5-15-1994

Blocking of logarithmic temporal relaxation of magnetic remanence by piezomagnetically induced domains in $Fe_{1-x}Zn_xF_2$

J. Kushauer
Angewandte Physik, University of Duisburg, Germany

Christian Binek
University of Nebraska-Lincoln, cbinek@unl.edu

Wolfgang Kleemann
Angewandte Physik, University of Duisburg, Germany, wolfgang.kleemann@uni-due.de

Blocking of logarithmic temporal relaxation of magnetic remanence by piezomagnetically induced domains in $Fe_{1-x}Zn_xF_2$

J. Kushauer, C. Binek, and W. Kleemann
Angewandte Physik, University of Duisburg, 47048 Duisburg, Germany

Faraday rotation and superconducting quantum interference device magnetometry were used to investigate the relaxation of the weak-field induced remanence, μ, of the random-field Ising model system $Fe_{1-x}Zn_xF_2$. The stretched logarithmic decay law, as predicted by Nattermann and Vilfan for the case of compact domains with fractal surfaces, was confirmed for freezing fields of $3 \leq B \leq 5$ T. Virtually constant remanence, μ, was found at low fields, $0.0002 \leq B \leq 1.5$ T. This ferrimagnetic moment is due to the piezomagnetic effect acting on random-stress induced immobile domains. Chemical etching, which removes surface stress originating from the sample preparation, proves to decrease the remanence by about 50%, part of which is restored after renewed optical polishing.

I. INTRODUCTION

Diluted uniaxial antiferromagnets (AFs) exposed to a uniform external magnetic field, B, are well known to exhibit the critical behavior of the random-field Ising model (RFIM).[1,2] Recently, considerable interest has been focused onto their nonequilibrium behavior after field cooling (FC) to below $T_{eq} \gtrsim T_c(B)$. Metastable domain states[3] arise, which are characterized by finite correlation lengths of the AF order parameter[4] and excess magnetization, ΔM, which is primarily concentrated on the domain walls.[3] After removing B a small portion of about 10% remains as long-lived thermoremanent magnetization (TRM), μ. In the absence of random-field (RF) pinning at $B=0$, μ evolves with time via random-exchange Ising model (REIM) dynamics.[5] This was demonstrated on prototypical systems like $Fe_{0.7}Mg_{0.3}Cl_2$[6] and $Fe_{0.47}Zn_{0.53}F_2$[7] as well as in Monte Carlo simulations.[8] It was noticed that no significant growth of the correlation length, after removing B, is visible in neutron scattering experiments.[4] This means that the reduction of the long-lived TRM arises merely from local rearrangements of the spins on the AF domain boundaries. For this situation stretched logarithmic decay with time, t,

$$\mu(t)=a[\ln(t/\tau)]^{-\psi}, \quad (1)$$

was predicted by Nattermann and Vilfan (NV)[5] and confirmed by measurements on $Fe_{0.7}Mg_{0.3}Cl_2$[6] and $Fe_{0.47}Zn_{0.53}F_2$[9] after FC in weak fields, $B/|J| \approx 0.1$, to low temperatures, $T/T_N \approx 0.1$. J, T_N, and τ are the leading exchange constant, the Néel temperature, and a microscopic relaxation time, respectively. In accordance with theory[5] exponents $\psi=0.4\cdots0.55$ were observed. At larger fields, $B/|J| \lesssim 1$, and higher temperatures, $T/T_N \lesssim 1$, however, the polydispersivity of the TRM decay function is better described by ordinary or generalized power laws.[7] This is related to volume relaxation processes of interpenetrating fractal domains, which render the system spin-glass like.[3]

Surprisingly, recent TRM experiments on $Fe_{0.47}Zn_{0.53}F_2$[9] did not reveal better agreement with the NV prediction, Eq. (1), in the very low B range. On the contrary, constant weak TRM emerges, which is virtually independent of $B<1.5$ T and t. Its temperature dependence approximately follows that of the AF order parameter. It was argued[9] that this unexpected effect is due to the formation of random shear-stress induced AF domains. They are expected to carry weak ferrimagnetic moments owing to symmetry-allowed piezomagnetism of the rutile-type crystal structure.[10] In fact, the formation of surface-stress induced AF domains was evidenced previously[11] on MnF_2 via neutron topography. On the other hand, residual magnetization persisting in poling fields as low as $B=10^{-3}$ mT were recently observed.[12] Very probably, this hitherto unexplained[12] ferrimagnetism is related to residual stress originating from sample growth and preparation.

