First-Order Transitions at H_{c1} and H_{c2} in Type II Superconductors*

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It is predicted that, for type-II superconductors with κ values near $1/\sqrt{2}$ at temperatures below T_c, the transition at H_{c1} is first order when $\xi_0/l \lesssim 50$ and the transition at H_{c2} is first order when $\xi_0/l \gtrsim 50$. It may not be possible to observe the latter transition because of experimental difficulties but the former transition should be observable. We also show that the vanishing of the normal-superconducting wall energy is irrelevant for determining if a given superconductor is type I or type II.

It is well known[1] that the Ginzburg-Landau-Abrikosov-Gor'kov (GLAG)[2-4] theory of type-II superconductors predicts that the transitions at H_{c1} and H_{c2} are of second order; the magnetization curve is continuous and there is no latent heat. The GLAG theory is, however, limited to $T=T_c$ and in this article we show that superconductors with $\kappa \approx 1/\sqrt{2}$ can have a first-order transition at H_{c1} or H_{c2}, depending on the ratio of the mean free path l to the coherence length ξ_0.

In the GLAG theory, the critical value of κ above which type-II behavior is obtained is $\kappa_c = 1/\sqrt{2}$; there are at least four distinct reasons for this and we now review them.

(a) The condition for the existence of bounded solutions of the linearized Ginzburg-Landau (GL) equation for the order parameter is that the applied field H_a be less than the upper critical field H_{c2}, where $H_{c2}=\sqrt{2}\,\kappa H_c$. Thus κ must be greater than $1/\sqrt{2}$ for type-II behavior.

(b) The expression for the Gibbs free-energy difference between the mixed and normal states near H_{c2} is[1,3]

$$G_m(H_a)-G_n(H_a)=\frac{-(H_a-H_{c2})^2}{8\pi(2\kappa^2-1)\beta_A}, \tag{1}$$

where β_A depends only on the lattice structure; κ must be greater than $1/\sqrt{2}$ for the mixed state to be stable.

(c) Calculations[1,5] of the lower critical field show that H_{c1} is less than H_c only if $\kappa < 1/\sqrt{2}$. For $\kappa = 1/\sqrt{2}$, $H_{c1}=H_c$ for all values of p, the number of flux quanta per isolated vortex.

(d) The normal-superconducting wall energy vanishes[2] for $\kappa=1/\sqrt{2}$ and is greater than or less than zero according to whether κ is less than or greater than $1/\sqrt{2}$.

The GLAG theory is, however, limited to $T=T_c$ and the question of the value of κ_c at lower temperatures is of interest.[6] We calculate κ_c using the simplest model of a type-II superconductor and restrict our attention to the temperature region just below T_c.

(a) The calculations of Tewordt[7,8] for H_{c2} give the result

$$H_{c2}=\sqrt{2}\,\kappa_1 H_c, \tag{2}$$

where

$$\kappa_1=\kappa[1+(1-t)f_1(\alpha)], \tag{3}$$

$t=T/T_c$, and $\alpha=0.882\xi_0/l$; Tewordt[7] has given an analytic expression for $f_1(\alpha)$ and has calculated $f_1(\alpha)$ for some values of α.

(b) Near H_{c2}, the Gibbs free-energy difference between the mixed and normal states is[8]

$$G_m-G_n=\frac{-(H_a-H_{c2})^2}{8\pi(2\kappa_2^2-1)\beta_A}, \tag{4}$$

where

$$\kappa_2=\kappa[1+(1-t)f_2(\kappa,\alpha)]. \tag{5}$$

The function $f_2(\kappa,\alpha)$ is given by Neumann and Tewordt.[8]

(c) H_{c1}/H_c for $p=1$ has been calculated by Neumann and Tewordt[9] in the form

$$\frac{H_{c1}}{H_c}=\left.\frac{H_{c1}}{H_c}\right|_{\mathrm{GL}}[1+(1-t)\delta_1(\kappa,\alpha)], \tag{6}$$

where $\delta_1(\kappa,\alpha)$ has been calculated for a mesh of (κ,α) values. H_{c1}/H_c for $p=2$ has been calculated by the author,[10] and the results have been expressed in the form of Eq. (6) with $\delta_1(\kappa,\alpha)$ replaced by $\delta_2(\kappa,\alpha)$.

