Modification of the Isotope Effect Due to Pair Breaking

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We have calculated the effect of pair breaking on the isotope-effect coefficient (β) of a superconductor. We find that, as the pair-breaking scattering rate is increased, β also increases in absolute value. Values of β much larger than the canonical value of $\frac{1}{2}$ can easily be achieved even in models where the electron-phonon interaction contributes only a very small amount to the value of the intrinsic critical temperature.

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The isotope effect gives information on the change in the critical temperature T_c with isotopic mass M. Early measurements of $\beta_0 = -d\ln T_{c0}/d\ln M$ helped greatly in understanding the mechanism responsible for superconductivity in conventional materials. In BCS theory, the scale of the critical temperature is set by the Debye energy from which it follows directly that T_c is inversely proportional to the square root of M and therefore $\beta_0=\frac{1}{2}$. The observed small and even negative values of β_0 in some very-low-T_c materials,[1] while initially controversial, can be explained[1-3] as due to the electron-phonon interaction provided Coulomb repulsions are additionally introduced. In this case, β_0 changes sign when the Coulomb pseudopotential μ^* is significant compared with the electron-phonon interaction. This only occurs when T_c itself is small in magnitude.

In the high-T_c copper-oxide superconductors, the small value of the oxygen isotope effect β_0 observed in $La_{1.85}Sr_{0.15}CuO_4$ and near zero value in $YBa_2Cu_3O_7$ (Refs. 4–16) was taken by some to be convincing evidence against the electron-phonon interaction at least as the dominant mechanism in these materials. The large energy scale for T_c also points to an electronic mechanism. Recent experiments have revealed a much richer and complex situation. Crawford *et al.*[17] have found an amazingly strong variation of β_0 with strontium concentration (x) in $La_{2-x}Sr_xCuO_4$ with the value of β_0 larger than $\frac{1}{2}$ for some values of x. Other experiments by Franck *et al.*[18] in $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ as a function of praseodymium (Pr) concentration (x) show a rapidly increasing β_0 (with increasing x) towards values around $\frac{1}{2}$.

There are several known mechanisms whereby β_0 can be increased above the canonical value of $\frac{1}{2}$. The energy dependence in the electronic density of states $[N(\epsilon)]$, on a scale of importance for superconductivity, can reduce or increase β_0 depending on the position of the chemical potential with respect to the structure in $N(\epsilon)$.[18-22] In particular, Tsuei *et al.*[22] have used a Van Hove-singularity model to explain the La-Sr-Cu-O data. Anharmonic effects[23] associated with a soft mode near a structural phase transition can also affect β_0. In this paper, we consider the effect of pair breaking due, for example, to paramagnetic impurities.

To compute the critical temperature of a superconductor with electron-phonon interaction given by the spectral density $\alpha^2F(\omega)$, Coulomb repulsions described by a pseudopotential μ^*, and magnetic impurities characterized by a scattering time τ_p, we need use only the linearized Eliashberg equations. They involve a frequency-dependent gap $\Delta(i\omega_n)$ and renormalization function $Z(i\omega_n)$ written on the imaginary frequency axis,[24,25] with $i\omega_n = i\pi T(2n-1)$, $n=0,\pm1,\pm2,\ldots$:

$$\Delta(i\omega_n)Z(i\omega_n)=\pi T\sum_m\left([\lambda(m-n)-\mu^*\theta(\omega_c-|\omega_m|)]\frac{\Delta(i\omega_m)}{|\omega_m|}\right)-\frac{\pi t\Delta(i\omega_n)}{|\omega_n|} \tag{1}$$

and

$$Z(i\omega_n)\omega_n=\omega_n+\pi T\sum_m\lambda(m-n)\operatorname{sgn}(\omega_m)+\pi t\operatorname{sgn}(\omega_n)\,. \tag{2}$$

In Eqs. (1) and (2), ω_c is a cutoff on the Matsubara sum over ω_m, t is a pair-breaking amplitude related to τ_p by $t=1/2\pi\tau_p$, and $\lambda(m-n)$ is given by

$$\lambda(m-n)=2\int_0^\infty\frac{\Omega\alpha^2F(\Omega)d\Omega}{\Omega^2+(\omega_n-\omega_m)^2}\,. \tag{3}$$