This idea will be pursued in the present paper. First, we recall[9] the crossover from logarithmically relaxing to piezomagnetically frozen TRM on a sample of $Fe_{0.47}Zn_{0.53}F_2$ when decreasing B from 4.86 to 0.001 T. Second, on a sample of $Fe_{0.6}Zn_{0.4}F_2$ with smaller stress-induced TRM, a weak B dependence, $\mu \propto B^{0.05}$, is found for $B<0.1$ T. This is explained by the inhomogeneity of the built-in stress fields. Furthermore, by chemical etching of the surface the TRM is asymptotically reduced to its bulk value. Subsequent mechanical polishing partially restores the surface-stress induced TRM. Its time independence is demonstrated.

II. EXPERIMENTAL

Relaxation of the field-induced TRM was observed[9] on the same diluted sample of $Fe_{0.47}Zn_{0.53}F_2$ used previously.[7] In a longitudinal magnetic field, parallel to the crystallographic c direction, the Faraday rotation (FR) angle θ, $\propto M$, was measured with light of wavelength $\lambda=442$ nm using a compensation circuit with a resolution of $\Delta\theta \sim 5\times10^{-4}$ deg and previously described modulation techniques.[6] The data were taken after FC from $T=2T_N(\sim 80$ K) to $T=2.8$ K. After stabilizing the temperature T to within $\Delta T=1$ mK, the field was switched off. The switch-off time was properly[7] taken into account when fitting to the different decay laws.

Commercial superconducting quantum interference device (SQUID) magnetometers (Quantum Design MPMS2 and MPMS5S) were used to determine the TRM of two samples, $Fe_{0.47}Zn_{0.53}F_2$[9] and $Fe_{0.6}Zn_{0.4}F_2$, after FC in a constant field, $0.00024 \leq B \leq 5$ T, from $T \sim 2T_N$ to $T=10$ and (for obtaining the paramagnetic response) 55 K. Furthermore, the low-field TRM, $\mu(0.24$ mT), of the $Fe_{0.6}Zn_{0.4}F_2$ sample was repeatedly measured both at $T=10$ and 55 K after stepwise etching with diluted nitric acid (4 vol % HNO_3 in H_2O at

 0021-8979/94/75(10)/5856/3/$6.00

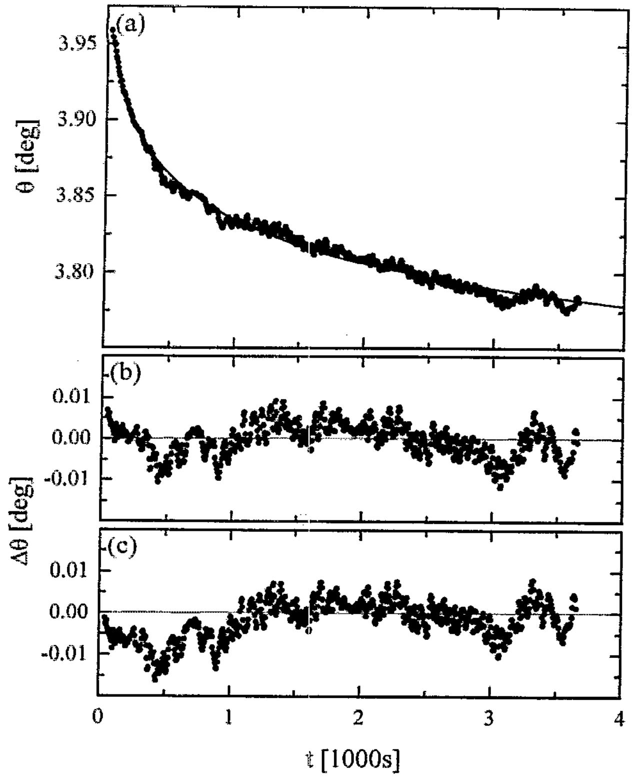

FIG. 1. Remnant Faraday rotation angle, θ, vs time, t, (a) measured on $Fe_{0.47}Zn_{0.53}F_2$ with λ=442 nm at T=2.8 K after FC with B=4 T (Ref. 9). The solid line is a best fit to the NV function, Eq. (1). Deviations of the experimental data, θ vs t (a), from the best-fitted NV function and the power law approximation, $\theta \propto t^{-x}$, are shown as $\Delta\theta$ vs t [(b) and (c), respectively].

