Isotope Effect in d-Wave Superconductors

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Based on recently proposed antiferromagnetic spin fluctuation exchange models for $d_{x^2-y^2}$ superconductors, we show that coupling to harmonic phonons *cannot* account for the observed isotope effect in the cuprate high-T_c materials, whereas coupling to strong anharmonic multiple-well lattice tunneling modes *can*. Our results thus point towards a strongly enhanced *effective* electron-phonon coupling and a possible breakdown of Migdal-Eliashberg theory in the cuprates.

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A growing, but not uncontroverted body of experimental evidence currently suggests that $YBa_2Cu_3O_7$ and, possibly, other cuprates are $d_{x^2-y^2}$ superconductors [1,2]. On the theoretical side, d-wave superconductivity would probably rule out *conventional* phonon-mediated pairing [3], while supporting an electronic pairing mechanism, most prominently antiferromagnetic (AF) spin fluctuation exchange models [2,4,5]. Yet, except for certain "optimal" doping concentrations, many cuprates, including $YBa_2Cu_3O_7$, exhibit a quite noticeable isotope effect [6], the classical hallmark of the superconducting electrons' coupling to the lattice vibrational degrees of freedom.

Starting from recent diagrammatic AF spin fluctuation models [2,4,5], we will show here that the observed *order of magnitude* of the isotope exponent [6] $\alpha \sim 1$ does, in fact, imply an extraordinarily large electron-phonon (EP) coupling parameter $\lambda \gg 1$. For coupling to harmonic phonon systems, such coupling strengths would be well in excess of upper limits imposed by structural stability arguments and, independently, by the observed T_c and normal-state transport scattering rates. However, the required λ's *are* consistent with a coupling to local, large-amplitude lattice tunneling excitations, arising in a strongly anharmonic multiple-well lattice potential. Barring experimental problems in the isotope measurements [6], our results thus point towards a very strong enhancement of the *effective* EP coupling, strong enough, in fact, to cause a breakdown of the very approximation upon which the conventional Migdal diagrammatic theory is based. As such, our results point towards the possibility that lattice vibrational degrees of freedom play a central role in the low-energy electronic properties of the cuprates.

We start from the linearized Migdal-Eliashberg equations [3] for the self-energy Σ and pair wave function Φ at wave vector k and Matsubara frequency $i\nu = (2m + 1)\pi iT$ (in energy units) at the transition temperature $T = T_c$.

$$\Sigma(k', i\nu') = -\frac{T}{N}\sum_{k,i\nu} V_-(k' - k, i\nu' - i\nu)G(k, i\nu),$$

$$\Phi(k', i\nu') = -\frac{T}{N}\sum_{k,i\nu} V_+(k' - k, i\nu' - i\nu) \times |G(k, i\nu)|^2\Phi(k, i\nu), \quad (1)$$

Here $k \equiv (k_x, k_y)$ is summed over an $N \equiv L \times L$ grid covering the Brillouin zone of a two-dimensional (2D) square lattice. The single-electron Green function G is obtained self-consistently from $G(k, i\nu) = [i\nu - \epsilon_k - \Sigma(k, i\nu)]^{-1}$, assuming a single, 2D electron band $\epsilon_k = -2t_1[\cos(k_x) + \cos(k_y)] - 4t_2\cos(k_x)\cos(k_y) - \mu$ with chemical potential μ and first and second neighbor hopping t_1 (= 250 meV) [5(a)] and t_2, respectively. For singlet d-wave pairing, the effective electron-electron interaction potentials in Eqs. (1) are $V_\pm(q, i\omega) = \pm g_s^2\chi_s(q, i\omega) + V_p(q, i\omega)$. This includes a spin-fluctuation term with coupling constant g_s^2 and dynamical spin susceptibility $\chi_s(q, i\omega)$ [2,4,5], and a phonon term of the general form $V_p(q, i\omega) = -U_p f(q)\Omega_q^2/(\omega^2 + \Omega_q^2)$, assuming a single phonon branch with an energy dispersion Ω_q and a form factor $f(q)$, normalized so that $U_p = -N^{-1}\sum_q V_p(q, 0)$. The ratio $\bar{\lambda} \equiv U_p/B$ provides then a rough estimate for the dimensionless Eliashberg parameter λ [3], averaged over the electronic bandwidth $B = 8t_1$. We emphasize that the isotopic mass (M) dependence of V_p enters *only* via Ω_q, but *not* via U_p [3].

