Upper critical field for a high-T_c electron-phonon superconductor: Regime of $T_c/\omega_{ln} \sim 1$

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We have calculated the upper critical field H_{c2} for an electron-phonon superconductor in the limit when the critical temperature T_c is of the same order as a typical phonon energy ω_{ln}. This extends well beyond the conventional range for which $T_c/\omega_{ln} \lesssim 0.25$. In this very-strong-coupling regime, the strong-coupling corrections to H_{c2} are found to be much less than one, in sharp contrast to the usual case for which they are invariably greater than one. On the other hand, the value of the reduced critical field h_{c2} at zero temperature can be larger than in Bardeen-Cooper-Schrieffer theory by more than a factor of 3. Also, as a function of reduced temperature (t), $h_{c2}(t)$ displays an unusual positive curvature for t in the middle of its range and $h_{c2}(0)$ acquires a very strong impurity dependence.

While the electron-phonon interaction has been very successful in describing, in detail, the properties of conventional superconductors,[1-6] it may turn out not to be the mechanism responsible for superconductivity in the heavy-fermion materials and in the recently discovered high-T_c oxides. Perhaps the simplest general argument against a pure electron-phonon interaction in the case of the high-T_c oxides is the drastic difference in the scale for T_c, which can be as large as 100 K in the oxides, to be compared with a maximum of 23.2 K for Nb_3Ge. Another argument is the lack of isotope effect.[7] Several theories based on purely electronic mechanisms have already been put forward,[8-13] but no consensus has, as yet, been reached. In fact, one can find many papers, both experimental[14-19] and theoretical,[20,21] that argue for a conventional mechanism although, presumably, with unusually large values of the electron-phonon interaction.

In this work we do not propose to review and, even less, critically examine all the accumulated evidence for or against the electron-phonon interaction or for some other mechanism, or even for a combination of both. Instead we want to study the behavior of the upper critical magnetic field H_{c2} that is predicted within Eliashberg theory when the critical temperature becomes comparable in magnitude to a typical phonon energy. The thermodynamic properties, in this extreme limit, have recently been worked out by Marsiglio, Akis, and Carbotte,[22] and were found to be quite unusual. We stress that, in our approach, no attempt is made to justify the large values of electron-phonon spectral density that will be required. Instead we simply assume them to be given, and work out their consequences for superconducting properties. The reader should keep in mind that there are criteria of lattice stability that must limit the size that the electron-phonon interaction can have in a given material. It is not the purpose of this work to address this important issue.

The equations for the upper critical magnetic field $H_{c2}(T)$, as a function of temperature T, that form the basis of our work were first given by Schossmann and Schachinger.[23] They can be written, with no Pauli limiting, in the form

$$\tilde{\Delta}(i\omega_n) = \pi T \sum_m [\lambda(\omega_m - \omega_n) - \mu^*] \times \frac{\tilde{\Delta}(i\omega_m)}{\chi^{-1}[\tilde{\omega}(i\omega_m)] - \pi t^+} , \quad (1)$$

and

$$\tilde{\omega}(i\omega_n) = \omega_n + \pi T \sum_m \lambda(\omega_m - \omega_n)\mathrm{sgn}(\omega_m) + \pi t^+ \mathrm{sgn}(\omega_n) , \quad (2)$$

with

$$\chi[\tilde{\omega}(i\omega_n)] = \frac{2}{\sqrt{\alpha}} \int_0^\infty dq\, e^{-q^2} \tan^{-1}\left[\frac{\sqrt{\alpha}}{|\tilde{\omega}(i\omega_n)|} q \right] . \quad (3)$$

In Eqs. (1) and (2),

$$\lambda(\omega_m - \omega_n) = \int \frac{2\Omega\alpha^2F(\Omega)\, d\Omega}{\Omega^2 + (\omega_m - \omega_n)^2} , \quad (4)$$

with the Matsubara frequencies $i\omega_n \equiv i\pi T(2n-1)$ and $n = 0, \pm 1, \pm 2, \ldots$. The parameter t^+, which appears both in the equation for the pairing energy $\tilde{\Delta}(i\omega_n)$ and in the renormalized frequency $\tilde{\omega}(i\omega_n)$, is related to the impurity lifetime τ by $t^+ \simeq 1/(2\pi\tau)$. The parameter μ^* is the Coulomb pseudopotential, which is a parameter that we fix at some convenient value, usually 0.1. In Eq. (3), the parameter α is defined by $\alpha = \frac{1}{2} eH_{c2}(T)v_F^2$ with e the electron charge and v_F the Fermi velocity. Finally, $\alpha^2F(\omega)$ in Eq. (4) is the electron-phonon spectral density.

