The intermediate type-I superconductors in the mesoscopic scale

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M. Tinkham and P. G. de Gennes, described in their books[1, 2] the existence of an intermediate type-I superconductor as a consequence of an external surface that affects the well known classification of superconductors into type-I and II. Here we consider the mesoscopic superconductor where the ratio volume to area is small and the effects of the external surface are enhanced. By means of the standard Ginzburg-Landau theory the Tinkham-de Gennes scenario is extended to the mesoscopic type-I superconductor. We find new features of the transition at the passage from the genuine to the intermediate type-I. The latter has two distinct transitions, namely, from a paramagnetic to diamagnetic response in descending field and a quasi type-II behavior as the critical coupling $1/\sqrt{2}$ is approached in ascending field. The intermediate type-I phase proposed here, and its corresponding transitions, reflect intrinsic features of the superconductor and not its geometrical properties.

I. INTRODUCTION

The classification of superconductors on the basis of their magnetic properties has been a hard earned knowledge. Nearly twenty five years after the discovery of superconductivity by H. Onnes, the experimental measurements of L. Shubnikov showed that the magnetic properties of alloys were very different from those of the pure metals[3]. The explanation had to wait for more than twenty years until the development of the theoretical work of A. Abrikosov[4], based on a new and unknown at the time phenomenological theory, the Ginzburg-Landau (GL) theory. Nowadays the GL theory enjoys enormous recognition for its applications in various fields, ranging from phase transitions to particle theory since the Higgs model may be regarded as a relativistic generalization of the GL theory[5]. The classification of superconductors is straightforwardly obtained from the ratio between two fundamental measurable lengths, namely, the London penetration length ($\lambda$) and the coherence length ($\xi$). A. Abrikosov found that the single coupling of the GL theory, $\kappa \equiv \lambda/\xi$, splits the superconductors into two classes, namely, type-I and II, and the critical value separating them is $\kappa = 1/\sqrt{2}$. Although his simplified geometry is beyond reality since there are no boundaries, this choice is useful in the sense that excludes any geometrical factor from entering the classification scheme. This critical coupling was also found by E. Bogomolny[6, 7] in the context of string theory thus rendering the transitions in $\kappa$ obtained here of possible interest to other areas of physics besides superconductivity. The magnetic difference between type-I and II stems from the existence of a vortex state in the type-II that disappears when the normal state sets in at the upper critical field $H_{c2}$. The type-I simply does not sustain a vortex state and goes to the normal state at the thermodynamic field $H_c$, where the normal and the superconducting Gibbs’ free energies become equal. Superconductivity was discovered in the pure elements, known to be type-I with the exception of Nb, V, and Tc which are type-II. Distinctively, from Shubnikov’s days to now, superconductivity has been discovered in alloys and other composite materials[8], which are mostly type-II. The many new families of high-$T_c$ superconductors[9] fall in the latter case, such as the cuprates, fullerenes, MgB$_2$, pnictides and many others. Nevertheless unexpected type-I superconductivity has been found in some alloys, such as TaSi$_2$[10], the heavily boron-doped silicon carbide[11], YbSi$_2$[12], and more recently in the ternary intermetallics YNiSi$_3$ and LuNiSi$_3$[13], rendering them of great interest, and worth

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of study. The coupling $\kappa$ varies greatly among superconductors ranging from very high, for the high-$T_c$ materials YBaCuO$_{2+\delta}$ ($\kappa = 95$), to very low values, for the pure metals [2] (Al (0.03), In (0.11), Cd (0.14), Sn (0.23) and Ta (0.38)) and also for some of the alloys [12, 13], such as (YbSb$_2$ (0.05), YNiSi$_3$ (0.1)). Since $\xi$ decreases with disorder, doping the material by impurities can produce adjustable $\kappa$ compounds that allow for the study of transitions in this coupling.

