

## Domain Structure in a Superconducting Ferromagnet

M. Fauré<sup>1</sup> and A. I. Buzdin<sup>1,2</sup>

<sup>1</sup>Centre de Physique Moléculaire Optique et Hertzienne, Université Bordeaux I, UMR 5798, CNRS, F-33405 Talence Cedex, France

<sup>2</sup>Institut Universitaire de France, Paris, France

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The domain structure is inherent to all ferromagnets and the recent discovery of the superconducting ferromagnets raises the question of the modification of this domain structure by superconductivity. In the framework of the general London theory, applicable to both singlet and triplet superconductors, we demonstrate that superconductivity leads to a dramatic shrinkage of the domain width. The presence of this dense domain structure has to be taken into account for all magnetic measurements on superconducting ferromagnets, and the study of the domain structure evolution could provide important information on the mechanisms of superconductivity and magnetism interplay.

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Superconductivity and ferromagnetism are two antagonistic orderings in the sense that a magnetic field can destroy conventional superconductivity in two ways, via the orbital effect and via the paramagnetic effect, and therefore they usually tend to avoid each other. The competition between these two orderings and their (im)possible coexistence have always been a subject of great interest. In a pioneer work, the electromagnetic mechanism (orbital effect) of the superconductivity destruction in ferromagnetic systems was considered as soon as 1956 by Ginzburg [1]. The recent discovery of superconducting properties in the ferromagnet  $\text{UGe}_2$  when high pressure is applied [2] is obviously of primary importance for the understanding of superconductivity and ferromagnetism coexistence mechanisms. Besides, note that in such a compound, superconductivity appears in the ferromagnetic phase, so its critical temperature  $T_C$  is lower than the Curie temperature  $\Theta$ . This situation is therefore different from the previously studied case of the reentrant ferromagnetic superconductors  $\text{ErRh}_4\text{B}_4$  and  $\text{HoMo}_6\text{S}_8$  where the magnetic order emerges in the superconducting phase (see as a review [3]). In these materials, a nonuniform magnetic structure appears instead of ferromagnetism, as predicted by Anderson and Suhl [4]. Another superconducting ferromagnet (SFM) is  $\text{URhGe}$  [5] while  $\text{ZrZn}_2$  [6] and  $\text{Fe}$  under pressure [7,8] might also be promising candidates. Note also that the superconducting transition has been previously observed in the weak ferromagnet  $\text{R}_{1.5}\text{Ce}_{0.5}\text{RuSr}_2\text{Cu}_2\text{O}_{10}$  [9]. Moreover, it should be underlined that the type of pairing in SFMs is not yet known for sure. Although the singlet pairing has been proposed (see [10,11]), strong arguments support the triplet character of the pairing. For instance, superconductivity appears in  $\text{URhGe}$  below  $T_C = 0.3$  K in the ferromagnetic phase with a Curie temperature  $\Theta = 9.5$  K [5], which is hardly compatible with the singlet pairing.

The domain structure (DS) is inherent to all ferromagnets and the main purpose of this Letter is to understand how superconductivity influences it. Indeed, it is of primary importance to identify the mechanism of superconductivity and ferromagnetism interaction in SFMs. The

question was first addressed by Sonin [12] who came to the conclusion that the DS is impossible in the Meissner state. We demonstrate here that this may be valid only in the nonrealistic limit of the vanishing London penetration depth. But otherwise, correct theoretical result implies that, on the contrary, the DS becomes more dense.

The influence of superconductivity on the domain structure has been considered in the case of superconductor-ferromagnet bilayers [13–16]. In contrast with SFMs, the superconductivity influence in these systems is rather small.

The studied system is a thin SFM film of thickness  $2L_z$  along the  $z$  axis. It is parallel to the  $(x, y)$  plane and presents a DS (see Fig. 1). We use the notation  $\ell_N$  for the domain width in the normal state and the new equilibrium domain width of the SFM will be noted  $\ell$ . We suppose the easy axis anisotropy along the  $z$  axis and the magnetization  $\vec{M}$  uniform. This infers that the domain wall (DW) thickness,  $w$ , is much smaller than the domain width, i.e.,  $\ell \gg w$ . The domain width in the normal state  $\ell_N$  is determined by the minimization of the energy taking into account the contributions of the magnetic field and the DW [17,18].