In the approach we wish to take here $\alpha^2F(\omega)$ can, in a sense, be chosen at will and it is not essential to attempt to choose some model that might be realistic for the high-T_c oxides. Indeed, as we have already mentioned, it may well turn out that a different mechanism applies or perhaps that it is the combination of two such mechanisms. Nevertheless, to be specific, we will start with the $\alpha^2F(\omega)$ calculated by Weber[20] for the case of $La_{1.85}Sr_{0.15}CuO_4$. His spectral density gives a value of T_c around 36 K. The

characteristic Allen-Dynes phonon energy ω_{1n} associated with this spectrum is ~ 14.0 meV and is defined by[24]

$$\omega_{1n} = \exp\left[\frac{2}{\lambda}\int_0^\infty \ln(\omega)\frac{\alpha^2F(\omega)}{\omega}\,d\omega\right] . \quad (5)$$

If Weber's $\alpha^2F(\omega)$ is multiplied by a constant value B so as to get a T_c of 96 K instead of 36 K, we find a value of $T_c/\omega_{1n} \cong 0.6$, which is very much greater than the values found in the conventional superconductors. As we have said, these all fall in the range $(0 \leq T_c/\omega_{1n} \lesssim 0.25)$. This implies that we are now in a very different regime for which no expansion in powers of T_c/ω_{1n} makes sense and we need to proceed numerically.

To be able to systematically span the small and high T_c/ω_{1n} region, we suggest the model

$$\alpha^2F(\omega) = B\alpha^2F^0(\omega\gamma) , \quad (6)$$

where the superscript zero on the right-hand side of Eq. (6) denotes our base spectrum for $La_{1.85}Sr_{0.15}CuO_4$ and $\alpha^2F(\omega)$ on the left is our *model* spectrum. The two constants B and γ are actually not independent in what follows. For a given choice of γ, which can be used to soften of stiffen the spectrum because[25]

$$\omega_{1n} = \omega_{1n}^0/\gamma , \quad (7)$$

the value of B is fixed to get a T_c of 96 K for $\mu^* = 0.1$. We could think of changing the value of T_c but we argue that this is unnecessary. For a given shape of $\alpha^2F(\omega)$, the quantities which we wish to calculate can be shown to depend only on the ratio T_c/ω_{1n},[25] and not on each factor separately. Hence, one can alternatively use some base spectrum and form model spectra by simply multiplying by some overall factor B leaving ω_{1n} fixed and continuously increasing T_c, thus, spanning the same range of T_c/ω_{1n} as with model (6). Since the properties of interest are universal functions of T_c/ω_{1n}, for a given shape, nothing new is generated. In our study of the upper critical field there is a potential complication since the impurity parameter t^+ also enters the equations. One can maintain the property of universality in this case as well by also scaling $t^+ \rightarrow t^+/\gamma$.

Having specified our model for $\alpha^2F(\omega)$, we can now present results for $H_{c2}(T)$. As is done conventionally, we introduce a strong-coupling correction factor $\eta_{H_{c2}}(t^+,T)$ through the definition[26]

$$H_{c2}(t^+,T) = \eta_{H_{c2}}(t^+,T)H_{c2}^{\rm BCS}(t^+,T) , \quad (8)$$

where we have made the impurity dependence of H_{c2} and $\eta_{H_{c2}}$ explicit, and $H_{c2}^{\rm BCS}$, on the right-hand side, is the BCS result[27,28] to which we compare. Note that the Fermi velocity drops out of $\eta_{H_{c2}}$ so that it is not an added parameter. This is so because we have not included Pauli limiting in our calculations. In Fig. 1(a) we show results for $\eta_{H_{c2}}(t^+,0)$ at T_c, and at zero temperatures for two values of t^+, namely, the clean limit $t^+ = 0$ and a reasonably dirty sample with $t^+ = 100$ meV. First note that the difference between our results for $t^+ = 0.0$ and $t^+ = 100$ meV are not large and we will not need to stress this further. Since $\eta_{H_{c2}}$ measures deviations from BCS in the limit $T_c/\omega_{1n} \rightarrow 0$ the correction factors $\eta_{H_{c2}}$ will tend towards