(d) The normal-superconducting wall energy (σ_{NS}) has been calculated by the author[10]; the result is

$$4\pi\sigma_{\mathrm{NS}}/H_c^2\lambda=g_0(\kappa)+(1-t)g_1(\kappa,\alpha), \tag{7}$$

where g_0 and g_1 have been tabulated.

The above calculations can be used to determine the first order corrections (in $1-T/T_c$) to the Ginzburg-Landau result $\kappa_c=1/\sqrt{2}$. From the five criteria $\kappa_1=1/\sqrt{2}$, $\kappa_2=1/\sqrt{2}$, $H_{c1}=H_c$ for

Table I. $d\kappa_c/dt$ at $T=T_c$ as a function of α for five different definitions of κ_c.

α	$\kappa_1=1/\sqrt{2}$	$\kappa_2=1/\sqrt{2}$	$H_{c1}=H_c$ $p=1$	$H_{c1}=H_c$ $p=2$	$\sigma_{NS}=0$
0	0.2881	0.7694	-0.0654	-0.0424	0.0191
0.2	0.2506	0.6456	-0.0393	-0.0205	0.0300
0.5	0.2131	0.5226	-0.0140	+0.0007	0.0403
1.0	0.1758	0.4012	+0.0104	+0.0212	0.0500
2.0	0.1394	0.2827	0.0341	0.0410	0.0593
4	0.1118	0.1920	0.0530	0.0568	0.0670
10	0.0922	0.1228	0.0697	0.0712	0.0751
20	0.0861	0.0976	0.0776	0.0782	0.0797
50	0.0834	0.0826	0.0840	0.0840	0.0839
100	0.0831	0.0779	0.0869	0.0866	0.0860
∞	0.0838	0.0740	0.0910	0.0906	0.0893

$p=1$, $H_{c1}=H_c$ for $p=2$, and $\sigma_{NS}=0$, we obtain κ_{c1}, κ_{c2}, κ_{c3}, κ_{c4}, and κ_{c5}, respectively, as functions of α. We define

$$\kappa_{ci}=1/\sqrt{2}-(1-t)[d\kappa_{ci}/dt]_{t=1}, \quad (8)$$

and give in Table I values of $d\kappa_{ci}/dt$ at $t=1$ as functions of α. By Eq. (8), however, the table also gives the values of $1/\sqrt{2}-\kappa_{ci}$ extrapolated to $T=0°K$, and I have chosen to discuss the results in terms of the latter interpretation since the effects are largest at low temperatures. The extrapolation of κ_2 to $T=0$ is poor for clean superconductors since κ_2 diverges logarithmically as $T\to 0$,[11] but this does not qualitatively affect our results; we use only the fact that $\kappa_2>\kappa_1$ for clean to moderately clean superconductors. Because of the extrapolation, however, the following results are only qualitatively correct. Table I has a number of interesting features which we now discuss.

(i) The five different criteria give different results for κ_c.

(ii) For $\alpha\lesssim 50$, κ_{c3} is greater than κ_{c4}; this means that flux penetration in the form of a doubly quantized isolated vortex is energetically preferable to flux penetration in the form of a singly quantized isolated vortex for these values of α.

(iii) For $\alpha\lesssim 40$ we have the inequalities $\kappa_{c2}<\kappa_{c1}<\kappa_{c4}$. We discuss the case $\alpha=0$ ($\kappa_{c1}=0.4190$ and $\kappa_{c4}=0.7495$) in detail, but similar results are obtained for all $\alpha\lesssim 40$. For $\kappa<0.4190$ there are no bounded solutions of the linearized order parameter equation, and we can conclude that the material is type I. For $0.4190<\kappa<0.7495$, the bounded solutions exist and are stable with respect to the normal solutions; hence the mixed state exists in such a substance for a range of applied field values between H_c and H_{c2}. For this range of κ values, however, $H_{c1}>H_c$ for both $p=1$ and $p=2$. Since the area under the magnetization curve gives the free-energy difference between the superconducting and normal states, the initial flux penetration must occur at a value of H_a which is less than H_c if the flux penetration is incomplete at fields greater than H_c; we can conclude that the initial flux penetration is not in the form of an isolated vortex with either $p=1$ or $p=2$. There are many possibilities for the form of the initial flux penetration but the most reasonable form is that of a lattice of singly quantized vortices with finite spacing; the magnetization would then be discontinuous at $H_a=H_{c1}$ and the transition would be of first order. It should be possible to extend the GLAG theory of the interacting vortices[3,12,13] to temperatures less than T_c to see if this conjecture is correct; work in this direction is in progress. The contradiction between the definitions of κ_{c1} from $\kappa_1=1/\sqrt{2}$ and H_{c1}/H_c was first noticed by Tewordt.[14] It is important to note that the range of κ values given above is only the range for which a contradiction occurs; the (conjectured) stability of the vortex lattice with finite spacing with respect to the state with an isolated vortex may extend to values of κ significantly larger than 0.7495.