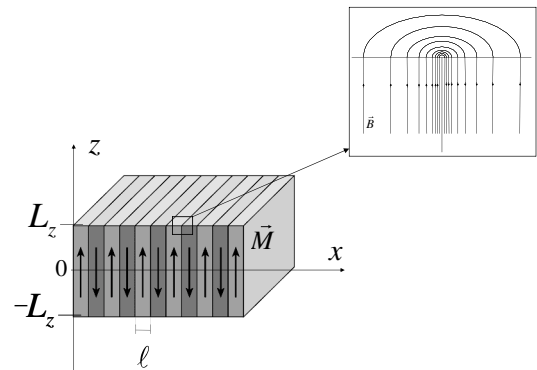


FIG. 1. Geometry of the considered domain system. The domain width is  $\ell$  and the thickness of the film is  $2L_z$ . The inset shows the field distribution near a DW.

The superconductivity appearance may strongly modify the magnetic field distribution and therefore influence the equilibrium DS. Bearing in mind the conditions  $\ell, \lambda \gg w$ , the magnetization  $\mathbf{M}$  of the DS can be approximated by the steplike function:  $\mathbf{M} = M(x)\mathbf{e}_z$ , with  $M(x) = \pm M_0$ . The magnetic field  $\mathbf{B}$  satisfies  $\nabla \mathbf{B} = \mathbf{0}$ , the London equation in the SFM:  $\Delta(\mathbf{B} - 4\pi\mathbf{M}) = \lambda^{-2}\mathbf{B}$  and  $\Delta\mathbf{B} = \mathbf{0}$  outside. Performing the Fourier transform and using the continuity of the magnetic field at the interface, the field distribution in SFM ( $z < L_z$ ) near the upper surface is

$$\begin{aligned} \mathbf{B}(x, z) = & \sum_{k=0}^{\infty} -C(q)e^{qz(z-L_z)} \cos(qx)\mathbf{e}_x \\ & + \sum_{k=0}^{\infty} \left( -\frac{q}{q_z} C(q)e^{qz(z-L_z)} + \frac{q}{q_z^2} \frac{16\pi M_0}{\ell} \right) \\ & \times \sin(qx)\mathbf{e}_z, \end{aligned} \quad (1)$$

and, in the vacuum for  $z > L_z$ ,

$$\begin{aligned} \mathbf{B}(x, z) = & - \sum_{k=0}^{\infty} C(q)e^{-q(z-L_z)} \cos(qx)\mathbf{e}_x \\ & + \sum_{k=0}^{\infty} C(q)e^{-q(z-L_z)} \sin(qx)\mathbf{e}_z, \end{aligned} \quad (2)$$

where  $q = (2k+1)\pi/\ell$ ,  $q_z = \sqrt{q^2 + \lambda^{-2}}$ , and  $C(q) = \frac{q}{q_z} \frac{1}{q+q_z} \frac{16\pi M_0}{\ell}$ . The field distribution near the bottom surface of the film is easily obtain by symmetry. Since the condition  $q_z L_z \gg 1$  is fulfilled, the two magnetic field distributions can be treated independently.

The DW energy and its thickness are determined by the energy balance between the exchange interaction and the anisotropy cost [17]. In a DW, the exchange energy (per unit area) increase is due to the modulation of the magnetic moment and, if the thickness of the wall is noted  $w$ , it may be estimated as  $\Theta/(wa)$ , where  $a$  is the interatomic distance. On the other hand, the anisotropy cost stems from the rotation of the magnetic moment in the wall and is of the order of  $(K/a^2)(w/a)$  where  $K$  is the anisotropy parameter. The minimization of the sum of these two terms gives the equilibrium thickness  $w = \sqrt{\Theta/K}a$  and the DW energy is  $\mathcal{E}_{\text{DW}} = 2wK/a^3$ . Moreover,  $K$  is usually related to the magnetodipole interaction and it is of the order of the electromagnetic energy  $\Theta_{\text{em}} = 2\pi M_0^2 a^3$ . Therefore, the DW energy may be written in the convenient form  $\mathcal{E}_{\text{DW}} = M_0^2 \tilde{w}$  with  $\tilde{w}$  being the effective DW thickness. In the general case,  $\tilde{w} = 4\pi w K / \Theta_{\text{em}}$  and is higher than  $a$ .

