Low-Field Magnetic Response of Complex Superconductors

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Solutions of the Ginzburg-Landau equations for a complex superconducting phase are presented for regions close to a domain wall and to the surface. The finite local magnetization found at these inhomogeneities yields a small magnetic response via a movement of domain walls in an external magnetic field. We examine line defects in the walls which are vortices enclosing a fraction of the universal flux quantum.

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In this Letter we examine the low-field magnetic properties of superconductors in the intrinsically complex phase. Such superconducting phases are distinguished by the property that they are not eigenstates of the time-reversal operator. Volovik and Gor'kov have previously pointed out that they can have unique properties such as domain walls with persistent currents and vortices with a nonuniversal flux quantum.[1] There has been much speculation about the possibility of similar states in the literature on high-T_c superconductivity.[2] Mota and co-workers report that high-T_c, heavy fermion and organic superconductors,[3,4] show unusual behavior at low magnetic fields ($H < H_{c1}$), i.e., strongly time- and history-dependent magnetization with continuously varying magnetic flux. With these motivations we examined in detail a domain wall (DW) between two domains related by time reversal and a fractional vortex, analogous to a Bloch line defect in the DW. Individual domains have net magnetic moments and a net force results on a DW in an external magnetic field. Further, the intersection of a DW with a surface leads to unusual magnetic structures which can be modified to incorporate a net magnetic flux.

As an illustration we consider a complex (even-parity) superconducting phase in a tetragonal crystal (point group D_{4h}) with a gap matrix $\hat{\Delta}(\mathbf{k}) \propto i\sigma_y k_z(k_x \pm ik_y)$. It has a twofold degeneracy because of broken time-reversal symmetry.[5] Its order parameter belongs to the Γ_5^+ representation of D_{4h} with basis functions

$$\psi_1(\mathbf{k}) = k_z k_x\,,\quad \psi_2(\mathbf{k}) = k_z k_y,\quad \hat{\Delta}(\mathbf{k}) = i\sigma_y \lambda_j \psi_j(\mathbf{k})\,. \tag{1}$$

We use the complex coefficients λ_j to formulate the Ginzburg-Landau (GL) theory. The GL free-energy expansion in dimensionless variables is

$$\begin{aligned} f = &-\tfrac{1}{2}(|u|^2+|v|^2)+(\tfrac{1}{8}+\tfrac{1}{2}\beta_2)(|u|^2+|v|^2)^2-\tfrac{1}{2}\beta_2(u^*v-uv^*)^2+\tfrac{1}{8}\beta_3(|u|^2-|v|^2)^2 \\ &+K_1(|d_xu|^2+|d_yv|^2)+K_2(|d_yu|^2+|d_xv|^2)+K_3[(d_xu)^*(d_yv)+(d_yu)^*(d_xv)+\text{c.c.}] \\ &+K_4(|d_zu|^2+|d_zv|^2)+\kappa^2\mathbf{b}^2\,, \end{aligned} \tag{2}$$

with $F_S - F_N = (H_c^2\xi^3/4\pi)\int d^3r f(\mathbf{r})$, $d_j = \partial_j - ia_j$ ($j = x,y,z$). Here $\mathbf{r}$ is the space variable in units of the average coherence length ξ and the vector potential $\mathbf{A} = \sqrt{2}H_c\delta\mathbf{a}$ (δ, London penetration depth) and $\mathbf{b} = \partial \times \mathbf{a}$. The real coefficients β_i and K_i are $O(1)$ and satisfy the relations $4\beta_2 > \beta_3$, $\beta_2 > 0$ and $K_1 + K_2 = 1$. The order parameter $(u,v) \propto (\lambda_1,\lambda_2)$ is defined to be $(u,v) = (1, \pm i)$ in the homogeneous phase.

