

## Theory of Upper Critical Fields in Highly Disordered Superconductors: Localization Effects

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(Received 21 October 1983)

The authors compute the temperature  $T$  dependence of the upper critical field  $H_{c2}$  arising from incipient localization effects in highly disordered three-dimensional superconductors. In agreement with experiments,  $H_{c2}(T)$  is enhanced over the standard result for dirty superconductors. This enhancement is due to the field-induced suppression of localization. It is demonstrated that properties of  $H_{c2}(T)$  are intimately connected to those of the magnetoresistance. This connection and other predictions of the authors' theory can be tested experimentally.

PACS numbers: 74.70.Nr, 71.55.Jv, 74.60.Ec, 74.70.Lp

Recent measurements<sup>1,2</sup> of the upper critical magnetic field  $H_{c2}(T)$  of amorphous transition-metal-based alloys and similar materials have shown a significant lack of agreement with the standard Werthamer-Helfand-Hohenberg (WHH) theory of dirty superconductors.<sup>3</sup> The low-temperature values of the upper critical field  $H_{c2}$ , for a given  $T_c$ , plotted as a function of temperature  $T$  show consistent enhancement over the corresponding WHH curve. Carter *et al.*<sup>4</sup> have proposed a phenomenological model based on spatial inhomogeneities which may explain such deviations. However, Tenhover, Johnson, and Tsuei<sup>1</sup> have ruled out this mechanism for their samples. In addition they have noted that the magnitude of the deviation of  $H_{c2}$  from WHH theory seems to have a systematic correlation with the room-temperature normal-state resistivity  $\rho_N$  of the material. In the measurements of Refs. 1 and 2 this resistivity is greater than  $100 \mu\Omega \text{ cm}$ , where effects of localization may be important.

In the present paper we calculate  $H_{c2}(T)$  for highly disordered superconductors and show that the deviations from the WHH curve can be explained as a localization effect. Our work emphasizes that the study of  $H_{c2}(T)$  provides a relatively good handle on determining the importance of localization effects on superconductivity, and may also help clarify the effects of magnetic fields on localization phenomena.

Maekawa, Ebisawa, and Fukuyama<sup>5</sup> have recently calculated some effects of localization on

$H_{c2}(T)$  and  $T_c$  perturbatively in two dimensions in the weak-disorder regime  $E_F\tau \gg 1$ . However, in three dimensions, the effects of incipient localization only become appreciable for strong disorder ( $E_F\tau \sim 1$ ) and therefore cannot be examined with perturbation theory. The present formalism allows us to examine this regime.

Our impurity eigenstate approach follows the work of deGennes<sup>6</sup> and more directly that of Anderson, Muttalib, and Ramakrishnan.<sup>7</sup> These latter authors have shown that in the presence of (incipient) localization the Coulomb interaction becomes highly retarded because of the anomalous scale-dependent diffusion associated with localization. This enhances the Coulomb pseudopotential  $\mu^*$  and hence reduces  $T_c$ . In order to sort out how important localization phenomena are in superconductivity as opposed to nonlocalization effects on  $T_c$ ,<sup>8</sup> it appears that studies of the temperature dependence of  $H_{c2}$  may be particularly useful. In  $H_{c2}$ -vs- $T$  measurements, variations with  $T$  (or  $H$ ) of the density of states and the electron-phonon interactions are expected to be relatively weak. This allows us to focus on the Coulombic interactions as the dominant effect.

The main result of the present work (which will be derived below) is that in highly disordered superconductors, the primary effect of localization on  $H_{c2}$  can be incorporated into a renormalized Coulomb pseudopotential  $\mu^*(H)$ . The upper critical field satisfies the equation

$$1 = [N(0)V_{\text{ph}} - \mu^*(H_{c2})] \left[ \ln(1.14\hbar\omega_D\beta) + \psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \beta\hbar\bar{D}H_{c2}/2\varphi_0\right) \right], \quad \beta = (k_B T)^{-1} \quad (1)$$

where  $\varphi_0 = \hbar c/2e$ ,  $\psi$  is a digamma function, and  $N(0)$  is the density of states per spin at the Fermi energy which, like the phonon interaction  $V_{\text{ph}}$ , in general, includes localization and field effects.  $\bar{D}$  is the (long-time) field-dependent diffusion coefficient. An explicit expression for  $\mu^*(H)$  will be presented below. The major difference between Eq. (1) and the usual WHH expression is that the Coulomb pseudopotential, which appears explicitly in our formalism, contains a dependence on both the normal-state resistivity  $\rho_N$  and the magnetic field  $H$ . Physically the enhancement of  $H_{c2}$  arises because the applica-

tion of a magnetic field diminishes the effects of incipient localization.<sup>9</sup> As a consequence, for a given sample,  $\mu^*(H)$  decreases with increasing field so that the enhancement over the WHH prediction increases with decreasing  $T$ , as is frequently observed experimentally.<sup>1</sup>