Global structural stability of the undoped, $\frac{1}{2}$-filled-band cuprate parent compounds requires the phonon-mediated on-site attraction U_p to be less than the Coulombic on-site repulsion, of the order of the on-site Hubbard U parameter [7]. If $U_p > U$, the conduction electrons would form local pairs (bipolarons), rather than local magnetic moments. At $\frac{1}{2}$ filling, such local pairs would undergo ordering in a charge superlattice [8], rather than forming the AF spin superlattice observed [1,2] in the undoped cuprates. This transition from local moment AF spin density order to local pair charge density order has been established in recent studies of Holstein-Hubbard and related EP models [8].

Local structural stability in the doped systems, i.e., stability of the dopant induced carriers against polaronic self-localization [8], requires an even more stringent upper limit $U_p < U_p^{(\mathrm{loc})} < U$. If $U_p > U_p^{(\mathrm{loc})}$, polaron formation would destroy (or, at the very least, strongly renormalize and broaden) the delocalized quasiparticle state whose existence is essential for the Migdal-Eliashberg

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approach. Holstein-Hubbard estimates [8(a)] give $U_p^{(\mathrm{loc})} \sim (2-3)t_1$ for a Hubbard $U \sim (8-12)t_1$ in the cuprates [7], or, equivalently, $\overline{\lambda}^{(\mathrm{loc})} \equiv U_p^{(\mathrm{loc})}/B \sim 0.25-0.4$ and $\overline{\lambda}^{(\mathrm{glob})} \equiv U/B \sim 1-1.5$.

Our results for the $d_{x^2-y^2}$ superconducting transition temperature T_c and its isotope exponent $\alpha \equiv -d\log(T_c)/d\log(M)$ are illustrated by Figs. 1 and 2 for the case of an Einstein phonon model with $\Omega_q \equiv \Omega_0 = \text{const}$, local coupling $f(q) \equiv 1$, and assumed isotopic mass dependence $\Omega_0 \propto M^{-1/2}$. Such a model could roughly represent, for example, the local coupling(s) to high-energy optical modes which are dominated primarily by the lightest atomic species, i.e., oxygen, in the cuprates with $\Omega_0 \lesssim 100$ meV [9]. The χ_s in the Millis-Monien-Pines (MMP) model [5], used in Figs. 1 and 2, is given by

$$\mathrm{Im}\chi_s(q,\omega + i0^+) = \mathrm{Im}[\chi_Q/(1 + \xi^2|q - Q|^2 - i\omega/\omega_s)]\theta(\omega_c - |\omega|), \tag{2}$$

with $Q + (\pi,\pi)$ for $q_x, q_y \geq 0$. *Except for values explicitly stated* in Figs. 1 and 2, we have used $U_p = 1$ eV, $\Omega_0 = 50$ meV, and the band and T-independent spin fluctuation parameters from Table II of Ref. [5(a)], hereafter referred to as MP-II, for a hole doping concentration $x \equiv 1 - n = 25\%$. Equations (1) are solved accurately, without further approximations, by fast Fourier transform techniques [10].

Figure 1 shows T_c and α as functions of the electron concentration n. To model roughly the doping dependence of χ_s, $\omega_s(x)$, and $\xi(x)$ are varied with $x \equiv 1 - n$, like [5(a)] $\omega_s \propto \xi^{-2} \propto x$, and attain their MP-II values at $x = 25\%$. Figure 2 shows α vs T_c for the same model at fixed $x = 25\%$, with T_c varied by increasing U_p from 0 to 2 eV. Figure 2 also shows results for coupling to anharmonic phonon and a Hubbard-based spin fluctuation model, discussed below. The figures clearly demonstrate the *very general result* that coupling to harmonic phonons will substantially suppress the d-wave T_c without, however, causing a large enough isotope effect to account for the *order of magnitude* of the experimental results, illustrated here by the (Y,Pr)$Ba_2Cu_3O_7$ data [6(a)].

