Size and Fluctuation Effects on the Dynamics of Linear Domain Walls in an Ising Ferromagnet

J. Kötzler, M. Grahl, and I. Sessler
Institute für Angewandte Physik, Universität Hamburg, 2000 Hamburg 36, Federal Republic of Germany

J. Ferré
Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay, France
(Received 5 October 1989)

Approaching the Curie temperature of $LiTbF_4$ ellipsoids of thicknesses between 0.5 and 7 mm up to $T_c - T = 3\times10^{-4}T_c$, the first clear evidence for critical fluctuations *and* macroscopic size effects on the domain-wall relaxation is presented. An analog to the Landau-Lifshitz model proposed recently and additional optical data on the domain width suggest associating both phenomena with the existence of linear (Bulaevskii-Ginzburg) walls and with domain branching near the surface.

PACS numbers: 75.60.Ch, 64.60.Ht, 75.40.Gb, 75.50.Dd

The domain dynamics of ferromagnets is determined by two essentials, both of substantial complexity: the domain structure and the local forces acting on the spins of a given structure during nonequilibrium. Even uniaxial ferromagnets, characterized by a simple two-phase domain state and relaxational spin dynamics, are still under debate, despite a long-standing history dating back to the pioneering work of Landau and Lifshitz.[1] Recently, the occurrence of stripe, bubble, and branched domains in the phase diagram of strongly uniaxial ("Ising") ferromagnets depending on temperature, magnetic field, and sample (slab) thickness has been evaluated[2-4] in agreement with experimental results.[5] Current theoretical work on the dynamics, based on phenomenological coefficients of wall kinetics, predicts exponential[6] and logarithmic[7] decays of the magnetization for striped and branched domains, respectively. Existing data on Ising ferromagnets[8-11] clearly indicate exponential behavior, and within the limited ranges of temperatures under examination all relaxation rates have been fitted by an Arrhenius law. However, in neither case could a physical significance of the fitted energy barriers nor a relation to concrete domain parameters be substantiated.

Very recently, an investigation of domain-wall dynamics close to the Curie temperature of the weakly uniaxial ferromagnet $GdCl_3$ confirmed the exponential behavior, however, the observed speeding up of the relaxation rate with temperature was ascribed to critical fluctuations[12] rather than to a thermal activation. This interpretation relied on the existence of so-called linear walls of width $\xi_-(T)$ (magnetic correlation length below T_c) proposed by Bulaevskii and Ginzburg,[13] in which the magnitude of the local magnetization $|\mathbf{M}|$ is changing from $+M_s$ to $-M_s$. This type of wall should be preferred in uniaxial ferromagnets, where the susceptibility along the easy axis $\chi_\parallel$ exceeds the transverse one $\chi_\perp$ because their energy density $M_s^2/\mu_0\chi_\parallel$ is smaller than $M_s^2/\mu_0\chi_\perp$, corresponding to that of the conventional Bloch wall. However, since, to date, neither a direct observation of the linear wall nor any experimental information on the domain structure of $GdCl_3$ exists, the interpretation of the critical wall dynamics in Ref. 12 is somewhat speculative. Moreover, the available relaxation rates Γ_d cover only one decade in reduced temperature not too close to T_c, i.e., $0.02 \le t \equiv 1 - T/T_c \le 0.2$,[12] where evidence of critical effects is not quite obvious.

