Disorder-dependent slopes of the upper critical field in nodal and nodeless superconductors

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We study the slopes of the upper critical field $S = \partial H_{c2}/\partial T$ at the superconducting transition temperature T_c in anisotropic superconductors with transport (nonmagnetic) scattering employing the Ginzburg-Landau theory, developed for this case by Pokrovsky and Pokrovsky [Phys. Rev. B **54**, 13275 (1996)]. We find unexpected behavior of the slopes for a d-wave superconductor and, in a more general case, of materials with line nodes in the order parameter. Specifically, the presence of line nodes causes S to *decrease* with increasing nonmagnetic scattering parameter $P = \hbar/2\pi T_{c0}\tau$ (T_{c0} is for the clean limit, τ is the scattering time), unlike the nodeless case where the slope *increases*. In a pure d-wave case, the slope changes from decreasing to increasing when the scattering parameter approaches $P \approx 0.91\, P_{\rm crit}$, where $P_{\rm crit} \approx 0.28$, at which $T_c \to 0$, which implies the existence of a "gapless" state in d-wave superconductors with transport scattering in the interval, $0.91\, P_{\rm crit} < P < P_{\rm crit}$. Furthermore, we consider the mixed $(s + d)$-wave order parameter with four nodes on a cylindrical Fermi surface when the d part is dominant, or no nodes at all when the s-wave phase dominates. We find that the presence of nodes causes the slope $S(P)$ to decrease initially with increasing P, whereas in the nodeless state, $S(P)$ monotonically increases. Therefore relatively straightforward measurements of the disorder dependence of the slope of H_{c2} at T_c can help distinguish between nodal and nodeless order parameters, which is particularly useful for quickly assessing newly discovered superconductors.

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I. INTRODUCTION

A brief literature survey finds many reports on the upper critical field slope evaluated at the superconducting transition temperature T_c as a function of nonmagnetic disorder. A consistent picture emerges—superconductors with line nodes exhibit decreasing slope, whereas those without nodes show increasing $\partial H_{c2}/\partial T$ with increasing disorder scattering. For example, the increase was experimentally observed in fully gapped $s_\pm$ iron-based superconductors where nonmagnetic disorder was introduced by ball-milling [1], irradiation with 2.5-MeV electrons [2], or fast neutrons [3]. On the other hand, a decrease of the slope concomitant with the decrease of T_c was found in a nodal pnictide superconductor [4].

The problem of slopes $S = \partial H_{c2}/\partial T$ at T_c can be addressed with the help of Ginzburg-Landau (GL) theory. To provide reasonable theoretical guidance, the theory should apply to various order parameter symmetries and anisotropic Fermi surfaces. There were a few attempts to develop such a version of GL, some confirming major features of the experimental information on the slope's dependence on disorder for superconductors with nodes and without [5,6]. In this work we employ a most general version of the GL theory applicable to anisotropic order parameters in the presence of transport scattering ascribed to Pokrovsky and Pokrovsky [7].

Below, we first outline the theory and then we apply it to the d-wave symmetry of the order parameter and show that, in fact, the slopes S are indeed suppressed by a relatively weak disorder but increase when the impurity scattering approaches the critical value at which $T_c = 0$. Next we consider the order parameter, which is a superposition of s and d that allows us to study slopes S in cases of nodes present or not and shows that the nodes change $S(P)$ from the node-free increase to decrease. We end up with a short discussion of existing and possible experiments.

To simplify the formalism, the effective coupling is commonly assumed factorizable [8], $V(\boldsymbol{k}, \boldsymbol{k}') = V_0\, \Omega(\boldsymbol{k})\, \Omega(\boldsymbol{k}')$, where $\boldsymbol{k}$ is the Fermi momentum. One then looks for the order parameter in the form

$$\Delta(\boldsymbol{r}, T; \boldsymbol{k}) = \Psi(\boldsymbol{r}, T)\, \Omega(\boldsymbol{k}). \tag{1}$$

The factor $\Omega(\boldsymbol{k})$ for the order parameter change along the Fermi surface is conveniently normalized:

$$\langle \Omega^2 \rangle = 1, \tag{2}$$

where $\langle ... \rangle$ stands for the Fermi surface average. This normalization corresponds to the critical temperature T_{c0} of a clean material given by the standard isotropic weak-coupling model with the effective interaction V_0.