Our calculation of  $H_{c2}$  follows the work of deGennes,<sup>6</sup> who noted that the phonon contribution to the real-space superconducting order parameter depends on the correlation function

$$g(rr';t) = \langle \sum_j \varphi_i^*(r) \varphi_i(r') \varphi_j^*(r) \varphi_j(r') \exp[i(E_i - E_j)t] \rangle_i, \quad (2)$$

where  $\langle \dots \rangle_i$  denotes an impurity average and  $E_i$  is the eigenvalue for the  $i$ th state  $\varphi_i$ . We may readily<sup>10</sup> generalize Ref. 6 to include the Coulomb contribution to the superconducting order parameter which is given in terms of the kernel

$$K_c(rr';\omega - \omega') = \frac{V_c \bar{g}(rr';\omega - \omega')}{1 + 2N(0)V_c \int_{\omega_D}^{\omega_F} d\bar{\omega} \int_{\omega_D}^{\omega_F} d\bar{\omega}' \bar{g}(rr';\bar{\omega} - \bar{\omega}') / (\bar{\omega} + \bar{\omega}')}. \quad (3)$$

Here  $\bar{g}$  is the Fourier transform of  $g$  and  $V_c$  is the coefficient of the simplified ( $\delta$  function) Coulomb potential.<sup>10</sup> In deriving Eq. (3) we have used the fact (which we will prove below) that  $\bar{g}(rr';\omega - \omega')$  falls off exponentially with  $|r - r'|$ .

We first calculate  $\bar{g}$  when the diffusion coefficient is not anomalous. In the presence of a field  $\bar{g}$  satisfies a modified diffusion equation,

$$i\Omega \bar{g}(rr';\Omega) = D(\nabla_r - 2ieA/c)^2 \bar{g}(rr';\Omega), \quad (4)$$

where  $D$  is the diffusion constant. To calculate the critical field, the low-frequency contribution to  $\bar{g}$  is obtained<sup>11</sup> by replacing the frequency argument by the lowest eigenvalue of Eq. (4),  $\epsilon_0 = 2DeH/c$ . The Fourier transform of  $\bar{g}$  with the initial condition  $g(rr';0) = \delta(r - r')$  is given by

$$g(rr';t) = \frac{1}{Q(t)} \exp \left\{ \frac{-1}{4Dt} \left[ \frac{(x-x')^2 + (y-y')^2}{\tanh(\omega_H t/2)/(\omega_H t/2)} + (z-z')^2 + i\omega_H t(x'y - y'x) \right] \right\}. \quad (5)$$

Here  $Q(t) = (4\pi Dt)^{3/2} \sinh(\omega_H t/2)/(\omega_H t/2)$  and  $\omega_H = 4DeH/hc$ . Finally we derive Eq. (1) by adding the phonon contribution and using the fact [derived from Eq. (5)] that  $\bar{g}$  falls off exponentially with  $r - r'$ . We can identify

$$\mu^*(H) = \frac{N(0)V_c \bar{g}(rr';\epsilon_0)}{1 + 2N(0)V_c \int_{\omega_D}^{\omega_F} d\bar{\omega} \int_{\omega_D}^{\omega_F} d\bar{\omega}' \bar{g}(rr';\bar{\omega} - \bar{\omega}') / (\bar{\omega} + \bar{\omega}')}. \quad (6)$$

In the limit  $H=0$  this reduces to the usual Coulomb pseudopotential.

It is important to note that the quantity  $\bar{g}(rr';\omega)$  which determines the  $H$  dependence of  $\mu^*$  also determines the localization contribution to the field dependence of the magnetoconductivity through the equation<sup>9</sup>

$$\frac{\delta\sigma(H,\omega)}{\sigma_0} = \frac{-1}{\pi N(0)} \int \bar{g}(rr';\omega) \frac{d^3r}{v}, \quad (7)$$

where  $\sigma_0$  is the Drude conductivity. Hence there is an intimate relation between localization contributions to  $\sigma(H)$  and the behavior of  $H_{c2}$ . We now include the anomalous diffusion<sup>12</sup> associated with localization. Equations (6) and (7) are generally valid for any  $\bar{g}$ . In the absence of an exact theory we proceed approximately by obtaining  $g(rr';t)$  from Eq. (5) after replacing  $D$  by the (space- and time-dependent) diffusion coefficient. We also replace  $\sigma_0$  by  $\sigma(H=0)$  which guarantees that in the spatially uniform and the  $H=0$  limit our approach is identical to that of Vollhardt and

Wölfle.<sup>13</sup> The diffusion coefficient, which is not known precisely, will be modeled with scaling arguments. We expect that our results, although not fully self-consistent,<sup>14</sup> are qualitatively insensitive to this modeling.

