Evidence for Critical Fluctuations in Bloch Walls near Their Disordering Temperature

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The temperature variation of the relaxation rate Γ_w of the domain walls about their transition at $T = T^*$ from the Bloch to the linear Bulaevskii-Ginzburg structure has been investigated in the uniaxial ferrimagnet $SrFe_{12}O_{19}$. The significant suppression of T^* below the mean-field approximation (MFA) value along with the speeding up of $\Gamma_w(T \leq T^*)$, which is much stronger than that predicted by the MFA, reveals the first evidence for critical fluctuations in a domain wall. The speeding up can be almost quantitatively explained by taking two dimensional fluctuations of the transverse magnetization in the wall into account.

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A quite recent investigation [1] of the domain wall dynamics near the Curie temperature of the well-known hexaferrite $BaO{\cdot}6Fe_2O_3$ [2] revealed the first clear signatures for the structural transformation of the Bloch wall (BW) to the so-called linear wall (LW). This continuous phase transition of the BW has been predicted by earlier work of Bulaevskii and Ginzburg to occur at some elevated temperature T_0^* in uniaxial ferromagnets and ferroelectrics [3] and also in ferrimagnets [4], where the walls link adjacent domains with up and down orientations of the order parameter $M_s(T)$. Basically, the instability of the BW arises from the nonsingular behavior of the transverse susceptibility in uniaxial materials, $\chi_\perp = M_s/H_A$, which in contrast to cubic systems remains constant when passing the Curie temperature T_C. As a result of the diverging longitudinal susceptibility the energy density associated with the longitudinal magnetization $M_z(x)$ in the wall, $M_s^2/2\chi_\parallel(T)$, may fall below that of the transverse component $2M_s^2/\chi_\perp$. Then, according to the mean-field approach (MFA) used by Bulaevskii and Ginzburg [3] the LW, which is characterized by a zero transverse magnetization, $M_y(x) = 0$, becomes stable above the transition temperature T_0^* given by $4\chi_\parallel(T_0^*) = \chi_\perp$. Below T_0^*, the transverse magnetization M_y becomes finite and assumes the maximum $M_y(0)$ in the center of the wall ($x = 0$). We consider the quantity $M_y(0)/M_s \equiv m_B$ as the order parameter of the BW, since approaching saturation at low temperatures, $m_B \to 1$, the pure circular structure usually associated with the BW is attained, whereas at finite temperatures the wall structure becomes elliptical; see Fig. 1.

Experimental evidence for the existence of LW's close to T_C was provided by previous investigations of the wall relaxation in uniaxial ferromagnets with low Curie temperatures, like $GdCl_3$ [5] and $LiTbF_4$ [6]. Based on a relaxational ansatz, $\dot{M}_z(x) = L_\parallel \Delta H(x)$, for the local magnetization in the LW of width δ and spacing d the resulting kinetic coefficient of the total magnetization,

$$L_w = 3\ L_\parallel(\delta_L/d)\ , \tag{1}$$

could quantitatively describe the observed [5–7] variations of the wall damping, $\Gamma_w = L_w N_\parallel$, with temperature, magnetic field, sample thickness D, and sample shape, i.e., demagnetization coefficient $N_\parallel$. The most significant signal of the LW was the thermal critical behavior, $\Gamma_w \sim (T_C - T)^{-0.8}$, which is due to the facts that its width is determined by the diverging correlation length [3] $\delta_L = \xi_0(1-T/T_C)^{-\nu}$ with $\nu = \gamma/(2-\eta) \approx \gamma/2$ for three dimensional systems, and that the domain period shrinks as $d \approx (D^2\delta_L)^{1/3}$ if branching at the surface is taken into account. We should note that the slight variation of $d(T)$ on approaching T_C has been considered by Stauffer [8] as a signal of the LW; however, experimental efforts to detect this temperature effect on the orthoferrit $YFeO_3$ [9] and on $LiTbF_4$ [6] using the Faraday-rotation method yielded no clear evidence. The