If we start by neglecting pair-breaking effects (i.e., take $\tau_p\to\infty$), approximate $\lambda(m-n)$ in (1) and (2) by a constant value $\lambda\equiv\lambda(0)$ equal to the mass renormalization due to the electron-phonon interaction, and cut off all frequency sums at ω_c, we obtain the simple, but approximate, equation for the critical temperature T_{c0}:

$$T_{c0}=1.13\omega_c e^{-(1+\lambda)/(\lambda-\mu^*)}, \tag{4}$$

where the subscript zero denotes $\tau_p \to \infty$. If we take ω_c to be a phonon energy which is inversely proportional to the square root of the ionic mass M, and note that μ^* is related to a pure electronic screened Coulomb potential μ by $\mu^*=\mu/[1+\mu\ln(\epsilon_F/\omega_c)]$, where ϵ_F is the Fermi energy, we arrive at the equation

$$\beta_0=-\frac{d\ln T_{c0}}{d\ln M}=\frac{1}{2}\left\{1-\frac{1+\lambda}{(\lambda-\mu^*)^2}\mu^{*2}\right\} \tag{5}$$

for the isotope coefficient β_0. For $\mu^*=0$, $\beta_0=\frac{1}{2}$, but for finite μ^*, β_0 can be smaller than $\frac{1}{2}$ and even zero or negative when λ is small.

If, as a start, we use the same model for $\lambda(m-n)$ as just described but now include pair breaking, we arrive at a new equation for T_c which involves the critical temperature T_{c0} (without pair breaking). It is[26,27]

$$\ln(T_{c0}/T_c)=\psi(\rho+\tfrac{1}{2})-\psi(\tfrac{1}{2}), \tag{6}$$

with $\psi(x)$ the digamma function and $\rho\equiv 1/2\pi\tau_p(1+\lambda)k_BT_c$, where k_B is the Boltzmann constant. Differentiation of Eq. (6) to obtain $\beta\equiv -d\ln T_c/d\ln M$ gives

$$\beta=\frac{\beta_0}{1-\rho\psi'(\rho+\frac{1}{2})}, \tag{7}$$

from which we conclude that $|\beta|\geq|\beta_0|$ and so pair breaking increases the absolute value of the isotope effect. As T_c is reduced due to pair breaking, a small change in pairing potential is seen to have a relatively strong effect on the value of T_c. For small pair breaking, i.e., $\rho\to 0$ or $\tau_p\to\infty$,

$$\beta=\frac{\beta_0}{1-0.7\tau_p^c/\tau_p}, \tag{8}$$

and for $\tau_p\to\tau_p^c$, where τ_p^c is the critical concentration scattering time,

$$\beta=0.236\beta_0(T_c^0/T_c)^2. \tag{9}$$

As $T_c\to 0$, $\beta\to\pm\infty$ depending on the sign of the underlying value of β_0—the isotope coefficient without pair breaking. It is clear then that large values of β, above $\frac{1}{2}$, can be obtained in a BCS superconductor when some pair-breaking interaction is introduced in the system. This is shown in Fig. 1 where we plot T_c/T_{c0} (solid curve) as a function of normalized impurity concentration n/n_c as well as β (dashed curve).

To get more accurate results for T_c and, consequently, for β, it is, of course, necessary to solve numerically the full Eliashberg equations (1) and (2) rather than rely on the BCS-type equation (6). To illustrate the main point of this paper, it will be sufficient to take a model spectral density for $\alpha^2F(\omega)$ consisting of an Einstein spectrum at

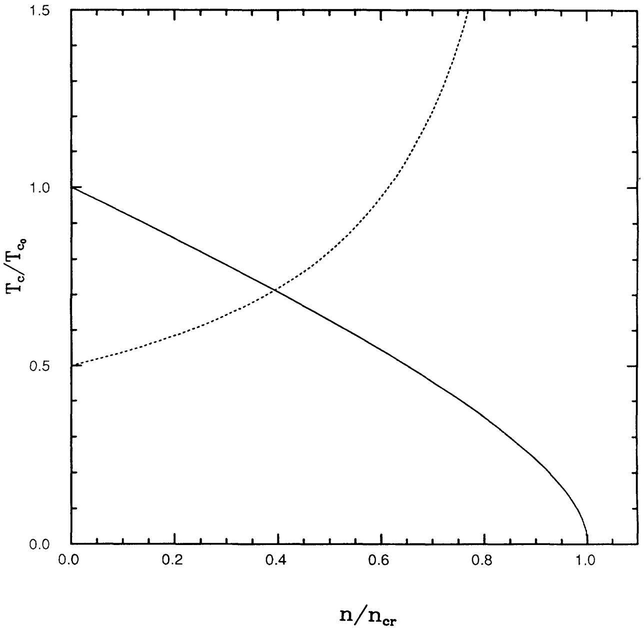

FIG. 1. T_c/T_{c0} as a function of normalized impurity concentration n/n_c (solid line) and the isotope-effect coefficient β (dashed line).

