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# A modified scaling law for 180° stripe domains in ferroic thin films

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The periodicity of 180° stripe domains has been calculated analytically by assuming a linear change in magnetization/polarization in the domain wall. A modified scaling law was proposed between the domain period and the film thickness for ferroelectric and ferromagnetic thin films. Both the slope and intercept of this scaling line can be used to get domain wall thickness in a consistent manner, which enriches the understanding of domain walls. Theoretical results have been used to compare with recent experimental data and suggest a good agreement. © 2009 American Institute of Physics. [DOI: 10.1063/1.3055355]

## I. INTRODUCTION

Ferroic ultrathin films, i.e., ferroelectric, ferromagnetic, or ferroelastic thin films, attracted much attention from both scientific and application points of view.<sup>1–3</sup> As the film thickness  $t$  approaches tens of unit cell length, the films show significantly different physical properties from those of bulk materials. In particular, it is found that the evolution of the domains plays important roles in the switching process and phase transitions,<sup>1–4</sup> which is vital to the application in nanoscale memories, capacitors, and tunnel junctions.

Ferroic materials usually display domain structures due to elastic, magnetic, or mechanical boundary conditions. The theory of domain structure that minimizes the total energy was worked out analytically for ferromagnetic crystals by Kittel in 1946.<sup>5,6</sup> The magnetization in the crystal has been approximated by a square wave of amplitude  $Q_0$  and period  $D$ , as shown in Fig. 1. The magnetic field in the space is given by the solution of the Laplace equation, with  $z$  and  $x$  components given by<sup>5</sup>

$$H_z = \sum_n c_n \sin kx \exp(-kz), \quad (1)$$

$$H_x = \sum_n c_n \cos kx \exp(-kz), \quad (2)$$

where  $n$  is an odd number,  $k = 2n\pi/D$ , and  $c_n = 8Q_0/n$ .

The total energy per unit area is given by<sup>5</sup>

$$F = 0.85Q_0^2 D + 2\sigma_w t/D, \quad (3)$$

where the first term denotes the contribution from the surface energy while the second term is the domain wall energy.  $\sigma_w$  is the domain wall energy density. The domain period  $D$  given by minimization of the total energy [Eq. (3)] could be obtained as

$$(D^k)^2 = 4\sigma_w t/1.7Q_0^2. \quad (4)$$

Thus the equilibrium value of the stripe period  $D$  is directly proportional to the square root of the crystal thickness  $t$ , which is called as Kittel's law in literature. Here a superscript  $k$  is used to denote the parameters given by the Kittel's law. This law was extended by Mitsui and Furuichi in 1953 to ferroelectric crystals<sup>7</sup> and by Roytburd in 1976 to epitaxially ferroelastic ones.<sup>8</sup> Catalan *et al.*<sup>9</sup> lately showed that the domain size of multiferroic BiFeO<sub>3</sub> scales with an exponent of around 0.59 rather than exactly 1/2. In all of the above, the domain wall is assumed to be negligible in comparison with the domain period and a square wave approximation was adopted.

## II. A LINEAR WALL MODEL

As a matter of fact, equilibrium 180° stripe domains were recently observed in ultrathin films, where the stripe

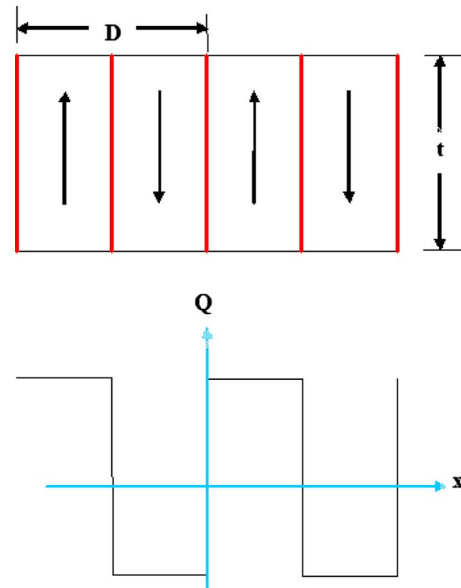


FIG. 1. (Color online) Schematic demonstration of periodic stripe domains for thick films, where the domain wall is marked by red. The domain wall width  $d$  is negligible in comparison with the domain period  $D$  and a square wave approximation adopted by Kittel (bottom panel) is valid.