By examining the Ising ferromagnet $LiTbF_4$, the present work is intended to perform a much more thorough check of the supposed linear walls and their dynamics. In addition to the extremely small $\chi_\perp$ [$=0.03$ (Ref. 14)], $LiTbF_4$ offers two fortunate features for this purpose due to the high quality of the crystals: (i) The critical temperature can be approached to $t \approx 10^{-4}$ (Ref. 15) without running into rounding phenomena and (ii) the material may be shaped to spheroids of thickness D between 0.1 and 10 mm with well-defined demagnetization coefficients N. This allows a first serious test of the significant size effect on Γ_d predicted by the relaxational-wall model.[12] Also of great interest is a detailed knowledge of the domain structure from experiment and theory summarized in Fig. 1. Above T_c, the needle-shaped magnetic fluctuations of extension $\xi_+(T)$, seen on $LiTbF_4$ by diffuse neutron scattering, are characterized by the dipolar wave number $q_d = 1.1$ Å^{-1}.[16] Below T_c, this short-range order condenses into the needle domains forming the up and down phases [see Fig. 1(a)], separated by linear walls of width ξ_-. For "thick" samples, $D \gg 40q_d^{-1}$,[4] branching is expected to occur near the surface, which reduces the dipolar stray fields at the expense of additional wall and internal magnetostatic energies. Because of this effect, the domain width in the bulk has been predicted[3,4] to grow as

$$d = (D^2 l)^{1/3}, \tag{1}$$

with a characteristic magnetic length $l = 1/q_d^2\xi_-$.[4,12] This $D^{2/3}$ dependence was discovered on thin Co platelets at low temperature, $T \ll T_c$.[17]

Because of the strong Faraday effect of $LiTbF_4$,[18] we could observe directly the domain pattern [Fig. 1(b)] by a technique described in a previous paper.[5] Apparently,

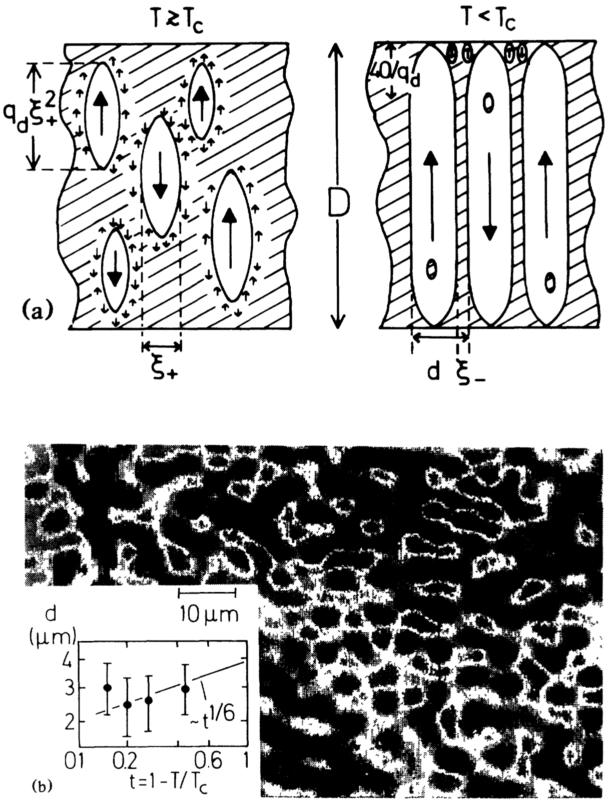

FIG. 1. (a) Cross sections through a plate of a strongly uniaxial ferromagnet illustrating short-range order above T_c and needle-shaped long-range order below T_c, both imbedded by (hatched) seas of disorder. (b) The domain pattern in the bulk of a $LiTbF_4$ plate of thickness 0.4 mm observed by Faraday rotation at $T=2.12$ K and $H=0$. Black and grey regions represent the two domain types.