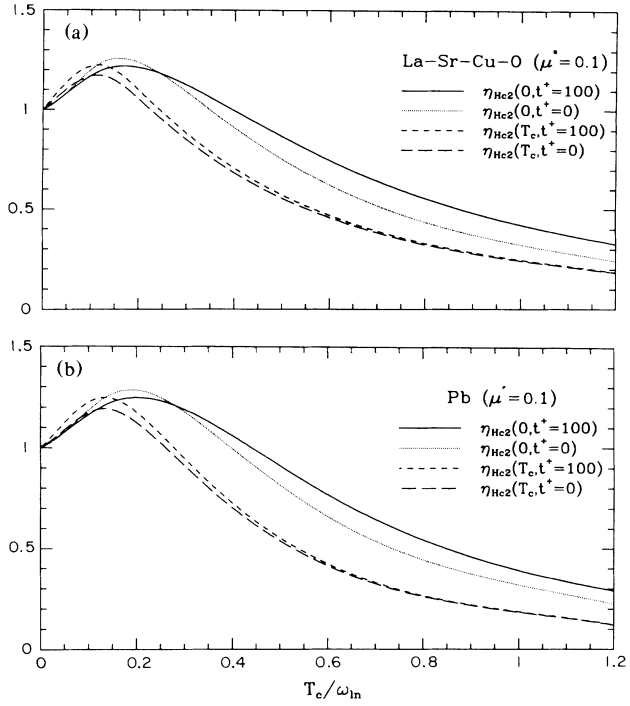

FIG. 1. (a) The strong-coupling correction parameter, $\eta_{H_{c2}}(t^+,T)$ is displayed for $T=0$ and $T=T_c$, and for $t^+=0$ and 100 meV. We have used the spectrum calculated by Weber for $La_{1.85}Sr_{0.15}CuO_4$ with $\mu^*=0.1$. T_c was held fixed at 96 K by scaling the spectrum in height while the abscissa was scaled in order to sweep through the values of T_c/ω_{1n} displayed in the figure. Note that all the correction parameters display the same qualitative trend. In the conventional strong-coupling regime $(T_c/\omega_{1n} \lesssim 0.2)$, all the corrections are greater than one, and modest. However, in the very-strong-coupling regime $(T_c/\omega_{1n} \approx 1)$, the corrections differ substantially from unity, and are less than one. No significant qualitative difference is noticeable between $t^+=0$ meV and $t^+=100$ meV. (b) The same results are displayed as in (a), but for a Pb spectrum. No qualitative change from (a) is observed, indicating that the results noted in (a) are not very model dependent.

one, for any temperature and impurity content. As T_c/ω_{1n} increases $\eta_{H_{c2}}$ increases and a T_c/ω_{1n} expansion should apply.[6] This is the conventional strong-coupling regime where all known electron-phonon superconductors fall. As T_c/ω_{1n} goes beyond 0.2 to 0.25 the situation reverses. The coefficient $\eta_{H_{c2}}$ first exhibits a maximum and then starts to drop. For values of T_c/ω_{1n} of order 1, $\eta_{H_{c2}}$ takes on a value well below one; this surprising result could not have been guessed at from an extrapolation of what is known in the conventional case for which $\eta_{H_{c2}}$ is always greater than one.

In experiments one often measures the slope at T_c of the upper critical magnetic field $H_{c2}(T)$ and uses this slope to get a measure of the zero-temperature Sommerfeld constant $\gamma(0)$. In the analysis, a BCS relationship is almost always used[15,16] to relate these two quantities. Convenient relationships that apply in the clean, dirty, and intermediate impurity case have been given in many places; among them is the useful recent compilation of Or-

lando, McNiff, Foner, and Beasely.[29] For the dirty limit we can write, assuming that BCS theory applies,

$$\left[-\frac{dH_{c2}(T)}{dT}\right]_{T_c}=4.48\times10^4\gamma(0)\rho_{\Omega\,\mathrm{cm}}\,\mathrm{Oe\,K^{-1}}\ , \quad (9)$$

where $\rho_{\Omega\,\mathrm{cm}}$ is the resistivity of the sample in Ω cm. Strictly speaking, only the impurity scattering contribution is to be included. For a strong-coupling superconductor Eq. (9) should be modified to include a factor of $\eta_{H_{c2}}(t^+,T_c)$. Also, we should take $t^+\rightarrow\infty$ but this can be ignored since $\eta_{H_{c2}}$ is not very dependent on impurity content. For superconductors in the limit $T_c/\omega_{1n}\sim1$ it is clear from Fig. 1 that ignoring the η correction could lead to a gross underestimate of $\gamma(0)$. This possibility should be kept in mind in the analysis of experiment.