The slope of the upper critical field $H_{c2} = \phi_0/2\pi\xi^2$ at T_c (ϕ_0 is the flux quantum) is determined by the T dependence of the coherence length $\xi = \xi_{GL}/\sqrt{1 - T/T_c}$:

$$\frac{\partial H_{c2}}{\partial T}\bigg|_{T_c} = -\frac{\phi_0}{2\pi \xi_{GL}^2 T_c}. \tag{3}$$

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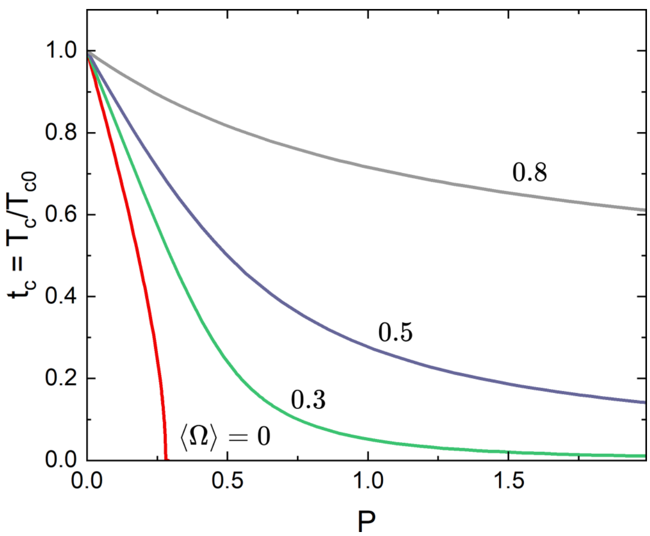

FIG. 1. $t_c = T_c/T_{c0}$ vs scattering parameter P for $\langle\Omega\rangle = 0.8, 0.5, 0.3, 0$ in top-down order. Note that for the d wave $\langle\Omega\rangle = 0$ and T_c turns zero at the critical scattering $P_{\rm crit} = 1/2e^{\gamma} = 0.28$.

The length ξ_{GL} is of the order of the BCS coherence length $\xi_0 = \hbar v_F/\pi\Delta(0)$ but differs from ξ_0—actually, it depends on the coupling, on the impurity scattering, on the order parameter symmetry, and the Fermi surface anisotropy. All these dependencies can, in principle, be found within the microscopic BCS theory. This has been done at early stages of the theory for the anisotropic order parameter only in the *clean* limit by Gor'kov and Melik-Barkhudarov [9],

$$\left(\xi_{GL}^2\right)_{ik} = \frac{7\zeta(3)\hbar^2}{16\pi^2 T_c^2}\langle\Omega^2 v_i v_k\rangle. \tag{4}$$

The transport scattering was included by Helfand and Werthamer [10], but only for the *isotropic* order parameter and Fermi surface:

$$\xi_{GL}^2 = \frac{\hbar^2 v^2}{24\pi^3 T_c^2 P^2}\left[\frac{\pi^2 P}{4} - \psi\left(\frac{1+P}{2}\right) + \psi\left(\frac{1}{2}\right)\right]. \tag{5}$$

Here ψ is the di-gamma function, and the scattering parameter

$$P = \hbar/2\pi T_{c0}\tau, \tag{6}$$

with τ being the scattering time and T_{c0} the critical temperature for the clean sample. It is easy to see that the slope $S = -\partial_T H_{c2}|_{T_c}$ grows, being roughly proportional to P. It is worth paying attention that the material parameter P does not depend on actual T_c, unlike the scattering parameter $\rho = \hbar/2\pi T_c\tau$ often employed in literature.

The critical temperature of materials with the anisotropic order parameter is suppressed even by nonmagnetic impurities. This was established by Openov [11], see also [12]:

$$-\ln\frac{T_c}{T_{c0}} = (1 - \langle\Omega\rangle^2)\left[\psi\left(\frac{P/t_c + 1}{2}\right) - \psi\left(\frac{1}{2}\right)\right], \tag{7}$$

where $t_c = T_c/T_{c0}$. Examples of $t_c(P)$ for a few values of Ω are given in Fig. 1.

Clearly, $T_c = T_{c0}$ for the isotropic order parameter with $\Omega = 1$ and arbitrary scattering rate P, as well as for the clean limit and any anisotropic Ω. For the d wave $\langle\Omega\rangle = 0$, and we have the standard Abrikosov-Gor'kov result [13], so that the transport scattering in the d wave affects T_c as the pair-breaking scattering in isotropic materials does.

