

amorphous Si and Ge that the atoms carry charges. However, they have interpreted the spectra as giving the average displacements of the atoms from their positions in a crystal, an admittedly unrealistic picture. We suggest the following alternative procedure: The vibrational modes of a specific model can be calculated, as we have done previously.<sup>1</sup> Given the vibrational eigenfunctions and the atomic charges (computed as we have or by an equivalent method) for that model, the dipole matrix elements can be evaluated, and the infrared spectrum can be calculated by averaging over a suitable frequency interval. The result would be subject to the double uncertainties introduced by the incomplete knowledge of the interatomic potential function and by deficiencies in the electronic solution, but the former could be practically eliminated if accurate neutron inelastic-scattering experiments could be performed. The infrared spectrum would thus be a rather direct probe of the electronic structure.

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## Upper Critical Field of Nb: Calculated Temperature Dependence and Anisotropy

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The magnitude, anisotropy, and temperature dependence of the  $H_{c2}$  for Nb are calculated from the underlying electronic structure. Excellent agreement between theory and experiment results if experimental Fermi velocities are used and if Fermi-velocity-anisotropy, strong-coupling, and impurity effects are included.

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The highest magnetic field which a type-II superconductor can sustain in the superconducting state is denoted  $H_{c2}$ , the upper critical field. For a given material  $H_{c2}$  depends upon sample purity, temperature, and the direction of the applied field relative to the crystal axes. In this paper, the magnitude, anisotropy, and temperature dependence of  $H_{c2}$  of Nb are calculated from its electronic structure.

In order to calculate  $H_{c2}$  one should include (1) an extension of the usual Ginzburg-Landau-Abrikosov-Gor'kov<sup>1-4</sup> theory to handle the non-

local effects which enter at temperatures below  $T_c$ ,<sup>5-12</sup> (2) finite-mean-free-path effects due to impurity scattering,<sup>5-9,11-14</sup> (3) strong-coupling effects,<sup>8,9,14</sup> (4) the wave-vector dependence of the Fermi velocity,<sup>6,10-14</sup> and (5) the anisotropy of the superconducting energy gap.<sup>11-13</sup> The only previous attempt to calculate  $H_{c2}$  with use of a realistic model of the band structure was that of Mattheiss,<sup>15</sup> who calculated the enhancement of  $H_{c2}(0)$  and of the ratio  $\langle H_{c2}(0) \rangle / T_c (dH_{c2}/dT)_{T_c}$  due to Fermi-velocity anisotropy.

Effects (1)-(5) are all included in the following

generalization of an equation derived (but not solved) by Eilenberger and Ambegaokar<sup>8</sup>:

$$\varphi_l = 2 \sum_{l' \geq 0} \frac{\lambda_{\text{eff}}(l, l')}{Z_{l'}} \left[ x f^{-1} \left( \frac{2l' + 1 + \alpha t}{x} \right) - \alpha t \right]^{-1} \varphi_{l'}, \quad (1)$$

In this equation,  $\varphi_l$  is the frequency-dependent gap function evaluated at the Matsubara frequencies  $i(2l+1)\pi k_B T$ . The electron-phonon interaction kernel  $\lambda_{\text{eff}} \equiv \frac{1}{2}[\lambda_{l-l'} + \lambda_{l+l'+1} - 2\mu^*]$  is defined in terms of frequency-dependent coupling parameters  $\lambda_l$  and the Coulomb pseudopotential  $\mu^*$ , both of which are defined by Allen and Dynes.<sup>16</sup>  $Z_l$  is the renormalization function  $Z_l = 1 + (\lambda_0 + 2\lambda_1 + \dots + 2\lambda_l)/(2l+1)$ . The parameter  $x$  contains the  $H_{c2}$  dependence  $x = [\langle v_F^2 \rangle e \hbar H_{c2}/c]^{1/2}/2\pi k_B T$ ,  $t$  is the reduced temperature  $t = T/T_c$ , and the impurity parameter  $\alpha \equiv \hbar/2\pi k_B T_c \tau_{\text{tr}}^*$  is inversely proportional to the transport lifetime  $\tau_{\text{tr}}^*$ . The function  $f(\eta)$  is defined by

$$f(\eta) = \int_0^\infty e^{-\eta y} \langle \Delta_k \exp(-\frac{1}{2} y^2 u_\perp^2) \rangle dy / \langle \Delta_k \rangle, \quad (2)$$

where angular brackets denote a Fermi-surface average,  $\Delta_k$  is the anisotropic energy gap, and  $u_\perp$  is the normalized component of the Fermi velocity perpendicular to the magnetic field  $u_\perp^2 = [v^2 - (\vec{v} \cdot \hat{e}_H)^2] / \langle v^2 \rangle$ .