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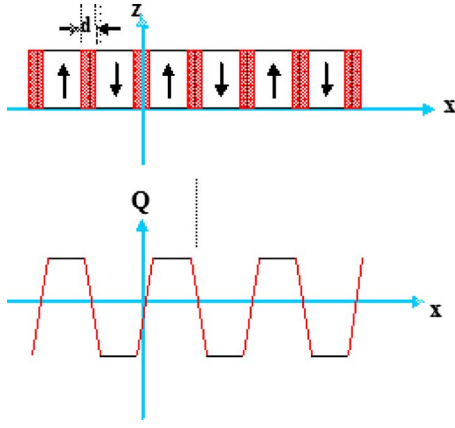


FIG. 2. (Color online) Schematic demonstration of periodic stripe domains for ultrathin films where  $d \sim D$  and the square wave approximations are not appropriate. The bottom panel shows the periodic change of magnetization  $M$  adopted in the present work.

period is of the same order as the thickness.<sup>3,6</sup> As illustrated in top panel of Fig. 2, the domain wall width  $d$  of these ultrathin films is comparable to domain period  $D$  and cannot be ignored for this “wide” domain case. Conventionally, wide acceptance for  $d \ll D$  were taken in “dense” or “narrow” domain approximations.<sup>10</sup> As a result, the square wave approximation illustrated in Fig. 1 is not appropriate for discussions for ultrathin films.

On the other hand, the one-dimensional domain wall structure with free boundary condition was worked out by Landau and Lifshitz (LL):<sup>11</sup>

$$Q = Q_0 \tanh(2x/d), \quad (5)$$

where  $d$  is the domain wall width. The LL domain structure as shown in Fig. 3 describes one domain wall of infinite crystal and has been adopted recently by many to explore both the static<sup>12</sup> and dynamic<sup>13</sup> behavior of domain wall in granular materials. However, it does not reflect the periodicity of domain structure in thin films. In this letter we make a compromise between Kittel’s simple square wave approach and the more complicated LL wall by assuming a zigzag

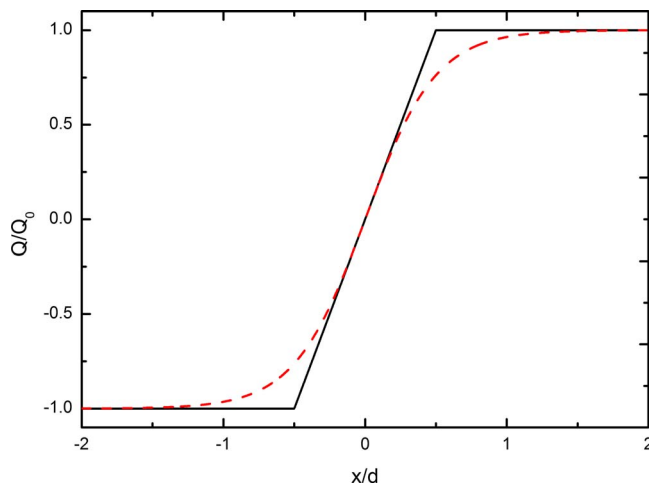


FIG. 3. (Color online) Comparison of the LL domain wall (dashed line) and the linear domain wall approximation adopted in the present work.

domain structure of period  $D$ . The square wave has been kept at the domain center, i.e.,

$$Q = -Q_0 \left( \frac{d-D}{2} < x < -\frac{d}{2} \right),$$

$$Q = Q_0 \left( \frac{d}{2} < x < \frac{D-d}{2} \right).$$

On the other hand, the core of the LL expression is approximated by a linear change in magnetization (Fig. 2),

$$Q = 2Q_0 \frac{x}{d} (|x| < d/2),$$

$$Q = -2Q_0 \frac{x-D/2}{d} \left( \left| x - \frac{D}{2} \right| < d/2 \right). \quad (6)$$

Such zigzag domain structure is repeated in the  $x$  direction of the film. This linear domain wall has been observed experimentally.<sup>14</sup> Such approximation separates the domain center and the wall clearly, which offers direct physical understanding for the contribution of the domain wall.