We assume that there exists a field-dependent scaling length  $L_s(H)$ , upon which the spatially dependent diffusion constant  $D(r)$  depends. The time dependence associated with this  $r$  dependence enters through a change<sup>7</sup> in  $Q(t)$  in Eq. (5). It follows from the form of magnetoconductivity<sup>15</sup> (which we have evaluated for finite-size systems) that as in the case  $H=0$ ,  $D(r)$  has a  $1/r$  dependence for small  $r$  and is constant for  $r$  greater than the characteristic magnetic length. Hence  $D(r)$  can always be written in the form

$$D(r) = D_0 l_e / r, \quad l_e < r < L_s(H), \quad (8a)$$

$$D(r) = \bar{D} = D_0 l_e / L_s(H), \quad L_s(H) < r, \quad (8b)$$

where  $l_e$  is the elastic mean free path and  $D$  is

the low-frequency limit of the diffusion coefficient which must also appear in the generalization of Eq. (4). The effects of the *inelastic* mean free path  $l_i \gg L_s(H)$  will be discussed below. To calculate  $L_s(H)$  we use the zero-field result ( $L_s/l_c) = (\rho_N/\rho_c)$ . Here  $\rho_c$  is the characteristic resistivity which determines the importance of localization effects. We make the natural *Ansatz* that for  $H \neq 0$ ,  $L_s(H)/l_c = (\rho_N/\rho_c)_H$ . After solving Eq. (7), using Eqs. (8) and (5), we find that  $1/L_s(H) - 1/L_s \propto \sqrt{H}$  in strong fields and  $\propto H^2$  in weak fields. Our results are similar to those found elsewhere,<sup>9</sup> except that now we have included an explicit dependence on  $\rho_N/\rho_c$  in  $\delta\sigma(H)$ .<sup>14</sup> Furthermore even in the nonanomalous regime we have avoided inaccuracies arising from a series truncation, by using an exact solution of the field-dependent diffusion equation [Eq. (5)]. To compute  $D(\nu)$  [or  $L_s(H)$ ] we use our calculated results for the high- and low-field limits and interpolate between these following Ref. (9).

It is important to note that because the field dependence of the scaling length approaches zero as  $H^2$ , the derivative  $\partial\mu^*/\partial H|_{H=0}$  also vanishes. As a consequence  $\partial H_{c2}/\partial T|_{T_c}$  reduces to the usual WHH expression. This is in accord with most experiments<sup>1,2</sup> which suggest that  $H_{c2}(T)$  can be fitted by WHH theory in the vicinity of  $T_c$ . Furthermore it serves to reinforce our *Ansatz* which relates the magnetoresistance to  $L_s(H)$ . As long as the change in magnetoresistance vanishes as  $H^{1+\delta}$ , where  $\delta > 0$ , this result for  $dH_{c2}/dT|_{T_c}$  will be valid.

Localization effects are most important for high frequencies corresponding to  $\omega\tau > (\rho_N/\rho_c)^{-3}$ . We estimate these frequencies to be considerably greater than the Debye frequency  $\omega_D$  [for  $E_F\tau \sim O(1)$ ]. Consequently the electron-phonon interactions, which are dominated by  $\omega < \omega_D$ , are not expected to be as strongly effected by localization as is  $\mu^*$ , which contains frequency contributions from 0 to  $E_F$ . For this reason and as in Ref. (5), we will ignore localization and its related field-dependent effects on  $V_{ph}$ . We have calculated explicitly the changes in  $N(0)$  arising from localization. These lead to extremely weak field variations which can be neglected in obtaining the  $T$  dependence of  $H_{c2}$ .

