From Dendrites to Labyrinths: The Morphology of Magnetic Flux Patterns in Superconductors

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Type-I Superconductivity

- What happens in a thin film?

- Such a configuration is energetically unfavorable. The sample breaks up into normal and superconducting regions→intermediate state.
Fig. 1. Intermediate-state structures for $H = 2'23'$ and (left to right) $H = 0.395$, $0.330$, $0.294$, $0.196$, $0.189$, $0.184$. Top row is for decreasing field, showing "loopy" structures similar to Fig. 4. Bottom row is for increasing field, showing "honeycomb" structure. @A. K. and E. H. (Ref: 5.4.1)
Issues:
- Many of the patterns involve sharp interfaces between two phases: either normal/superconducting, superconducting/vortex liquid, etc. Focus on developing models for the interface dynamics.
- Can we understand
  - length scales?
  - topology?
  - dynamics?

Outline:
- Interface dynamics without demagnetizing effects.
  - Growth of the superconducting phase—a free boundary model.
  - Instabilities of the interface motion and analogies with dendritic growth.
  - Studies using the time-dependent Ginzburg-Landau equations.
  - Experiments.
- Including demagnetizing effects—the intermediate state.
  - Landau's model of the intermediate state.
  - The current loop model of the intermediate state.
  - Numerical studies of branching instabilities.
  - The laminar state—energetics, fluctuations, and defects.
  - Experiments.


Finally, there is a most remarkable coincidence: The equations for many different physical situations have exactly the same appearance. Of course, the symbols may be different—one letter is substituted for another—but the mathematical form of the equations is the same. This means that having studied one subject, we have a great deal of direct and precise knowledge about the solutions of the equations of another.
Interface Dynamics in Superconductors

How is the magnetic flux expelled from the Meissner phase? Consider a type-I superconductor (without demagnetizing effects).

Free boundary model for interface motion

- In the normal phase eddy currents are produced due to flux motion. Faraday’s law + Ampère’s law + Ohm’s law ($\mathbf{J} = \sigma \mathbf{E}$) leads to the diffusion equation for the $B$ field ($\mathbf{B} = B(x,y)\hat{z}$), with $D_B = \frac{c^2}{4\pi \sigma}$:

$$\partial_t B = D_B \nabla^2 B \quad \text{(normal regions).}$$

- In the superconducting regions $B = 0$.

- On the normal side of the S/N interface, $\mathbf{E} = -\mathbf{v} \times \mathbf{B}$, so that $E_t = v_n B_i$ (t = tangent, n = normal). Combine with Ohm’s law and Ampere’s law to arrive at the boundary condition on the moving boundary:

$$B_i v_n = -D_B (\partial B/\partial n)|_i.$$

- For an equilibrium, planar S/N interface, $B = B_i$ as the interface is approached from the superconducting side. If the interface has curvature $\mathcal{K}$, and moving with a normal velocity $v_n$, this becomes

$$B_i = H_c (1 - d_0 \mathcal{K} - \beta v_n),$$

where $d_0$ is the capillary length and $\beta$ is the kinetic coefficient.

- Far from the S/N interface the magnetic field is the applied field.

- The diffusion equations + boundary conditions constitute a free boundary problem for the moving interface. It is highly nonlinear and nonlocal; analytic solutions are only known for zero surface tension and in special circumstances.
Dendritic growth of a pure substance

- Place a piece of a solid into its supercooled liquid. The conversion of liquid into solid produces latent heat \( L \), which must diffuse away from the interface in order for the solid to continue growing:

\[ \partial_t T = D_T \nabla^2 T. \]

- At the interface,

\[ \frac{L \nu_B}{\text{rate of heat production}} = \frac{[D_T \rho_c (\partial T / \partial n)_{\text{solid}} - D_T \rho_c (\partial T / \partial n)_{\text{liquid}}]}{\text{rate at which heat flows into liquid and solid}}. \]

- The temperature at the planar solid/liquid interface is the melting temperature \( T_m \); for a curved, moving interface we have the Gibbs-Thomson boundary condition:

\[ T_i = T_m (1 - d_0 \mathcal{K} - \beta \nu_n), \]

with \( d_0 \) the capillary length, \( \mathcal{K} \) the curvature, and \( \beta \) a kinetic coefficient.