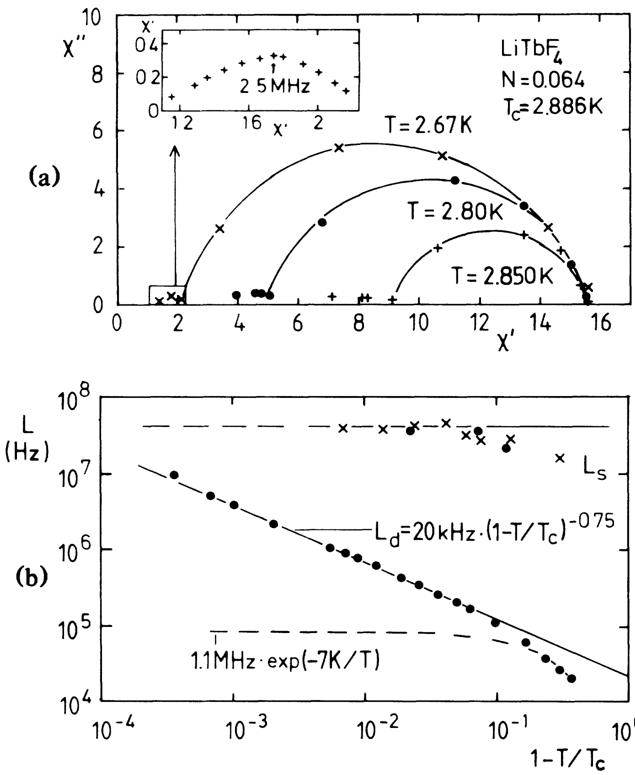

FIG. 2. (a) Argand diagrams of $\chi(\omega)$ for a needle-shaped sample at different temperatures below T_c. Frequencies from right to left: 26.8 Hz, 214 Hz, 858 Hz, 1.717 kHz, 6.867 kHz, 479 kHz, 1.76 MHz, 3.52 MHz, and 7.03 MHz. Full lines are fits with Eq. (2). Inset: Frequencies between 400 kHz and 50 MHz. (b) Kinetic coefficients for domain-wall motion L_d and spin-spin relaxation L_s vs reduced temperature: ●, needle [from (a)], ×, sphere. The dotted line represents an Arrhenius law fitted to L_d at low temperature.

there is a tendency towards bubble formation rather than to stripes, which we associate with two facts: (i) the absence of a preferred magnetic direction perpendicular to the easy axis, i.e., in the tetragonal plane; and (ii) the energies of stripe and bubble structures are very close to each other.[19] Hence, we may discuss the results in terms of this stripe model modified by branching near the surface. The domain widths d, shown in the inset of Fig. 1(b), have been determined by averaging across various stripelike regions. To compare them with the estimate from Eq. (1), we use the general relation between the isothermal susceptibility and correlation length, $\chi_\parallel = (q_d\xi)^2$, confirmed for $LiTbF_4$ at $T>T_c$.[20] Taking $\chi_\parallel(T<T_c)=1.7(1-T/T_c)^{-1.06}$ from Ref. 21 we obtain $d(T)=d_0(1-T/T_c)^{0.17}$ with $d_0=2.1(4)$ μm for the given thickness of the slab. With regard to the approximate nature of Eq. (1), which combines macroscopic and atomic lengths resulting from rather crude estimates of the magnetostatic and wall energies, the agreement to the experimental number $d_0=3.9(1.0)$ μm is quite remarkable. Hence, it might be justified to accept the presence of linear walls as a working hypothesis for the following discussion of the domain dynamics.

The complex susceptibility $\chi(\omega)$, measured between 20 Hz and 50 MHz along the easy axis of one of the six $LiTbF_4$ ellipsoids under investigation, is shown by Argand diagrams in Fig. 2(a). Both the low- and the high-frequency dynamics deviate slightly from the semicircle shape, i.e., exponential (Debye) relaxation. Reaching the maximum possible susceptibility, i.e., the demagnetization plateau $\chi_d = 1/N$, the slow process arises from the domain-wall motion well described by the form

$$\chi(\omega)=\frac{\chi_d-\chi_s}{1+(i\omega/\Gamma_d)^{1-\alpha}}+\chi_s\,. \tag{2}$$

