Intermediate type-I superconductors in the mesoscopic scale

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M. Tinkham [*Introduction to Superconductivity*, 2nd ed. (Dover, Mineola, NY, 2004), Chap. 4, pp. 135–138] and P. G. de Gennes [*Superconductivity of Metals and Alloys* (Benjamin, New York, 1966), Chap. 6, pp. 199–201] described 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 volume-to-area ratio 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 additional 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.

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I. INTRODUCTION

The classification of superconductors on the basis of their magnetic properties has been a hard earned knowledge. Nearly 25 years after the discovery of superconductivity by Onnes, the experimental measurements of Shubnikov showed that the magnetic properties of alloys were very different from those of pure metals [1–3]. The explanation had to wait another 20 years until the development of the theoretical work of Abrikosov [4] based on a new and at the time unknown phenomenological theory, namely 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 (λ) and the coherence length (ξ). Abrikosov found that the single coupling of the GL theory, $\kappa \equiv \lambda/\xi$, splits the superconductors into two classes, namely type I and type 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 it excludes any geometrical factor from entering the classification scheme. This critical coupling was also found by Bogomolny [6,7] in the context of string theory, thus rendering the transitions in κ 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 type II that disappears when the normal state sets in at the upper critical field H_{c2}. 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 time until 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], $YbSb_2$ [12], and more recently in the ternary intermetallics $YNiSi_3$ and $LuNiSi_3$ [13], rendering them of great interest and worthy of study. The coupling κ varies greatly among superconductors ranging from very high, for the high-T_c materials $YBaCuO_{7-\delta}$ ($\kappa = 95$), to very low values, for the pure metals [9] [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) and $YNiSi_3$ (0.1). Since ξ decreases with disorder, doping the material by impurities can produce adjustable κ compounds that allow for the study of transitions in this coupling.

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According to Tinkham [1] and 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 κ, as discussed below. There has been an intense search for this *genuine-intermediate* transition both theoretically [14,15] and experimentally [16–19] in the 1970s, 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 has 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, 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 nonzero. 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 ξ around the external boundary. Saint-James and de Gennes [21] found the critical field $H_{c3} > H_{c2}$ whose value in the case of a flat interface is $H_{c3} = 1.695 H_{c2}$ [21–27]. H_{c3} is also present in type-I superconductors [28] and signals several processes, e.g., the expulsion of magnetic flux [29,30]. In fact, it is H_{c3} that gives rise to the *genuine* and *intermediate* type-I superconductors associated with $H_{c2} < H_{c3} < H_c$ and $H_{c2} < H_c < H_{c3}$, respectively, such that κ_c is obtained from $H_{c3} = H_c$.

In this paper, we report properties of the *genuine-intermediate* transition such as its critical κ and also other transitions in κ in the *intermediate* phase, as seen by isothermal magnetization $M(H)$ curves. These several transitions can be experimentally investigated using the ballistic Hall magnetometry technique [31] applied to submicron-sized superconductors. Type-I mesoscopic superconductors have been investigated both theoretically [32] and experimentally [33], however the *genuine-intermediate* transition in κ is first considered here and found to acquire new properties. As one goes from the macroscopic to the mesoscopic scale, κ_c gives rise to κ_{c1} and also to the transitions κ_{c2} and κ_{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 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. [34,35]). We remark on 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 the Supplemental Material), and so they 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 nonzero and the superconducting state sets in. As shown here, these fields, as well as the $M(H)$ curves, are strongly dependent on κ 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, which has been a fortuitous coincidence thus far. The transition to the *quasi*-type-II class takes place for $\kappa_{c3} < 1/\sqrt{2}$.

Our numerical analysis was carried out on a very long needle with a 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 κ_{ci} may be affected by it. We look at several cross-section sizes, namely $L = \rho\lambda$ and $\rho = 8,\ 12,\ 16,\ 24,$ 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]. If the next-to-leading-order corrections are included [37], an intermediate phase emerges in the diagram κ versus T in between the type-I and -II domains. However, the analysis of Vagov *et al.* [36,37] 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 us to see intrinsic effects. Geometrical factors [39–42] 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 κ values where transitions take place but not their existence, which only reflects the ordering among the critical fields.