To be sure that the results of Fig. 1(a) are not strongly base dependent, we have carried out additional calculations using the Pb $\alpha^2F^0(\omega)$ as the base electron-phonon spectral density instead of that for $La_{1.85}Sr_{0.15}CuO_4$. These additional results are presented in Fig. 1(b). It is clear that the differences between the Pb-based results and those based on $La_{1.85}Sr_{0.15}CuO_4$ are never great. While acknowledging that there are indeed small quantitative differences that come from the very different shapes of the two base $\alpha^2F^0(\omega)$, we can safely conclude that shape is not as essential feature in determining the qualitative behavior of $\eta_{H_{c2}}$ as a function of T_c/ω_{1n}. Thus, we expect that any reasonably shaped spectral density which corresponds to a value $T_c/\omega_{1n}\cong1$ will give a value of $\eta_{H_{c2}}$, which is much smaller than one and near 0.2.

In Fig. 2(a) we show results for the reduced temperature ($t\equiv T/T_c$) dependence of the normalized upper critical field $h_{c2}(t,t^+)$ defined by[23,27,28]

$$h_{c2}(t,t^+)\equiv H_{c2}(t^+,T)\Big/T_c\left|\left(\frac{dH_{c2}(t^+,T)}{dT}\right)_{T_c}\right|. \quad (10)$$

The normalization with the slope of H_{c2} at the critical temperature T_c means that h_{c2} itself is independent of the choice of Fermi velocity and, therefore, depends only on $\alpha^2F(\omega)$ and the value used for $\mu^*=0.1$. We see in Figs. 2(a) and 2(b), which apply respectively to the clean limit $t^+=0$ and to $t^+=100$ meV, that for large values of T_c/ω_{1n}, the curves start to deviate substantially from a weak-coupling BCS behavior, which is also shown for comparison, and is typical of the small T_c/ω_{1n} limit of Eliashberg theory. In particular, the curves acquire a positive curvature at intermediate values of T. This feature means that any extrapolated values for $h_{c2}(0,t^+)$ based on its behavior near $t=1$ and the BCS predictions[27,28] as a guide, will greatly underestimate the zero-temperature reduced critical magnetic field.

In Fig. 3 we look more closely at the value of $h_{c2}(t,t^+)$ for $t=0$ (zero-temperature limit). Curves are shown for $h_{c2}(0,t^+)$ as a function of T_c/ω_{1n} for a Pb-base spectrum (dotted curve), as well as for a $La_{1.85}Sr_{0.15}CuO_4$ base (solid lines), and for two values of the impurity content, namely $t^+=0$, the clean limit, and $t^+=100$ meV, a rather larger impurity concentration. We first note that in the very-strong-coupling region of $T_c/\omega_{1n}\sim1$, the results do depend on the base spectrum used, with those for a Pb-base considerably larger. At $T_c/\omega_{1n}=1.2$, $h_{c2}(0,t^+=0)\cong1.3$ and $h_{c2}(0,t^+=100)\cong1.56$. These values are very much greater than the BCS values of 0.73 for the clean and 0.69 for the dirty limit, respectively. Even larger values can be obtained when t^+ is increased further, as is illustrated in the inset of Fig. 3, which shows the unusually large impurity dependence of $h_{c2}(0,t^+)$ when $T_c/\omega_{1n}=1.14$. Note that it is only around $t^+=10000$ meV that we begin to see signs of saturation in the curves and that $h_{c2}(0,t^+)$ has reached ~2.5 for the dotted curve based on a PB spectrum.