It is useful to consider the quantity

$$h_{c2}(T) \equiv H_{c2}(T)/(-T_c dH_{c2}/dT)_{T_c}.$$

With these reduced units the WHH curve plotted versus  $T/T_c$  is a universal function, i.e., independent of disorder. Observed deviations of

$h_{c2}(T)$  from this universal curve cannot be attributed to changes in the simple electronic properties [such as<sup>8</sup>  $N(0)$ ] of the system. We discussed earlier the physical mechanism for the enhancement of  $H_{c2}(T)$  over the (nonuniversal) WHH curve, for a fixed  $T_c$ . This mechanism also gives rise to an enhancement in  $h_{c2}(T)$  with respect to the universal WHH curve. The curves for  $h_{c2}(T)$  shown in Fig. 1 correspond to varying degrees of disorder. For definiteness we chose  $N(0)V_{ph} = 0.59$ ,  $\hbar\omega_D/E_F = 10^{-2}$ ,  $E_F\tau = 1.0$ , and  $\theta_D = 300$  K. These are fairly typical numbers for strongly disordered transition-metal alloys such as amorphous MoRe.<sup>4</sup> With this parametrization we find that the curve  $\rho_N/\rho_c = 2.0$  corresponds to a sample with  $T_c = 8.4$  K,  $H_{c2}(0) = 197$  kG, and  $D = 0.38$  cm<sup>2</sup>/sec, for  $H = 0$ , which appears to be appropriate for amorphous MoRe.<sup>1</sup> While the  $T = 0$  and  $T = T_c$  limits are unaffected, the intermediate temperature dependence of  $H_{c2}(T)$  depends on the *inelastic* mean free path  $l_i$ . We found we could get a reasonable fit to the MoRe data of Ref. 1 by choosing  $l_i = 5 \times 10^3 l_c / T^{3/2}$ . These data are indicated in the figure. We stress that our theory is only qualitative so that quantitative agreement with experiment is not very meaningful. As shown in the inset of Fig. 1, for higher values of  $\rho_N/\rho_c$  we find a small upward curvature

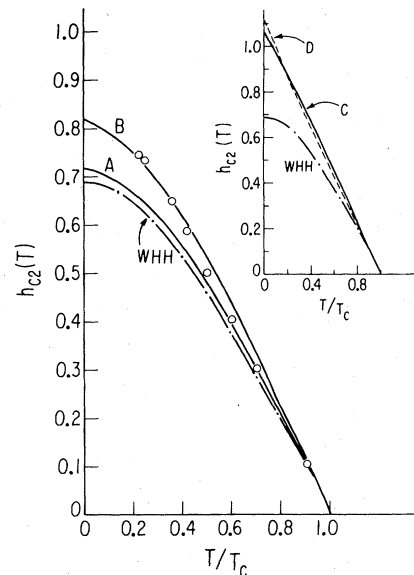


FIG. 1. Calculated temperature dependence of upper critical field (in reduced units) for various  $\rho_N/\rho_c = 1.5$  (curve A) and  $2.0$  (curve B) and in the inset  $\rho_N/\rho_c = 2.9$  (curve C) and  $3.2$  (curve D). The standard dirty-superconductor result (labeled WHH) and experimental data (circles) from Ref. 1 are shown for comparison.

in  $H_{c2}(T)$ . This curvature has been reported in  $\text{Mo}_{0.75}\text{Si}_{1.25}$ .<sup>2</sup> Our numerical studies indicate that the smaller the ratio  $l_i/l_e$  the more pronounced the upward curvature.

In WHH theory  $h_{c2}(0)$  is independent of  $\rho_N$ . By contrast we find that because the effect of a magnetic field on  $\mu^*$  is larger for larger resistivity,  $h_{c2}(0)$  increases with  $\rho_N/\rho_c$ . These features have also been observed experimentally in Ref. 1 and should be tested more systematically. It may be possible to use  $h_{c2}(0)$ -vs- $\rho_N$  measurements as a means of distinguishing between localization and in homogeneity contributions to  $H_{c2}$ . However, it should be noted that unlike the  $T$  dependence of  $H_{c2}$  (for fixed  $\rho_N$ ) the variations of  $H_{c2}$  with disorder may contain significant contributions other than those deriving from localization.

Our work suggests that systematic measurements of  $T_c$ ,  $H_{c2}$ , and the magnetoresistance in highly disordered superconductors will clarify the effects of localization on superconductivity as well as help further elucidate the effects of magnetic fields on localization phenomena.

We thank T. F. Rosenbaum and T. H. Geballe for helpful conversations. This work was supported by National Science Foundation Grants No. DMR 81-15618 and No. MRL 82-16892, and is part of a Ph.D. dissertation submitted by one of us (L.C.) to the Department of Physics, The University of Chicago.

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<sup>14</sup>As in previous work (see for example Ref. 9), in contrast to the work of Y. Ono, D. Yoshioka, and H. Fukuyama, *J. Phys. Soc. Jpn.* **50**, 2465 (1981), we used a non-self-consistent procedure to compute  $\delta\sigma(H)$ . Furthermore in the magnetoresistance Eqs. (8) were evaluated at  $H=0$ . Our theory should be viewed as semiquantitative.

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