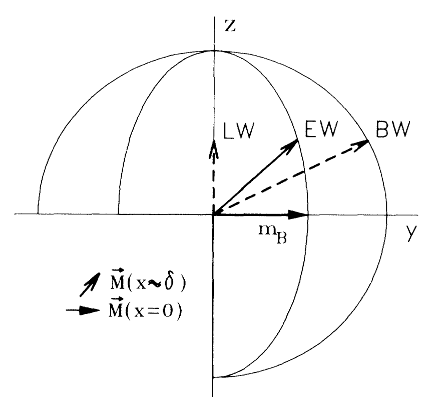

FIG. 1. Magnetization profiles $\mathbf{M}(x)$ in domain walls of uniaxial materials: Bloch wall (BW, $m_B = 1$), elliptic wall (EW, $0 < m_B < 1$), linear wall (LW, $m_B = 0$) with the order parameter of the Bloch wall, $m_B = M_y(0)/M_s$, defined by the reduced magnetization in the wall center.

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direct measurement of δ_L, usually ranging between 1 nm and 100 nm, is still lacking. A promising technique on this route may be the tunneling stabilized magnetic force microscopy, by which quite recently at room temperature on the same material as in Ref. [1], $BaO{\cdot}6Fe_2O_3$, a lateral resolution of 50 nm has been achieved [10]. In Ref. [1] a deep minimum of Γ_w in Ba-hexaferrite was observed at $T^* = 0.99T_C$, which, resting on a recent general theory on the wall dynamics about T_0^* [11,12], could qualitatively be related to the transition from the LW to the BW structure.

The present work has been motivated by two interesting but unexplained phenomena in Ref. [1]: (i) below T^*, the relaxation rate increased much faster than predicted by the existing kinetic theory [11,12] based on the MFA for elliptical walls [3] and (ii) the observed disordering transition of the BW occurred at a significantly lower temperature than the MFA value T_0^*. One rather general conjecture in Ref. [1] attributed these features to critical fluctuations near the wall transition. Fluctuation effects were considered before in theoretical work by Lawrie and Lowe [13], who predicted the BW order parameter to increase like that of the strongly fluctuating two dimensional Ising system, $m_B \sim (T^* - T)^{1/8}$, being accompanied by the strongly divergent susceptibility, $\chi_B \sim |T^* - T|^{-7/4}$.

In order to search for these fluctuations, we have chosen a high-quality single crystal of $SrO{\cdot}6Fe_2O_3$ [14], the static susceptibility of which obeyed the critical divergence, $\chi_\| \sim t^{-1.18}$, to reduced temperatures as small as $|t| = |1 - T/T_C| = 2 \times 10^{-4}$ without showing rounding effects. We also extended the frequency range by almost 2 orders of magnitude over that of Ref. [1] to investigate the speeding up of the wall relaxation in more detail. The present ac susceptibility data shown in Fig. 2(a) have been obtained using a well balanced mutual inductance system allowing measuring frequencies up to more than 20 MHz. Filled and empty sytems were immersed in a shielded, Cs-driven heat pipe [15], which allowed a temperature stabilization of better than 0.02 K about the Curie temperature of Sr-hexaferrite, $T_C = 750.0(2)$ K. For excitation amplitudes of $H_{\mathrm{ac}} = 0.7$ Oe applied here, the sample response proved to be linear. The frequency dependence of $\chi'(\omega)$, shown in Fig. 2(b), could be fairly well described by the sum of two Debye functions because the wall relaxation rate Γ_w is much slower than that of the homogeneous phase, Γ_d :

$$\chi(\omega) = \frac{N_\|^{-1} - \chi_T}{1 + i\omega/\Gamma_w} + \frac{\chi_T}{1 + i\omega/\Gamma_d}. \quad (2)$$

Γ_d undergoes the thermodynamic slowing down, $\Gamma_d = L_\|/\chi_T$, as expected for uniaxial ferromagnets [16], and the kinetic coefficient of the longitudinal magnetization, $L_\| = 60(2) \times 10^6$ s^{-1}, proved to be the same on both sides of T_C. The internal susceptibilities diverge as $\chi_\|^\pm = [\chi_T^{\pm\,-1} - N_\|]^{-1} = C \pm |1 - T/T_C|^{-\gamma}$ with $\gamma = 1.19(1)$ being close to exponents measured on Ba-hexaferrite, $\gamma = 1.18(3)$ [1], and the uniaxial ferromagnet Gd, $\gamma = 1.19(4)$ [17]. The amplitudes are $C_+ = 1.4 \times 10^{-4}$ and $C_- = 0.7 \times 10^{-4}$ above and below T_C, respectively.