Single domain wall.— We consider degenerate phases separated by a DW in the y-z plane $[(u,v) \to (1, \pm i)$ for $x \to \pm\infty]$. Varying Eq. (2) with regard to the order parameter and the vector potential leads to seven nonlinear coupled differential equations. We find self-consistent solutions with periodic boundary conditions (many parallel DW) by a numerical relaxation algorithm similar to that used by Thuneberg.[6] In Fig. 1(a) the order parameter, parametrized as $(u,v) = [|u|, |v|\exp(i\gamma)]\exp(i\phi)$, is shown. Note $|u|$ and $|v|$ are reduced slightly from their bulk value 1. The relative phase γ changes from $-\pi/2$ to $\pi/2$ in a width $\tilde{\xi} \sim (K_2/\beta_2)^{1/2}$ which corresponds to the approximate solution of Volovik and Gor'kov.[1] The total or "Josephson" phase ϕ also becomes finite, but tends to the same value ($\phi = 0$) on both sides for large $|x|$; i.e., there is no current through the DW.

The complex structure of the gradient terms in Eq. (2) leads to an unusual expression for the supercurrents in the London equation $\partial \times (\partial \times \mathbf{a}) = \mathbf{j}$:

$$\begin{aligned} \kappa^2 j_{x(y)} = &(K_1|u|^2+K_2|v|^2)(\partial_{x(y)}\phi - a_{x(y)})+K_{2(1)}|v|^2\partial_{x(y)}\gamma+K_3(\partial_{y(x)}\phi - a_{y(x)})|u|\,|v|\cos\gamma \\ &-\tfrac{1}{2}K_3[(|u|\partial_{y(x)}|v|-|v|\partial_{y(x)}|u|)\sin\gamma+|u|\,|v|\cos\gamma\,\partial_{y(x)}\gamma]\,. \end{aligned} \tag{3}$$

We neglect the z component, since the geometry used assumes homogeneity in this direction. With the condition $j_x = 0$

for all x, j_y (neglecting the vector potential) has a term

$$K_3|u|\,|v|\cos\gamma\,\partial_x\gamma\left(\frac{K_2|v|^2}{K_1|u|^2+K_2|v|^2}-\frac{1}{2}\right), \quad (4)$$

which generates a spontaneous supercurrent centered on the DW parallel to the y axis, because $\cos\gamma\,\partial_x\gamma$ is finite near $x=0$. In Fig. 1(b) the result of the self-consistent solution is given for j_y and the generated magnetic field b_z. Screening countercurrents lead to a decay of b_z with the length scale δ ($=\kappa\xi$). The approximate solution gives a maximal value of $b_z \sim K_3(K_2-0.5)/\kappa^2$ [$\approx 1.25\times10^{-3}$ in the case of Figs. 1(a) and 1(b)]. Since b_z is an odd function of x there is no net magnetization.

Vortex on a domain wall.—The DW shown in Fig. 1 is twofold degenerate. The relative phase γ passes from $-\pi/2$ to $\pi/2$ through $\gamma=0$, but the way through $\gamma=\pi$ is clearly degenerate. If on one DW these two structures are present, then they are separated by a line defect similar to a Bloch line in a ferromagnet. Let us choose this line to be parallel to the z axis. If we go around it, the relative phase γ winds once:

$$\oint \partial\gamma\cdot ds = 2\pi[-\pi/2 \to 0 \to +\pi/2 \to \pi \to 3\pi/2(\equiv -\pi/2)]\,.$$

Hence this line must be a special singularity of the order parameter where one component vanishes to ensure the topological stability of this structure. The magnetic flux can be calculated by integrating $\int(\partial\phi-\mathbf{a})ds$ along a rectangular path around the line where the parts parallel to the DW are so distant that $\int a_y\,dy$ gives no contribution. The perpendicular part is obtained by the solution of the single DW problem using the fact that $j_z=0$;

$$\partial_x\phi - a_x = -\frac{K_2|v|^2}{K_1|u|^2+K_2|v|^2}\partial_x\gamma + \frac{K_3|u|\,|v|}{K_1|u|^2+K_2|v|^2}a_y\cos\gamma\,. \quad (5)$$