The solution of the Laplace equation gives the same formula for the magnetization field as that of Eqs. (1) and (2), but with a different coefficient

$$c_n = \frac{8Q_0 \sin(n\eta\pi)}{n \eta\pi}. \quad (7)$$

Here  $n$  is an odd number and  $\eta = d/D$  with  $2\eta$  corresponding to the volume occupation of the domain wall. One could check that when  $d=0$ ,  $c_n = 8Q_0/n$ , and the Kittel’s law could be recovered.

The surface energy density could be obtained as

$$\sigma_s = \sum_n \left( \frac{\sin n\eta\pi}{n\eta\pi} \right)^2 \sigma_n^k, \quad (8)$$

where  $\sigma_n^k$  is the energy given by Kittel through a square wave approximation, i.e.,  $\sigma_n^k = (8/\pi^2 n^3) Q_0^2 D$ . The higher order terms ( $n > 1$ ) are trivial, which could be dropped, thus Eq. (8) could be simplified as

$$\sigma_s = 1.05 \left( \frac{\sin \eta\pi}{\eta\pi} \right)^2 \sigma_1^k, \quad (9)$$

where a factor of 1.05 has been adopted to compensate the dropped higher order terms.<sup>5</sup> For small  $\eta$ , the Taylor expansion of  $\sigma_s$  at  $\eta=0$  leads to

$$\sigma_s = 1.05 \left( 1 - \frac{\eta^2 \pi^2}{3} \right) \sigma_1^k, \quad (10)$$

where only the lowest-order term has been kept. The reduced energy  $\sigma_s/\sigma_1^k$  as functions of  $\eta$  given by Eqs. (8) and (10) have been shown in Fig. 4. As  $\eta$  increases from 0 to 0.5,  $\sigma_s$  given by Eq. (8) decreases from  $1.05 \sigma_1^k$  to  $0.42 \sigma_1^k$ . For  $\eta < 0.3$ , the linear approximation given by Eq. (10) agrees well with Eq. (8), justifying the validity of Eq. (10). For larger  $\eta$  values, the two curves deviate from each other significantly and Eq. (8) has to be used instead. Substituting the first term

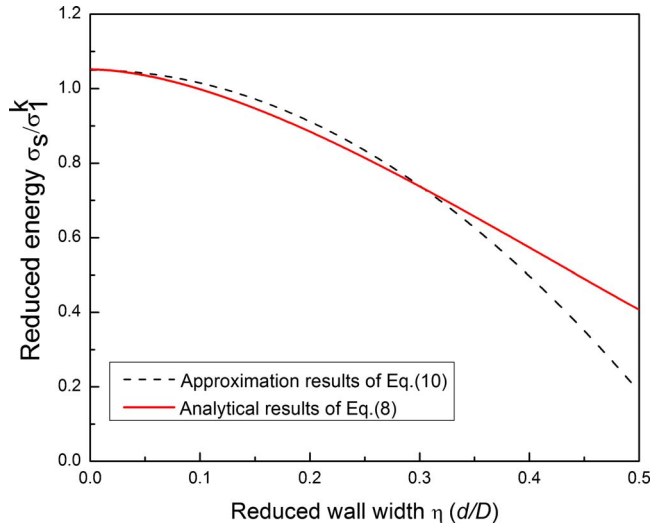


FIG. 4. (Color online) Reduced surface energy as a function of the reduced domain wall width  $d/D$ . Shown for comparison (dotted line) is the approximation.