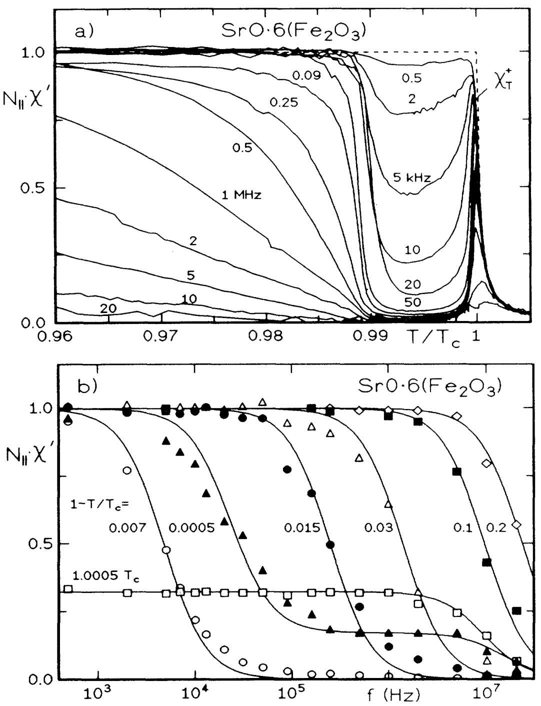

FIG. 2. Real part of the dynamic susceptibility measured along the easy (hexagonal) axis of a $SrO{\cdot}6Fe_2O_3$ crystal with demagnetization coefficient $N_\| = 0.43$: (a) temperature dependence about $T_C = 750.0(2)$ K and (b) frequency dependence. Full curves in (b) are fits by Eq. (2).

Below T_C, the data can be more accurately fitted to the so-called Cole-Cole function $\chi(\omega) \sim 1/[1 + (i\omega/\bar{\Gamma}_w)^{1-\alpha}]$ used in previous analyses of wall relaxation [5,6]. Since $\alpha \leq 0.1$ the mean relaxation rate $\bar{\Gamma}_w$ does not differ from Γ_w defined by Eq. (2). A deviation from the relaxational shape due to wall inertia (see, e.g., Ref. [2]) is not seen. As in the previous work [1,5–7] we extract from the measured rate Γ_w the quantity of central interest, i.e., the kinetic coefficient of the wall damping [5], $L_w = \Gamma_w/[N_\|(1 - N_\|\chi_T^-)]$. Figure 3 shows a deep minimum of $L_w(T)$ at $T^* = 0.99\,T_C$ produced by a weaker (stronger) speeding up towards higher (lower) temperatures. Based on the previous results [1,5,6], the critical behavior near T_C, $L_w = 428$ s$^{-1}(1 - T/T_C)^{-0.89}$, is to be associated with the presence of LW's, and the exponent indicates the presence of closure domains at the surface terminating the up and down domains [18]. On the other hand, the speeding up of L_w observed below T^* cannot be described by a power law in $(T_C - T)$: Figure 3 shows for comparison the classical result by Landau and

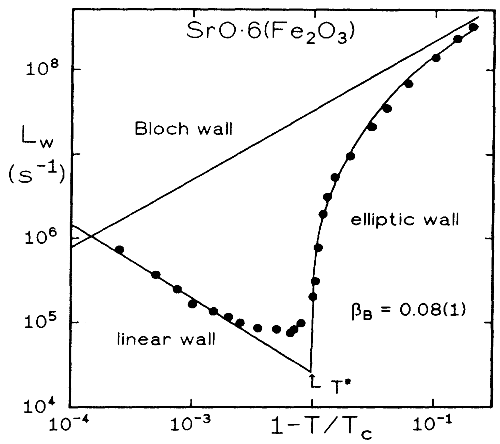

FIG. 3. Temperature variation of the kinetic coefficient of the wall relaxation. Full curves correspond to fits of the linear wall ($T > T^*$) and elliptical wall dynamics, Eq. (3), including fluctuations of the transverse wall magnetization ($T < T^*$).