An approximate calculation of the finite flux enclosed leads to ($|u|\approx|v|$)

$$\Phi = \oint \mathbf{a}\,ds \approx n\Phi_0 + K_2\Phi_0 + O(K_3^2/\kappa^2)\,, \quad (6)$$

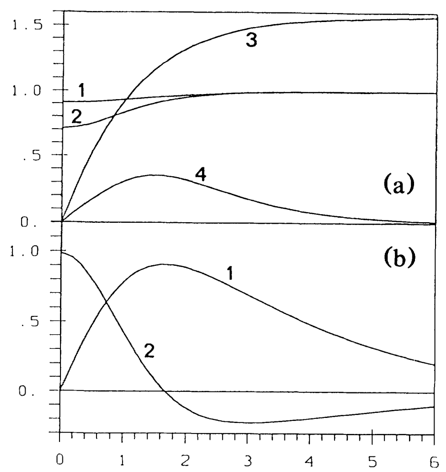

FIG. 1. Self-consistent numerical solution of the GL equations for the domain wall at $x=0$ with $\beta_2=0.1$, $\beta_3=-0.6$, $K_1=0.7$, $K_3=0.1$, and $\kappa=4$ (Ref. 7). (a) Order parameter $(u,v)=[|u|,|v|\exp(i\gamma)]\exp(i\phi)$. 1, $|u|$; 2, $|v|$; 3, γ; 4, $10\times\phi$. $\gamma(x)$ is an odd and $\phi(x)$ is an even function of x. (b) Magnetic field and current distribution. 1, $10^3 b_z$; 2, $10^3 j_y$. $b_z(x)$ is an odd and $j_y(x)$ is an even function of x.

where n is the winding number of the total phase ϕ. Φ_0 is the universal flux quantum in dimensionless units ($\Phi_0=2\pi$). Since $0<K_2<1$, this new type of vortex can contain a magnetic flux which is a fraction of Φ_0: $\Phi\approx\pm K_2\Phi_0$, $\pm(K_2-1)\Phi_0$ ($\pm$ depends on the winding sense of γ).[8] Two neighboring lines on a DW enclose together an integer number of Φ_0. The winding sense of γ is opposite in the two vortices so that the only winding variable is ϕ which leads to the Abrikosov result $\Phi=n\Phi_0$. However, the creation and annihilation of fractional vortices can be different. It is not unlikely that a usual vortex trapped by a DW will decay into two fractional vortices. This is indicated in the calculations by Schenstrom *et al.* for the analogous superconducting phase of hexagonal UPt_3, where a spatial splitting of the vortex into two parts is observed.[9] A rough estimate of the energy expense $\tilde{\epsilon}$ to create a fractional vortex in the case $K_{1,2}=0.5$, $K_3=0.5$, $n=0$, and $\xi=1\ll\tilde{\xi}\ll\kappa$ gives $\tilde{\epsilon}=\ln(\kappa\tilde{\xi})/4$, whereas a usual vortex needs $\tilde{\epsilon}'=\ln\kappa$ in di-

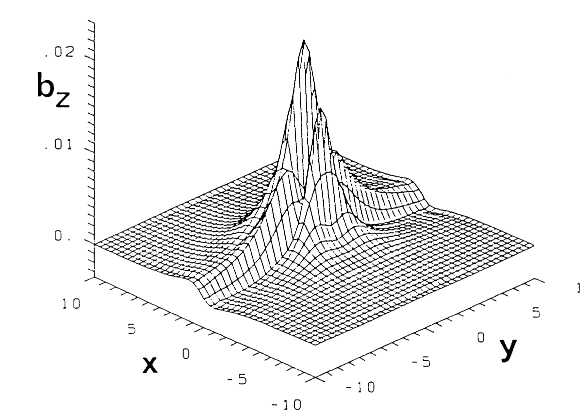

FIG. 2. The magnetic field distribution $b_z(x,y)$ in the self-consistent numerical solution of the GL equations for a line defect on the DW with the parameters of Fig. 1. The magnetic field b_z has two peaks due to the superposition of the DW and vortex currents. The calculated critical field is $\tilde{h}\approx 2.6\times10^{-3}$ and the enclosed magnetic flux is $\Phi\approx 1.6$.

