Current-Loop Model for the Intermediate State of Type-I Superconductors

Raymond E. Goldstein,[1] David P. Jackson,[1,*] Alan T. Dorsey[2]
[1] *Department of Physics, Joseph Henry Laboratories, Princeton University, Princeton, New Jersey 08544*
[2] *Department of Physics, University of Virginia, McCormick Road, Charlottesville, Virginia 22901*
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A theory is developed of the intricately fingered patterns of flux domains observed in the intermediate state of thin type-I superconductors. The patterns are shown to arise from the competition between the long-range Biot-Savart interactions of the Meissner currents encircling each region and the superconductor-normal surface energy. The energy of a set of such domains is expressed as a nonlocal functional of the positions of their boundaries, and a simple gradient flow in configuration space yields branched flux domains qualitatively like those seen in experiment. Connections with pattern formation in amphiphilic monolayers and magnetic fluids are emphasized. [S0031-9007(96)00250-5]

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When a thin film of a type-I superconductor is placed in a magnetic field normal to the sample, the large demagnetizing effects associated with the film geometry preclude the establishment of the Meissner phase (with magnetic induction $\mathbf{B} = 0$). The sample instead accommodates the field by breaking up into a large number of superconducting ($\mathbf{B} = 0$) and normal ($\mathbf{B} \neq 0$) regions, usually forming very intricate patterns [1]. As has been understood since Landau's pioneering work [2] these structures arise from the competition between the magnetic field energy of the domains and the surface energy between the superconducting and normal regions. All existing theories of these patterns [2,3] have explored this competition with variational calculations that assume *regular* geometries of the flux domains. The hypothesized parallel stripes, ordered arrays of circles, etc. are rarely seen, the norm instead being the disordered patterns well documented in experiments [1,4]. Moreover, the temperature and magnetic field history of the sample strongly influence the patterns, suggesting that they are likely not in a global energetic minimum.

Recent work has emphasized that diffusion of magnetic flux in the normal phase can influence the domain morphology [5]. Asymptotic methods applied to the time-dependent Ginzburg-Landau model [6] yield a free-boundary dynamics of superconductor-normal (S-N) interfaces nearly identical to that for the growth of a solid into a supercooled liquid, where the interface motion is unstable (e.g., forming dendrites). By analogy it was suggested, and confirmed by numerical studies [5], that the growth of the superconducting phase into the supercooled normal phase should be dynamically unstable, leading to highly ramified domain shapes. While such diffusive instabilities may play a role in the pattern formation in the intermediate state, these studies are not directly applicable as they have all ignored demagnetizing effects.

Here we ignore entirely any diffusional instabilities and focus instead on the role of demagnetizing fields in producing the observed patterns. This is done in a completely general way, without imposing a predetermined ordered flux structure. We ask the basic question: *What is the energy of a thin multiply connected superconducting domain, the normal regions of which are threaded with a magnetic field?* A central issue is whether the interactions between the Meissner currents flowing along the S-N interfaces within the film are screened by the superconducting regions. Pearl [7] made the important observation that, unlike in bulk, vortices in a thin film interact with an *unscreened* potential $V(r) \sim 1/r$ for large separations r, while for small r, $V(r) \sim \ln(\Lambda/r)$, with Λ an appropriate cutoff. The lack of screening reflects the dominant role of the electromagnetic fields in the vacuum above and below the film. This suggests the simple model developed here: domains bounded by current loops interacting as in free space, endowed with line tension, and subject to the constraint of constant total magnetic flux through the sample. When applied to the stripe phase our model predicts equilibrium lengths close to those found by Landau [2] and seen in experiment [1], suggesting that it captures the essential physics. More importantly, having formulated the model for arbitrary domain shapes, we can address the origin of the disordered patterns which are so prevalent. A simplified *dynamical* model for the evolution of domain boundaries is used to show that the long-range interactions destabilize flux domains of regular shape, producing branched, fingered structures as seen in experiment.

Apart from the global flux constraint, this model is equivalent to one for domains of magnetic fluids in Hele-Shaw flow [8], which exhibit patterns like the intermediate state [9], with history dependence like that noted earlier. Thin magnetic films [10] and monolayers of dipolar molecules [11] exhibit similar behavior, and are described by such models through the underlying correspondence between electric and magnetic dipolar phenomena [12]. This model is also very similar to a reaction-diffusion system [13] in which chemical fronts move in response to line tension and a nonlocal coupling, producing labyrinthine patterns seen in experiment [14].

