Intermediate state in a type-I superconducting sphere: pinning and size effect

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Abstract

Simulations based on the time-dependent Ginzburg–Landau equations show that the magnetization and spatial structure of the intermediate state are strongly affected by both the radius of the sphere and by the concentration of pinning centers. The intermediate state undergoes a transformation from a one-domain structure for a small sphere to a multi-domain structure in big spheres. In spheres where part of the superconducting material is replaced by the 0.5% randomly distributed normal phase (dirty case), the intermediate state demonstrates a pronounced turbulence behavior.

Keywords: intermediate state, flux turbulence, type-I superconductors

(Some figures may appear in colour only in the online journal)

1. Introduction

The subject of bulk superconductivity in a magnetic field has been studied for many years and is well described in numerous text books [1]. Long before Bardeen–Cooper–Schrieffer (BCS) [2], some very successful phenomenological theories of superconductors had been conceived. The most powerful tool was the Ginzburg–Landau (GL) theory, which was conceived in 1950 [3]. Considering superconductivity as a phase transition, GL constructed a gauge invariant field theory. GL theory yields the Abrikosov parameter \( \kappa = \lambda / \xi \), where \( \lambda \) and \( \xi \) are the magnetic field penetration length and the coherence length, respectively. In their original publication, GL showed that the solutions of their equations behave quite differently when \( \kappa < 1/\sqrt{2} \) and \( \kappa > 1/\sqrt{2} \), corresponding to what have come to be called type-I and type-II superconductors. The infinite type-I superconducting cylinder in parallel to the axis magnetic field expels the magnetic field from its interior for fields smaller than the thermodynamic critical field \( H_c \). Meanwhile, for applied fields larger than \( H_c \), the sample is in the normal state, fully penetrated by the magnetic field. Magnetization in type-I superconductors demonstrates hysteresis at small \( \kappa \) (\( \kappa < 0.42 \)) [4]. In a real system geometry, the external magnetic field reaches its critical value in some parts of the system while remaining smaller in the rest (diamagnetic factor). This factor leads to the appearance of an intermediate state, in which regions of both normal and Meissner states coexist. Due to the proximity effect, these regions overlap, blurring the borders between domains. The superconducting sphere with the diamagnetic factor (\( n = 1/3 \)) is the oldest studied example of such a system [5]. Experimental and theoretical efforts are growing to obtain a general understanding of the problem [6–12]. The fundamental problem is that in a finite system, it is generally impossible to predict the topology of the intermediate state based solely on the energy minimization [7–19]. In fact, in this approach we have to guess the topology of the intermediate state and then minimize the energy that is not feasible for restricted geometry. From this point of view, exact numerical simulation of the time-dependent GL equations in mesoscopic samples is the only way to study the dynamics of the intermediate state from first principles with no initial assumption on the spatial distribution of the order parameter and magnetic field inside the sample. There are several factors that affect and often determine the topology of the intermediate state. Among them are sample size (size effect) and the inclusions of the normal phase (pinning centers) [20–25]. In spite of efforts the problem is far from solved for the following reasons. First of all, most of the numerical simulations were performed for a large GL...
parameter ($\kappa = 0.42$ for Pb in [26]), and therefore the dependence of the intermediate state on $\kappa$ was ignored along with the hysteresis behavior of the magnetization. Secondly, the size effect and pinning effect affecting the intermediate state structure have not been systematically studied. (In fact, in the overwhelming majority of published papers, flux pinning was omitted completely). Our calculation method, in contrast to others, allows learning of the dynamics of the intermediate state inside the sphere. Here, we numerically study the intermediate state in two spheres ‘big’ and ‘small’ in a wide ranges of magnetic fields at small GL parameter $\kappa = 0.18$ typical for clean Sn. $\kappa$ changes to 0.22 by the non-magnetic randomly distributed impurities in dirty spheres. We study the pinning effect on the intermediate state and find a topologically induced change in magnetization in the big spheres, where the intermediate state adopts a multi-domain form. Hysteresis in magnetization recorded in the clean spheres was practically absent in dirty samples.