Dendritic growth \( \xleftrightarrow{\text{flux expulsion}} \)

- There is a close analogy between flux expulsion and dendritic growth:

<table>
<thead>
<tr>
<th>Flux expulsion</th>
<th>Solidification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flux diffuses away from the interface.</td>
<td>Heat diffuses away from the interface.</td>
</tr>
<tr>
<td>Flux diffusion: ( \partial_t B = D_B \nabla^2 B )</td>
<td>Thermal diffusion: ( \partial_t T = D_T \nabla^2 T )</td>
</tr>
<tr>
<td>( D_B = \frac{c^2}{4 \pi \sigma} \sim 10 \text{ cm}^2 \text{ s}^{-1} )</td>
<td>( D_T \sim 10^{-3} \text{ cm}^2 \text{ s}^{-1} )</td>
</tr>
<tr>
<td>Faraday's law: ( B \nu_n = -D_B (\partial B / \partial n)_i )</td>
<td>Heat flux: ( L \nu_n = -D_T \rho_c (\partial T / \partial n)_i )</td>
</tr>
<tr>
<td>( B_i = H - (1 - d_0 \mathcal{K} - \beta \nu_n) )</td>
<td>Gibbs-Thomson: ( T_i = T_m (1 - d_0 \mathcal{K} - \beta \nu_n) )</td>
</tr>
</tbody>
</table>

- There should be a dynamic instability of the flux front, which is only stabilized at short wavelengths due to surface tension effects.

Dynamic instabilities of the interface

- In the solidification problem, the growth is known to be unstable; highly ramified patterns are formed ("dendrites"). Therefore we expect the growing superconducting nucleus to also be dynamically unstable!

\[ H_a < H_c \]

- \( \partial B / \partial n \) is largest near the bump; recalling that \( B_i v_n = -D_B (\partial B / \partial n)_i \), we see that bumps grow faster.
A linear stability analysis for a planar interface shows that the growth rate for long wavelength perturbations is positive, and is stabilized at short wavelengths by the surface tension:

- In the solidification problem, the crystalline anisotropy will "focus" the instability, leading to dendritic patterns ("snowflakes").

- Mass conservation gives
  \[ \partial_t h = -h \nabla \cdot \mathbf{v}. \]

- Combine with Darcy's law,
  \[ \mathbf{v} = -k \mu \nabla p \]
  (for Poiseuille flow \( k = h^2/12 \)); assuming gradients in \( h \) are small,
  \[ \nabla^2 p = \frac{\mu}{k} \left[ \frac{1}{h} \frac{\partial h}{\partial t} \right]. \]

- At the interface, \( v_n \propto -\partial p/\partial n \).

\(^3\)Special thanks to Chris Lobb.
Time-dependent Ginzburg-Landau (TDGL) theory

- The TDGL equations for the order parameter $\psi$ and the vector potential $A$ are
  \[ \hbar \gamma \left( \partial_t + \frac{ie^*}{\hbar} \phi \right) \psi = -\frac{\delta F}{\delta \psi^*} = \frac{\hbar^2}{2m} \left( \nabla - \frac{ie^*}{\hbar} A \right)^2 \psi + a \psi - b|\psi|^2 \psi, \]
  \[ \nabla \times \nabla \times A = 4\pi (J_n + J_s), \]
  where $J_n$ and $J_s$ are the normal and supercurrents,
  \[ J_n = \sigma (-\nabla \phi - \partial_t A) \]
  \[ J_s = \frac{\hbar e^*}{2m} (\psi^* \nabla \psi - \psi \nabla \psi^*) - \frac{e^2}{m} |\psi|^2 A. \]
  The parameter $a = a_0 (1 - T/T_c)$ and controls the correlation length $\xi = \hbar/(2m|a|)^{1/2}$ and penetration depth $\lambda = [mb/4\pi e^2 |a|]^{1/2}.
  The magnetic field is $H = \nabla \times A$.\[ \]
- Important dimensionless parameters: $\kappa = \lambda/\xi$ (ratio of length scales), $\sigma = 4\pi \kappa^2 (\hbar \sigma / 2m \gamma)$ (ratio of time scales).
- Can be derived from the microscopic BCS theory in the appropriate limit.
- Can derive interface model from TDGL equations using matched asymptotic expansions.\[ \]