of Eq. (3) by the right hand of Eq. (10) and minimizing the energy yields

$$D = \sqrt{(D^K)^2 - \frac{\pi^2 d^2}{3}}, \quad (11)$$

where  $D^K$  denotes the domain period predicted by Kittel.<sup>5</sup> A more general formula for  $D^K$  has been derived by Kooy and Enz<sup>15</sup> with the susceptibility anisotropy considered as

$$(D^K)^2 = (1 + \sqrt{\mu})\sigma_w t / (0.85Q_0^2), \quad (12)$$

where  $\mu = 1 + 2\pi Q_0^2/K$  and  $K$  is the anisotropy constant. Equation (11) shows that the linear relationship between  $D^2$  and  $t$  sustains when a linear domain wall is incorporated. But the  $D^2$  versus  $t$  curve does not pass the origin anymore. Rather, it has an intercept of  $-\pi^2 d^2/3$ . This is because for a domain wall profile we used has a finite width (with a linear change in the order parameter across the wall), the bulk energy term is calculated to be lower than the Kittel's value (with a zero-width domain wall profile), by a factor that depends on the ratio of the wall width to the stripe domain period. This introduces an extra term into the equilibrium relationship between stripe period and film thickness, so that the stripe period (for films not too thin relative to the wall thickness) should extrapolate to a negative domain period at zero film thickness.

For ferroelectric materials Eq. (11) holds as well, with  $D^K$  given by

$$(D^K)^2 = [\xi\epsilon_{\text{ex}} + (\epsilon_a\epsilon_c)^{1/2}]\sigma_w t / (0.85Q_0^2), \quad (13)$$

where  $\xi$  is a dimensionless parameter between 0 and 1,  $\epsilon_{\text{ex}}$  is the substrate's dielectric constant,  $\epsilon_a$  and  $\epsilon_c$  are the dielectric constants of the crystal in the  $x$  and  $z$  directions respectively. For a ferroelectric thin film with symmetric substrates at both surfaces,  $\xi$  is 1 and the formula given by Streiffer *et al.*<sup>16</sup> and Bjorkstam and Oettel<sup>17</sup> could be recovered. On the other hand, for a ferroelectric thin film in free space  $\xi\epsilon_{\text{ex}}=1$  and Eq. (13) reduces to the formula obtained by Mitsui and Furuichi.<sup>7</sup>

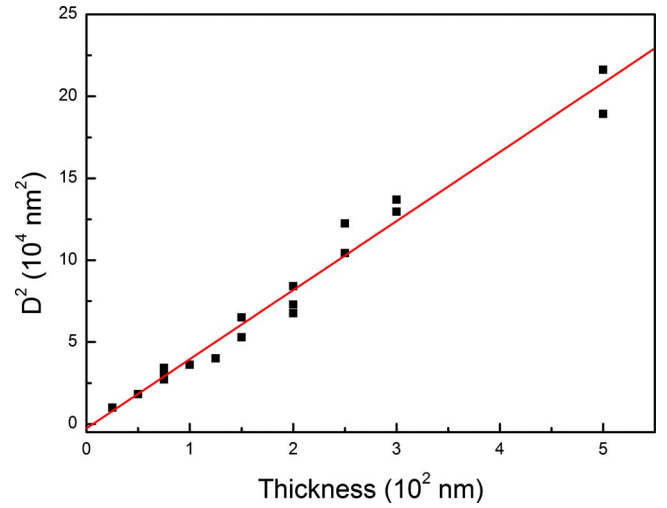


FIG. 5. (Color online) Thickness dependence of  $D^2$  for cobalt thin films. The straight line is the linear fit of the experimental data.