At $\omega \gg \Gamma_d$, the walls are fixed and the adiabatic susceptibility of the disordered phase within the domains is recovered,[22] which displays the much faster spin dynamics illustrated by the inset to Fig. 2(a). Since the deviation parameter α, traditionally being attributed to a distribution of relaxation rates,[23] remains small in the present case, $\alpha = 0.10(5)$, we can restrict our discussion to the (mean) domain relaxation rate Γ_d. As an intrinsic dynamical quantity, we consider the so-called Onsager coefficient of wall motion, $L_d = \dot{m}_d/\Delta h$, determining the magnetization change as linear response to a small nonequilibrium field Δh, and related to Γ_d by the following form:[12]

$$\Gamma_d = \frac{L_d}{\chi_d}\left(1 - \frac{\chi_s}{\chi_d}\right). \tag{3}$$

The temperature dependence of L_d depicted in Fig. 2(b) reveals a speeding up which can be fitted to a power law,

$$L_d(T) = L_d(0)(1 - T/T_c)^{-z}, \tag{4}$$

with $z = 0.75(5)$. This fit covering almost *three* decades of the reduced temperature provides the strongest evidence on the effect of critical fluctuations on domain-wall motion. At $t \approx 0.1$, L_d starts to drop much faster, which we relate to the fact that there the correlation length ξ_- is decreased to one lattice constant, so that collective dynamics passes to single-spin processes. Indeed, fitting these data to an Arrhenius law one finds an activation energy of about $2kT_c$ [see Fig. 2(b)], which represents the maximum energy required for a single spin flip in the ferromagnetic state. Figure 2(b) also illustrates that any thermally activated dynamics cannot explain our data, wherever a fit is attempted.

First, to discuss the critical effect on L_d we start with the prediction obtained from a relaxational *Ansatz* for the kinetics of the local magnetization in the linear wall:[12]

$$L_d = 3L_\parallel(\xi_-/d), \tag{5}$$

where $L_\parallel$ and the ratio ξ_-/d represent the microscopic flip rate and the relative number of wall spins, respectively. Inserting d from Eq. (1), we find

$$\frac{\xi_-}{d} = \left(\frac{\chi_\parallel}{q_d D}\right)^{2/3}, \tag{5a}$$

which because of $\chi_\parallel \sim t^{-1.06}$ (Ref. 22) yields $\chi_\parallel^{2/3} \sim t^{-0.71}$. This is the main result which fully explains the observed critical speeding up of L_d.

The other specific prediction of the relaxational wall model, the size effect on L_d, has been tested by investigating various sample spheroids of thickness D between 0.5 and 7 mm along the easy magnetic direction. In fact, the inset to Fig. 3 clearly demonstrates that the kinetic coefficient is suppressed by increasing D while the critical law $L_d \sim t^{-0.75}$ remains unchanged. As a principal result, Fig. 3 shows the rather satisfying agreement between the critical amplitudes $L_d(0)$ and the $D^{2/3}$ dependence predicted by Eq. (5a) for stripe domains in thick plates. Note that this comparison employs an effective thickness of the spheroids, $D_{\text{eff}}^{2/3} \equiv \overline{D_{\text{loc}}^{2/3}}$, to include the variation of the local free energy[3,4] and of the domain

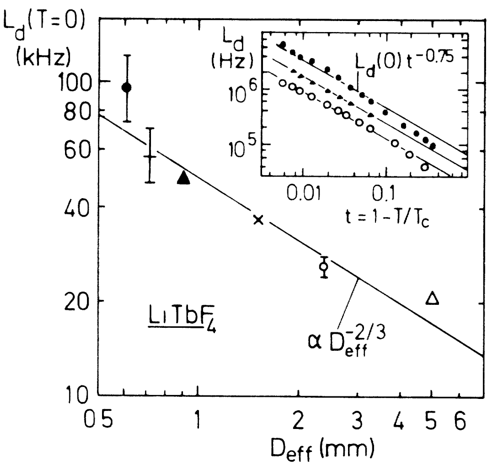

FIG. 3. Critical amplitude of the kinetic coefficient for domain-wall relaxation vs effective thickness of various sample ellipsoids. Inset: $L_d(T)$ for $D_{\text{eff}} = 0.6$ (●), 0.9 (▲), and 2.2 (○) mm.

