed at four temperatures between 240 and 270 K. A spectrum and a time series are plotted in Figs. 3(a) and 3(b). Here, an Arrhenius plot, Fig. 3(c), was obtained from the frequencies of the peak, yielding an E_a of 0.9 ± 0.2 eV and an f_0 of $1\times10^{17\pm4}$ Hz, also slightly too high to be consistent with a temperature-independent barrier.

In addition to the easily resolved individual switchers, apparently given by individual domain-wall motion, the equilibrium noise showed many bumps as a function of both f and T, as illustrated in Fig. 4(a). The features in the T dependence generally matched well with deviations of the slope from $\gamma=1$, as expected for thermally activated processes.[22] The temperatures at which features appeared in a given frequency range varied from sample to sample and to some extent from one cooldown to the next. (There was a similar variation of spectral features in 300-times-larger samples.[10]) At 75 K the noise was slightly non-Gaussian, with a variance in successive measures of the power in an octave around 0.5 Hz being 1.10 ± 0.02 of the Gaussian value.[14]

The peak in α vs T at 40 K behaved differently from the other peaks, in that the temperature and frequency dependence were nearly identical in samples 1 and 2, and the noise was Gaussian to within experimental uncertainty. Figures 4(b)–4(d) show that, as the sample was cooled through $\sim$70 K, spectra became nearly power law, i.e., featureless.

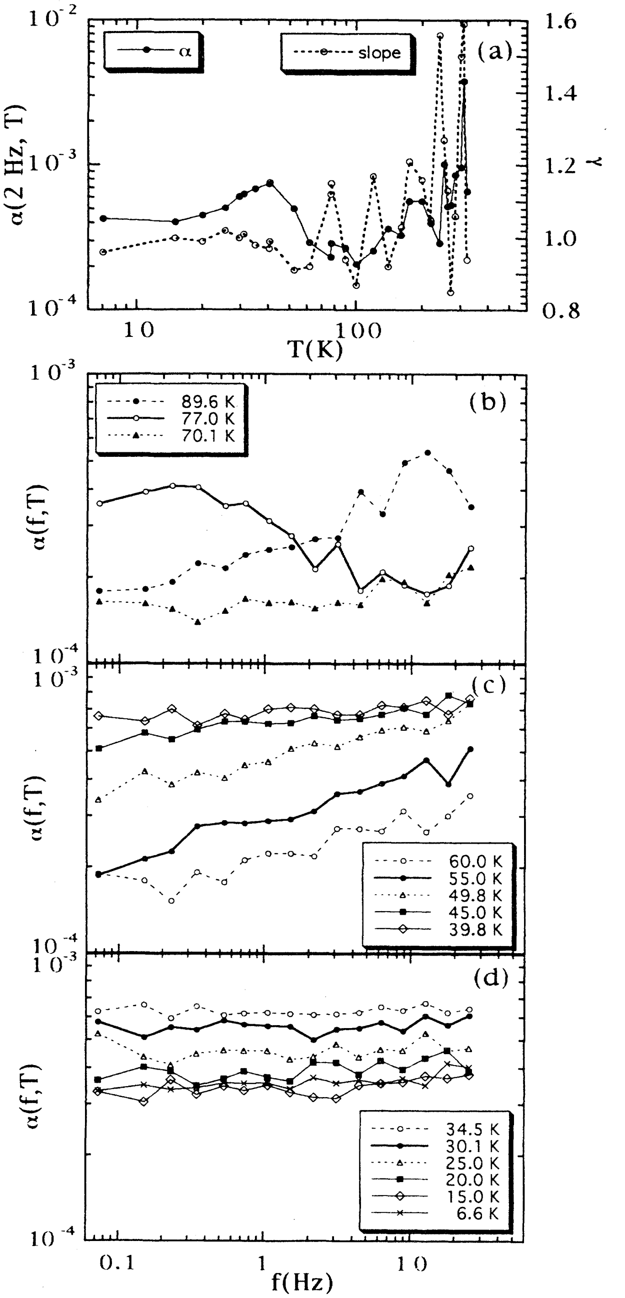

FIG. 4. (a) α vs T and γ vs T of sample 1 in the range 0.41–10.6 Hz. (b)–(d) Spectra of sample 2 at several different temperatures. Note the large spectral features at 89.6 and 77.0 K. Spectra became approximately power law below 70 K. The temperatures given do not include corrections for Joule heating of the sample, which are significant only at 6.6 K.

B. Barkhausen noise

Since some domain walls jump via thermal activation, it is reasonable to expect the appearance of Barkhausen jumps when domain walls are forced to move by an applied magnetic field. We observed such jumps when a sinusoidal field (at frequency f_H and amplitude H_{ac}) was applied to sample 2. At some other temperatures, spikes in the derivative were absent, and domain-wall motion either occurred in smaller jumps or in a nearly continuous motion.