II. THEORETICAL FORMALISM

The basis of our dimensionless treatment of the GL theory is λ and $H_{c2} = \Phi_0/2\pi\xi^2$, which renders the free energy,

$$G = \int \left[\left| \left(-\frac{i}{\kappa}\nabla - \mathbf{A} \right)\psi \right|^2 - |\psi|^2 + \frac{1}{2}|\psi|^4 \right] d^3r + \int (\mathbf{h} - \mathbf{H})^2\, d^3r, \tag{1}$$

in reduced units. Lengths are in units of λ; the order parameter ψ is in units of $\psi_\infty = \sqrt{\alpha/\beta}$, where α and β 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

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

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

where $\mathbf{J_s} = \mathbb{R}[\bar{\psi}(-\frac{i}{\kappa}\nabla - \mathbf{A})\psi]$ is the superconducting current density.

The GL equations were solved numerically within a suitable relaxation method and upon using the link-variable method as presented in Ref. [43]. For this, we used a meshgrid 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, κ 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 the presence of an applied external field has been studied by Zharkov *et al.* [44–47]. 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 κ_{ci} transitions is beyond the scope of their framework since they are limited to a subset 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 with specific κ ranges, and to determine them we have performed a series of numerical simulations varying κ in steps of $\Delta\kappa = 0.01$. Within this precision, we were able to numerically obtain κ_{c1}, κ_{c2}, and κ_{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.

Figure 1 features the *genuine-intermediate* transition through the number of vortices, N, trapped at H'_{c3}. The superconductor response is markedly distinct according to κ, and this is exemplified here for $L = 16\lambda$ and 24λ. Above κ_{c1}, which is equal to 0.28 for $L = 16\lambda$, and 0.19 for $L = 24\lambda$, N varies according to κ, thus corresponding to the *intermediate* type-I class. However, below κ_{c1}, N drops to zero showing that no vortex enters the needle at H'_{c3}, which characterizes the *genuine* type-I class. The insets of Fig. 1 depict the magnetization curve of two selected κ values belonging to the two

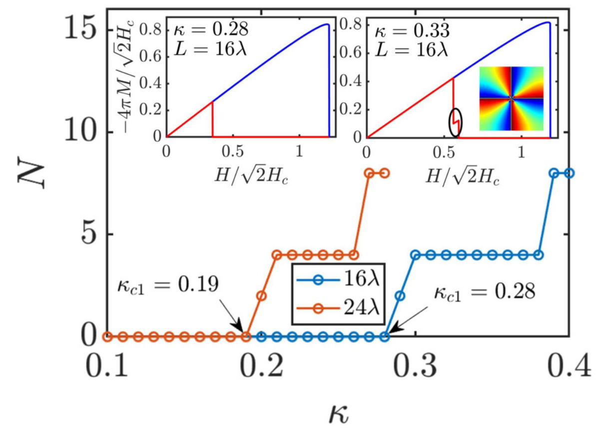

FIG. 1. The vorticity N at H'_{c3}, the field where the superconducting state sets in decreasing field, is plotted as a function of κ. The transition κ_{c1} corresponds to $N = 0$ marking the onset of the *genuine* type-I class and the end of the *intermediate* type-I. The insets show typical magnetization curves for the up (blue) and down (red) branches, above and below this transition. The black ellipse highlights the spike state, which is the last possible vortex state in the *intermediate* type-I class. In the spike state, a vortex nucleates and is immediately spelled from the superconductor.

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 the 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.

Figure 2 features the *dia-para* transition. The main panel presents κ_{c2} as a function of L. The small red circles indicate the numerically obtained κ_{c2} values of 0.4425, 0.4175, 0.4025, 0.4025, 0.4075, 0.415, and 0.415 for L/λ equal to 8, 9, 10, 12, 16, 24, and 32, respectively. Although κ_{c2} is a nonmonotonic

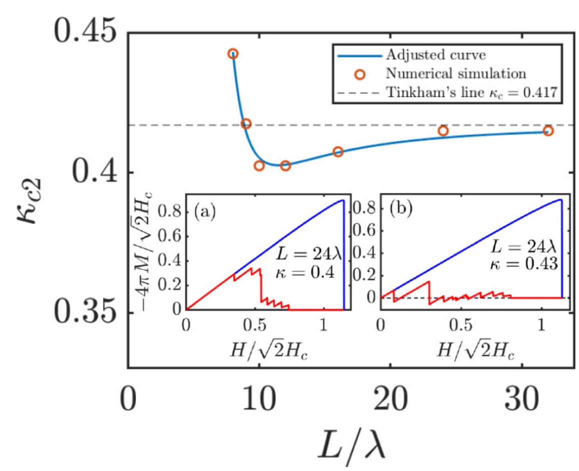

FIG. 2. The magnetization in descending field reveals the transition at κ_{c2}, shown in the main panel as a function of L, from paramagnetic to diamagnetic response. Insets (a) and (b) display typical magnetization curves below and above the transition. Inset (b) shows a still paramagnetic magnetization, while in panel (a) it has become totally diamagnetic.