width through the variation of the actual thickness D_{loc} across the sample. No significant differences between D_{eff} and D resulted for the oblate spheroids ($D < 2$ mm), while the maximum change was found for the longest needle, $D_{\text{eff}} - 0.8D = 5.2$ mm. At this point considering Eq. (5a) one may argue whether the variation of D_{loc} around D_{eff} causes a distribution of L_d and therefore of domain relaxation rates. However, the present data do not display any relevant correlation between the variance of D_{loc} and the (small) distribution parameter α in Eq. (2). We also emphasize that the demagnetizing effect on Γ_d is fully accounted by $\chi_d = 1/N$ in Eq. (3).

A final remark should be devoted to the magnitude of the local flipping rate $L_\parallel$, which remains essentially constant in the critical region. Adjusting the L_d data for $t < 0.1$ to Eq. (5) with $\xi_-/d = 3\times10^{-5}t^{-0.71}[D/(1\text{ mm})]^{2/3}$,[24] one obtains $L_\parallel = 0.5$ GHz, which does not correspond to the other kinetic coefficients known for $LiTbF_4$. First, $L_\parallel$ is larger than $L_s = \Gamma_s\chi_s = 0.05$ GHz [see inset to Fig. 2(b)], determining the spin relaxation within the domains and, second, $L_\parallel$ is much smaller than $\Gamma_p(\omega \to 0)\chi_\parallel = 18$ GHz,[25] describing the low-frequency relaxation in the paramagnetic region. From Ref. 25 it is known that the spin dynamics are dominated by a complicated interplay between dipole-dipole and crystal-field interactions, the details of which are not yet explored below T_c.

In conclusion, the discussion of the novel fluctuation and size effects on the domain-wall relaxation reported here together with optical images of the domains provide rather strong arguments for (i) the existence of linear walls in uniaxial ferromagnets and (ii) the influence of domain branching near the surface on the domain width even for samples of mm size close to T_c. The critical and size effects have been analyzed using a recent relaxational model;[12] however, the magnitude of the inherent local spin-flip rate remains unexplained. The data also imply a narrow relaxation spectrum consistent with the dominance of a single domain type, while the branching and nonplanar shape of the samples do not cause any significant broadening. One may argue whether thinner samples, where surface effects gain more importance, display the modification of the dynamics considered by Gabay and Garel[3,7] for the various, possible branching schemes. These effects can perhaps be realized better by the present method than by optical means.

The authors are indebted to W. Assmus, Frankfurt-am-Main, for providing them with the crystals and to R. Pommier and P. Meyer, Orsay, for help with the optical measurements.

[1]L. Landau and E. Lifshitz, Phys. Z. Sowjetunion **8**, 153 (1935).

[2]T. Garel and S. Doniach, Phys. Rev. B **26**, 325 (1982).

[3]M. Gabay and T. Garel, J. Phys. (Paris) **46**, 5 (1985).

[4]W. A. Barker and G. A. Gehring, J. Phys. C **19**, 259 (1986); G. A. Gehring, J. Magn. Magn. Mater. **54–57**, 843 (1986).

[5]J. Pommier, P. Meyer, J. Ferré, and I. Laursen, J. Phys. (Paris), Colloq. **49**, C8-915 (1988).

[6]J. Rajchenbach, J. Phys. C **21**, L447 (1988).

[7]M. Gabay and T. Garel, Phys. Rev. B **33**, 6281 (1986).

[8]A. Schlachetzki and J. Eckert, Phys. Status Solidi (a) **11**, 611 (1972).

[9]W. L. C. Rutten and J. C. Verstelle, Physica (Amsterdam) **86–88B**, 564 (1977).

[10]P. H. Müller, M. Schienle, and A. Kasten, Phys. Status Solidi (b) **119**, 239 (1983).

[11]J. Kötzler, D. Sellmann, and W. Assmus, J. Magn. Magn. Mater. **45**, 245 (1984).