mensionless quantities. From this energy we obtain a critical magnetic field $\tilde{h} = \tilde{\epsilon}/2\kappa^2\Phi = \ln(\kappa\xi)/4\kappa^2$, which is smaller than the standard lower critical field $h_{c1} = \ln\kappa/2\kappa^2$. This agrees with very recent calculations by Izyumov and Laptev for a simplified theory of fractional vortices.[10]

The currents in the DW distort the vortex due to the Lorentz force. Figure 2 shows the result of a self-consistent numerical calculation of the magnetic field distribution. The unusual form arises from a complicated superposition of the DW and the circular currents.

Surface magnetization.—It is not simple to find the correct boundary condition (BC) for the order parameter at the surface. Different surface scattering properties of the two basis states in Eq. (1) produce a different suppression of the components u and v. Using group-theoretical methods we introduce general BC by adding all second-order terms to Eq. (2) allowed by the lower symmetry at the surface. For an infinite planar surface, $y=0$, the remaining symmetry of D_{4h} is the group $C_{2v}(x)$ and the surface terms are

$$f_s = \delta(y)[g_1(|u|^2+|v|^2)+g_2(|v|^2-|u|^2)]. \quad (7)$$

The coefficients g_i are real. An additional BC prohibits current flow through the surface: $(\partial\times\mathbf{b})\cdot\hat{\mathbf{y}}(y=0)=0$. With the assumption $g_1 \approx g_2 > 0$, $|u|$ is not affected at $x=0$, but $|v|$ is strongly suppressed. Neglecting a and setting $|u| \equiv 1$ we find

$$\partial_y^2|v| + 2\alpha^2(|v| - |v|^3) = 0, \quad (8)$$

$$(K_1\partial_y|v| + \tilde{g}|v|)|_{y=0} = 0, \quad (9)$$

with $\alpha^2 = (1+4\beta_2-\beta_3)/8K_1$ and $\tilde{g} = g_1+g_2$. The solution is

$$|v| = \tanh[\alpha(y-y_0)], \quad 2\alpha y_0 = \sinh^{-1}(2K_1\alpha/\tilde{g}). \quad (10)$$

The fourth term of Eq. (3) again generates a supercurrent parallel to the surface;

$$j_x = \frac{K_3}{2\kappa^2}\partial_y|v|\sin\gamma \approx \frac{K_3}{2\kappa^2}\frac{\alpha\sin\gamma}{\cosh^2[\alpha(y-y_0)]}. \quad (11)$$

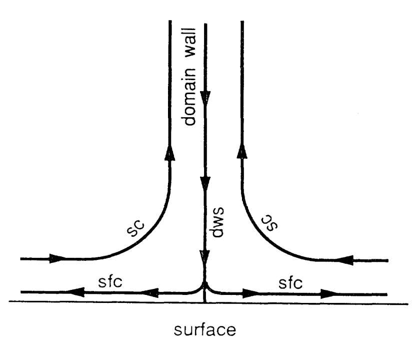

FIG. 3. The simplest current distribution when a DW intersects a surface. dwc, domain-wall current; sfc, surface current; sc, screening current.

The magnetic field due to this current is screened towards the interior, but now there is a finite net magnetization concentrated near the surface, $m_z \approx K_3\sin\gamma(1-K_1\alpha/\tilde{g})/2\kappa$ per unit surface area (ξ^2). Thus a single domain sample has a finite magnetization produced spontaneously by the broken-time-reversal symmetry of the superconducting phase.