Figure 5(a) shows the time-dependent part of the rms sample voltage (measured with an applied ac current to avoid direct electrical pickup). Most of the response was at frequency f_H or $2f_H$. After subtracting the fundamental from the top trace, the voltage scale could be expanded to reveal jumps [Fig. 5(b)] or higher harmonics of f_H. As seen in Figs. 5(c)–5(f), the spikes in the time derivative (from the difference of successive points) clearly illustrate sharp jumps in resistance. Using Eq. (2), the volume affected by the largest jumps of magnetization was

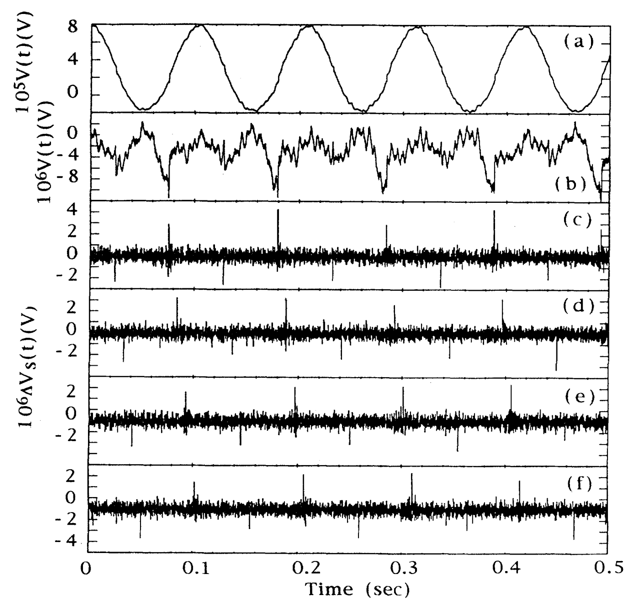

FIG. 5. Barkhausen noise of sample 2 with $T=88$ K, $H_{ac}=53$ Oe, $f_H=9.5$ Hz, average rms sample voltage of 0.234 V, and time between successive points of 10^{-4} sec. (a) Time-dependent part of sample voltage, $V(t)$. (b) $V(t)$ with the fundamental subtracted away. (c)–(f) One long time trace of the difference between successive points, proportional to the time derivative, of data plotted in (b).

around 1×10^{-14} cm^3, somewhat larger than the thermally activated switcher at 310 K.

These spikes reproduced on every cycle, although with slightly fluctuating sizes. The upward and downward spikes under particular field and temperature conditions were not symmetrical, although there is no reason to expect any systematic asymmetry. Just before and after each upward spike in Fig.5, the noise increased.

The size of the jumps increased with increasing H_{ac}, i.e., with increasing dH/dt. A small H_{ac} (5 Oe) produced no perceptible jumps larger than background noise. At other combinations of T and H_{ac}, there were no obvious spikes in the time derivative of the magnetoresistance. Sometimes there were no discontinuities in R at all, but only harmonics of f_H.

V. DISCUSSION

The most obvious explanation of the many features in the equilibrium noise above 70 K is spontaneous thermal fluctuation of ferromagnetic domains. Thermally activated domain-wall motion was previously proposed[10] based on the overall suppression of such noise in larger samples in fields of about 1 kOe. Here, we have explicitly shown that this noise includes discrete two-state fluctuators. By showing the very strong dependence of its duty cycle on the applied magnetic field, we found that one such fluctuator must come from a small ferromagnetic region, probably the consequence of the motion of a single ferromagnetic domain wall. This fluctuating region has a volume of about 2×10^{-15} cm^3. From the size of spectral features, following the techniques of Ref. 10, we estimate that the volume of typical fluctuating domain regions at $T>70$ K was somewhat smaller, about 3×10^{-17} cm^3. In larger samples fluctuating domain clusters with volumes up to 4×10^{-14} cm^3 (Ref. 10) have been found.

Although the kinetics of these domain motions are thermally activated, the attempt rates inferred by assuming fixed barriers were unphysically high. Most likely the barriers to rotation or domain-wall movement shrank over a considerable range of T as T_C was approached.

The loss of the spectral features in these small samples below 70 K is consistent with FM domain motion freezing below the ferro-spin-glass crossover temperature. Such FM domain stiffening or slowing down was also seen in neutron scattering,[19] Lorentz microscopy,[23] ferromagnetic resonance,[24] Barkhausen noise,[25] and the onset of large remanence in dc magnetization data.[21] Furthermore, it probably was reflected in low-field ac magnetic susceptibility measurements, where the FM response nearly vanished below the reentrant spin-glass temperature.[17,18]

In their repeatability as H was cycled, the Barkhausen jumps were similar to other data showing partially reproducible jumps in dM/dt of multidomain samples while measuring hysteresis loops with an ac field.[26] Such a result is obviously consistent with standard pictures based on individual domain walls moving in a fixed environment of defects, e.g., Ref. 5. Since the domains see the same potential barriers, or defects, each time the field is cycled, jumps tend to occur repeatedly at the same part of the cycle. However, the enhanced noise which both precedes and follows some of the large jumps suggests an avalanchlike process, perhaps having something in common with the phenomenon dubbed self-organization,[27] without any criticality.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation through Grant No. DMR 93-05763 and, through the facilities and staff of the Materials Research Laboratory, including R. S. Averback's film deposition apparatus, Grant No. DMR 89-20538.

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form of $(\Delta\rho/\rho)(T)$. SRA in *Fe*Zr [H. Ma, Z. Wang, H. P. Kunkel, G. Williams, and D. H. Ryan, J. Appl. Phys. **67**, 5964 (1990)] and several Ni alloys [Yu. V. Vasílev, Phys. Status Solidi **38**, 479 (1970)] are similar. Temperature dependence of μ_a is discussed by Abdul-Razzaq *et al.* in Ref. 21. To within less than a factor of 2, we can approximate it to be proportional to $M(T)$ except for T very near T_C.

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