[12]M. Grahl and J. Kötzler, Z. Phys. B **75**, 527 (1989).

[13]L. W. Bulaevskii and V. L. Ginzburg, Zh. Eksp. Teor. Fiz. **45**, 772 (1963) [Sov. Phys. JETP **18**, 530 (1964)].

[14]L. Holmes, T. Johansson, and H. J. Guggenheim, Solid State Commun. **12**, 993 (1973).

[15]R. Frowein, J. Kötzler, and W. Assmus, Phys. Rev. Lett. **42**, 739 (1979).

[16]J. Als-Nielsen, Phys. Rev. Lett. **37**, 1161 (1976).

[17]R. Gemperle, A. Gemperle, and I. Bursuc, Phys. Status Solidi **3**, 2101 (1963).

[18]J. A. Griffin and J. D. Litster, Phys. Rev. B **19**, 3676 (1979).

[19]J. Kaczér, IEEE Trans. Magn. **6**, 442 (1970).

[20]J. Kötzler, J. Magn. Magn. Mater. **54–57**, 649 (1986).

[21]From magnetization data published by R. Frowein and J. Kötzler, Phys. Rev. B **25**, 3292 (1982).

[22]A. Froese and J. Kötzler, in *Proceedings of the Seventeenth International Conference on Low Temperature Physics,* edited by U. Eckern *et al.* (Elsevier, New York, 1984), p. 183.

[23]K. S. Cole and R. H. Cole, J. Chem. Phys. **9**, 341 (1941).

[24]This ratio implies that, even for the present minimum values of t and D, there are well-defined domains with $\xi_- \approx 0.02d$, so that interactions between walls (Ref. 4) play no role.

[25]J. Kötzler, H. Neuhaus-Steinmetz, A. Froese, and D. Görlitz, Phys. Rev. Lett. **60**, 647 (1988).

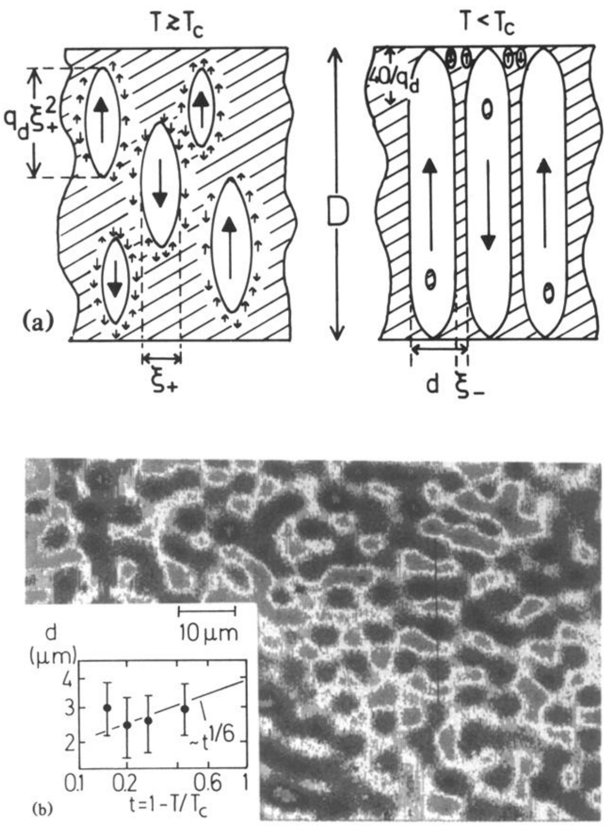

FIG. 1. (a) Cross sections through a plate of a strongly uniaxial ferromagnet illustrating short-range order above T_c and needle-shaped long-range order below T_c, both imbedded by (hatched) seas of disorder. (b) The domain pattern in the bulk of a $LiTbF_4$ plate of thickness 0.4 mm observed by Faraday rotation at $T = 2.12$ K and $H = 0$. Black and grey regions represent the two domain types.