Numerical solution of the TDGL equations

- Computational lattice:

- Discretize TDGL equations. Put gauge fields on the links of the lattice to insure gauge invariance ($\mu = x, y$):
  \[ U^\mu(x) = \exp[-i\kappa A_\mu(x)]. \]
  Then derivatives become
  \[ \left( -\frac{1}{2i\kappa} \partial_\mu - iA_\mu \right) \psi \rightarrow \frac{1}{i\kappa a} \left[ U^\mu(x) \psi(x + a\hat{\mu}) - \psi(x) \right]. \]
  \[ \left( \nabla \times A \right)_z \rightarrow -\frac{1}{i\kappa a^2} \left[ U^x(x) U^y(x + a\hat{y}) U^z(x + a\hat{y})^{-1} U^z(x)^{-1} - 1 \right]. \]
  Becomes a lattice gauge theory.
- Iterate equations of motion.

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The process repeats itself once more. The field at the interface increases to H₁, and the field at the wall. The field at the interface is now lower than in the previous case. The field at the interface is now lower than in the previous case.

Negative surface tension is AFL phase. What happens to interface?
Propagating Front Solutions in One Dimension

- Dimensionless TDGL equations in one dimension ($\psi = f e^{i\theta}$, $q = A - \nabla \theta / \kappa$):

\[
\begin{align*}
\partial_t f &= \frac{1}{\kappa^2} \partial_x^2 f - q^2 f + f - f^3, \\
\bar{\theta} \partial_t q &= \partial_x^2 q - f^2 q.
\end{align*}
\]

Both diffusive ($v \sim t^{-1/2}$) and propagating ($v = \text{constant}$) solutions exist.

![Figure 1: Numerical values (open squares) of the front speed as a function of $Q_m$ for $\kappa = 1$ and $\theta = 1$.](image)

- For propagating solutions, for small flux the problem reduces to *Fisher-KPP equation* (population biology); $v = 2/\kappa$. For large flux can use matched asymptotic expansions.
- For $\kappa = 1/\sqrt{2}$ and $\bar{\sigma} = 1/2$ the equations can be solved exactly, with ($v = \sqrt{2}$).

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Some experimental results


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Demagnetizing effects and the intermediate state

- In the film geometry the sample cannot expel the flux, so the superconducting and normal phases coexist.
- What sets the characteristic size of a domain? Need to account for
  - demagnetizing energy (bending of field lines), which favors a finely divided structure (energy $\sim a$);
  - surface energy of the interfaces, which favors a coarse structure (energy $\sim 1/a$).
- Minimizing, we find $a = \sqrt{\Delta / f(h)}$, with $d$ the film thickness, $\Delta$ the interfacial width (microscopic), and $f(h)$ a model dependent function of the reduced magnetic field $h = H_a / H_c$. Gives the correct order of magnitude.

Fig. 8. Laminar structure observed in 35 mil silver disk at 5.4 K, 0.8 turn. Applied field makes an angle of 1$\degree$ with the surface. All photographs except one show normal regions on edge. (After Sharon.)