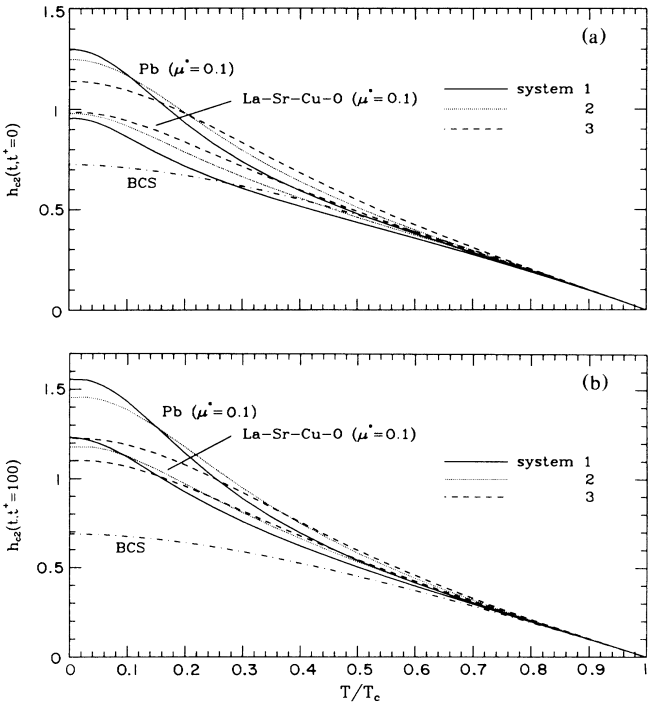

FIG. 2. (a) The reduced upper critical magnetic field, in the clean limit, is displayed as a function of reduced temperature T/T_c. We show curves for three systems in the very-strong-coupling regime for both Pb and $La_{1.85}Sr_{0.15}CuO_4$. For Pb they can be characterized by $T_c/\omega_{1n}=1.14$ (system 1), $T_c/\omega_{1n}=0.86$ (system 2), and $T_c/\omega_{1n}=0.57$ (system 3). For $La_{1.85}Sr_{0.15}CuO_4$ the scaled spectra have $T_c/\omega_{1n}=1.19$ (system 1), $T_c/\omega_{1n}=0.83$ (system 2), and $T_c/\omega_{1n}=0.60$ (system 3). Also shown is the BCS result, for comparison. Note that in the very-strong-coupling regime the curves have developed a large positive curvature, a feature missing in the BCS model. Also note that for Pb the value of $h_{c2}(0,t^+=0)$ is still increasing as T_c/ω_{1n} is increasing above unity, whereas, for $La_{1.85}Sr_{0.15}CuO_4$ $h_{c2}(0,t^+=0)$ has attained a maximum value near $T_c/\omega_{1n}\approx0.6$ and is slightly decreasing as T_c/ω_{1n} increases further (see Fig. 3). (b) The same results are displayed as in (a), but for impurity parameter $t^+=100$ meV.

In conclusion, we have calculated the upper critical magnetic field $H_{c2}(t^+,T)$ for two model electron-phonon spectral densities within the Eliashberg formulation of the theory. With T_c fixed by choice at 96 K and the Coulomb pseudopotential μ^* set equal to 0.1 we have spanned the region from small values of T_c/ω_{1n} to large values, near and even above one. This was accomplished through a