The energy per unit length along the  $y$  axis  $F$  results from the contributions of the magnetic field  $F_M$ , the superconducting currents  $F_{\text{SC}}$  and the DW  $F_{\text{DW}}$ :

$$\begin{aligned} F(\mathbf{B}, \mathbf{M}) &= F_M(\mathbf{B}, \mathbf{M}) + F_{\text{SC}}(\mathbf{B}, \mathbf{M}) + F_{\text{DW}}, \\ F_{\text{SC}}(\mathbf{B}, \mathbf{M}) &= \frac{1}{8\pi} \int dx dz [\lambda^2 (\nabla \times (\mathbf{B} - 4\pi\mathbf{M}))^2], \\ F_M(\mathbf{B}, \mathbf{M}) &= \frac{1}{8\pi} \int dx dz (\mathbf{B} - 4\pi\mathbf{M})^2. \end{aligned}$$

$F_{\text{SC}}$  describes the contribution from the superconducting current expressed through the London equation. Direct calculations lead to the following expression of the energy per unit area

$$\mathcal{F} = 32\pi M_0^2 \sum_{k=0}^{\infty} \frac{L_z l^2}{\pi^4 \lambda^2 (2k+1)^2 ((2k+1)^2 + \frac{\ell^2}{\pi^2 \lambda^2})} + 32\pi M_0^2 \sum_{k=0}^{\infty} \frac{\ell}{\pi^3 ((2k+1)^2 + \frac{\ell^2}{\pi^2 \lambda^2})^{3/2} (1 + \sqrt{1 + \frac{\ell^2}{\pi^2 \lambda^2} \frac{1}{(2k+1)^2})}} + \mathcal{F}_{\text{DW}}, \quad (3)$$

where the contribution of the DW  $\mathcal{F}_{\text{DW}}$  is

$$\mathcal{F}_{\text{DW}} = \mathcal{E}_{\text{DW}} L_z / \ell = \tilde{w} M_0^2 L_z / \ell. \quad (4)$$

The domain width in the normal state  $\ell_N$  can be deduced from the minimization of  $\mathcal{F}$  when  $\lambda \rightarrow \infty$ . One obtains  $\frac{\partial \mathcal{F}}{\partial \ell} = 0$  for  $\ell_N = \sqrt{\tilde{w} L_z \sqrt{\pi^2 / (14\zeta(3))}} \sim \sqrt{\tilde{w} L_z}$ . Then, the DW energy  $\mathcal{F}_{\text{DW}}$  (4) may also read  $\mathcal{F}_{\text{DW}} = \frac{14}{\pi^2} \zeta(3) M_0^2 \frac{\ell_N^2}{\ell}$ . The new domain equilibrium width  $\ell$  is obtained by the minimization of  $\mathcal{F}$ . We consider two limiting cases whether  $\ell \ll \lambda$  or  $\ell \gg \lambda$  when the formula (3) can be simplified so that analytical expressions for  $\ell$  can be found. First, if  $\ell \ll \lambda$ , the total energy becomes

$$\mathcal{F} = 32\pi M_0^2 \left( \frac{1}{96} \frac{L_z \ell^2}{\lambda^2} + \frac{7}{16} \zeta(3) \frac{\ell}{\pi^3} \right) + \frac{14}{\pi^2} \zeta(3) M_0^2 \frac{\ell_N^2}{\ell}. \quad (5)$$

If  $\lambda \gg (\tilde{w} L_z^3)^{1/4}$ , the new equilibrium domain width dif-

fers a little from that in the normal state and can be written as

$$\ell = \ell_N \left( 1 - \frac{\pi^3}{42\zeta(3)} \frac{L_z \ell_N}{\lambda^2} \right). \quad (6)$$