- Laminar's structure is only observed in oblique fields.
- Laminar state the exception, not the rule.
- Need a more general approach for disordered patterns; dynamics.
Landau's theory of the intermediate state

- Assume a laminar structure:

![Diagram showing SC and Hs with field lines]

- Top view

- The laminar structure is only observed experimentally in oblique fields.
- Global flux conservation: \( H_n A = H_n A_n \), with \( A = A_n + A_s \).
- Area fraction: \( \rho_n = A_n / A = H_n / H_n \).
- Energy balance (bulk):
  \[
  F = -(H_s^2 / 8\pi)A_d + (H_n^2 / 8\pi)A_n d.
  \]
  condensation energy field energy
- Minimum at \( \rho_n = H_n / H_c \); i.e., \( H_n = H_c \).
- Leaves out surface energy and demagnetizing effects. These require a model of the laminae shape.

Determining the shape of the laminae

- Problem: find the shape of the laminae, determine the unknown \( f(h) \).
- Two dimensional magnetoostatics→ complex variable methods.
- Boundaries are unknown. Solve using the hodograph method, developed in the context of fluid mechanics:

<table>
<thead>
<tr>
<th>Free streamline flow around a plate</th>
<th>Laminae in superconductors</th>
</tr>
</thead>
<tbody>
<tr>
<td>Complex potential ( w = \phi + i\psi )</td>
<td>Complex potential ( w = \phi + iA_p )</td>
</tr>
<tr>
<td>Fluid velocity ( u - iv = -dw/d\zeta )</td>
<td>Magnetic field ( B = B_x - iB_y = -dw/d\zeta )</td>
</tr>
<tr>
<td>Streamlines</td>
<td>Field lines (lines of force)</td>
</tr>
<tr>
<td>Free streamline</td>
<td>Superconducting-normal interface</td>
</tr>
<tr>
<td>Free streamline velocity ( U )</td>
<td>Superconducting critical field ( H_s )</td>
</tr>
<tr>
<td>Region of fluid flow</td>
<td>Normal phase with nonzero magnetic field</td>
</tr>
<tr>
<td>Cavity behind plate</td>
<td>Superconducting phase</td>
</tr>
<tr>
<td>Riabouchinsky flow</td>
<td>Lamina in a finite thickness plate</td>
</tr>
</tbody>
</table>
The current loop (CL) model\(^7\)

\[ E[\{r_i\}] = V \frac{H_c^2 \rho_n}{8\pi} + V \frac{H_a H_n}{4\pi} (1 - \rho_n) \]

\[ + \frac{H_c^2}{8\pi} \Delta d \sum_i L_i - \frac{1}{2} M^2 \sum_{ij} \int ds_i \int ds_j \int ds \int ds \frac{\hat{r}_i \cdot \hat{r}_j}{R_{ij}} \]

- What is the energy of a collection of normal domains of magnetization \( M = -H_n/4\pi \)?

- Assume the dynamics is overdamped:

\[ \eta \dot{r}_i(s) = -\frac{1}{\sqrt{\delta}} \frac{\delta E}{\delta r_i(s)} \]

- Can solve numerically using intrinsic coordinates (arc length and tangent angle).


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Disordered patterns and instabilities

- Competition between long-range, repulsive interaction among the currents, and the surface tension, which is short range and attractive. For sufficiently small surface tension this results in a branching instability.

- Elongational instability of a circular flux domain, \( h = 0.38 \).

- Branching instability for \( h = 0.45 \).
Energy of the laminar state in the CL model

- The CL model captures many of the features of the disordered patterns. It can also be applied to the laminar state (observed in oblique fields).
- The function $f(h)$ calculated in the CL model is very close to $f(h)$ in the Landau model.
- Can use CL model to study the dynamics of the laminar state.

Figure 5.10 - Image analysis of laminar pattern: (a) typical laminar pattern ($H=270$ Gauss, $\beta=20^\circ$). Normal regions are bright, superconducting regions are dark; (b) 2D fast-Fourier-transform of (a); (c) spectral intensity - experimental points (+), fitted curve (—).
Fig. 4 Periodicity of laminar structures as a function of the reduced field, Landau nonbranching model (—) and GJD CL model (—) for Sharvin's geometry. The points are the experimentally observed periodicities scaled using $\Delta_{c}$ (•) and $\Delta_{c1}$ (•) (ND$_{1}$ transition).