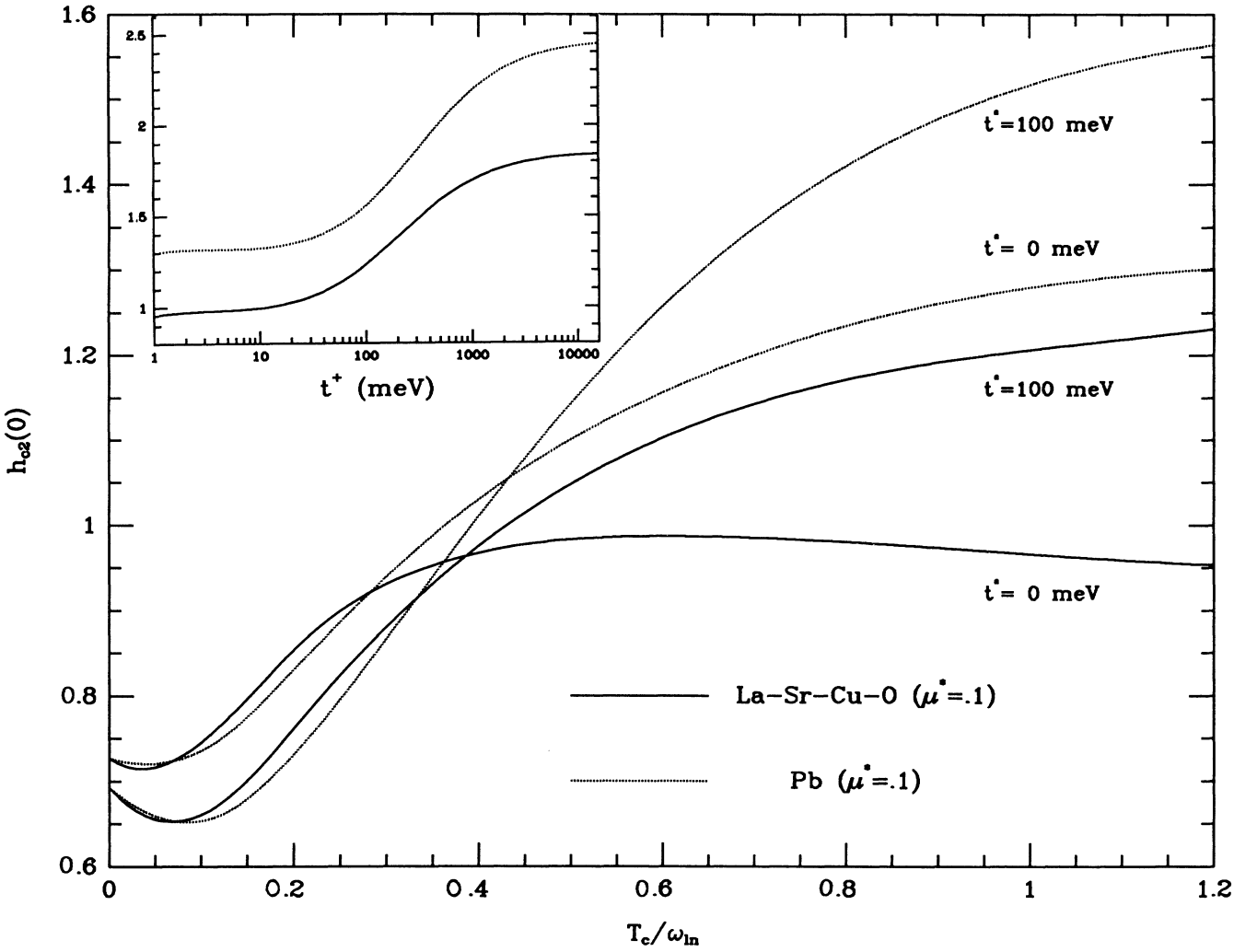

FIG. 3. The reduced upper critical magnetic field at zero temperature, $h_{c2}(0)$, is shown for the scaled spectra as a function of T_c/ω_{1n}, for impurity parameters $t^+=0$ and 100 meV. All curves show a modest decrease below their respective BCS values as T_c/ω_{1n} increases beyond BCS. As T_c/ω_{1n} increases even further, however, the corrections become larger than unity, and show significant deviations from BCS in the very-strong-coupling regime. Note that the difference between $t^+=0$ and 100 meV becomes much more pronounced as T_c/ω_{1n} increases (although a value of T_c/ω_{1n} exists in the intermediate strong-coupling regime for which there is no difference). The striking enhancement of the difference between the clean and dirty limits in the very-strong-coupling limit, is illustrated in the inset, where the dirty limit deviates from the clean limit by roughly 100%, as compared to 5% in the BCS model.

change of the vertical and horizontal scale in the spectral density. When T_c is comparable in size to a typical phonon energy (ω_{1n}), which we call the extreme strong-coupling regime to distinguish this regime from the conventional usage of the words "strong coupling," which applies to the case $T_c/\omega_{1n} \lesssim 0.25$, we find that the strong-coupling corrections $\eta_{H_{c2}}$ to H_{c2} are much smaller than one, in sharp contrast to the conventional case for which $\eta_{H_{c2}}$ is larger than one. Also, the zero-temperature value of the reduced upper critical magnetic field h_{c2} can take on values much larger than in BCS theory, sometimes by a factor as large as, and even larger than, 3 and it displays a very strong impurity dependence. As well, the temperature dependence of $h_{c2}(t^+,t)$ becomes very different from that of a BCS superconductor and exhibits a positive curvature at intermediate values of t.