the energy ω_E, of weight A, i.e., take $\alpha^2F(\omega)=A\delta(\omega-\omega_E)$. For definiteness, consider the specific case of $YBa_2Cu_3O_7$ for which it is well known that the total isotope effect is small, less than 0.1 and probably of order $\beta_0=0.05$. For a T_{c0} value of 96 K, such a small isotope-effect coefficient cannot be obtained in a pure phonon model with μ^* of order 0.1 (the canonical value in conventional superconductors). On the other hand, if we are willing to consider anomalously large values of $\mu^*(\omega_E)$ of order 0.5, such a small value of β_0 can be accommodated provided the coupling is predominantly to very-high-energy phonons with $\omega_E\sim 80$ meV.[28] A more attractive model, which we shall adopt here, is to assume that in $YBa_2Cu_3O_7$, only a small fraction of T_{c0} is due to the electron-phonon interaction and that some other electronic mechanism is involved. If we take this second mechanism to be due to a very-high-energy boson exchange process of some kind, we can treat it approximately as a BCS constant pairing potential, which acts oppositely to the Coulomb pseudopotential μ^* and overtakes it. Alternately, we could introduce a second piece to the spectral density centered around some high energy compared with T_c. That is, we can use an effective value of μ^* in Eq. (1) with $\mu^*_{\rm eff}$ negative. The effective $\mu^*_{\rm eff}$ then leads, on its own, to a finite value of critical temperature when the electron-phonon contribution is neglected. We will call this a joint mechanism. In our simplified model, we can rewrite Eqs. (1) and (2) in a dimensionless form by introducing normalized quantities $\bar{Q}$

$\equiv Q/\omega_E$ for any energy variable Q. We have

$$\bar{\Delta}(i\bar{\omega}_n)Z(i\bar{\omega}_n)=\pi\bar{T}\sum_m\left[\frac{\lambda}{1+(\bar{\omega}_m-\bar{\omega}_n)^2}-\mu^*_{\rm eff}\right]\frac{\bar{\Delta}(i\bar{\omega}_m)}{|\bar{\omega}_m|}-\pi\bar{t}\frac{\bar{\Delta}(i\bar{\omega}_n)}{|\bar{\omega}_n|}, \tag{10}$$

$$\bar{\omega}_nZ(i\bar{\omega}_n)=\bar{\omega}_n+\pi\bar{T}\sum_m\frac{\lambda}{1+(\bar{\omega}_m-\bar{\omega}_n)^2}\text{sgn}(\bar{\omega}_m)+\pi\bar{t}\,\text{sgn}(\bar{\omega}_n), \tag{11}$$

for which we can conclude immediately that $T_c=\omega_E\mathcal{F}(\lambda,\mu^*_{\rm eff},\bar{t})$, where $\mathcal{F}$ is a universal functional form given by Eqs. (10) and (11). If pair-breaking effects are ignored the pure material T_{c0} is given by $T_{c0}=\omega_E\mathcal{F}(\lambda,\mu^*_{\rm eff},0)$, so that the ratio T_{c0}/ω_E depends only on two parameters, namely, λ and $\mu^*_{\rm eff}$. If, in addition to requiring $T_{c0}=96$ K, we insist on a β_0 value of 0.075, both λ and $\mu^*_{\rm eff}$ are fixed for a given choice of ω_E. After determining λ and $\mu^*_{\rm eff}$ in this way, we can turn on the pair breaking $\bar{t}$. This reduces the critical temperature from T_{c0} to T_c. In Fig. 2, we plot β as a function of T_c for various choices of phonon energy ω_E, namely, 40.0 meV (long-dash-dotted curve), 30.0 meV (dash-dotted curve), 20.0 meV (dashed curve), 10.0 meV (dotted curve), and 5.0 meV (solid curve). All curves go through the point $\beta_0=0.075$ at $T_c=T_{c0}=96.0$ K. As the amount of pair breaking is increased, the value of the critical temperature is reduced and β increases. We see that the increase is steepest when the coupling is to lower-frequency modes, i.e., small ω_E. For $\omega_E=5.0$ meV, this corresponds to a rather large value of electron-phonon mass renormalization $\lambda=2.2$. On the other hand, and we need to emphasize it, a very significant increase towards values greater than 0.5 can also be obtained in a model with small λ, for example, $\lambda=0.095$, for $\omega_E=30.0$ meV. In Fig. 2, the solid and open circles are the data points given by Franck *et al.*[18] We see that they fall closest to the curve with $\omega_E=20.0$ meV for which $\lambda=0.17$. In this case, little of the total critical temperature of 96 K is due to the electron-phonon interaction. While some authors think that the reduction in T_c observed in $Y_{1-x}Pr_xBa_2Cu_3O_7$ as a function of increasing x can be modeled by a pair-breaking mechanism, others believe it is due to electronic changes.[29-31] Thus, the results found by Franck *et al.*[18] may require a different explanation, and so, we have not tried to get a best fit to their data. All we really want to do here is to point out that pair-breaking effects, when they exist, can greatly increase the absolute value of the isotope-effect coefficient as T_c is reduced towards zero and that the theory developed here can give results similar to those observed without requiring a large phonon contribution to T_{c0}.