The energy density of the domain wall in Eq. (11) could be calculated, which is

$$\sigma_w = \frac{2}{3}dQ_0^2/\chi_c, \quad (14)$$

$$\sigma_w = dQ_0^2/\chi_c, \quad (15)$$

for ferroelectric and ferromagnetic material, respectively.<sup>6</sup> Thus Eq. (11) could be rewritten as

$$D^2 = \rho dt - \frac{\pi^2 d^2}{3}, \quad (16)$$

where  $\rho = 4.9[\xi\chi_{\text{ex}}/\chi_c + (\chi_a/\chi_c)^{1/2}]$  for ferroelectric materials and  $\rho = 7.4(\sqrt{\mu} + 1)/(\mu - 1)$  for ferromagnetic materials.  $\chi_a$  and  $\chi_c$  are the susceptibilities perpendicular and parallel to the film surface, while  $\chi_{\text{ex}}$  is the susceptibility of the substrate. This scaling law has obvious physical meaning and holds for both ferroelectric and ferromagnetic materials.

### III. DISCUSSIONS AND CONCLUSIONS

Figure 5 shows the experimental data for Co thin film obtained by Hehn *et al.*<sup>18</sup> It can be seen that a linear relationship does exist between the square of the domain period  $D^2$  and the film thickness  $t$  when  $t$  changes from 25 to 500 nm. Similar experimental data were obtained by Dumas *et al.*<sup>19</sup> The slope and intercept of the fitted line are  $4.2 \times 10^2$  and  $-2.6 \times 10^3 \text{ nm}^2$ , respectively. One could derive the domain wall width of cobalt from the intercept directly, which is about 28 nm, which is in between the previously reported experimental and theoretical values.<sup>6,20</sup> The domain wall energy could be derived from the slope also based on Eq. (12). Taking  $K = 4.6 \times 10^6 \text{ erg/cm}^3$  and  $Q_0 = 1.4 \text{ kOe}$ , one arrives at  $\sigma_w = 23 \text{ erg cm}^{-2}$ , in good agreement with those given by Hehn *et al.* ( $\sigma_w = 25 \pm 3 \text{ erg cm}^{-2}$ ) (Ref. 18) and by Dumas *et al.* ( $\sigma_w = 22 \pm 3 \text{ erg cm}^{-2}$ ).<sup>19</sup> From this wall energy we could obtain the wall width, which varies for different kind of domain walls. For an LL wall, the typical value of  $d$  is  $\sigma_w/(2k)$ , i.e., 25 nm derived from the slope of the Fig. 5, which is close to that derived from the intercept, demonstrating the self-consistency of the present model.

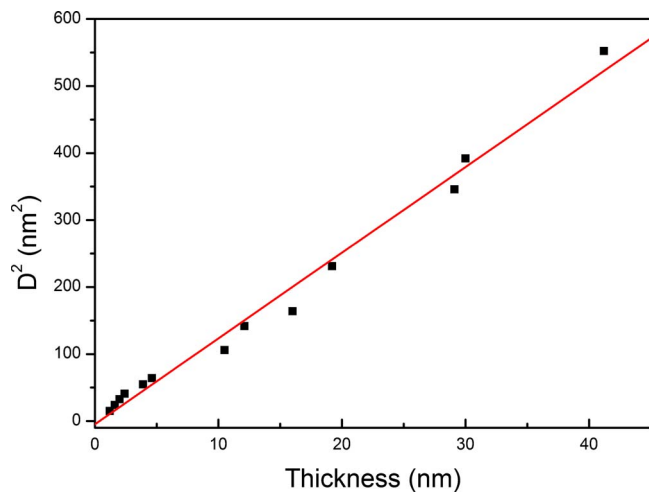


FIG. 6. (Color online) Thickness dependence of  $D^2$  for PbTiO<sub>3</sub> thin films. The straight line is the linear fit of the experimental data.

Figure 6 shows the experimental data of PbTiO<sub>3</sub> thin film ( $\beta$ -phase) given by Fong and Streiffer *et al.*<sup>3,16</sup> A good linear relationship exists between the square of the domain period  $D^2$  and the film thickness  $t$  from  $t=41$  nm down to  $t=1.2$  nm. The slope and intercept of the fitted line are 13 nm and  $-4.6$  nm<sup>2</sup>, respectively. One could derive the domain wall width of PbTiO<sub>3</sub> from the intercept, which is about 1.2 nm (three unit cells).