II. LINEARIZED GINZBURG-LANDAU EQUATIONS

Thus according to Eq. (3), the slope of H_{c2} at T_c is determined by the coherence length which enters the linearized GL equation for the order parameter Ψ,

$$-(\xi^2)_{ik}\Pi_i\Pi_k\Psi = \Psi, \tag{8}$$

with the tensor of squared coherence length $(\xi^2)_{ik}$; $\boldsymbol{\Pi} = \nabla + 2\pi i\boldsymbol{A}/\phi_0$, where $\boldsymbol{A}$ is the vector potential and ϕ_0 is the flux quantum. For arbitrary Ω and any Fermi surface in the presence of transport scattering, Pokrovsky and Pokrovsky [7] evaluated the tensor

$$(\xi^2)_{ik} = \zeta_{ik}/a. \tag{9}$$

Here (in our notation)

$$a = \frac{T_c - T}{T_c}\left[1 + (1 - \langle\Omega\rangle^2)\psi'\left(\frac{P/t_c + 1}{2}\right)\right]. \tag{10}$$

The tensor $\hat{\zeta}$ is given by

$$\zeta_{ik} = \frac{\hbar^2}{16\pi^2 T_c^2}\left[h_{3,0}\langle\Omega^2 v_i v_k\rangle + \frac{P}{t_c}h_{3,1}\langle\Omega v_i v_k\rangle\langle\Omega\rangle + \frac{P^2}{4t_c^2}h_{3,2}\langle v_i v_k\rangle\langle\Omega\rangle^2\right]. \tag{11}$$

The quantities $h_{\mu,\nu}$ are functions of $x = P/2t_c$, defined as

$$h_{\mu,\nu}(x) = \sum_{n=0}^{\infty}(n + 1/2 + x)^{-\mu}(n + 1/2)^{-\nu}, \tag{12}$$

so that

$$h_{3,0} = -\frac{1}{2}\psi''\left(\frac{1}{2} + x\right),$$
$$h_{3,1} = \frac{1}{x^3}\left[\psi\left(\frac{1}{2} + x\right) - \psi\left(\frac{1}{2}\right) - x\psi'\left(\frac{1}{2} + x\right) + \frac{x^2}{2}\psi''\left(\frac{1}{2} + x\right)\right],$$
$$h_{3,2} = \frac{1}{2x^4}\left\{\pi^2 x - 6\left[\psi\left(\frac{1}{2} + x\right) - \psi\left(\frac{1}{2}\right)\right] + 4x\psi'\left(\frac{1}{2} + x\right) - x^2\psi''\left(\frac{1}{2} + x\right)\right\}. \tag{13}$$

It is straightforward to see that for the isotropic order parameter with $\Omega = 1$ on the Fermi sphere these formulas reduce to the BCS form,

$$\xi^2 = \frac{7\zeta(3)}{48\pi^2 T_c^2\delta t}\chi(P), \qquad \delta t = 1 - \frac{T}{T_c}, \tag{14}$$

with the Gor'kov function

$$\chi(P) = \frac{1}{7\zeta(3)}\sum_{n=0}^{\infty}\frac{1}{(n + 1/2 + P)(n + 1/2)^2}. \tag{15}$$

This limit can also be checked by comparison with H_{c2} slopes at T_c given by Helfand and Werthamer [10].

It is worth noting that for the clean limit $x=P/2t_c \to 0$, whereas $h_{3,0} \to 7\zeta(3)$, $h_{3,1} \to \pi^4/6$, and $h_{3,2} \to 31\zeta(5)$. Hence in this limit only the first term on the right-hand side of Eq. (11) survives, in agreement with [8]. In the opposite limit $x = P/2t_c \gg 1$, the last term in Eq. (11) dominates [7], as is seen in Fig. 2 (however, this limit does not apply for the d wave since both the second and third terms in Eq. (11) are zeroes due to $\langle\Omega\rangle = 0$).

A. General order parameter

In the general case we obtain for the slope of H_{c2} along the c axis of a uniaxial material

$$\frac{\partial H_{c2}}{\partial T}\Big|_{T_c} = -\frac{8\pi\phi_0 T_{c0}}{\hbar^2}\frac{t_c\left[1+(1-\langle\Omega\rangle^2)\psi'\left(\frac{1+P/t_c}{2}\right)\right]}{h_{3,0}\langle\Omega^2 v_a^2\rangle + 2(P/2t_c)h_{3,1}\langle\Omega\rangle\langle\Omega v_a^2\rangle + (P/2t_c)^2 h_{3,2}\langle\Omega\rangle^2\langle v_a^2\rangle}. \tag{16}$$

Here all coefficients $h_{\mu,\nu}(x)$ are taken at $x = P/2t_c$.