$H_{c2}$  is obtained by finding the largest value of  $x$  (for given  $\alpha$ ,  $t$ , and field direction) for which Eq. (1) has a nontrivial solution. Equation (1) is equivalent to Eq. (A4) of Ref. 8. The new features included here enter through Eq. (2) in which  $f(\eta)$  has been generalized to include the effects of Fermi surface and gap anisotropy. The effect of gap anisotropy is included in the manner suggested by Teichler,<sup>11</sup> who assumed that the electron-phonon coupling parameter was separable and energy independent. I have not attempted to improve upon these approximations because I believe the entire effect of gap anisotropy on  $H_{c2}$  to be very small in Nb. This belief is based upon calculations<sup>17</sup> of  $\Delta_k$  which predict an rms deviation of only 3% and upon the present calculations which show good agreement with the experimental  $H_{c2}$  when gap anisotropy is neglected entirely.

I found effects (1) and (4) to be the most important in relatively clean Nb. Fortunately, a very simple pair of parametric equations can be derived which determine  $H_{c2}$  in the clean, weak-coupling approximation. The effects of strong coupling and of impurities can then be described in terms of enhancement factors of order unity which multiply the clean, weak-coupling result.

The clean limit is obtained by setting  $\alpha = 0$  in Eq. (1) and the weak-coupling limit is obtained by a standard but somewhat subtle procedure which involves neglect of the  $l$  dependences of  $\varphi_l$ ,  $Z_l$ , and  $\lambda_{\text{eff}}(l, l')$ . The resultant divergent sums are handled by subtracting off the  $T_c$  equation ( $x \rightarrow 0$  limit) so that Eq. (1) becomes<sup>10</sup>

$$-\ln t = 2 \sum_{l \geq 0} \left[ \frac{1}{2l+1} - \frac{1}{x} f \left( \frac{2l+1}{x} \right) \right]. \quad (3)$$

Equation (3) is equivalent to the clean limit of Eqs. (5) and (26) of Ref. 6. The sum over  $l$  may be performed exactly and yields a function  $F(x, \hat{e}_H)$  which completely determines  $H_{c2}$  in this approximation:

$$F(x, \hat{e}_H) = \frac{1}{x} \int_0^\infty dy \frac{\langle 1 - \exp(-\frac{1}{2} y^2 u_\perp^2) \rangle}{\sinh(y/x)}. \quad (4)$$

Thus,  $H_{c2}$  as a function of  $t$  is entirely determined by the following pair of parametric equations with parameter  $x$ :

$$t = \exp[-F(x, \hat{e}_H)], \quad (5)$$

$$H_{c2} = \frac{(2\pi k_B T_c)^2}{\langle v_F^2 \rangle e \hbar / c} x^2 t^2. \quad (6)$$

I calculated  $F(x, \hat{e}_H)$  using a Fermi surface and Fermi velocities calculated previously in connection with studies of superconductivity and transport in Nb,<sup>18</sup> and used Eqs. (5) and (6) to determine  $H_{c2}(t, \hat{e}_H)$  in the absence of strong-coupling and impurity effects.

Strong-coupling effects were calculated by solving Eq. (1) in the clean limit by use of coupling constants  $\lambda_l$  generated from the spectral function  $\alpha^2(\omega)F(\omega)$  shown in Fig. 7 of Ref. 17. The Coulomb pseudopotential was set equal to 0.176 in order to obtain the experimental  $T_c$ . The effect of strong coupling is an enhancement of  $H_{c2}$  primarily through the renormalization of the Fermi velocity [ $v_F \rightarrow v_F^* = v_F/(1+\lambda)$ ] but also through an additional factor  $e_{sc}$  coming from the energy dependence of the gap function. The calculated enhancement factor  $e_{sc}$  is about 1.104 at  $T_c$  and is approximately independent of temperature and field direction [Fig. 1(a)]. Werthamer and McMillan<sup>9</sup> noted the temperature independence of  $e_{sc}$  but did not give its magnitude while McMillan and Hohenberg<sup>19</sup> estimated  $e_{sc}$  to be 1.4 for Nb, substantially higher than the present result. As a check on the numerical procedures, I also calculated the enhancement factor in the dirty limit and obtained 1.18 in agreement with the results of Rainer and Bergmann.<sup>20</sup>