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Note first a crucial separation of length scales between the typical size of the flux domains ($\simeq$0.1 mm) and the penetration depth ($\leq$1 μm) [1]. Thus *on the scale of the patterns the superconductor-normal interface is sharp, and we may view the order parameter magnitude as piecewise constant*. It follows that the energy is determined solely by the locations of the S-N interfaces.

In this macroscopic approach, the energy $\mathcal{F}$ of a configuration of flux domains arises from the condensation energy, the boundaries between the domains, and the magnetic field energy,

$$\mathcal{F} = \mathcal{F}_{\rm cond} + \mathcal{F}_{\rm wall} + \mathcal{F}_{\rm field}\,. \tag{1}$$

Suppose the film has thickness d, total area A, volume $V = Ad$, and contains a set of normal domains i with area A_i, length L_i, and whose boundary positions are $\mathbf{r}_i(s)$ (Fig. 1). We assume $\mathbf{r}_i$ is independent of z, neglecting the "fanning out" of the domains near the film surfaces [2]. The two phases occupy volumes V_s and $V_n = d\sum_i A_i$, with $V_s + V_n = V$. Their bulk free energy densities F_s and F_n define the critical field $H_c(T)$ as $F_n - F_s = H_c^2/8\pi$. With $\rho_n = A_n/A$ the area fraction of the normal phase, and $\sigma_{\rm SN} = (H_c^2/8\pi)\Delta$ the S-N interfacial tension [$\Delta(T)$ being the interfacial width], we have

$$\mathcal{F}_{\rm cond} = V\frac{H_c^2}{8\pi}\rho_n, \qquad \mathcal{F}_{\rm wall} = \sigma_{\rm SN} d\sum_i L_i\,, \tag{2}$$

where $\mathcal{F}_{\rm cond}$ is measured with respect to the purely superconducting state.

The complexity of this problem lies entirely in the computation of the field energy. An applied field $\mathbf{H} = H_a\hat{\mathbf{e}}_z$, produces a field $H_n\hat{\mathbf{e}}_z$ in the normal regions, where $AH_a = A_nH_n$ by flux conservation, so

$$H_n = \frac{H_a}{\rho_n}\,. \tag{3}$$

The requirement of tangential continuity of $\mathbf{H}$ across a S-N interface gives the field in the superconducting region as $\mathbf{H}_s = H_s\hat{\mathbf{e}}_z = H_n\hat{\mathbf{e}}_z$. The superconducting regions are perfectly diamagnetic, with magnetization $\mathbf{M} = -(H_n/4\pi)\hat{\mathbf{e}}_z$. The discontinuous magnetization at the S-N boundaries implies that the sample consists of a collection of current *loops* (of strength $H_n/4\pi$) encircling the domains. The field energy thus has two contributions: the first is simply that of the magnetization $\mathbf{M}$ associated with current loops in the presence of the external field ($-\int d^3r\,\mathbf{H}_a\cdot\mathbf{M}$). The second contribution is the self-induction and mutual induction of those loops. We obtain

$$\mathcal{F}_{\rm field} = V\frac{H_aH_n}{4\pi}(1-\rho_n) - \frac{1}{2}\left(\frac{H_n}{4\pi}\right)^2\sum_{i,j}\int_0^d dz\int_0^d dz'\oint ds\oint ds'\,\frac{\hat{\mathbf{t}}_i\cdot\hat{\mathbf{t}}_j}{R_{ij}}\,, \tag{4}$$

where $\hat{\mathbf{t}}_i = \hat{\mathbf{t}}_i(s)$ is the tangent vector, and $R_{ij} = \{[\mathbf{r}_i(s) - \mathbf{r}_j(s')]^2 + (z-z')^2\}^{1/2}$.

A first application of this model is to the laminar state, a periodic structure of alternating superconducting and normal domains. Landau's calculation treated the cross-sectional shape of the domain walls as a free-boundary problem. Exploiting the two-dimensional nature of this geometry, he found using conformal mapping techniques that deep within the slab the walls are indeed straight, but near the surface the normal lamina flare out, leading to a reduction in the magnetic field energy. The free energy density $F = \mathcal{F}/V$ in Landau's model is

$$F = \frac{H_c^2}{8\pi}\left(\rho_n + \frac{h^2}{\rho_n}\right) + \frac{H_c^2}{4\pi}\left[\frac{\Delta}{a} + \frac{a}{d}f(\rho_n)\right], \tag{5}$$