2. Model and numerical simulations

The method of minimal energy proposed by Landau in his pioneering paper [7, 8] is based on the initial assumption of a spatially distributed intermediate state and is robust only for a simple geometry. For example, it is natural to assume for a thick superconducting film the existence of an alternating normal and superconducting domains with different sizes, calculate the energy, and then minimize it by taking the domain’s sizes as trial parameters. Even in this case, there is no evidence that this state really has the lowest energy. Moreover, in samples with more complicated geometry, the spatial structure of the intermediate state is a-priori unknown and this method fails. The problem is related to an irreversible (dissipative) force that determines the evolution of the system and the topology of the solution. The time-dependent Ginzburg–Landau (TDGL) theory just allows us to solve the problem from the first principles. Starting from an arbitrary state, the system relaxes to the steady state without any assumptions on its shape.

We consider here the magnetization as a function of the external magnetic field for type-I superconducting spheres with radii in the range $\lambda \ll \xi_{\text{eff}} \ll R$, where $\lambda$ is the magnetic penetration depth, and $\xi$ is the coherence length of the clean materials. Meanwhile, $\xi_{\text{eff}}^{-1} = \xi^{-1} + l^{-1}$ (1 is the mean free electron path) is the effective coherence length in dirty superconductors. In our model the normal phase inclusions (‘dirty case’) amount to 0.5% of the total volume of the sphere. In fact both the coherence and the penetration lengths are changed by the impurities. In the GL theory we are interested in the modified $\kappa_d$ parameter. Using the well-known relation $\kappa_d \approx \kappa(1 + \xi/1.35\xi)$ [1] for the effective $\kappa_d$ and estimating the mean free path for electrons in the dirty sphere as $l = h(N_{\text{imp}}/N_{\text{total}})^{-1/3}$ (where $h = 0.5\xi$ is the step of the grid, $N_{\text{imp}}$, $N_{\text{total}}$ are the number of grid points where the order parameter is zero (impurities) and the total number of grid points, respectively), one obtains $l = 3$ (in our case this ratio is $N_{\text{imp}}/N_{\text{total}} = 1/200$). As a result, $\kappa_d$ varies from 0.18 for a clean sphere to 0.22 for a dirty one. The magnetic moment of a sphere with radius R subjected to an external magnetic field $E$ was studied by numerically solving the time-dependent GL equations [27]. Starting from the dimensionless GL Hamiltonian [28],

\[
G = \int \int d^3r \left( \left( \frac{\partial}{\partial t} - iA \right) \psi \right)^2 + \left( \frac{1}{2} |\psi|^2 + \kappa_d^2 (\partial \times A - H)^2 \right).
\]

Here, $\theta = T/T_\text{c}$, $T$ is the temperature, $T_\text{c}$ is the critical temperature, $\psi$ is the superconducting order parameter, and $A$ is the vector potential in $\sqrt{2}\lambda H_c$ units. $H_c$ is the bulk thermodynamic magnetic field; the applied magnetic field $H$ is in $\sqrt{2}\lambda H_c$ units, while the coordinates are scaled by the coherence length [29]. The TDGL is constructed in the following way. In equilibrium, the GL energy has a minimum with respect to $A$ and $\psi(\psi^* \frac{\delta G}{\delta \psi} = 0, \frac{\delta G}{\delta A} = 0)$. If the superconductor is driving out of equilibrium, both the order parameter and the vector potential should relax back to their equilibrium values.
rate of relaxation depends on the deviation from equilibrium and the relaxation equations \[27, 30\]:

\[
\frac{\partial \psi}{\partial t} = -\frac{\delta G}{\delta \psi^*} + f_\psi, \quad \frac{\partial A}{\partial t} = -\frac{1}{2} \frac{\delta G}{\delta A} + f_A.
\] (2)

Two dimensionless TDGL equations are as follows (see details in \[28\]):

\[
\frac{\Gamma}{\theta} \frac{\partial \psi}{\partial t} = (1 - \theta) \psi - |\psi|^2 \psi - (i \nabla + A)^2 + f_\psi, \quad (3)
\]

\[
\frac{\partial A}{\partial t} = -\nabla \times \nabla \times A - \frac{i}{2\kappa^2} (\psi^* \nabla \psi - \psi \nabla \psi^*) - \frac{1}{\kappa^2} [\psi]^2 A + f_A,
\]

where \(f_\psi, f_A\) are the magnitudes of the order parameter and magnetic vector potential random noise. We use a link-variable approach, a rectangular Cartesian grid \[31\], and the boundary conditions for the order parameter at the sphere border \((i \nabla + A) = 0\) and for magnetic field \(H = H_z\) far from the sphere. A random number generator with a uniform distribution of the zero magnitudes of the order parameter at grid points (normal phase) was used: 0.5\% of the grid points were randomly replaced by the order parameter zeros. The set of equation (2) was solved numerically until a stationary state was reached. The stationary state solution provides the local magnetic field inside the sphere \(H(r)\) and the magnetic induction \(B = \int H(r) d^3r / \int d^3r\). The magnetic moment has the usual form: \(M(H) = (B - H) / 4\pi\) (the magnetic moment \(M\) is in \(\sqrt{2} H_c\) units). The topology of the intermediate state and its characteristics strongly depend on the radius of the sphere, and the order parameter inside the sphere is calculated for various magnetic fields at the magnetization curve. We consider Abrikosov parameter \(\kappa = 0.18\) (in the clean case) in