According to M. Tinkham [1] and P. G. de Gennes [2] surface states split type-I superconductors into genuine and intermediate classes that comprise the coupling ranges $\kappa < \kappa_c$, and $\kappa_c < \kappa < 1/\sqrt{2}$, respectively, where $\kappa_c = 0.417$. Although this transition is driven by surface effects it solely reflects an intrinsic property of the superconductor, namely, $\kappa$, as discussed below. There has been an intense search for this genuine-intermediate transition both theoretically [14, 15] and experimentally [16, 19] in the seventies, but after this period it became an elusive topic. The existence of the transition was even questioned [20] and its study no longer pursued. Now that type-I alloys were found, the classification of superconductors acquired a renewed interest. In this paper we pursue this study in the mesoscopic scale, which offers a unique framework since surface effects are enhanced there. In the bulk (no surface) the relation $H_{c2} = \sqrt{2\kappa}H_c$ elucidates the difference between types I and II since for $\kappa < 1/\sqrt{2}$ ($\kappa > 1/\sqrt{2}$) $H_{c2} < H_c$ ($H_{c2} > H_c$), thus a type-I (type-II) superconductor. Interestingly M. Tinkham has stressed in his book [1] that both fields $H_{c2}$ and $H_c$ are directly measurable in type-I superconductors. The normal state is retained below the thermodynamic field $H_c$ until the lower field $H_{c2}$ is reached [17], where the order parameter (magnetization) abruptly becomes non-zero. From the other side evolving from the superconducting state this lasts beyond $H_{c2}$ until $H_c$ is reached and the normal state is recovered. However the presence of a surface modifies the above bulk analysis since superconductivity is extended beyond $H_{c2}$ to exist with thickness $\xi$ around the external boundary. Saint-James and de Gennes [21] found the critical field $H_{c3} > H_{c2}$ whose value in case of a flat interface is $H_{c3} = 1.695H_{c2}$ [21–27]. $H_{c3}$ is also present in type-I superconductors [28] and signals several processes, e.g., the expulsion of magnetic flux [29, 31]. In fact it is $H_{c3}$ which gives rise to the genuine and intermediate type-I superconductors, associated to $H_{c2} < H_{c3} < H_c$, and $H_{c2} < H_c < H_{c3}$, respectively, such that $\kappa_c$ is obtained from $H_{c3} = H_c$.

In this paper, we report properties of the genuine-intermediate transition such as its critical $\kappa$ and also other transitions in $\kappa$ in the intermediate phase, as seen by isothermal magnetization $M(H)$ curves. These several transitions can be experimentally investigated by the ballistic Hall magnetometry technique [31] applied to submicron size superconductors. Type-I mesoscopic superconductors have been investigated both theoretically [32] and experimentally [33], however the genuine-intermediate transition in $\kappa$ is firstly considered here and found to acquire new properties. As one goes from the macroscopic to the mesoscopic scale, $\kappa_c$ gives rise to $\kappa_{c1}$ and also to the transitions $\kappa_{c2}$ and $\kappa_{c3}$ inside the intermediate phase. In descending field starting from the normal state the magnetization can display paramagnetic regions but it becomes diamagnetic at any applied field provided that $\kappa < \kappa_{c2}$. Hereafter we call this as the dia-para transition. In ascending field the magnetization ($-M$) has a nearly linear growth (Meissner state) up to a maximum and next undergoes an abrupt fall and a residual magnetization regime is reached that only exists if $\kappa > \kappa_{c3}$. The Meissner state is followed by the disappearance of the magnetization for $\kappa < \kappa_{c3}$. This residual magnetization signals the quasi type-II class and is caused by giant vortices (for a discussion about giant vortices in mesoscopic superconductors see for instance Refs. [34] and [35]).

We remark the presence of several notable fields in our study. In the up branch there are $H'_c$ (the peak of $-M$) and $H''_c$ (the vanishing of the magnetization). They are very near to each other for $\kappa < \kappa_{c3}$ but not for $\kappa > \kappa_{c3}$. Both critical fields fall above $H_c$ (see supplementary material), and so are inside a region of metastability since the superconducting state there has higher Gibbs free energy than the normal state. In the descending branch we define $H_{c3}$ where the magnetization becomes non-zero and the superconducting state sets in. As shown here these fields, as well as the $M(H)$ curves, are strongly dependent on $\kappa$ in the type-I domain. We bring numerical evidence that the genuine-intermediate mesoscopic transition takes place at a coupling lower than the macroscopic one, $\kappa_{c1} < \kappa_c$. Interestingly the dia-para transition occurs at $\kappa_{c2} \approx \kappa_c$, thus near to the macroscopic transition a so far fortuitous coincidence. The transition to the quasi type-II class takes place for $\kappa_{c3} < 1/\sqrt{2}$.

Our numerical analysis was carried on a very long needle with square cross section of size $L^2$ in the presence of an applied field parallel to its major axis. The needle is sufficiently long such that the top and the bottom surfaces can be ignored and just a transverse two-dimensional cross section needs to be considered. The square cross section is the most suited to our numerical procedure which is done on a square grid. The boundary conditions are smoothly implemented in this geometry, namely, of no current exiting the superconductor and that at the surface the local field meets the external applied field. Nevertheless our major findings hold independently of the selected cross section geometry, though the value of the critical fields and of the delimiting $\kappa_{c1}$ may be affected by it. We look at several cross section sizes, namely $L = \rho \lambda$, $\rho = 8, 12, 16, 24,$ and $32$.