Note here that the MP-II spin fluctuation parameters, with $g_s = 640$ meV, were carefully tuned [5(a)] to reproduce the observed NMR data, resistivity, *and* $T_c \cong 90$ K in $YBa_2Cu_3O_7$, *excluding* the effect of phonons. If we now *include* phonons, with a strength of only $U_p \cong 1$ eV (i.e., $\overline{\lambda} \cong 0.5$), say, we need to roughly double g_s^2 (see Fig. 1) in order to maintain a $T_c \cong 90$ K. Thus we will also double, roughly, the normal-state model resistivity from spin fluctuation scattering (not even counting phonon scattering), which already seriously compromises the agreement with the experimental resistivity data. So, in the MMP model [5], Eq. (2), the observed *maximum* $T_c \cong 90$ K, combined with the resistivity data for $YBa_2Cu_3O_7$, will again limit U_p to $U_p \lesssim 1$ eV.

We have generated overwhelming numerical evidence that the foregoing results are generic and robust, (i) by systematically varying, over wide ranges, all spin fluctuation, electron-phonon and band parameters, (ii) by using different types of form factors $f(q)$, different types of and/or multiple q-dependent phonon branches (including, e.g., 3D acoustic phonons), (iii) by modifying the AF spin fluctuation model (introducing, e.g., spin gap effects into the MMP model or using other proposed types of spectral models [2,4(a),(b),11], and (iv) by adding impurity scattering [11,12]. While such modifications of the model may substantially affect T_c, they do not increase the overall magnitude of α significantly. We thus conclude that the

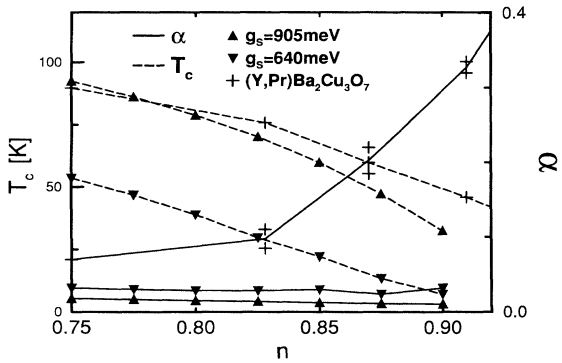

FIG. 1. $d_{x^2-y^2}$ transition temperature T_c and isotope exponent α vs electron concentration n for coupling to Einstein phonons in the MMP model, Eq. (2), with $\omega_s \propto \xi^{-2} \propto 1 - n$. Also shown are the T_c and oxygen isotope data for (Y,Pr)$Ba_2Cu_3O_7$, from Ref. [6(a)], with $n = 0.75$ assumed at the maximum T_c.

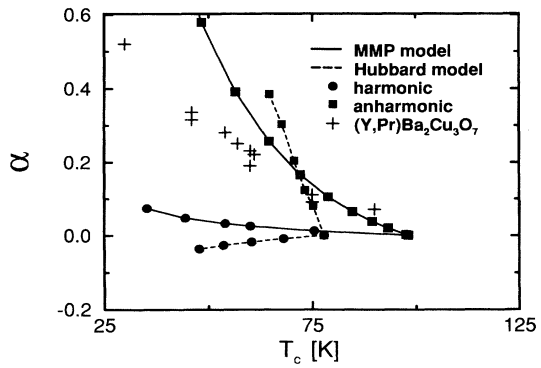

FIG. 2. Isotope exponent α vs $d_{x^2-y^2}$ transition temperature T_c in the Hubbard model, Ref. [4(a),(b)], with $U/t_1 = 6$, $t_1 = 250$ meV, $t_2 = 0$, and $n \cong 0.86$, and in the MMP model, Eq. (2), with MP-II parameters, for coupling to *harmonic* Einstein phonons, and for coupling to *anharmonic* tunneling excitations, with EP parameters given in text. Also shown are the oxygen-isotope vs T_c data for (Y,Pr)$Ba_2Cu_3O_7$, from Ref. [6(a)].

observed magnitudes of T_c *and* α in the cuprates cannot possibly be explained by a conventional Migdal-Eliashberg treatment of the conduction electrons' coupling to AF spin fluctuations and harmonic phonons [13].