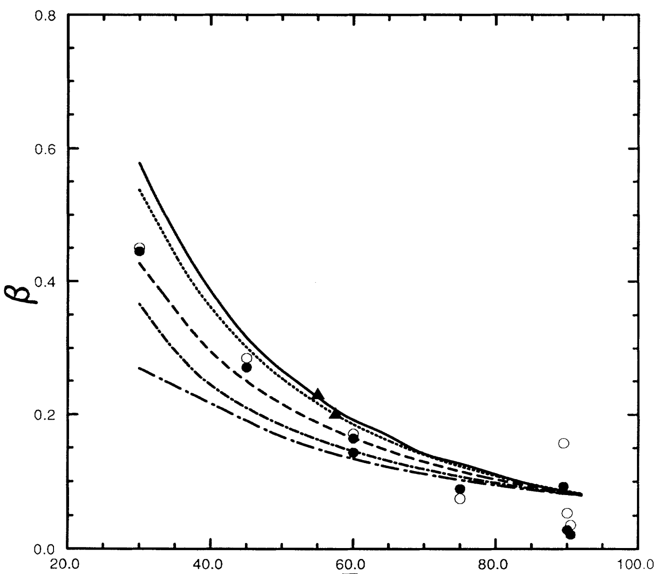

FIG. 2. The isotope-effect coefficient $\beta\equiv -d\ln T_c/d\ln M$ for a system with pair breaking as a function of sample critical temperature. The amount of pair breaking does not appear explicitly as the reduced value of critical temperature is used on the horizontal axis. As the value of T_c decreases, β increases rapidly above its unperturbed value of $\beta_0=0.075$. The rate of increase depends on the choice of phonon Einstein frequency ω_E in the model spectral density. The solid curve is for $\omega_E=5.0$ meV; dotted curve, 10.0 meV; dashed curve, 20.0 meV; dash-dotted curve 30.0 meV; and long-dash-dotted curve, 40.0 meV. The open and solid circles are data from Franck *et al.*

In conclusion, we have shown that pair-breaking effects in a superconductor can, in some cases, drastically modify the value of the isotope coefficient β. Values of β larger than $\frac{1}{2}$ are easily obtained even in models where only a very small part of the intrinsic T_{c0} is due to a phonon mechanism. Thus, large values of β cannot be interpreted unambiguously as due to a large electron-phonon interaction. Experiments in conventional superconductors with paramagnetic impurities should be carried out to verify our theoretical predictions.

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[1] J. W. Garland, Jr., Phys. Rev. Lett. **11**, 114 (1963).
[2] J. C. Swihart, IBM J. Res. Dev. **6**, 14 (1962).
[3] P. B. Allen, Nature (London) **335**, 396 (1988).

[4]T. A. Faltens, W. K. Ham, S. W. Keller, K. J. Leary, J. N. Michaels, A. M. Stacy, H. zur Loye, D. E. Morris, T. W. Barbee, III, L. C. Bourne, M. L. Cohen, S. Hoen, and A. Zettl, Phys. Rev. Lett. **59**, 915 (1987).

[5]L. C. Bourne, S. Hoen, M. F. Crommie, W. N. Creger, A. Zettl, Marvin L. Cohen, L. Bernardez, J. Kinney, and D. E. Morris, Solid State Commun. **67**, 707 (1988).