Figure 3. Density of the superconducting electrons inside a small sphere in the Y–Z projection for clean \((\kappa = 0.18)\) and dirty \((\kappa_d = 0.22)\) superconducting spheres. The magnetic field is 0.05. The magnetic field is in the z direction.

Figure 4 Density of superconducting electrons inside a small sphere in the Y–Z projection for clean (left) and dirty (right) spheres. The magnetic field is 0.06.
two spherical samples: a small sphere with radius $R = 6\xi$, and a large sphere with $R = 15\xi$ (here, $\xi$ is the coherence length; all calculations were performed for step $h = 0.5\xi$ and the reduced temperature $\theta = 0.5$).

3. Small sphere $R = 6\xi$

The initial state of the superconducting sphere in zero magnetic field is plotted in figure 1. The color scale denotes the magnitude of the superconducting electrons density changing from 0.5 (the red color corresponds to the uniform superconducting state where $|\psi|^2 = 1 - \theta = 1/2$) to 0 (black).

The magnetization curve $M(H)$ (see figure 2) was calculated both when the external magnetic field increased from zero to the critical field $H_{ab}$ (denoted as the forward direction; here, $M(H_{ab}) = 0$) and when the magnetic field decreased from $H_{ab}$ to zero (i.e. in the backward direction). The lower, deeper curve depicts the magnetization of the clean superconducting sphere while the upper, more shallow curve shows the magnetization of the sphere with normal phase inclusions (‘dirty case’) amounting to 0.5% of the total volume of the sphere.

This figure demonstrates weak hysteresis for the clean sphere superconductor (where the red circles indicate forward magnetization and blue squares indicate backward magnetization) and for the dirty sample. The forward magnetization (green circles) coincides with the backward magnetization (blue triangles). Conversely, hysteresis is completely absent for the dirty sample. The magnetization curves for a small sphere demonstrates reversibility at most magnetic field values. The intermediate state inside the sphere shows a strong difference between the clean and dirty samples. This is illustrated in figure 3, where the density of superconducting electrons $n_s = |\psi|^2$ is plotted for the same magnitudes of the magnetic fields in both $x$-$y$ and $y$-$z$ projections.

4. Large sphere $R = 15\xi$

The magnetic moment of the superconducting sphere in this case is more complicated. It demonstrates the hysteretic behavior typical for bulk type-I superconductors with a small $\kappa$ parameter [29]. The magnetic moment as a function of the external magnetic field was calculated in four different protocols: in the forward direction for (a) clean and (b) dirty spheres subjected to an external magnetic field increasing from zero to the critical field $H_{cb}$, and (c, d) for a magnetic field decreasing from $H_{cb}$ to zero for clean and dirty samples (the backward direction). Results are presented in figure 5.

The magnetization demonstrates both Meissner behavior at a small magnetic field and a well-pronounced intermediate state evidenced by the long tail that extends to $H_{bc}$, where the superconducting state is suppressed by the applied magnetic field and $M(H_{ab}) = 0$. Magnetization of the clean spheres demonstrates the well-pronounced hysteresis typical for bulk type-I superconductors where the metastable normal state appears in magnetic fields in the range $H_{ce} < H < H_{ab}$; here, $H_{ce}$ is the surface critical magnetic field ($H_{ce} = 0.69H_{c2}$, and $H_{c2}$ is the second critical magnetic field). In our case, however, the unstable, hysteretic region extends down to zero magnetic field ($H_{ce} \to 0$). The intermediate state in large spheres is complicated and topologically diverse. It contains the multi-domain superconducting to normal structure where the proximity effect smooths the boundaries between domains typical for macroscopic type-I superconductors; meanwhile, the density of superconducting electrons is spatially modulated and there is no sharp border between the domains. Typical spatial structures of the intermediate state domains are presented in figures 6 and 7.