Since the Fermi velocity is not a constant at anisotropic Fermi surfaces, we normalize velocities on some value v_0 for which we choose [14]

$$v_0^3 = 2E_F^2/\pi^2\hbar^3 N(0), \tag{17}$$

where E_F is the Fermi energy and $N(0)$ is the total density of states at the Fermi level per spin. One easily verifies that $v_0 = v_F$ for the isotropic case.

The slope expression (16) remains the same except for a changed prefactor:

$$-\frac{8\pi\phi_0 T_{c0}}{\hbar^2} \to -\frac{8\pi\phi_0 T_{c0}}{\hbar^2 v_0^2}, \tag{18}$$

and the velocity v_a is now dimensionless (although we leave for it the same notation).

B. d wave

The case of the d-wave symmetry of the order parameter with $\langle\Omega\rangle = 0$ is relatively simple. We have the coherence length relevant for H_{c2} along the c axis of a uniaxial crystal:

$$\xi_{aa}^2 = \frac{\zeta_{aa}}{a} = \frac{\hbar^2\langle\Omega^2 v_a^2\rangle h_{3,0}}{16\pi^2 T_c^2[1+\psi'(P/2t_c+1/2)]\delta t}. \tag{19}$$

For a Fermi cylinder, with $\Omega = \sqrt{2}\cos 2\varphi$, the average $\langle\Omega^2 v_a^2\rangle = v^2/2$. Hence,

$$\xi_{aa}^2 = -\frac{\hbar^2 v^2\, \psi''(P/2t_c+1/2)}{64\pi^2 T_c^2[1+\psi'(P/2t_c+1/2)]\delta t}, \tag{20}$$

and we obtain

$$\frac{dH_{c2,c}}{dT}\Big|_{T_c} = \frac{32\pi\phi_0 T_{c0}}{\hbar^2 v^2}\, t_c \frac{1+\psi'(P/2t_c+1/2)}{\psi''(P/2t_c+1/2)}. \tag{21}$$

For numerical work it is convenient to use the reduced slope,

$$s = -\frac{dh_{c2}}{dt}\Big|_{t_c} = -\frac{\hbar^2 v^2}{8\pi\phi_0 T_c}\frac{dH_{c2}}{dT}\Big|_{T_c}. \tag{22}$$

While the actual slope $\partial H_{c2}/\partial T|_{T_c}$ is negative, we are interested in its magnitude and use a positive quantity, as given by Eq. (22).

The behavior of the slope as a function of P according to this result is shown in Fig. 3. As is known, the maximum scattering parameter P for which the d-wave superconductivity survives is $P_{\rm crit} = 1/2e^{\gamma} = 0.28$ (double the critical value for the spin-flip magnetic scattering [13]). Hence, similar to materials with magnetic scatterers [15], the slope $\partial H_{c2}/\partial T$ at T_c for the d wave decreases with increasing transport scattering. It is worth noting that this behavior changes to increase near $P \approx 0.91\, P_{\rm crit} = 0.25$; this estimate coincides with that given by Abrikosov and Gor'kov for the low bound of the gapless state. This suggests that d-wave materials can also be gapless if the transport scattering parameter lays in the interval $0.25 < P < 0.28$. Thus $S(P)$ dependence might be a macroscopic manifestation of the gapless superconductivity in impure d-wave materials, a speculation worthy of further study.

Figure 4 shows the slopes of Fig. 3 plotted vs the critical temperature $t_c(P)$.