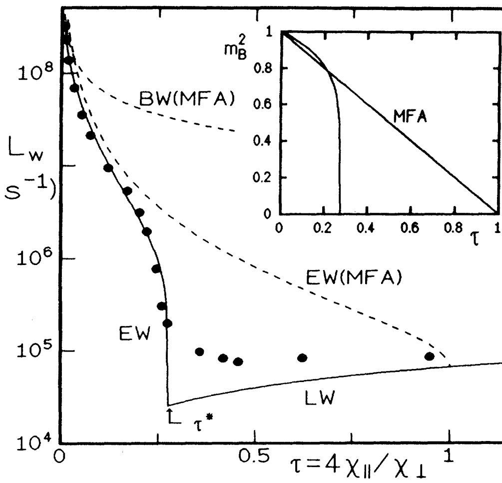

FIG. 4. Variation of the kinetic coefficient about the transition from the linear (LW) to the elliptical (EW) wall structure in terms of the reduced longitudinal susceptibility (using $\chi_\perp = 0.25$ from Ref. [20]). Note the strong suppression of the actual transition τ^* against the MFA value $\tau = 1$ and the significant deviation between the L_w's of the Bloch walls (BW, Ref. [19]) and the EW's as predicted by Eq. (3) using the MFA , Eq. (4), and the present critical law, Eq. (4a), for m_B.

Lifschitz (LL) [19] for the BW, $L_w = 2(\gamma M_s)^2\delta_B/L_\perp d$, calculated with parameters determined below. There is an extremely striking deviation from the data, reaching more than 2 orders of magnitude near the transition temperature T^*.

In order to associate this essential observation with the disordering of the BW we refer to the recent kinetic theory [11,12] based on the LL-Bloch equations, which has determined the wall mobility of the domain walls μ_w in the entire temperature range below T_C. Using the relation $L_w = 2M_s\mu_w/d$ [5], the result of the theory can be written as

$$L_w = \frac{2L_\parallel \delta/d}{\frac{2}{3} + \frac{m_B^2}{3} - \frac{m_B}{\sqrt{1-m_B^2}}\tan^{-1}\left(\frac{\sqrt{1-m_B^2}}{m_B}\right) + \frac{L_\parallel}{L_\perp} m_B f}, \tag{3}$$

where

$$f = 1\Big/\sqrt{1-m_B^2}\,\tan^{-1}\left(\sqrt{1-m_B^2}/m_B\right) - m_B\Big/\sqrt{(\alpha_\perp^2+m_B^2)(1-m_B^2)}\,\tan^{-1}\left[\sqrt{(1-m_B^2)/(\alpha_\perp^2+m_B^2)}\right]$$

with the Gilbert parameter $\alpha_\perp = L_\perp/\gamma M_s$ (for Sr-ferrite $\alpha_\perp \ll 1$). Above T^*, where $m_B = 0$ and $\delta = \delta_L$, the speeding up associated with the LW's is reproduced [see Eq. (1) and for more details Refs. [1,18]]. In the other limit, $m_B \to 1$, one finds $L_w \sim [\alpha_\parallel^{-1}(8/15)(1-m_B)^2 + \alpha_\perp/(1+\alpha_\perp^2)]^{-1}$. This is the classical result by LL [19] supplemented by an ellipticity term, containing the longitudinal Gilbert parameter $\alpha_\parallel = L_\parallel/\gamma M_s$. As a result of $\alpha_\parallel \le \alpha_\perp$ and $\alpha_\perp \ll 1$ in Sr-hexaferrite, the ellipticity provides a significant contribution to the wall relaxation even for $m_B \approx 1$. Physically this arises from the fact that during the passage of elliptic walls (EW's) at a plane at some $x = x_0$, a change of the modulus of the local $\mathbf{M}(x_0)$ determined by small $\alpha_\parallel$ is required. This implies that except for very small $1 - m_B$ the classical LL mechanism, corresponding to the last term in the denominator in Eq. (3), is dominated by the longitudinal relaxation mechanism.