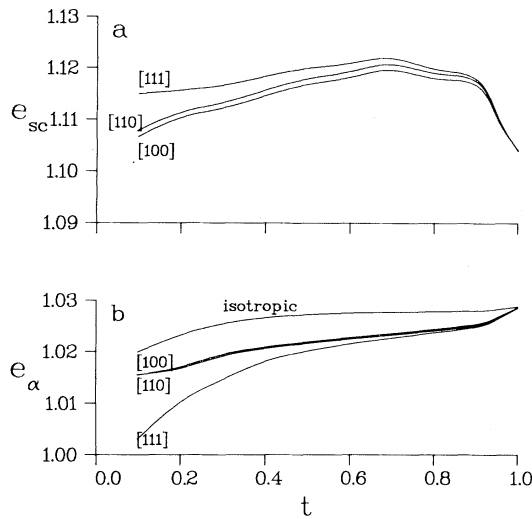


FIG. 1. Enhancements of  $H_{c2}$  in Nb over weak-coupling isotropic theory. (a) Enhancement due to strong-coupling effects for three symmetry directions. (b) Enhancement due to an impurity parameter of 0.03 for three symmetry directions and for an isotropic Fermi surface.

Although our primary concern in this paper is with clean Nb, experiments must be performed on samples with some impurity scattering. I investigated the effect of impurities on  $H_{c2}$  by solving the weak-coupling version of Eq. (1) retaining velocity anisotropy and impurity effects. Near  $T_c$ ,  $H_{c2}$  is enhanced by an isotropic factor,  $1 + 0.965\alpha$  as shown by Gor'kov<sup>3,4</sup> but at lower temperatures, the enhancement is much less and depends upon

$$H_{c2} = \frac{4\pi ck_B^2 T_c^2 e_{sc}(T_c)}{\langle v^{*2} \rangle e\hbar} \{ 0.71306\theta + [2.18521\langle u_{\perp}^4(\hat{e}_H) \rangle - 1.0696]\theta^2 \}. \quad (7)$$

Consider the first term in Eq. (7). Experimental values of  $(-dH_{c2}/dt)_{t=1}$  for clean Nb when corrected for impurity enhancement range from 0.420 T (Ref. 21) to 0.435 T.<sup>22, 23</sup> The value calculated here is 0.420 in excellent agreement with experiment. The overall scale of  $H_{c2}(T, \hat{e}_H)$  depends crucially on the renormalized mean square Fermi velocity  $\langle v^{*2} \rangle$  which is difficult to calculate precisely because of the limitations of band theory and the possible existence of several types of many-body enhancements.  $v_k$  was renormalized by use of slightly different values of  $\lambda$  for each sheet of Fermi surface. They were chosen so as to yield the correct sheet-to-sheet variation in the density of states as determined experimentally by Crabtree *et al.*<sup>17</sup> and the correct total density of states as determined by specific-heat

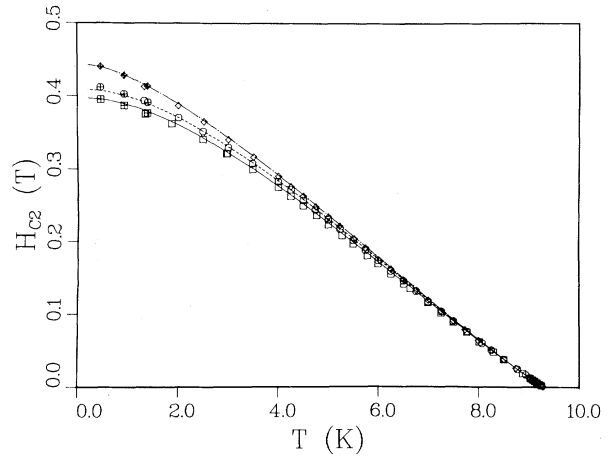


FIG. 2. Calculated and experimental  $H_{c2}$  for clean Nb. Solid, dashed, and dash-dotted curves are calculations for [100], [110], and [111] symmetry directions, respectively. Squares, circles, and diamonds are from experiments of Refs. 21 (open) and 22 (with crosses) for [100], [110], [111] symmetry directions, respectively. Units for  $H_{c2}$  are teslas.