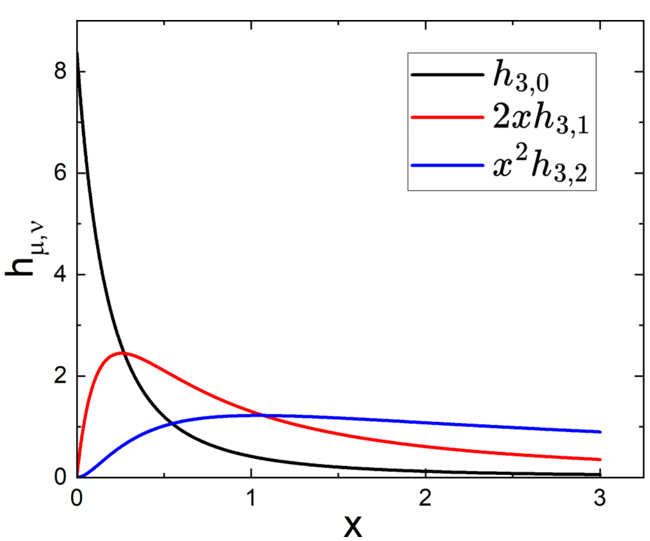

FIG. 2. The coefficients $h_{3,0}(x)$, $2xh_{3,1}(x)$, and $x^2h_{3,2}(x)$ vs x.

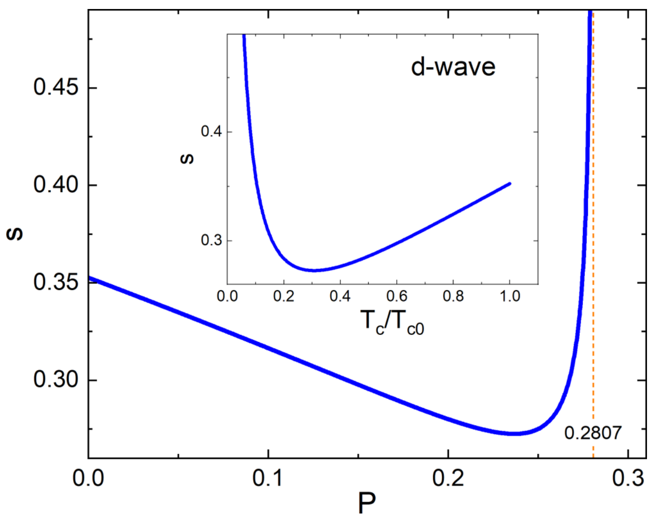

FIG. 3. The slope $s = -\partial h_{c2}/\partial t$ at t_c according to Eq. (22) vs P for a d-wave material. Inset: The same slopes vs $t_c = T_c/T_{c0}$.

C. $(s + d)$ wave

Obviously, the major interest in the community is to determine whether the easily accessible measurements of the H_{c2} slope near T_c may provide some insight into the structure of the order parameter. Here we examine the simplest case of a $(s + d)$-wave state where the order parameter is the isotropic s wave in one limit and a standard 2D d wave in the other. Keeping in mind the normalization $\langle\Omega^2\rangle = 1$, a convenient order parameter can be written as

$$\Omega = \sqrt{1 - r^2/2} + r\cos(2\varphi). \tag{23}$$

If $r = 0$, $\Omega = 1$ and when $r = \sqrt{2}$, $\Omega = \sqrt{2}\cos(2\varphi)$, which are the required limits. We choose this order parameter not because it may describe any particular real material, rather, we intend to check whether or not there is a connection between the *microscopic* anisotropy of the order parameter and the *macroscopic* dependence of H_{c2} slopes on the degree of disorder P.

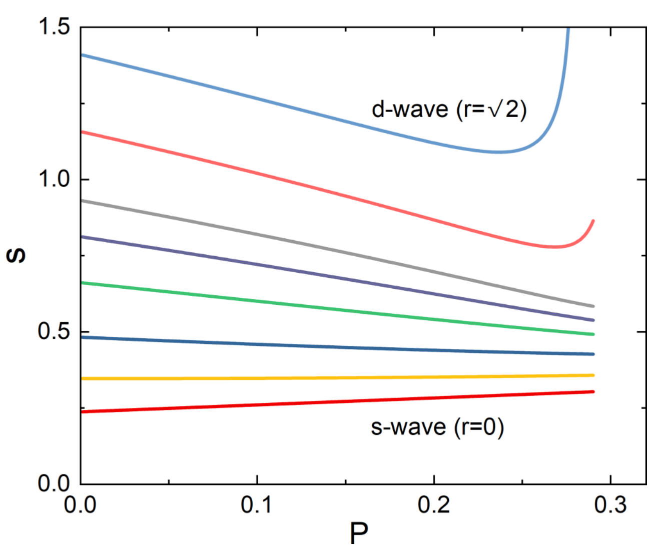

FIG. 4. $s(P)$ according to Eqs. (22) and (16) for a set of parameters $r = 0, 0.75, 1, 1.2, 1.3, 1.35, 1.4$ and $\sqrt{2}$. The approach to a gapless regime is characterized by an upturn starting with $P \approx 0.25$ for a d-wave order parameter with $r = \sqrt{2}$. The calculations were performed for a cylindrical Fermi surface.