To compare this prediction with the data we start in a first approximation from the MFA results [3] for the order parameter,

$$m_B^{MF}(T) = [1-\tau(T)]^{1/2}, \quad \tau(T) = 4\chi_\parallel(T)/\chi_\perp, \tag{4}$$

and for the width of the wall, $\delta = \delta_B = \delta_L(T_0^*)$. The temperature independence of δ implies the same for the domain width d. Figure 4 shows that Eq. (4) fits the data much better than the classical LL result for pure BW's. The sharp drop of L_w for $T \nearrow T^*$ corresponding to $\tau \nearrow \tau^* = 0.27$, however, is not reproduced and quite naturally this discrepency suggests an improved approach: We consider the effect of fluctuations on the order parameter of the BW m_B but keep the width δ constant because δ_B does not depend on m_B in the MFA. In this spirit, we expect the leading effect of fluctuations on L_w [Eq. (4)] to arise from

$$m_B(T) = [1-\tau/\tau^*]^{\beta_B}, \quad \tau^* = 4\chi_\parallel(T^*)/\chi_\perp, \tag{4a}$$

with $\beta_B < \beta_B^{MF} = 1/2$ and the transition point $\tau^* < \tau^*_{MF} = 1$. In fact, Fig. 4 demonstrates that the fit of this ansatz to the data below $\tau^* = 0.27$ becomes almost perfect. Note that the critical exponent $\beta = 0.08(1)$ is the only parameter determining the temperature-dependent kinetic coefficient for EW's near the disordering "temperature" τ^*. The strong reduction of both τ^* and β_B with respect to the MFA values signals large fluctuation effects on m_B. At lower temperatures, the classical LL mechanism of the DW damping becomes dominant and we get $L_\perp = 1.2 \times 10^9$ s^{-1} from our fit, in good agreement with $L_\perp = 0.9 \times 10^9$ s^{-1} obtained from resonance data [20].

It is interesting to note that the exponent β_B turns out to be even smaller than $\beta_{2D} = 1/8$. This value was predicted by Lawrie and Lowe [13], who proposed the BW to belong to the universality class of the two dimensional Ising system. This assignment is plausible provided (i) the correlation length of m_B, ξ_B, is much larger than the width of the wall δ and (ii) the fluctuations of the transverse magnetization $M_x(x)$ are suppressed by the magnetostatic energy. We do not know $\xi_B(T)$, but in some vicinity of T^* one can safely assume $\xi_B > \delta_B$ due to the divergence of $\xi_B \sim (T^* - T)^{-\nu_B}$. The fluctuations against the magnetostatic self-field of the wall, $\mathbf{H} = -M_s\mathbf{e}_x$, are suppressed over those parallel to $\mathbf{e}_y$ (see Fig. 1) by an amount $(M_s + H_A)/H_A$ which, using $\chi_A = M_s/H_A = 0.25$ [20], is 1.25, so that such additional XY-like fluctuations can be ruled out rather close to T^*. However, they might affect m_B at the lower temperatures of our fits, which then would be one reason for the result $\beta_B < \beta_{2D}$.

Another possible source for the smallness of β can be the effect of the fluctuations on the width of the EW in Eq. (3) which has been fixed to $\delta_B^* = \delta_L(\tau^*)$ in the fit. Because of the large suppression of the transition, $\tau^* = 0.27$, this width is smaller than that of the pure Bloch wall, $\delta_B = \delta_L(\tau = 1)$, so that below τ^* the fit by Eq. (3) had implicitly accounted for the rise of $\delta_B(T)$ with lowering temperature from δ_B^* at τ^* to δ_B at $\tau = 0$, leading to a lower effective exponent β_B. Generally, one expects the effect of fluctuations of m_B on the widths of both BW's and LW's to be strongest very close to T^*, so that also the discrepancies between the data and the fit by Eq. (4) seen in Figs. 3 and 4 above T^* and τ^*, respectively, may be related to the presence of a short-range order of m_B. To our knowledge, a theoretical treatment of fluctuation effects on δ is not yet available. Such a theory should probably also consider the (perhaps minor) effect of the fluctuations on the generalized LL-Bloch equations [12] from which Eq. (3) was derived. We believe it would not be necessary to consider fluctuation effects on $L_\parallel$ since it was shown above that $L_\parallel$ is unchanged when passing T_C.

Future experimental investigations of fluctuation effects on domain walls should be devoted to pure uniaxial ferromagnets like Gd and also to ferroelectrics. On the hexaferrites, the present work will be extended to study the following effects on the wall dynamics: (i) of a transverse magnetic field H_y, which due to $m_B \sim H_y^{1/\Delta}$ with $\Delta \approx 15$ at $\tau = \tau^*$ should be very pronounced, (ii) of a static longitudinal field H_z, which increases the domain period d [2], and (iii) sample size effects using thinner crystals and small particles.

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