In the external magnetic field $H = 0.04$, the intermediate state in the big, clean sphere has a typical Meissner state shape while in the dirty case the intermediate state manifests turbulent behavior. In a larger magnetic field, $H = 0.06$, the Meissner state in a clean sphere is broken and the intermediate state contains a set of domains (tubes) separated by normal regions (where weak superconductivity is induced by the proximity effect). In the dirty sphere with random inclusions of the normal phase, the turbulent state becomes more
Figure 6. The intermediate state in the large, clean superconducting sphere \((R = 15\xi)\) for an external magnetic field \(H = 0.04\), where the Meissner state is well pronounced.

Figure 7. The intermediate state in the large, dirty superconducting sphere \((R = 15\xi)\) for the external magnetic field \(H = 0.04\), where flux turbulence is exhibited.

Figure 8. The intermediate state in a large, clean superconducting sphere \((R = 15\xi)\) in an external magnetic field \(H = 0.06\).
pronounced and normal state percolation domains cross the sample (figures 8 and 9).

For the magnetic field $H = 0.06$, the Meissner intermediate state in clean, big spheres is split by several domains (figure 8) while in the dirty samples the turbulence state becomes more pronounced (figure 9). The domains in the clean sphere form an Abrikosov lattice similar to that in type-II superconductors. At magnetic fields $H = 0.07$, $H = 0.08$ (the critical magnetic field $H_{c1} = 0.1$), the amplitude of the domains decreases dramatically while the shape of the intermediate state and number of domains are enhanced. The turbulent intermediate state in this case completely disappears while the sphere undergoes a transition to the normal state (figures 10 and 11).

5. Summary

The magnetic moment and intermediate state as a function of the external magnetic field for small (radius $R = 6\zeta$) and large ($R = 15\zeta$) type-I superconducting spheres were calculated numerically in the framework of the time-dependent GL equations. Both the clean and dirty limit spherical samples (dirty case $-0.5\%$ of the superconducting material of the spheres replaced by the normal phase playing the role of the pinning centers) were studied. It was demonstrated that the magnetic moments of the small sphere does not exhibit hysteresis and irreversibility (figure 2). In the large, clean sphere, magnetization behaves similarly to an infinite system with a well-pronounced hysteresis. The unstable normal hysteretic line (blue squares in figure 5) extends down to zero field due to the small Abrikosov $\kappa$ parameter [4]. Moreover, the intermediate state inside the spheres demonstrates (at the same external magnetic field) the domains (tubes) in the clean case and a turbulent structure in the dirty case.

These results are in good qualitative agreement with the experiment of [6], where a tubular structure in large, clean spheres was observed. In particular, figure 4 of [6] shows the

Figure 9. The intermediate state in large, dirty superconducting sphere ($R = 15\zeta$) in an external magnetic field $H = 0.06$.

Figure 10. Two projections (a) and (b) of the density of superconducting electrons inside the large, clean sphere in the intermediate state for applied magnetic field $H = 0.07$. 
The turbulent behavior in the intermediate state of small, clean sphere consists of just one superconducting domain. The domain extends along the magnetic field direc-

tion. The turbulent behavior in the intermediate state of a hemisphere. The flux tubes appear at the descending branch of the magnetization curve in a good agreement with our figure 10. The size of the sphere plays an extremely important role, strongly affecting the structure of the intermediate state. In particular, the intermediate state in a small, clean sphere consists of just one superconducting domain. The domain extends along the magnetic field direction. The turbulent behavior in the intermediate state of small, dirty sphere is shown in figures 3, 7 and 9. Note that while our calculation (following the general argument of [4]) for tin was performed at small parameter \( k_d \), the experiment in [6] was done on lead samples with a relatively large \( k_d = 0.42 \). This is reflected in a qualitative difference of the magnetization curve: hysteretic for tin versus reversible for lead.

The Abrikosov parameter \( k_d \) for the tin samples varies from 0.18 for clean spheres to 0.22 for dirty ones. Separate calculations [25] for a clean sphere with \( k_d = 0.22 \) demonstrated that the tube domain structure is well pronounced. Our calculation for \( k_d = 0.22 \) with randomly distributed pinning centers results in turbulence. Therefore, we conclude that disorder and its influence on the superconducting order parameter is largely responsible for transformation of the regular domain structure into a turbulent one.

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Figure 11. Density of superconducting electrons in two projections (a) and (b) inside the large, clean sphere (\( R = 15 \xi \)) in the intermediate state for applied magnetic field \( H = 0.08 \).
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