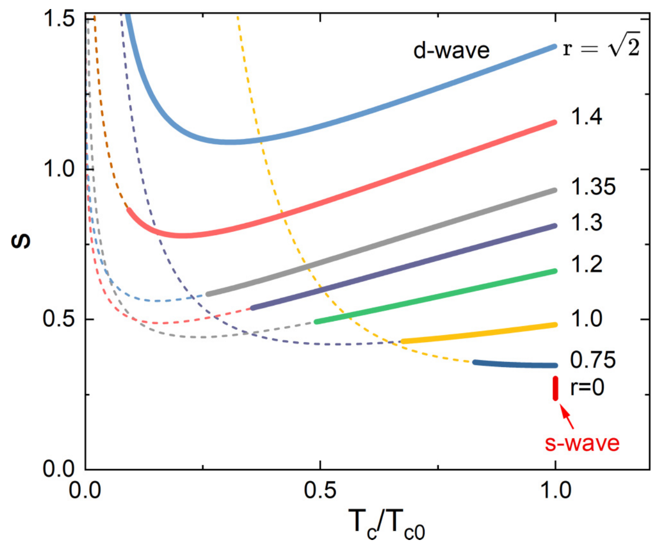

FIG. 5. Solid lines are the slopes s for the same values of the scattering parameters P as in Fig. 4 but plotted vs the transition temperature $t_c(P)$. Dashed continuations are for $P > 0.28$, for which the pure d wave phase does not exist. In the pure s wave for which $t_c = 1$, $S(P)$ does not depend on P and is shown by a vertical red line.

Figure 4 shows $s(P)$ calculated with the help of Eqs. (16) and (22), plotted for a few coefficients r of the Ω function, Eq. (23). At $r = 0$ the order parameter is the isotropic s wave, whereas at $r = \sqrt{2}$ it is a two-dimensional d-wave order parameter. The calculations were performed for a cylindrical Fermi surface, keeping in mind possible applications to high-T_c cuprates. Figure 4 shows that the slopes $s(P)$ for purely d wave are (a) nonmonotonic and decrease with increasing P up to about $P = 0.25$, followed by divergence for $P \to 0.28$, as we have seen in Fig. 3. With increasing fraction of s wave, however, the negative slope of $S(P)$ for small and intermediate P weakens and turns to a positive, nearly linear increase of S. To gain further insight, we plot $S(P)$ versus t_c in Fig. 5.

To analyze the overall trend of whether the slope s is increasing or decreasing with nonmagnetic scattering P, we utilize a numerical derivative χ, given by $\chi = [S(P + \Delta P) - S(P)]/\Delta P$. A negative value of χ indicates a decreasing slope, whereas a positive value indicates the opposite. Based on the results in Fig. 4, it can be observed that $s(P)$ is nearly linear, at least up to $P = 0.15$, within the entire range of $0 \leqslant \Omega \leqslant 1$. As a result the specific selection of P_0 and ΔP is not critical. To reflect a realistic degree of disorder, we opted for $P_0 = 0.1$ and $\Delta P = 0.01$, though other values were tested with no significant variation in the outcomes.

Figure 6 shows the numerical derivative χ plotted versus the coefficient r of the order parameter, Eq. (23). A straightforward algebra shows that the order parameter is nodeless (anisotropic s wave) for $r < \sqrt{2/3} \approx 0.82$ and has four line nodes for $r > \sqrt{2/3}$. Remarkably, Fig. 6 shows that the rate of