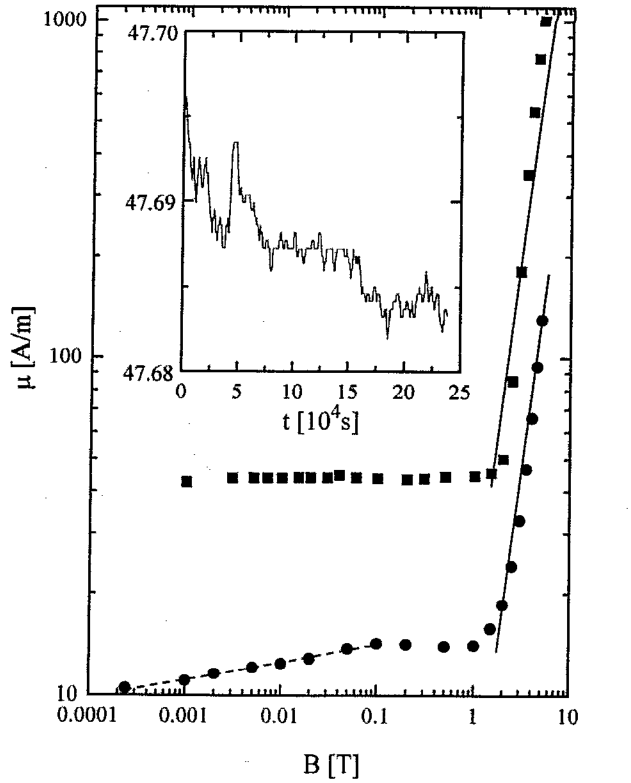

FIG. 2. Log–log plots of the TRM, μ, of $Fe_{1-x}Zn_xFe_2$ [x=0.53: squares (Ref. 9); x=0.4: circles] vs freezing field at T=10 K. The solid and dashed lines represent B^2 and $B^{0.05}$ dependences (see the text). The inset shows the t dependence of μ for the x=0.53 sample in zero field after FC with B=0.8 T to T=10 K.

T=293 K). After 5 h total etching time the sample was repolished with 7 and 1 μm diamond abrasive in order to demonstrate the surface stress induced ferrimagnetic moment. The loss of weight of the sample due to etching and polishing was monitored at each step.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the time dependence of TRM measured by FR angle, θ, in zero field, after cooling in a magnetic field of B=4.0 T down to T=2.8 K. On the experimental time scale up to 3500 s the magnetization drops by about 0.5°, while the scattering of the data is in the order of 5 mdeg and does not affect the resulting shape of the relaxation curve. Among the different fitting functions, NV, pure (PP), and generalized power law (GP), NV turns out to be the best choice.[9] This becomes evident from χ^2 tests[9] and from direct comparison of the deviations $\Delta\theta$ between experimental data and model functions. This is shown in Fig. 1, where $\Delta\theta$ clearly yields smaller rms values for NV (b) than for PP (c). Although the GP yields similar χ^2 numbers as NV, it does not qualify as the best fit function since an additional fitting parameter is involved. Another crucial argument in favor of the NV theory is the appearance of the predicted exponent, $\psi \sim 0.4$ [ψ=0.38 in the case of Fig. 1(a)]. This holds for intermediate freezing fields, $3 \leqslant B \leqslant 5$ T, at low temperatures.[9] At larger fields, $B>5$ T, the system crosses over into the spin-glass-like domain state,[3] where the PP or the GP are found to be valid.[7] At lower fields, $B<3$ T, perturbations due to piezomagnetic domain formation become important as will be discussed below.