crystal orientation. Figure 1(b) shows the enhancement of  $H_{c2}$  due to an impurity parameter of  $\alpha = 0.03$  for the three symmetry directions of Nb. For comparison, the corresponding enhancement due to an isotropic (i.e., spherical) Fermi surface is also shown.

When  $T$  is very near  $T_c$ ,  $x$  is small in Eqs. (4)–(6) and  $F(x, \hat{e}_H)$  may be expanded in powers of  $x^2$ . Keeping only terms through  $x^4$  and solving for  $H_{c2}$  as a function of  $\theta = 1 - t$  yields

measurements.<sup>24</sup> The average value of the mass enhancement factor was 2.25. Mattheiss<sup>15</sup> calculated a somewhat smaller value of  $(-dH_{c2}/dt)_{t=1}$  because of his use of a smaller mass enhancement factor and also noted the difficulties involved in obtaining  $v^*$ . The second term in Eq. (7) depends upon field direction through  $\langle u_{\perp}^4(\hat{e}_H) \rangle$  which Berthel and Pietrass<sup>10</sup> write as  $\langle u_{\perp}^4(\hat{e}_H) \rangle = \frac{8}{15} + A_1 + A_2(e_x^4 + e_y^4 + e_z^4 - \frac{3}{5})$ . By examination of the experimental orientation and temperature dependence of  $H_{c2}$  near  $T_c$ , they deduced  $A_1 = 0.29 \pm 0.01$  and  $A_2 = -0.11$  for Nb. The values calculated here are identical to two figures.

$H_{c2}$  as a function of temperature calculated for three symmetry directions is shown in Fig. 2. The calculations are for clean Nb but include

strong-coupling and Fermi-surface-anisotropy effects. The experimental data of Williamson<sup>22</sup> and Kerchner *et al.*<sup>21</sup> are also shown. Both sets of experimental data were corrected for impurity enhancement and the data of Kerchner *et al.* have been multiplied by a constant factor<sup>25</sup> of 1.015 to bring them into closer agreement with those of Williamson.

At low temperatures  $t \ll 1$ ,  $x$  is large, and  $F(x, \hat{e}_H)$  is determined by those parts of the Fermi surface for which  $u_{\perp}^2(\hat{e}_H)$  is very small. In the limit of very large  $x$ ,  $F(x, \hat{e}_H)$  can be shown to approach  $\frac{1}{2} \langle \ln(2x^2 \gamma_0 u_{\perp}^2) \rangle$  ( $\ln \gamma_0 = 0.577$ ) and Eqs. (4)–(6) reduce to Takanaka's result<sup>26</sup> for  $H_{c2}$  at  $t = 0$

$$H_{c2}(\hat{e}_H) = \frac{(2\pi k_B T_c)^2 c}{e \hbar 2 \gamma_0 \langle v^{*2} \rangle} \exp[-\ln u_{\perp}^2(\hat{e}_H)]. \quad (8)$$

The Fermi-surface average in Eq. (8) must be performed carefully since  $\ln(u_{\perp}^2)$  is singular when  $u_{\perp}^2$  vanishes. I have calculated  $H_{c2}(\hat{e}_H)$  in the  $(1\bar{1}0)$  plane. These results, which are in good agreement with experiment, will be published elsewhere.<sup>27</sup> The singular nature of  $\langle \ln u_{\perp}^2 \rangle$  in Eq. (8) causes an expansion of  $H_{c2}$  in cubic harmonics,  $H_{c2}(\hat{e}_H) = \langle H_{c2} \rangle [1 + \sum_{l=4,6,\dots} a_l H_l(\hat{e}_H)]$ , to converge rather slowly; nevertheless, values of  $a_l$  determined from experimental measurements of  $H_{c2}$  in the  $(1\bar{1}0)$  symmetry plane<sup>13,22</sup> are in good agreement with those calculated here [ $a_4 = -0.024$  (expt) vs  $a_4 = -0.0224$  (calc) and  $a_6 = 0.009 - 0.011$  (expt) vs  $a_6 = 0.013$  (calc)].