[6]H. C. zur Loye, K. J. Leary, S. W. Keller, W. K. Ham, T. A. Faltens, J. N. Michaels, and A. M. Stacy, Science **238**, 1558 (1987).

[7]B. Batlogg, R. J. Cava, A. Jagaraman, R. B. van Dover, G. A. Kourouklis, C. Sunshine, D. W. Murphy, L. W. Rupp, H. S. Chen, A. White, K. T. Short, A. M. Majsce, and E. A. Rietman, Phys. Rev. Lett. **58**, 2333 (1987).

[8]L. C. Bourne, M. F. Crommie, A. Zettl, H. C. zur Loye, S. W. Keller, K. L. Leary, A. M. Stacy, K. J. Chang, M. L. Cohen, and D. E. Morris, Phys. Rev. Lett. **58**, 2337 (1987).

[9]K. J. Leary, H. C. zur Loye, S. W. Keller, T. A. Faltens, W. K. Ham, J. N. Michaels, and A. Stacy, Phys. Rev. Lett. **59**, 1236 (1987).

[10]E. Morris, R. M. Kuroda, A. G. Markelz, J. H. Nickel, and J. T. T. Wei, Phys. Rev. B **37**, 5936 (1988).

[11]E. L. Benitez, J. J. Lin, S. J. Poon, W. E. Farneth, M. K. Crawford, and E. M. McCarron, Phys. Rev. B **38**, 5025 (1988).

[12]P. J. Yvon, R. B. Schwarz, C. B. Pierce, L. Bernardez, A. Conners, and R. Meisenheimer, Phys. Rev. B **39**, 6690 (1989).

[13]S. Hoen, W. N. Creager, L. C. Bourne, M. F. Crommie, T. W. Barbee, III, M. L. Cohen, and A. Zettl, Phys. Rev. B **39**, 2269 (1989).

[14]J. P. Franck *et al.*, in *Proceedings of the International Conference on Materials and Mechanisms of Superconductivity and High Temperature Superconductors, Stanford, 1989* [Physica (Amsterdam) **162-164C**, 753 (1989)].

[15]L. C. Bourne, A. Zettl, T. W. Barbee, III, and M. L. Cohen, Phys. Rev. B **36**, 3990 (1987).

[16]H. Katayama-Yoshida *et al.*, Physica (Amsterdam) **156C**, 481 (1988).

[17]M. K. Crawford, M. N. Kunchur, W. E. Farneth, E. M. McCarron, III, and S. J. Poon, Phys. Rev. B **41**, 283 (1990).

[18]J. P. Franck, J. Jung, M. A. K. Mohamed, S. Gygax, and G. I. Sproule, in Proceedings of the Nineteenth International Conference on Low-Temperature Physics, LT19, 1990 (to be published).

[19]S. G. Lie and J. P. Carbotte, Solid State Commun. **34**, 599 (1980).

[20]J. Labbé and J. Bok, Europhys. Lett. **3**, 1225 (1987).

[21]E. Schachinger, M. G. Greeson, and J. P. Carbotte, Phys. Rev. B **42**, 406 (1990).

[22]C. C. Tsuei, D. M. Newns, C. C. Chi, and P. C. Pattnaik, Phys. Rev. Lett. **65**, 2724 (1990).

[23]N. M. Plakida, Phys. Scr. **T29**, 77 (1989).

[24]E. Schachinger, J. M. Daams, and J. P. Carbotte, Phys. Rev. B **22**, 3184 (1980).

[25]M. Ashraf and J. P. Carbotte, J. Low Temp. Phys. **58**, 451 (1985).

[26]A. A. Abrikosov and L. P. Gorkov, Zh. Eksp. Teor. Fiz. **39**, 1781 (1961) [Sov. Phys. JETP **12**, 1243 (1961)].

[27]S. S. Skalski, O. Betbeder-Matibet, and P. R. Weiss, Phys. Rev. **136**, A1500 (1964).

[28]R. Akis and J. P. Carbotte, Phys. Rev. B **41**, 11 661 (1990).

[29]C. Infante *et al.*, Physica (Amsterdam) **16C**, 640 (1990).

[30]J. J. Neumeier *et al.*, Phys. Rev. Lett. **63**, 2516 (1989).

[31]G. Y. Guo *et al.*, Phys. Rev. B **41**, 6372 (1990).