To understand why harmonic phonon exchange V_p causes a *large* T_c suppression, but only a *small* isotope effect, consider first its effect on the self-energy Σ in a simple McMillan-type analysis [3,4], generalized to the case of d-wave pairing. In the real-frequency domain, $i\nu \rightarrow \nu + i0^+$, V_p contributes, via V_-, to the quasiparticle mass enhancement $Z = 1 - \partial_\nu \mathrm{Re}\Sigma|_{\nu=0} \equiv 1 + Z_s + Z_p$ and to the quasiparticle damping $\mathrm{Im}\Sigma \equiv \mathrm{Im}\Sigma_s + \mathrm{Im}\Sigma_p$, with "$s$" and "$p$" denoting the spin fluctuation and phonon contribution, respectively. Now $Z_p \cong \lambda \sim U_p/B$ is independent of isotropic mass and, in McMillan theory, enters into T_c roughly as [3,4] $T_c \sim \langle\omega\rangle \exp(-Z/\lambda_d)$ with a bare d-wave pairing strength [3,4] λ_d and an appropriate average energy $\langle\omega\rangle$ of the V_+ spectrum [3,4]. Thus Z_p causes a large *isotope-independent* reduction in T_c, by roughly a factor $\exp(-\lambda/\lambda_d)$, which is quite consistent with our numerical results for the U_p dependence of T_c. On the other hand, the phonon contribution to the quasiparticle damping [3], $|\mathrm{Im}\Sigma_p| \sim \lambda \max_q(\Omega_q)$, is comparable to the phonon energy scale Ω_q and thus much smaller than the typically electronic magnitude of Σ_s. Therefore $\mathrm{Im}\Sigma_p$ causes an additional T_c suppression, by pair breaking, which *does* depend on isotopic mass, but is much smaller than the "intrinsic" T_c suppression, due to Σ_s, thus causing only a small α [13]. Treating the phonons solely in terms of an Abrikosov-Gorkov pair-breaking theory [14] (*without* inclusion of the mass enhancement effect) is seriously flawed, since it severely underestimates the T_c suppression caused by the phonons, for a given magnitude of the isotope exponent. In this regard, the effect of phonons is quite different from that of impurities [11,12].

In the case of a q-dependent $V_p(q, i\omega)$, V_p may also contribute directly, via V_+, to the d-wave pairing potential. In McMillan theory [3,4], the dominant effect is again an *isotope-independent* phonon contribution $\lambda_{d,p}$ to the bare d-wave pairing strength $\lambda_d \equiv \lambda_{d,s} + \lambda_{d,p}$ entering into $T_c \sim \langle\omega\rangle \exp(-Z/\lambda_d)$. The *isotope-dependent* contribution $\langle\omega\rangle_p$ to $\langle\omega\rangle \equiv \langle\omega\rangle_s + \langle\omega\rangle_p$ is again of order of the phonon energy scale, i.e., small compared to the spin fluctuation contribution $\langle\omega\rangle_s$, thus, again, giving only a small contribution to α. A substantially larger α and smaller T_c suppression *may* be obtained in the case of *phonon-mediated* d-wave pairing, i.e., if $V_p(q, i\omega)$ is attractive in the $d_{x^2-y^2}$-pairing channel [12], with large enough $\lambda_{d,p} > 0$. Because of the expected cancellation effects in the presence of multiple phonon branches, this scenario seems unlikely, but should nevertheless be explored by first-principles $\lambda_{d,p}$ calculations.

Phonon renormalizations of the spin fluctuation exchange potential $g_s^2\chi_s$ also do not provide a viable mechanism for obtaining a significantly larger $|\alpha|$, as illustrated by the Hubbard model results in Fig. 2. Here both Σ and $g_s^2\chi_s$, as well as a less important charge fluctuation term $g_s^2\chi_c$ in $V_\pm$, Eq. (1), were calculated self-consistently, from $G(k, i\nu)$, in the fluctuation exchange approximation to the 2D Hubbard model [2,4(a)], using numerical renormalization group techniques [4(b)]. The phonon term V_p in Eq. (1) thus explicitly modifies χ_s and χ_c, via Σ. The result, $|\alpha| \ll 1$, is explained by noting that, again, the dominant effect of V_p on χ_s and χ_c comes from the *isotope-independent* mass renormalization Z_p.