Figure 2 shows the remnant magnetic moment, μ, in zero field ($B<0.2$ mT) as a function of the freezing field after FC to T=10 K for $Fe_{0.47}Zn_{0.53}F_2$ (squares)[9] and $Fe_{0.6}Zn_{0.4}F_2$ (circles). Straight interpolated lines in the range $1.5 \leqslant B \leqslant 5$ T are best fits to $\mu = bB^2 - c$ with adjustable parameters b and c. These parabolas indicate RF-like behavior corrected for relaxation loss in the vicinity of $T_c(B)$.[3,9] In the low-field range the μ vs B data level off to become virtually constant, $\mu_0 \sim 44$ A/m for x=0.53 within $0.001 \leqslant B \leqslant 1.5$ T and $\mu_0 \sim 15$ A/m for x=0.4 within $0.1 \leqslant B \leqslant 1.1$ T. These remanences are attributed[9] to piezomagnetic moments, $m_z \propto \lambda_2 \sigma_{xy} l_z/|l_z|$. They are generated by accidental built-in shear stress, σ_{xy}, and controlled by the external field, B_z, via minimization of the magnetostatic energy, $E_m = -m_z B_z$. As argued previously,[9] randomly distributed shear stress fields with alternating signs of the σ_{xy} components give rise to antiferromagnetic domains with alternating signs of the AF order parameter, $l_z/|l_z|$, while cooling through $T_c(B)$ with $B \| [001]$. All domains contribute to m_z with the sign of B_z. Only at very low or really vanishing freezing field μ=0 will be achieved,[12] where nonvanishing stress fields might still create piezomagnetic moments with zero average, $\langle m_z \rangle = 0$. The different μ_0 values found for both samples seem to re-

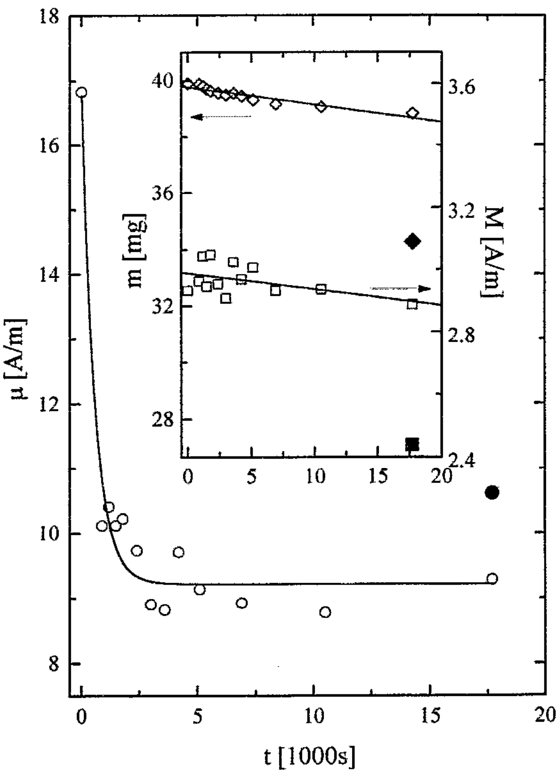

FIG. 3. Magnetization at $T=10$ K (μ; circles) and 55 K (M; squares) and mass (m; diamonds) of a sample with $x=0.4$, field cooled and measured in $B_0=0.24$ mT, as a function of etching time up to $t=1.6\times10^4$ s (open symbols) and after subsequent optical polishing (solid symbols).

flect different average stress fields, $\langle\sigma_{xy}\rangle$. The lower stress in the $x=0.4$ sample—about 1/3 of that of the $x=0.53$ sample—gives rise to a marked field dependence, $\mu_0\propto B^{0.05}$, at very low fields, $0.0002\leq B\leq0.1$ T, where the thermal energy comes close to the magnetostatic one, $\langle E_m\rangle$. Hence, thermal disorder will destroy the complete alignment of the domains and thus decrease $\langle m_z\rangle$.

It will be interesting to study the change of the TRM from relaxational to static behavior when decreasing B to below 1.5 T. Very weak time dependence is found in the crossover region. The inset of Fig. 2 shows the decrease of the TRM in the $x=0.53$ sample after FC to $T=10$ K with $B=0.8$ T. The rate, $-(d\mu/dt)/\mu\sim8\times10^{-10}\ \mathrm{s}^{-1}$, as measured over a period of 2.5×10^5 s is clearly above the noise level of the MPMS5S SQUID apparatus. Tentatively it is ascribed to NV-type relaxation of magnetized AF domain walls, which are preferentially formed at unstrained sample regions, where σ_{xy} and, hence, $l_z/|l_z|$ change sign from one domain to the other.