The above unusual features are not expected to depend qualitatively on the detailed shape of electron-phonon spectral density used. They should apply to any Eliashberg superconductor for which the critical temperature is of the order of the phonon energies. Of course, we have not addressed, in this work, the problem of stability of the lattice, which may well become a factor before the large values of electron-phonon interaction needed in this work are reached. Nor have we been concerned directly with the question of the actual mechanism responsible for the superconductivity in the oxides. Rather we have shown that an electron-phonon superconductor with high T_c will exhibit properties that are quite different from those of a BCS superconductor.

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[1]J. P. Carbotte, Sci. Prog. Oxford **71**, 329 (1987).
[2]W. L. McMillan and J. M. Rowell, in *Superconductivity,* edited by R. D. Parks (Dekker, New York, 1969), Vol. 1, p. 561.
[3]J. M. Daams and J. P. Carbotte, J. Low Temp. Phys. **43**, 263 (1981).
[4]B. Mitrović, E. Schachinger, and J. P. Carbotte, Phys. Rev. B **29**, 6187 (1984).
[5]B. Mitrović and J. P. Carbotte, Phys. Rev. B **33**, 591 (1986).
[6]F. Marsiglio and J. P. Carbotte, Phys. Rev. B **33**, 6141 (1986).
[7]B. Batlogg, R. J. Cava, A. Jayaraman, R. B. van Dover, G. A. Kourouklis, S. Sunshine, D. W. Murphy, L. W. Rupp, H. S. Chen, A. White, K. T. Short, A. M. Mujsce, and E. A. Rietman, Phys. Rev. Lett. **58**, 2333(1987).
[8]P. W. Anderson, Science **235**, 1196 (1987).
[9]A. Alexandrov and J. Ranniger, Phys. Rev. B **23**, 1796 (1981).
[10]B. K. Chakraverty, J. Phys. (Paris) Lett. **40**, L99 (1979).
[11]P. Prelovsek, T. M. Rice, and F. C. Zhang (unpublished).
[12]A. Allender, J. Bray, and J. Bardeen, Phys. Rev. B **7**, 1020 (1973).
[13]C. M. Varma, S. Schmitt-Rink, and E. Abrahams (unpublished).
[14]B. Batlogg, Physica **26B**, 275 (1984).
[15]R. J. Cava, R. B. van Dover, B. Batlogg, and E. A. Rietman, Phys. Rev. Lett. **58**, 408 (1987).
[16]W. K. Kowk, C. W. Crabtree, D. G. Hinks, D. W. Capone II, J. D. Jorgensen, and K. Zhang, Phys. Rev. B **35**, 5343 (1987).
[17]M. E. Hawley, K. F. Gray, D. W. Capone II, and D. G. Hinks, Phys. Rev. B **35**, 7224 (1987).
[18]J. R. Kinley, C. C. Tsuei, S. I. Park, C. C. Chi, J. Rosen, and M. W. Shafer, Phys. Rev. B **35**, 7216 (1987).
[19]B. D. Dunlap, M. V. Nevitt, M. Slaski, T. E. Klippert, Z. Sungaila, A. G. McKale, D. W. Capone II, R. B. Poeppel, and B. K. Flandermeyer, Phys. Rev. B **35**, 7210 (1987).
[20]W. Weber, Phys. Rev. Lett. **58**, 1371 (1987); **58**, 2154(E) (1987).
[21]W. E. Pickett, H. Krakauer, D. A. Papconstantopoulos, and L. L. Boyer, Phys. Rev. B **35**, 7252 (1987).
[22]F. Marsiglio, R. Akis, and J. P. Carbotte (unpublished).
[23]M. Schossmann and E. Schachinger, Phys. Rev. B **33**, 6123 (1986).
[24]P. B. Allen and R. C. Dynes, Phys. Rev. B **12**, 905 (1975).
[25]J. M. Coombes and J. P. Carbotte, J. Low Temp. Phys. **63**, 431 (1986).
[26]D. Rainer and G. Bergmann, J. Low Temp. Phys. **14**, 501 (1974).
[27]E. Helfand and N. R. Werthamer, Phys. Rev. **147**, 288 (1966).
[28]N. R. Werthamer, E. Helfand, and P. Hohenberg, Phys. Rev. **147**, 295 (1966).
[29]T. P. Orlando, E. J. McNiff, S. Foner, and M. R. Beasley, Phys. Rev. B **19**, 4545 (1979).