It should be underlined that the existence of the DS is confirmed by an energy point of view. Indeed, the energy in the Meissner state is  $\mathcal{F}_M = 4\pi M_0^2 L_z$  (that is the energy when there is no domain) while the energy of the system given by (5) is  $\mathcal{F} \sim M_0^2 \sqrt{\tilde{w} L_z}$ . In this regime, one can note that  $\mathcal{F} \ll \mathcal{F}_M$ . Therefore, the DS is obviously energetically more favorable than the single domain ferromagnet. Next, if  $\lambda \ll (\tilde{w} L_z^3)^{1/4}$ ,  $\mathcal{F} = \frac{1}{3} \pi M_0^2 \frac{L_z \ell^2}{\lambda^2} + \frac{14}{\pi^2} \zeta(3) M_0^2 \frac{\ell_N^2}{\ell}$  and its minimization gives rise to

$$\ell = \left( \frac{21}{\pi^3} \zeta(3) \right)^{1/3} \frac{\ell_N^{2/3} \lambda^{2/3}}{L_z^{1/3}} \sim \tilde{w}^{1/3} \lambda^{2/3}. \quad (7)$$

Note that in this limit  $\ell \ll \ell_N$ , and the main contribution to the magnetic field energy is due to the partial screening of the internal field near the DW. The energy is approximated by

$$\mathcal{F} \sim M_0^2(L_z \ell)/\lambda^2 \sim M_0^2 L_z (\tilde{w}/\lambda)^{2/3} \ll \mathcal{F}_M, \quad (8)$$

and the DS is still favorable provided  $\lambda > \tilde{w}$ .

Now, let us consider the situation  $\ell \gg \lambda$ . Then, the sums in Eq. (3) may be substituted for integrals and

$$\mathcal{F} = 4M_0^2 \pi L_z \left(1 - 2 \frac{\lambda}{\ell}\right) + \frac{14}{\pi^2} \zeta(3) M_0^2 \frac{\ell_N^2}{\ell}. \quad (9)$$

The second term describes the decrease of the magnetic energy due to the incomplete field screening at the distance  $\lambda$  near the DW. In the limit  $\lambda \rightarrow 0$ , this contribution disappears and the minimum of the total energy occurs for  $\ell \rightarrow \infty$ ; that means that the domains must be absent in the Meissner state. This is exactly the conclusion of the work [12] that is correct in the limit of small  $\lambda$  only. The formation of the DS is energetically favorable if the correction proportional to  $1/\ell$  in (9) is negative. This occurs at  $\lambda > \frac{7}{4} \frac{\zeta(3)}{\pi^3} \frac{\ell_N^2}{L_z} \sim \tilde{w}$  when the creation of the DW decreases the total energy of the system. Therefore, in the case  $\lambda > \tilde{w}$ , the DS should exist in the Meissner state, which contrasts with the statement of [12].

Note also that the condition  $\lambda > \tilde{w}$  allows us to neglect the change of the DW energy due to the superconducting screening. But to complete our analysis, we must consider separately the case  $\lambda < w$ . In such a case, the steplike approximation for the magnetic moment variation in the DW is not appropriate anymore. The simple estimate of the magnetic energy gain (per unit area) reads  $\delta \mathcal{E}_m \sim -(\Theta_{\text{em}}/a^3)(\lambda/w)^2 w$  and it is much smaller than the total energy of the DW  $\mathcal{E}_{\text{DW}} \sim (\Theta/a^2)(a/w)$  provided  $\lambda \ll w \sim \sqrt{\Theta/\Theta_{\text{em}}} a$ . This means that the existence of the domains is not energetically favorable if  $\lambda < w$ .

The values of  $\lambda$  in SFMs are not well known, but it may be roughly estimated as 900 nm for URhGe [5]. It seems unlikely that the DW thickness can be larger than  $\lambda$ . Therefore, SFMs may be appropriate candidates for the observation of the domains shrinkage in the superconducting state.

The minimization of  $\mathcal{F}$  can be made numerically in the general case and permits to determine  $\ell$ . The evolution of  $\ell$  with the temperature is related to the temperature dependence of the London penetration depth:  $\lambda \equiv \lambda(T)$ . Figure 2 describes the variation of  $\ell/\ell_N$  as a function of temperature. Note that the DS in SFM is always more dense than in the normal state.