Since in the domains $(u,v)=(1,\pm i)(\gamma=\pm\pi/2)$ this magnetization has opposite sign, there will generally be regions of different surface magnetization separated by DW. An external field $\mathbf{h}_{ext}$ introduces a difference in the Gibbs free-energy density $g = f - 2\kappa^2\mathbf{b}\cdot\mathbf{h}_{ext}$ between domains causing a force of magnitude $\sim 2\kappa^2 m_z\hat{\mathbf{z}}\cdot\mathbf{h}_{ext}$. So near the surface the DW moves to favor the domain with $m_z h_{ext,z} > 0$. This effect leads to a small decay of the diamagnetic response of the superconductor. The time scale can be rather long, if the DW is pinned on impurities that require thermal activation. It is clear that such slow processes can give rise to irreversible behavior in the magnetic field.

A domain wall at the surface.—When a DW intersects the surface the simplest structure is that its currents convert continuously into surface currents (Fig. 3) so that the current flows around each domain, positively for the domain $(1,+i)$ and negatively for $(1,-i)$.[1] Essentially this is confirmed by the self-consistent calculation. However, the solution shows more structure than expected (Fig. 4). In one of the two corners formed by the surface and the DW a peak in the magnetic field appears. It has no counterpart on the other side of the DW. The choice of DW (i.e., γ through 0 or π) determines the side on which this peak occurs. Therefore this peak has a similar origin as the line defect on a DW and can be considered as a precursor of a fractional vortex

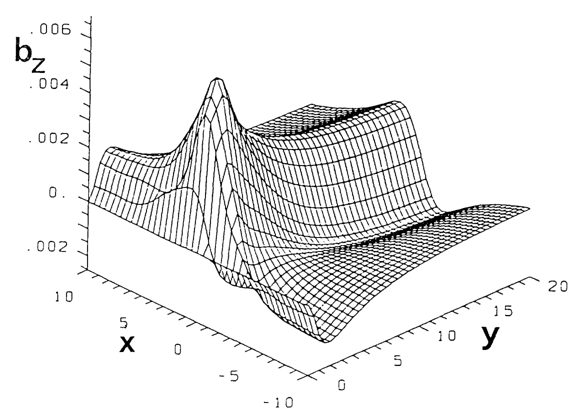

FIG. 4. The magnetic field distribution $b_z(x,y)$ in the self-consistent numerical solution of the GL equations for a DW at a surface with the parameters of Fig. 1. The magnetic field b_z varies smoothly from the DW to the surface, where it leads to a net magnetization. A significant peak occurs on one side of the DW depending on whether γ passes through 0 (shown above) or π.

which would appear in a parallel external field. Without such a field this peak is a magnetic flux trapped in a potential well due to the Lorentz force of the DW-surface currents and the screening countercurrents. The magnitude of this flux is not a topological charge, rather it depends on the strength of the trapping well.

In conclusion, we have shown that the low-field behavior of nonunitary superconducting phases is a complex problem with more ways of incorporating magnetic flux than by introducing Abrikosov vortices. This leads to various possibilities to identify such phases. The experiments of Mota and co-workers[3,4] are encouraging although all other possible mechanisms need to be carefully ruled out.

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[5]Note the anyon and generalized flux phases discussed in Ref. 2 may differ in some ways from the generalized BCS states discussed here.

[6]E. V. Thuneberg, Phys. Rev. B **36**, 3583 (1987).

[7]An increase of the parameter β_2 leads to a domain-wall structure of higher symmetry (second-order transition). Some of the properties mentioned below are not present in this type of wall.

[8]The enclosed magnetic flux also depends on the orientation of the DW. A rotation by an angle θ around the z axis would lead to a flux of $\Phi(\theta) \approx \tilde{K}(\theta)\Phi_0$, $[1-\tilde{K}(\theta)]\Phi_0$, where $\tilde{K}$ is approximately given by $\tilde{K} = K_1 \sin^2\theta + K_2 \cos^2\theta + K_3 \sin\theta\cos\theta$.

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