It is well-known that the GL theory is the leading term of an order parameter expansion derived from the microscopic BCS theory [2, 36, 37]. In case the next to leading order corrections are included [27] an intermediate phase emerges in the diagram $\kappa$ versus $T$ in between the type-I and II domains. However the analysis of A. Vagov et al. [36, 57] does not take into account surface effects whereas here the intermediate phase is
solely due to these surface effects \[^{38}\], and for this reason the intermediate phase is found already in the standard GL theory level. The choice of an infinitely long system, instead of being a limiting factor, really expands the importance of the present results, once it allows to see intrinsic effects. Geometrical factors \[^{29\text{–}42}\] hinder the observation of the intrinsic transitions observed here. It is well known that a sufficiently thin type I superconductor turns into a type II one by a change of its thickness. The geometry of the cross section affects the \(\kappa\) values where transitions take place but not their existence, that only reflects the ordering among the critical fields.

II. THEORETICAL FORMALISM

The basis of our dimensionless treatment of the GL theory is \(\lambda\) and \(H_{e2} = \Phi_0 / 2\pi \xi^2\), that renders the free energy,

\[
G = \int \left[ \left( \left( \frac{-i}{\kappa} \nabla - A \right) \right) \left( \left( \frac{-i}{\kappa} \nabla - A \right) \right) \left( \frac{1}{2} \right) \left( \psi \right)^2 \right] d^3r + \int \left( \mathbf{h} - \mathbf{H} \right)^2 d^3r , \tag{1}
\]

in reduced units. Lengths are in units of \(\lambda\); the order parameter \(\psi\) is in units of \(\psi_\infty = \sqrt{\alpha/\beta}\), where \(\alpha\) and \(\beta\) are the two phenomenological constants of the GL theory; magnetic fields are in units of \(\sqrt{2}H_c\); and the vector potential \(\mathbf{A}\) is in units of \(\sqrt{2}\lambda H_c\). The GL equations become,

\[
\left( \frac{-1}{\kappa} \nabla - A \right) \psi + \psi \left( 1 - |\psi|^2 \right) = 0 , \tag{2}
\]

\[
\nabla \times \mathbf{h} = J_s , \tag{3}
\]

where \(J_s = \Re \left[ \bar{\psi} \left( \frac{1}{\kappa} \nabla - A \right) \psi \right]\) is the superconducting current density.

The GL equations were solved numerically within a suitable relaxation method and using the link-variable method as presented in Ref. \[^{43}\]. For this, we used a mesh-grid size with \(\Delta x = \Delta y = 0.2\lambda\). The applied magnetic field is adiabatically increased in steps of \(\Delta H = 10^{-3}\sqrt{2}H_c\) for both up and down branches of the field. In each simulation, \(\kappa\) was held fixed and the stationary state at \(H \mp \Delta H\) was used as the initial state for \(H\) for up and down cycles, respectively. The emergence of superconductivity in a long mesoscopic cylindrical (\(R \sim \lambda\)) in presence of an applied external field has been studied by G.F. Zharkov et al. \[^{44\text{–}47}\]. Their approach is limited to the search of solutions of the Ginzburg-Landau differential equations with radial symmetry which limits the search to central vortex states. In our numerical search through the link variable method we find the presence of point vortices forming various geometrical patterns inside the superconductor that fall beyond their description. Hence the observation of the present \(\kappa_{c1}\) transitions are beyond the scope of their framework since they are limited to a sub set of the possible vortex states.

III. RESULTS AND DISCUSSION

The intermediate and the genuine type-I classes are distinguishable by their magnetic properties. In decreasing field the genuine class features a direct and abrupt change from the normal state to the Meissner state, \(i.e.,\) vortices are never trapped inside the superconductor. In the same situation the intermediate class displays vortices, which are trapped inside either as single or giant ones and then are gradually or suddenly expelled. The two classes are associated to specific \(\kappa\) ranges, and to determine them we have performed a series of numerical simulations varying \(\kappa\) in steps of \(\Delta \kappa = 0.01\). Within this precision we were able to numerically obtain \(\kappa_{c1}, \kappa_{c2}\) and \(\kappa_{c3}\) for all the \(L\)'s under investigation. In what follows, we report properties of the genuine-intermediate, dia-para and quasi type-II transitions, the latter two being inside the Intermediate type-I class.