In the previous analysis, the magnetization was implicitly supposed to be weak, i.e.,  $M_0 < H_{c1}(\lambda/w)^{2/3}$ , (as it will be shown below) where  $H_{c1}$  is the lower critical field of the superconductor. But in fact, SFMs may present rather high magnetizations (for instance,  $4\pi M_0 \sim 2$  kOe for UGe<sub>2</sub> [2]). Thus, superconductivity appears with cool-

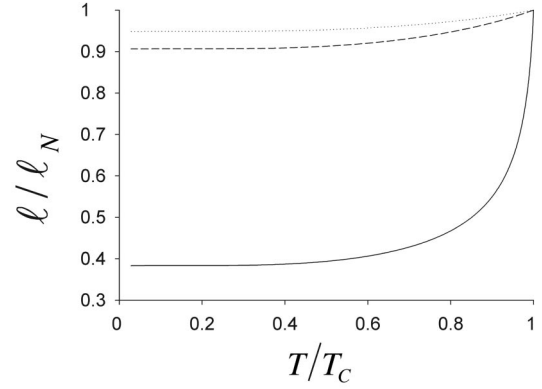


FIG. 2. Evolution of the domain width with the temperature for different values of the parameters. The standard BCS type formula has been used for the temperature dependence of  $\lambda(T)$ . The solid line corresponds to  $\ell_N/\lambda(0) = 0.5$  and  $\lambda(0)/w = 100$ , the dotted line to  $\ell_N/\lambda(0) = 0.1$  and  $\lambda(0)/w = 100$ , and the dashed line to  $\ell_N/\lambda(0) = 0.1$  and  $\lambda(0)/w = 200$ .

ing when the condition  $H_{c2}(T) = 4\pi M_0$  is fulfilled. The corresponding critical temperature  $T_C$  is smaller than the bare critical temperature  $T_{C0}$  of the superconducting transition in the absence of the orbital effect, and the difference is  $T_C - T_{C0} = -4\pi M_0/(dH_{c2}/dT)_{T_{C0}}$ . In the model of the electromagnetic interaction, the superconductivity occurrence is more favorable at the DW [19] and the condition of this localized superconductivity appearance is  $H_{c3}(T) = 1.69H_{c2}(T) = 4\pi M_0$  [20]. The corresponding critical temperature  $T_C^{\text{DW}}$  is defined by  $T_C^{\text{DW}} = T_{C0} - 4\pi M_0/(dH_{c3}/dT)_{T_{C0}}$  and is therefore higher than  $T_C$ .

When the temperature is just below  $T_C^{\text{DW}}$  ( $H_{c2} < 4\pi M_0 < H_{c3}$ ), the nucleation of superconductivity at the DW decreases its energy by  $\delta \mathcal{E}_{\text{DW}} \sim -H_c^2 \xi$ , where  $H_c$  is the thermodynamic critical field. This results in a slight shrinkage of the DS period. With further lowering of the temperature, a vortex state [3,21,22] gets organized when  $H_{c1} < 4\pi M_0 < H_{c2}$ .

If the temperature is slightly below  $T_C$  and  $(H_{c2} - 4\pi M_0)/4\pi M_0 \ll 1$ , three contributions to the energy may be listed: the magnetic energy, the DW energy, and a bulk term. The stray field energy whose expression was  $M_0^2 \ell$  has to be modified to take into account the type II superconductors magnetization  $M_s$  in the mixed state. It becomes  $\mathcal{E}_m \sim (M_0 + M_s)^2 \ell$  where  $M_s = -(H_{c2} - 4\pi M_0)/(4\pi \beta_A(2)\kappa^2)$  [23], and  $\kappa$  is the Ginzburg-Landau parameter while  $\beta_A$  is the Abrikosov parameter. Moreover, the local condensation energy  $-H_c^2 L_z \xi/\ell$  (assuming  $\xi > w$ ) must be added to the formula of the DW energy, which reads  $\mathcal{E}_{\text{DW}} \sim M_0^2 \frac{L_z w}{\ell} \left(1 - \frac{\xi}{w} \frac{1}{4\pi \kappa^2}\right)$ . There is also one more contribution due to the interaction between  $M_s$  and the ferromagnetic moment. It is a bulk term  $\mathcal{E}_{\text{bulk}} \sim 4\pi M_0 M_s L_z (\ell - \xi)/\ell$  because the vortices are at the distance  $\xi$  from the DW when  $4\pi M_0 \sim H_{c2}$ . Keeping in mind that  $\ell \sim \sqrt{L_z w}$  and assuming  $\xi > w$ , the comparison of the

three energy terms shows that the domain width decreases comparing with  $\ell_N$ .