The domain wall width could be derived from the slope also based on Eq. (13). The experiment was conducted at  $T_c - 250$  K,<sup>16</sup> with the average temperature as 600 K. At this temperature,  $\epsilon_{\text{ex}} \approx 1.4(\epsilon_a \epsilon_c)^{1/2}$  and  $\chi_a/\chi_c \approx 1.6$ .<sup>21</sup> Taking  $\xi = 1/2$  (only one surface of the crystal is covered by the substrate), we have  $\rho = 8.1$  and  $d = 1.6$  nm. This domain width obtained is in reasonable agreement with that from the intercept, justifying the present model.

It has been a challenging problem to directly measure the domain wall thickness  $d$  in ferroics, especially in ferroelectrics. Catalan *et al.*<sup>6</sup> recently proposed a method to predict  $d$  from the slope of the  $D^2$  and  $t$  curve based on the original Kittel's law. The present analysis provides a more robust way as we could calculate and compare  $d$  from both the intercept and the slope of the curves. Although the relative uncertainties in the intercepts are larger than those in the slopes, one could derive the domain wall width  $d$  from the intercept directly while  $d$  obtained from the slope are subject to the accuracy of many other parameters, as could be seen from Eqs. (12) and (13). Combining the two methods will increase the reliability.

For example, our predicted value of  $d$  for PbTiO<sub>3</sub> is much smaller than the domain wall width of the magnetic material, supporting the view that ferroelectric domain walls are very thin. However, it is much larger than that derived by Catalan *et al.*, which is  $d=0.49$  nm.<sup>6</sup> Using the formula given by Streiffer *et al.*<sup>16</sup> and Bjorkstam and Oettel<sup>17</sup> and the susceptibilities at room temperature, we could recover Catalan's result, which, however, overestimates the role of the substrate. The predicted value of  $d$  for PbTiO<sub>3</sub> could be further justified by comparing with the first principle calculations. Lai *et al.*<sup>22</sup> found that  $D^2$  and  $t$  scales linearly for

PbTiO<sub>3</sub> thin films when  $t > 1.6$  nm based on a first principle calculation. Although the authors claimed that  $D^2$  is exactly proportional to the thickness  $t$  in the paper. A close examination of their figures shows that the slope and intercept for  $D^2$  versus  $t$  curve are 5.9 nm and  $-2.0$  nm<sup>2</sup>, respectively. The domain wall widths obtained from the intercept and the slope are 0.8 and 0.9 nm, respectively, also consistent with each other. Moreover, this value is in agreement with our predicted value when a temperature correction factor of  $(T_c/250)^{1/2} \approx 1.75$  is included (the wall width is roughly inversely proportional to the square root of  $T_c - T$ ). Such linear relationship between  $D^2$  and  $t$  (with nonzero intercept) holds for other ferroic stripe domains as well. For example, linear relationship between  $D^2$  and  $t$  has been obtained experimentally by Catalan *et al.*<sup>6</sup> for BiFeO<sub>3</sub>, by Schilling *et al.*<sup>23</sup> for BaTiO<sub>3</sub>, and Wu *et al.* for LaSrMnO<sub>3</sub>.<sup>24</sup> Close inspection of the results shows that nonzero intercept exists in all these results, indicating the universality of the present scaling law.

It should be noted that the present scaling law, i.e., Eq. (16), is valid only when  $d/D < 0.3$ , which is satisfied in most ferromagnetic and ferroelectric ultrathin films. For example, although the actual dimensions of film thickness and domain width in Figs. 5 and 6 are quite different, the corresponding values of  $d/D$  are both in the region of  $0.04 \sim 0.23$ . For films in the immediate vicinity of the phase transition, however, the domain wall width could be very large and this scaling law is not suitable. On the other hand, the linear approximation of the domain wall, as given by Eq. (6), is not subject to the abovementioned restriction and could be used to investigate various physical properties for thin films, including switching process and phase transition.

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