Fig. 1 features the genuine-intermediate transition through the number of vortices, \(N\), trapped at \(H'_{c3}\). The superconductor response is markedly distinct according to \(\kappa\), and this is exemplified here for \(L = 16\lambda\) and \(L = 24\lambda\). Above \(\kappa_{c1}\), which is equal to 0.28 for \(L = 16\lambda\), and 0.19 for \(L = 24\lambda\), \(N\) varies according to \(\kappa\), thus corresponding to the intermediate type-I class. However, below \(\kappa_{c1}\), \(N\) drops to zero showing that no vortex en-
FIG. 2. (Color online) The magnetization in descending field reveals the transition at \( \kappa_{c2} \), shown in the main panel as a function of \( L \), from paramagnetic to diamagnetic response. Insets (a) and (b) display typical magnetization curves below and above the transition. Inset (b) shows a still paramagnetic magnetization while in panel (a) it has become totally diamagnetic.

ters the needle at \( H'_{c3} \), which characterizes the genuine type-I class. The insets of Fig. 1 depict the magnetization curve of two selected \( \kappa \) values belonging to the two classes, chosen as 0.28 and 0.33 for \( L = 16\lambda \). The left inset depicts a magnetization that goes directly from the normal \((M = 0)\) to the Meissner state, while the right inset shows a spike highlighted by the black ellipse that corresponds to the coalescence of flux in form of vortices and their subsequent exit at an infinitesimally lower field. The magnetization curves show the up (blue line) and down (red line) branches for both insets.

Fig. 2 features the dia-para transition. The main panel presents \( \kappa_{c2} \) as a function of \( L \). The small red circles indicate the numerically obtained \( \kappa_{c2} \) values of 0.4425, 0.4175, 0.4025, 0.4025, 0.4075, 0.415, and 0.415, for \( L/\lambda \) equal to 8, 9, 10, 12, 16, 24, and 32, respectively. Although \( \kappa_{c2} \) is a non-monotonic function of \( L \), it asymptotically approaches a limiting value for large \( L \), suggestively close to \( \kappa_c \), which is the value that delimits the genuine-intermediate transition in the macroscopic limit. Insets (a) and (b) of Fig. 2 depict the transition occurring at \( \kappa_{c2} \) for the case of \( L = 24\lambda \) through two selected values of \( \kappa \), each characterizing one side of the transition. Inset (a) shows the typical diamagnetic behavior for \( \kappa = 0.4 < \kappa_{c2} \), the magnetization is always negative for any value of the applied field. Inset (b) presents the magnetization for the case with \( \kappa = 0.43 > \kappa_{c2} \) at which paramagnetic regions exist in the down branch and alternates with diamagnetic ones, thus not qualifying as a totally diamagnetic response.

Fig. 2 shows the quasi type-II transition, signaled in the ascending magnetization by two notable fields, namely, \( H''_c \) and \( H'_c \). For \( \kappa < \kappa_{c3} \), the maximum of the magnetization is immediately followed by its sudden drop to zero, \( H''_c \approx H'_c \). However for \( \kappa > \kappa_{c3} \) the two fields depart from each other and for increasing \( \kappa \), \( H''_c - H'_c \) also increases and vortices are observed in the superconductor. The kink in the curve \( H''_c \approx \kappa \) shown in Fig. 3 defines \( \kappa_{c3} \). The insets of Fig. 3 display situations below (left) and above (right) the transition. The left one shows the magnetization curve going from the Meissner state directly to the normal state, whereas the right one, in contrast, presents a vortex state in between the Meissner and normal states. The black ellipse in this inset highlights the vortex state region. Remarkably, this transition is found to occur at \( \kappa_{c3} = 0.54 \) for any size \( L \). Interestingly, it is found that, apart from small numerical deviations, \( H''_c \approx H'_c \) for \( \kappa > \kappa_{c3} \). The Gibbs free energy of the quasi type-II class, it is negative though very close to zero. This small negative value is still sufficient to render it slightly below the normal state energy, which is zero (see the supplementary material). This vortex regime is subsequent to the peak of the magnetization, which lies in an energetically metastable regime. This is in contrast with the standard type-II superconductor, where the peak of the magnetization is within a totally stable regime. We find that \( \kappa = 0.8 \) is still quasi type-II behavior but not specify the upper boundary which ought to be connected to the stability of the magnetization peak.

Fig. 3 displays the \( \kappa \) versus \( L \) phase diagram containing all the transitions discussed here. The GL theory ceases to be valid at \( L = \xi \), and for this reason the
diagram features $\kappa \geq 0.125$, which guarantees $L > \xi$ for all $L$ considered. The delimiting curves separating any two regions are obtained by a fitting process. The $\kappa_c(L)$ lines are indicated at the right margin of the figure and they separate the four regions, namely Genuine and Intermediate, the latter split into sub classes known as dia, para, and quasi type-II. Hence Fig. 4 is the generalization for the mesoscopic superconductors of the de-Gennes-Tinkham transition found at $\kappa_c$ for the macroscopic superconductor.

IV. CONCLUSIONS

In summary, we show that mesoscopic type-I superconductors have intrinsic transitions in $\kappa$. The genuine type-I behavior is only possible below $\kappa_{c1}$, and above it, vortices exist in this so-called intermediate type-I class that has a rich structure with a transition from paramagnetic to diamagnetic response, in descending field ($\kappa_{c2}$), and a quasi type-II behavior, in ascending field ($\kappa_{c3}$).

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