where a is the stripe period, $h = H_a/H_c$, and f is a known d-independent function. Minimizing F with respect to ρ_n, we see that for large d the first ("bulk") term dominates, yielding the familiar result $\rho_n = h$; the field at the S-N interface is H_c. Minimization with respect to a then yields the equilibrium period

$$a^* = \left(\frac{\Delta d}{f_L(h)}\right)^{1/2}, \tag{6}$$

where f_L is the Landau function [15]. Thus, the characteristic domain size is set by the geometric mean of the microscopic wall thickness Δ and the slab thickness d. The current-loop (CL) model yields an energy of the form in Eq. (5), and hence an equilibrium width given by (6), but with $f_L(h)$ replaced by [17]

$$f_{\rm CL}(h) = \frac{1}{2\pi^3}\sum_{n=1}^{\infty}\frac{\sin^2(n\pi h)}{n^3}\,. \tag{7}$$

As shown in Fig. 2 the functions $f_L(h)$ and $f_{\rm CL}(h)$ have very similar forms over the entire range of h [16]. Analytically, at small h they have similar limiting behavior: $f_L \simeq (1/\pi)h^2\ln(0.56/h)$, and $f_{\rm CL} \simeq (1/2\pi)h^2\ln(0.71/h)$. The equilibrium widths a^* are therefore also close in the two models, and compare favorably with experiment [1,4], suggesting the validity of the current-loop model. Moreover, this model may be

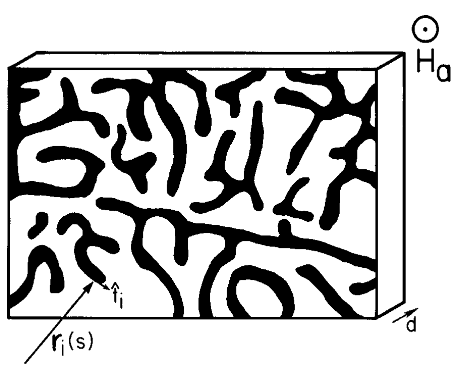

FIG. 1. A thin slab of type-I superconductor, viewed along the applied field $\mathbf{H}_a$. Normal regions are shown black. Adapted from [4].

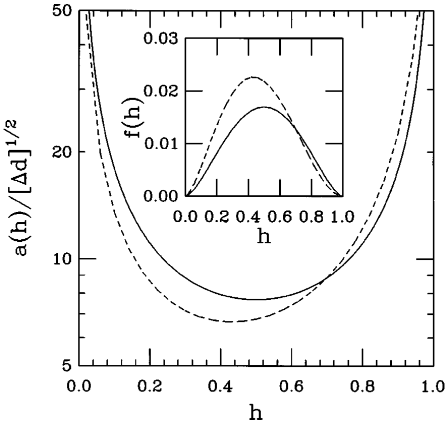

FIG. 2. Equilibrium stripe width in the laminar state as a function of reduced field h. Landau's model (dashed) and the current-loop approximation (solid) compare favorably. Inset: the Landau function $f_L(h)$ (dashed) compared with the current-loop function $f_{\mathrm{CL}}(h)$ (solid).

used to study perturbations about the laminar state, allowing for the calculation of elastic moduli and dislocation energies [17], which simply cannot be determined using Landau's methods.

Having thus "calibrated" the current-loop model, we turn to the most important new aspect of this work: domain shape instabilities arising from long-range current-current interactions. The dynamical evolution of flux domain shapes with energy (1)–(4) is a many-body problem of considerable computational complexity. Insight into its behavior comes from a mean-field description of a single current loop in which only its self-induction is treated in detail, the mutual induction of the surrounding loops contributing a bulk energy term analogous to that in the laminar calculation (5). For small departures from the minimizing area fraction the amplitude of the circulating currents is then set by H_c rather than the local field. The area fraction is defined by assigning the loop to a cell of area A_{cell}, with $\rho_n = A_n/A_{\mathrm{cell}}$. Appropriate rescaling of the spatial variables shows that the location of the system in the H-T plane is uniquely specified by the two dimensionless quantities h and $\Delta/A_{\mathrm{cell}}^{1/2}$.