Figure 2 also shows results obtained for coupling to an anharmonic phonon system, consisting of independent, local anharmonic oscillators with one lattice displacement degree of freedom u_j per lattice site j. Each u_j is subject to a local Hamiltonian $h_j \equiv (-\hbar^2/2M)\partial^2/\partial u_j^2 + w(u_j)$ with a double-well potential $w(u_j) = 4\Delta_B[2(u_j/d)^4 - (u_j/d)^2]$. Here d is the distance between the two local w minima. The chosen tunneling barrier height $\Delta_B = 26.89$ meV and rescaled atomic mass $\overline{M} \equiv Md^2/\hbar^2 = 0.3555\ \mathrm{meV}^{-1}$ give a double-well tunneling splitting $\Omega_t \equiv E_1 - E_0 \cong 7.25$ meV and a quasiharmonic (single-well "phonon") excitation energy $\Omega_h \equiv E_2 - E_0 \cong 35.5$ meV, where E_ℓ denotes the ℓth excited state energy of h_j. The q-independent V_p was calculated as [15] $V_p(i\omega) = -C^2 \int_0^\beta d\tau \langle u_j(\tau)u_j(0)\rangle \exp(i\omega\tau)$, with the local correlation function $\langle u_j(\tau)u_j(0)\rangle$ obtained from numerical solutions for the eigenstates of h_j. T_c in Fig. 2 was varied by raising the rescaled coupling $\overline{C} \equiv Cd$ from 0 to [8(a),15(a)] of 200 and 300 meV for the MMP [5] [Eq. (2)] and Hubbard [4] model, respectively. In contrast to harmonic phonon models, $\overline{\lambda} \equiv -V_p(0)/B \sim \overline{C}^2/B\Omega_t$, at $T = 0$ [8(a),15(a)], is strongly enhanced here and strongly isotope dependent, since the tunneling splitting Ω_t varies exponentially with the isotopic mass M. For given T_c, this model thus leads to a much larger α and, for given α, requires a much smaller bare coupling C than the harmonic phonon models discussed above. These larger values of α can be achieved without substantially increasing the model's dc resistivity, since the phonon contribution to the quasiparticle damping, $\mathrm{Im}\Sigma$, is, again, small compared to the spin fluctuation contribution. When studied as a function of doping concentration x, with ξ and/or ω_s varied with x as in Fig. 2, the isotope exponent in the anharmonic model typically increases in magnitude with decreasing T_c as one moves away from the optimal doping. This behavior appears to be generic to the model and is in qualitative agreement with the data in the cuprates.

Substantial experimental evidence for a strongly anharmonic multiple-well lattice dynamics in the cuprates exists [9]. Polaron formation is a possible mechanism for generating anharmonic tunneling modes of sufficient effective coupling strength to the conduction electron system within physically reasonable parameter limits [8]. The foregoing treatment extends the Migdal-Eliashberg approach to model the two essential features

of such a polaronic system, namely, multiple-well lattice anharmonicity and isotope-dependent electron mass renormalization [8,15(a)]. However, as polaronic instabilities may well cause a breakdown of the Migdal approximation, the present treatment should be reexamined in a more general strong-coupling framework [8].

In summary, based on a McMillan analysis and a large body of numerical evidence, we conclude that harmonic phonon exchange causes a large T_c suppression, but only a small isotope exponent α, in d-wave pairing instabilities driven by AF spin-fluctuation exchange. Physical constraints on the EP coupling strengths limit $|\alpha|$ to theoretical values below 0.1, which is an order of magnitude smaller than observed values in nonoptimally doped, reduced-T_c cuprate materials. By contrast, acceptable values of α at reasonably large T_c's *can* be obtained, with physically reasonable coupling constants, by exchange of strongly anharmonic lattice tunneling excitations. Such anharmonic lattice modes lead to a very strongly enhanced, isotope-dependent effective λ parameter. As such, our results, combined with the experimental isotope data [6], can be regarded as evidence for a very strongly enhanced effective EP coupling in the cuprates which may well cause a breakdown of the Migdal approximation. The further development of strong-coupling approaches [8], beyond Migdal-Eliashberg theory, will be crucial for gaining a deeper understanding of the role of phonons in the low-energy physics of the cuprate high-T_c systems. Experimentally, it would be of great interest to study the isotope dependence of other physical properties, for example, the specific heat, in order to test whether the cuprates indeed exhibit a strong isotope dependence of their electronic quasiparticle mass, as predicted by polaronic and anharmonic lattice models.

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