(iv) For $\alpha \gtrsim 50$, one has the inequalities $\kappa_{c3} < \kappa_{c4} < \kappa_{c1} < \kappa_{c2}$; the first of these implies that the initial flux penetration is in the form of a singly quantized rather than a doubly quantized isolated vortex. The case $\alpha \to \infty$ is typical, and we discuss it in detail. We then have $\kappa_{c3} = 0.6161$, $\kappa_{c1} = 0.6233$, and $\kappa_{c2} = 0.6331$. For $0.6161 < \kappa < 0.6233$, H_{c1} is less than H_c for $p = 1$, but bounded solutions of the linearized order parameter equation do not exist (and would not be stable if they did exist) for $H_a > H_c$. We can use the converse of the argument in (iii) above: The flux penetration must be incomplete for a range of fields greater than H_c if the flux exclusion is incomplete for a range of fields less than H_c. I suggest that there is a first-order transition at H_{c2}. For $0.6233 < \kappa < 0.6331$, H_{c1} is less than H_c and the bounded solutions exist but they are unstable. If one considers the shape of the magnetization curve as κ is decreased through $\kappa = 0.6331$ by varying the κ of the pure superconductor, one is lead by a continuity argument to the conclusion that the transition at H_{c2} is of first order.[15] Since it is difficult to make high-quality samples with short mean free paths, it is unlikely that the first-order transition at H_{c2} can be observed. Although the range of κ values given above is only the range for which a contradiction is obtained, the first-order transition probably exists only over a small range; hence the magnetization curve would be very steep for superconductors of interest and even if good samples could be made, it would be difficult to recognize a first-order transition.

(v) If we define κ_{c12} as the greater of κ_{c1} and κ_{c2} and κ_{c34} as the greater of κ_{c3} and κ_{c4}, then the table shows that κ_{c5} is larger than the greater of κ_{c12} and κ_{c34}. We can conclude that the condition $\sigma_{NS} = 0$ is irrelevant for determining the critical value of κ.

In the above discussion we have used the isotropic, weak-coupling model (which also neglects spin paramagnetism, spin-orbit coupling, the existence of two bands in transition metals, and anisotropic defect scattering); it is possible that the inclusion of these "real-metal" effects in the model would result in qualitative as well as quantitative changes in our predictions. A detailed consideration of these effects is quite difficult, and I suggest that an experimental investigation of the possibilities of first-order transitions in type-II superconductors, particularly at H_{c1}, would be more fruitful. A more detailed account of this work will be published elsewhere.

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[1]Recent reviews of the subject of type-II superconductivity from the theoretical and experimental standpoints, respectively, have been given by A. L. Fetter and P. C. Hohenberg, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker, New York, 1969), Chap. 14; B. Serin, *ibid.* Chap. 15.

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[15]First order transitions at H_{c2} have been found by G. Sarma [J. Phys. Chem. Solids 24, 1029 (1963)] for a uniform exchange field acting on the conduction-electron spins and by K. Maki [Physics (Long Is. City, N.Y.) 1, 127 (1964) and Phys. Rev. 148, 362 (1966)] for a superconductor with large Pauli-spin paramagnetism and moderate spin-orbit coupling. The discussion of Maki's work by Fetter and Hohenberg (Ref. 1) is more closely related to our argument for a first-order transition at H_{c2}.