Figure 3 shows the total magnetization, μ, of the $x=0.4$ sample obtained after FC with $B_0=0.24$ mT (=remanence of the supraconducting coil) to $T=10$ K versus etching time, t (open circles). In parallel, both the paramagnetic magnetization, M, induced by B_0 at $T=55$ K and the mass, m, of the sample have been determined as functions of t (inset: open squares and diamonds). It is seen that these quantities decrease linearly with t by about 3% within the total etching time of 1.6×10^4 s. This is a consequence of the loss of mass during the chemical attack. Far greater effects are found with the low-T magnetization (open circles), which is primarily due to stress induced piezomagnetic moments beside a nearly negligible unperturbed AF contribution [M^{AF}(10 K)$\sim M^{\mathrm{PM}}$(55 K)/6$\sim$0.5 A/m].[13] Within 10^4 s nearly 50% of the initial magnetization are removed. The μ vs t data are best fitted to an exponential decay law, $\mu=\mu_s e^{-t/\tau}+\mu_v$ with volume and surface contributions $\mu_v=9.2$ A/m and $\mu_s=8.0$ A/m, respectively, and a time constant $\tau=580$ s. Obviously, a large amount of stress centers is very efficiently removed from the surface by a first-order chemical reaction, $-d\mu/dt\propto\mu$. Very probably these stress fields are due to the surface treatment of the sample, which was optically polished at the beginning, $t=0$. In fact, repolishing of the slightly roughened surfaces after the etching procedure at $t=1.6\times10^4$ s recovers about 20% of μ_s (Fig. 3: full circle) despite a volume loss due to abrasion of about 12% (inset of Fig. 3: solid symbols).

IV. CONCLUSION

The field-induced thermoremanence of the prototypical domain state system $Fe_{1-x}Zn_xF_2$ ($x=0.53$) is confirmed to follow the stretched logarithmic temporal decay law of Nattermann and Vilfan.[5] At weak fields, $B\leq1.5$ T, the relaxation rate drops to nearly unmeasurable values. About 50% of the virtually constant residual magnetic moment of an $x=0.4$ sample is demonstrated to be due to surface stress by virtue of the longitudinal piezomagnetic effect.[10] Revision of results obtained previously[2,3] in the critical regime with fairly weak fields, $1\leq B\leq2$ T, appears desirable.

ACKNOWLEDGMENTS

Thanks are due to V. Jaccarino for the high quality crystals. Work supported by Deutsche Forschungsgemeinschaft through Sonderforschungsbereich 166.

[1] S. Fishman and A. Aharony, J. Phys. C **12**, L729 (1979).
[2] For a review see D. Belanger and P. Young, J. Magn. Magn. Mater. **100**, 272 (1991).
[3] For a review see W. Kleemann, Int. J. Mod. Phys. B **7**, 2469 (1993).
[4] R. A. Cowley, H. Yoshizawa, G. Shirane, and R. J. Birgeneau, Z. Phys. B **58**, 15 (1984).
[5] T. Nattermann and I. Vilfan, Phys. Rev. Lett. **61**, 223 (1988).
[6] U. A. Leitão and W. Kleemann, Phys. Rev. B **35**, 8696 (1987).
[7] S.-J. Han, D. P. Belanger, W. Kleemann, and U. Nowak, Phys. Rev. B **45**, 9728 (1992).
[8] U. Nowak and K. D. Usadel, Phys. Rev. B **39**, 2516 (1989).
[9] J. Kushauer, W. Kleemann, J. Mattsson, and P. Nordblad, Phys. Rev. B (in press).
[10] A. S. Borovik-Romanov, Sov. Phys. JETP **11**, 786 (1960).
[11] M. Schlenker and J. Baruchel, J. Appl. Phys. **49**, 1996 (1978); Acta Cryst. A **34**, S 255 (1978); J. Baruchel, M. Schlenker, and B. Barbara, J. Magn. Magn. Mater. **15–18**, 1510 (1980).
[12] T. Fries, Y. Shapira, A. Paduan-Filho, C. C. Becerra, and F. Palacio, J. Phys. Cond. Matt. **5**, L 107 (1993); M. Lederman, J. Hammann, and R. Orbach, Physica B **165 & 166**, 179 (1990).
[13] W. Kleemann, A. R. King, and V. Jaccarino, Phys. Rev. B **34**, 479 (1986).