Figure 5.9 - Image analysis of corrugated pattern: (a) typical corrugated pattern, $h=0.53$. Normal regions are bright, superconducting regions are dark; (b) 2D fast-Fourier-transform of (a); (c) spectral intensity - experimental points (•), fitted curve (—).
Fluctuations and defects

- Can use CL model to examine deformations and fluctuations of the laminae.

The *continuum elastic theory* is identical to that of a two-dimensional smectic liquid crystal:

\[ \mathcal{F}_{\text{elastic}} = \int d^2r \left[ \frac{B}{2} \left( u_x + \frac{1}{2} u_y^2 \right)^2 + \frac{K_1}{2} u_{yy}^2 \right]. \]

- Can also include defects into the model in the form of *edge dislocations*, the dislocation energy is finite and can be small.
Some other labyrinthine patterns

- There are many other systems in which surface tension competes with long-range dipolar interactions; the energy is generally of the form

\[ E = \Pi \sum_i A_i + \gamma \sum_i L_i - \frac{1}{2} \Omega \int ds \int ds' \hat{i}_i \cdot \hat{i}_{ij}(R_{ij}/\xi). \]

<table>
<thead>
<tr>
<th>System</th>
<th>( \Pi )</th>
<th>( \gamma )</th>
<th>( \Omega )</th>
<th>( \Phi )</th>
</tr>
</thead>
<tbody>
<tr>
<td>type-I superconductors</td>
<td>( (H^2/d/8\pi)(\rho_n + \hbar^2/\rho_n) )</td>
<td>( H^2d/8\pi )</td>
<td>( H^2d/8\pi )</td>
<td>( \sinh^{-1}(1/z) ) + ( z - \sqrt{1 + z^2} )</td>
</tr>
<tr>
<td>magnetic fluids</td>
<td>Lagrange multiplier</td>
<td>( d\sigma_{FW} )</td>
<td>( 2dM^2 )</td>
<td>( \sinh^{-1}(1/z) ) + ( z - \sqrt{1 + z^2} )</td>
</tr>
<tr>
<td>Langmuir monolayers</td>
<td>Lagrange multiplier</td>
<td>( \gamma_{LE-LC} (\Delta \mu)^2 )</td>
<td>( \Delta F )</td>
<td>( 1/2z )</td>
</tr>
<tr>
<td>FitzHugh-Nagumo model</td>
<td>( \Delta F )</td>
<td>( D )</td>
<td>( \rho )</td>
<td>( K_0(z) )</td>
</tr>
</tbody>
</table>

Explanation of symbols: \( \sigma_{FW} \), ferrofluid water surface tension; \( M \), ferrofluid magnetization; \( \gamma_{LE-LC} \), line tension between liquid expanded (LE) and liquid condensed (LC) phases in a Langmuir monolayer; \( \Delta \mu \), discontinuity in electric dipole moment density between LE and LC phases; \( d_{mol} \), a molecular cutoff – monolayer thickness.
Summary

- Growth of the superconducting phase after a quench from the normal phase.
  - Growth limited by diffusion of magnetic flux away from the interface.
  - Interfacial instabilities lead to ramified patterns. Analogies with dendritic growth.
  - Behavior contained in simple free-boundary model is contained in TDGL equations.
- Structure of the intermediate state in type-I superconductors.
  - Introduced a current-loop model for the intermediate state.
  - For certain parameters the Biot-Savart interaction produces a branching instability.
  - CL model can also be applied to ordered structures such as the laminar state.
- Future work.
  - Phase ordering kinetics for layered systems. Dynamic scaling?
  - Go beyond relaxational dynamics and include diffusive dynamics. Easiest case—FitzHugh-Nagumo model (with R. Goldstein).
  - Pattern formation in type-II superconductors—flux invasion.
From Carlos Duran
AT&T Bell Labs

Nb film
Duran et al., Phys. Rev. B

Normal phase (dark)
SC phase (light)

$\uparrow$

$T$

$H$