In the regime  $H_{c1} \ll 4\pi M_0 \ll H_{c2}$ ,  $4\pi M_s \sim -H_{c1}$  when the weak logarithmic correction is neglected, see [24]. The stray field energy is changed into  $\mathcal{E}_m \sim (M_0 - \frac{H_{c1}}{4\pi})^2 \ell = M_0^2 (1 - \frac{H_{c1}}{4\pi M_0})^2 \ell$ . The DW energy becomes  $\mathcal{E}_{DW} \sim M_0^2 \frac{L_z w}{\ell} (1 - \frac{H_{c1}}{4\pi M_0} \frac{\lambda}{w})$ . The volume contribution is  $\mathcal{E}_{bulk} \sim 4\pi M_0 M_s L_z (\ell - \lambda)/\ell \sim M_0 H_{c1} L_z (\ell - \lambda)/\ell$  because the vortex structure is absent at the distance  $\lambda$  near the DW. It can be inferred that in the limit  $\lambda > w$  the domain width still decreases. More precisely, the correction is  $(\ell_N - \ell)/\ell_N \sim (H_{c1}/M_0)(\lambda/w)$ .

The transition between the domains with vortices and the DS without vortices occurs when their state energies are equal. The energy of the DS without vortices is given by (8) while the DS energy in the mixed state may be estimated as  $M_0 L_z H_{c1}$ . Therefore, the vortex DS disappears when  $M_0 \leq H_{c1}(\lambda/w)^{2/3}$ .

Up to now, the exchange interaction between superconductivity and ferromagnetism has not been taken into account. However, in real compounds, both mechanisms determine the equilibrium size of the domains. For singlet pairing superconductivity, the paramagnetic effect always favors the nonuniform domainlike structures, so it only contributes to an additional shrinkage of the domains below  $T_C$  [3]. More precisely, if  $w < \xi$ , it is locally weakened near the DW (just like the orbital effect is), which gives more opportunity for superconductivity onset [20]. For triplet superconductivity, the orbital effect is still weakened by the inhomogeneous magnetization in the DW. But, we may expect the transition into a superconducting state with nonzero spin polarization, so the order parameters will be different in the adjacent domains. Therefore, the exchange interaction tends to suppress superconductivity in the DW ([19],[25]), leading to a DW energy increase below  $T_C$ . If  $w < \xi$ , this increase is about  $\delta\mathcal{E}_{DW} \sim H_c^2 \xi$ , and  $H_c^2(0)a^3 \sim T_C^2/\mathcal{E}_F$  where  $\mathcal{E}_F$  is the Fermi level energy. However, as  $T_C$  is very small in the known presumably triplet SFM, this positive contribution to the energy is extremely small.

Finally, just below  $T_C$ , the evolution of the domain width is qualitatively different and depends whether the compound is a singlet or a triplet superconductor. However, when the temperature is further decreased, the regime  $4\pi M_0 \ll H_{c2}$  is reached. In that case, the main interaction between superconductivity and ferromagnetism arises from the electromagnetic effect for both singlet and triplet pairing. The previous analysis is then fully applicable, which implies shrinking of the ferromagnetic domains. Therefore, the evolution of the domain width with temperature for a triplet pairing superconductivity is non-monotonous. This result clearly contrasts with the singlet superconductivity monotonous behavior.

In summary, we have demonstrated that the interplay between superconductivity and magnetism could strongly influence the DS. Contrary to the conclusion of [12], the

DS is fully compatible with the Meissner state. The evolution of the structure period below  $T_C$  is clearly different for the singlet and triplet cases, and provide an unambiguous determination of the pairing type in SFMs. In particular, movements of ferromagnetic domains caused by the vicinity of a superconductor have recently been identified [26]. This might indicate that DW are not pinned even at low temperature, and that similar experiments may be feasible for SFMs. The predicted effects could also be verified with atomic force microscopes or magneto-optical techniques.

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