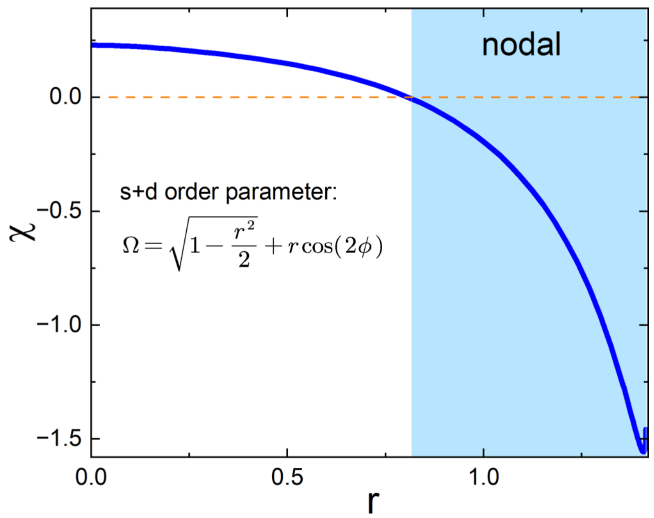

FIG. 6. The rate of change of the slope $S(P)$ defined as $\chi = S(0.11) - S(0.01)/0.1$, evaluated for different values of the r coefficient.

slope change χ becomes negative as soon as the nodes appear. In other words, if one would measure $S = -\partial H_{c2}/\partial T|_{T_c}$ as a function of nonmagnetic disorder, the increasing S would indicate nodeless superconductivity, whereas the slope decrease can be considered as evidence for the presence of nodes.

III. DISCUSSION

We have applied the GL theory developed by Pokrovsky and Pokrovsky [7] for anisotropic materials in the presence of nonmagnetic scattering to evaluate slopes $\partial H_{c2}/\partial T|_{T_c}$ for d-wave superconductors and for the case of mixed $(s+d)$-wave symmetry. For the d wave, we find that for weak and intermediate scattering rates $P = \hbar/2\pi T_{c0}\tau$, the slopes $S = -\partial H_{c2}/\partial T|_{T_c}$ are suppressed, similar to the situation of the magnetic pair-breaking disorder and opposite to the transport scattering enhancement of slopes $S(P)$ for the s wave. One example of such a behavior is in studies of slopes in YBaCuO by Antonov *et al.* [16].

Thinking along these lines, we have examined the mixed $(s+d)$-wave order parameter $\Omega(r) = \sqrt{1-r^2/2} + r\cos(2\varphi)$ such that $\Omega(0)$ corresponds to the pure isotropic s wave and $\Omega(\sqrt{2})$ is the pure d wave. We found a remarkable one-to-one correspondence between the presence or absence of nodes and the decrease or increase of the H_{c2} slopes at T_c. Practically, this is a highly useful observation, notwithstanding our oversimplified model.

Unexpectedly, however, if the scattering rate approaches the critical value $P_{\rm crit} = 0.28$ for which $T_c \to 0$, the slopes increase starting with $P \approx 0.25$ to diverge at $P_{\rm crit}$. The value $0.25 \approx 0.9$ of $P_{\rm crit}$, the fraction found by Abrikosov and Gor'kov for the low bound of the gapless domain in standard s-wave materials with magnetic impurities [13]. This analogy suggests strongly that in a d-wave superconductor with nonmagnetic disorder close to the maximum disorder possible we are dealing with a kind of "gapless" state. In our view this speculation deserves careful examination.

The slopes of the upper critical field at T_c are quite straightforward to measure. It has been done even for nearly-room-temperature hydride superconductors under extremely high pressures within diamond pressure cells *in situ*. If, indeed, such macroscopic measurement may hint at the microscopic symmetry of the order parameter, this is worth doing. One has to examine the rate of change of the slopes in samples of the same chemical composition but with different amounts of nonmagnetic disorder. Such disorder can be induced by ion implantation [16,17], and electron [2], proton [18], neutron [3,19], or gamma [20] and even alpha particle irradiation [21].

Since the upper critical field was measured for most superconductors, it is now possible and very important to check its evolution with disorder. However, one must be sure to analyze the true H_{c2} and not the irreversibility line, while also avoiding any major disturbance to the electronic band structure of the material due to introduced disorder. For instance, isovalent doping is preferred over charge doping, and, as mentioned above, proton or electron irradiations are even better. It is worth noting that there are only a few successful phase-sensitive experiments capable of differentiating between the same sign and sign-changing order parameter. These experiments require significant effort and special sample preparation, making them technically too challenging or even impossible for most superconductors, for example, due to cleaving issues. Our theory provides a relatively simple and straightforward qualitative prediction.

A word of caution. It is quite possible that complex multiband materials will not follow our simple scheme based on the generally accepted factorization of temperature and angular variations of the order parameter. It is also possible that other types of order parameters in materials other than uniaxial and in fields other than parallel to the c axis will not follow it either. However, our analysis is based on the universal Ginzburg-Landau theory, and hopefully, our conclusions are robust.

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