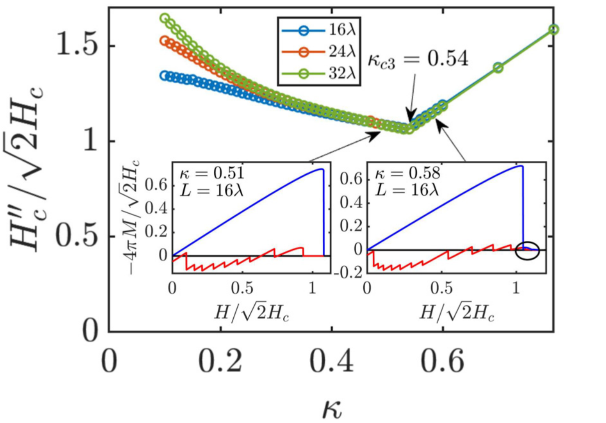

FIG. 3. The magnetization is a maximum at the field H_c' (the peak of $-M$) and vanishes at the field H_c'' where superconductivity is destroyed and the normal state is restored. H_c'' vs κ is shown here and clearly points to a κ_{c3} transition. The insets show typical magnetization curves below and above this transition. The left inset shows the situation that $H_c'' \approx H_c'$ and no vortices are present, whereas the right inset illustrates the situation $H_c'' > H_c'$ in which vortices do nucleate in the region highlighted by an ellipse.

function of L, it asymptotically approaches a limiting value for large L, suggestively close to κ_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 κ_{c2} for the case of $L = 24\lambda$ through two selected values of κ, each characterizing one side of the transition. Inset (a) shows the typical diamagnetic behavior for $\kappa = 0.4 < \kappa_{c2}$, and 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 it alternates with diamagnetic regions, thus not qualifying as a totally diamagnetic response.

Figure 3 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 κ, $H_c'' - H_c'$ also increases and vortices are observed in the superconductor. The kink in the curve H_c'' versus κ shown in Fig. 3 defines κ_{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, $H_c'' \approx H_{c3}'$ for $\kappa > \kappa_{c3}$. Concerning 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 Supplemental Material [48]). 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

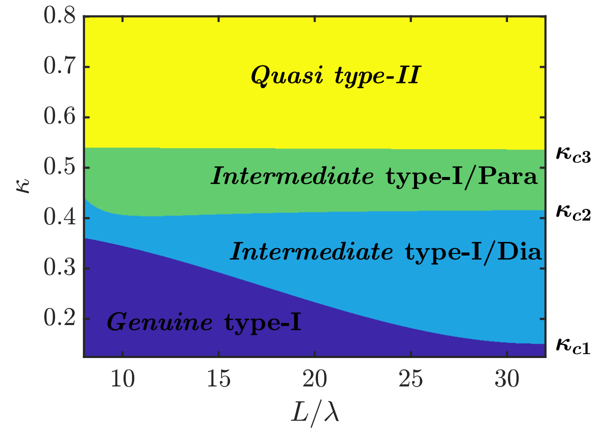

FIG. 4. κ-L phase diagram for the system under study in the ranges $0.125 < \kappa < 0.8$ and $8\lambda < L < 32\lambda$. As given in the figure, the dark blue, the light blue, the green, and the yellow regions correspond to *genuine* type-I, *intermediate* type-I with diamagnetism, *intermediate* type-I with the occurrence of paramagnetism in the down branch, and *quasi*-type-II, respectively.

regime. We find that $\kappa = 0.8$ is still *quasi*-type-II behavior, but the upper boundary, which ought to be connected to the stability of the magnetization peak, is not specified.

Figure 4 displays the κ 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 $\kappa \geqslant 0.125$, which guarantees $L > \xi$ for all L considered. The delimiting curves separating any two regions are obtained by a fitting process. The $\kappa_{ci}(L)$ lines are indicated at the right margin of the figure, and they separate the four regions, namely *genuine* and *intermediate*, the latter split into subclasses 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 κ_c for the macroscopic superconductor.

IV. CONCLUSIONS

In summary, we show that mesoscopic type-I superconductors have intrinsic transitions in κ. The *genuine* type-I behavior is only possible below κ_{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 (κ_{c2}), and a *quasi* type-II behavior, in ascending field (κ_{c3}).

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[48] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.103.014504 for additional magnetization curves, the energy curve for the vortex configuration in the quasi type-II phase and animations of the order parameter for different values of κ.