The magnitude, anisotropy, and temperature dependence of  $H_{c2}$  for Nb have been calculated accurately from first principles. The extraordinary agreement between theory and experiment is due to an accurate Fermi surface (adjusted slightly to obtain agreement with de Haas-van Alphen experiments), experimental mass enhancement factors, and careful attention to strong-coupling and impurity effects.  $\langle v^{*2} \rangle$  is the most important factor influencing the magnitude of  $H_{c2}$  and the shape of the Fermi surface is the most important factor affecting its anisotropy and temperature dependence. These calculations show no evidence for gap anisotropy in Nb (contrary to Ref. 13); however, small anisotropies consistent with the  $\lambda_k$ 's of Crabtree *et al.*<sup>17</sup> cannot be ruled out. No evidence is found for a breakdown in the weak anisotropy approximation of Refs. 6 and 7.

The  $H_{c2}$  of V is qualitatively very similar to that of Nb and an analysis along the lines presented here should work well. A calculation of  $H_{c2}$  for a high- $T_c$  A-15 compound should be very

interesting. I expect (based on the temperature dependence of the anisotropy of Nb) that such a calculation would show larger anisotropies at low temperatures than have been observed experimentally.

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## Post-Ionization of Field-Evaporated Ions

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It is shown that the final observed charge state of a field-evaporated ion can be explained by the occurrence of post-ionization whereby an ion being accelerated away from a metal surface in a strong electric field loses one or more electrons by tunneling into the substrate. Calculations using an approximate analytic formula derived from a simple model potential predict the probability of post-ionization for many different elements.

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Field evaporation<sup>1</sup> is the process of removal of an ion from a metal surface by the effect of a high electric field (typically, a few volts per angstrom). In this Letter we consider an ion being evaporated from a surface of the same element; the removal of an adsorbed impurity atom, usually known as field desorption, is not explicitly considered here but may be treated by the same methods.

Post-ionization is the process by which an ion being accelerated away from a surface by an electric field is further ionized by the tunneling of electrons from the ion into the substrate. These electrons may only tunnel to empty states at or above the Fermi level of the substrate and this requires the ion to be at least a critical distance  $z_c$  from the surface. This critical distance is the same as that occurring in field ionization<sup>2</sup> of a gas atom and is given approximately by

$$eFz_c = I - \Phi, \quad (1)$$

where  $F$  is the electric field,  $I$  the ionization potential of the ion, and  $\Phi$  the surface work function. It is vital to have a good estimate of the probability of post-ionization to distinguish between two different possible processes of field evaporation. The two processes are: (a) The ion being initially evaporated in its final charge state; (b) the ion being initially evaporated in some lower charge state and then being post-

ionized to its final charge state.

Current theories of field evaporation<sup>3</sup> cannot satisfactorily explain the finally observed charge state of field-evaporated ions by process (a). Calculations reported here show that process (b) can occur and indeed that it leads to the correct finally observed charge state. Previous theories of post-ionization have used one-dimensional tunneling models<sup>4</sup> or transfer Hamiltonian methods<sup>5</sup> and have predicted that post-ionization is not a significant effect except with work by Ernst<sup>6</sup> on the post-ionization of  $\text{Rh}^+$ . Ernst again used the demonstrably inaccurate one-dimensional tunneling method and his tunneling potential is not given. His electronic frequency factor and ion velocity may well be chosen to "fit" the experimental data. The model potential chosen in the present work for the post-ionization process is deliberately simple while retaining the physically important effects. We thus seek to demonstrate the predictions of the theory without introducing unnecessary complications. Work on the ionization rate of the hydrogen atom (Stark ionization) with use of various model potentials<sup>7</sup> has shown that an acceptable model potential when the ion is at distance  $z_0$  from the surface, is

$$V_{\text{model}} = -Z/r - Fr \cos\theta \quad (2)$$

with cutoff at  $r \cos\theta = z_0$ ,  $\theta = 0$  is the field direction. All quantities are expressed in atomic units