In this mean field model, we study the simplest dynamics for domain wall motion in which a generalized normal force $\hat{\mathbf{n}} \cdot \delta\mathcal{F}/\delta\mathbf{r}$ balances a local viscous drag $-\eta\hat{\mathbf{n}} \cdot \partial_t\mathbf{r}$. The resulting equation of motion is

$$\hat{\mathbf{n}} \cdot \frac{\partial \mathbf{r}(s)}{\partial t} = \frac{H_c^2 d}{8\pi\eta}\Big\{\Pi - \Delta\mathcal{K}_i(s) - \frac{1}{2\pi d}\oint ds'\,\hat{\mathbf{R}}(s,s') \times \hat{\mathbf{t}}(s')\Phi(R/d)\Big\}, \tag{8}$$

with $\mathcal{K}_i(s)$ the curvature, $\mathbf{R}(s,s') = \mathbf{r}(s) - \mathbf{r}(s')$, $\Phi(\xi) = 1 - (1 + \xi^{-2})^{1/2}$ is the Coulomb potential averaged over the thickness of the slab, and

$$\Pi = h^2/\rho_n^2 - 1\,. \tag{9}$$

The dynamics (8) is local in time; it neglects diffusion of magnetic flux in the normal phase [6], which is equivalent to setting the normal state conductivity to zero. The magnetic vector potential is then "slaved" to the order parameter, analogous to the fast-inhibitor limit for a reaction-diffusion system [13]. In this slaving limit, we estimate the kinetic coefficient η for strongly type-I *bulk* superconductors as [6] $\eta = (H_c^2 d\Delta/8\pi)\pi\hbar/8k_B T_c\xi_0^2$, where T_c is the critical temperature and ξ_0 is the zero-temperature correlation length [18].

Equation (8) reveals a competition between a magnetic pressure Π incorporating flux conservation, the Young-Laplace force from interfacial tension and the Biot-Savart force of circulating currents. The latter long-range contribution is well known for magnetic fluids [8] and monolayers [12,19]. Extensive analytical linear stability analyses [8,9,12] show branching instabilities of circular domains (on a length scale given by the laminar calculation) and buckling instabilities of stripes, phenomena which should carry over to the present problem with nonconserved area. Indeed, buckled domains are well known in type-I superconductors [4] and monolayers [20].

To see the effects of the Biot-Savart interaction on the stability of a circular flux domain, Fig. 3 shows the evolution of a domain prepared with an area significantly less than the equilibrium value. The long-range interactions result in the formation of a branched flux domain. We see that a transient fourfold coordinated vertex is unstable to fission into two threefold nodes, very similar to those seen in experiment [1,4]. In the early epoch the shape evolution is primarily a dilation with little change in shape, while the branching instabilities occur on a longer time scale. The inset of Fig. 3 shows that when the Biot-Savart coupling is omitted the weakly perturbed circle simply relaxes to a circle of larger radius driven primarily by the magnetic pressure. As found in previous studies [9], in the later stages of the fingered shape evolution the driving force for the interfacial motion becomes extremely small, with very small energy differences between rather differ-

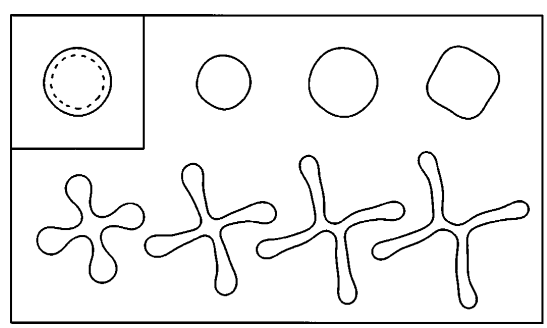

FIG. 3. Numerical results from the gradient-flow model for flux domain boundary motion. Initial condition is a circle of unit radius perturbed by low-order modes, evolving with $\Delta = 0.01$ and $h = 0.5$. Evolution displays rapid dilation followed by fingering. Inset: rapid relaxation to a circle without long-range Biot-Savart interactions.

ent shapes. This suggests that the interface motion would be extremely sensitive to external perturbations such as impurities or grain boundaries, which would then be effective in pinning the interfaces, not unlike the pinning of vortices in type-II superconductors. This may contribute to the history dependence of the patterns discussed in the introduction.

Given the connections outlined here between pattern formation in the intermediate state and in other systems, it would be of interest to extend experimental studies of flux domain shapes to probe systematically the branching instabilities as a function of field and temperature. Similarly, the results described here may be important for understanding the fingering instabilities observed during flux invasion into films of type-II superconductors [21].

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*Present address: Department of Physics, Dickinson College, Carlisle, PA 17013-2896.

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