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Electron-phonon interaction in $Ba_2YCu_3O_7$

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A realistic tight-binding theory, based on the energy-band results of Mattheiss and Hamann, is applied to study the electron-phonon interaction in $Ba_2YCu_3O_7$. In contrast to previous results for the 40-K superconductor $La_{2-x}(Ba,Sr)_xCuO_4$, the theoretical values for the electron-phonon coupling are much too small to yield superconducting transition temperatures in the 90-K range.

The recent discovery of high-temperature oxide superconductors[1,2] has led to a plenitude of theoretical models[3] for an attractive electron-electron interaction that is strong enough to explain the high transition temperatures. Among these is the conventional electron-phonon coupling model generally believed to be the origin of superconductivity in simple and transition metals and their compounds. Calculations of the electron-phonon interaction in $La_{2-x}(Sr,Ba)_xCuO_4$, based on realistic energy bands[4] and carried out in the framework of the nonorthogonal-tight-binding theory (NTB) of lattice dynamics, have shown that values of $T_c \sim 35$–40 K are attainable.[5] In principle, even values of $T_c \sim 80$–90 K might be possible in an oxide system, provided that the electron-phonon coupling constant $\lambda \sim 2$ and the average frequency of the strong-coupling phonons is of the order of the highest O stretch vibrations (~ 60–70 meV).

In this paper, we report NTB calculations of the electron-phonon (e-ph) interaction for $Ba_2YCu_3O_7$. These calculations, which utilize the energy-band results of Mattheiss and Hamann (MH),[6] have been performed using procedures that parallel those described in Ref. 5. Our main result is that, in contrast to the calculations for $La_{2-x}(Sr,Ba)_xCuO_4$, the theoretical values for T_c in $Ba_2YCu_3O_7$ are *much lower* than the experimentally observed values.

Energy-band results for $Ba_2YCu_3O_7$ have been presented by several workers.[6–8] The individual results differ to some extent (for instance, in the magnitude of the overall bandwidth), but are rather similar in regard to the number and nature of the conduction bands. Most likely, these differences are due in part to slight variations in the underlying crystal-structure models, and partly to differences in the computational methods used by the various groups.

While $La_{2-x}(Ba,Sr)_xCuO_4$ contains a single conduction band that originates from σ-bonded Cu $d(x^2-y^2)$ and O $p(x,y)$ orbitals in the CuO_2 plane, there are three analogous σ-bonding conduction bands in $Ba_2YCu_3O_7$, one for each of the three Cu–O layers in this structure.[9] Two of the conduction bands exhibit two-dimensional (2D) band dipersion that is similar to that in $La_{2-x}(Ba,Sr)_xCuO_4$. They consist mainly of orbitals localized in the two Cu(2)-O(2,3) layers. These have predominantly $d(x^2-y^2)$ symmetry at the Cu(2) sites and $p(x,y)$ symmetry at the O(2,3) sites, analogous to the situation in $La_{2-x}(Ba,Sr)_xCuO_4$. However, the two layers in the unit cell interact and this leads to a splitting and mixing of the two bands such that each subband contains orbital components of similar magnitude from the two layers. Most importantly, these interlayer-induced band splittings spoil the almost perfect Fermi-surface nesting geometry which was a prominent feature of the $La_{2-x}(Ba,Sr)_xCuO_4$ results near half-filling ($x \sim 0$).

The third $Ba_2YCu_3O_7$ band originates from the linear Cu(1)-O(4) chains of the middle layer. It consists of Cu(1) $d(z^2-y^2)$ orbitals hybridized with O(4) $p(y)$ and O(1) $p(z)$ orbitals. Because of interactions with the pair of 2D bands, the one-dimensional (1D) character of this band is diminished. In addition, this 1D band is less than half-filled so that the Fermi-surface nesting vectors are much smaller than the Brillouin-zone (BZ) boundary, namely, $q_{2k_F} \sim \frac{1}{2} q_{ZB}$.

In addition to these 1D and 2D bands, each of the calculations[6–8] predicts the presence of one or two additional flat bands near the Fermi energy E_F. These have mainly π character, i.e., they consist of π-bonding combinations of Cu $d(t_{2g})$ and neighboring O $p(\pi)$ orbitals. These bands increase the electronic density of states at E_F in comparison to $La_{2-x}(Ba,Sr)_xCuO_4$.

For the e-ph calculations, a 46-orbital NTB model (involving Cu s and d, O p, Ba s, and Y d states) has been applied to fit the $Ba_2YCu_3O_7$ valence and conduction bands. The NTB energy and overlap matrix elements have been determined from fits to the linearized augmented plane wave (LAPW) energy-band results of MH. Two separate fits have been carried out. In the first, the bands of the original calculation[6] were used, while the second fit involved LAPW results for a refined version of the crystal structure.[9] The rms quality of these fits has been less satisfactory than that for $La_{2-x}(Ba,Sr)_xCuO_4$, (0.15 eV versus 0.10 eV) probably because of the added complexity of the $Ba_2YCu_3O_7$ band structure and also, to some extent, because of the reduced convergence of the LAPW re-

sults. In general, the overall features of the fitted bands are similar in the two compounds, with dominating ($pd\sigma$) interactions in the antibonding conduction bands and an almost perfect hybridization, i.e., $E_d \sim E_p$. It should be noted, however, that the magnitude of ($pd\sigma$) was determined to be $\sim$10–20% smaller than that for $La_{2-x}(Ba,Sr)_xCuO_4$ because of the reduced bandwidth of the d-p manifold.

In the NTB theory of lattice dynamics,[10,11] the phonon dynamical matrix $D(q) = D_b + D_2$ is divided into D_b, a contribution representing the "bare" phonon frequencies, and D_2, the renormalization due to the conduction electrons. Using common notation, we have

$$D_2(\kappa\alpha,\kappa'\beta|q) = -\sum_{\substack{k\mu\mu' \\ k'=k+q}} \frac{f_{k'\mu'}-f_{k\mu}}{\varepsilon_{k\mu}-\varepsilon_{k'\mu'}} g^{\kappa\alpha}_{k\mu,k'\mu'} g^{\kappa'\beta}_{k'\mu',k\mu}, \quad (1)$$

where (k,μ) denote electronic states with wave vector k and band index μ, while q is the phonon wave vector. The electron ion form factors $g^{\kappa\alpha}$ (κ: particle, α: Cartesian index) contain first order gradients of the NTB integrals with respect to atomic displacements. When one sets the form factors $g^2 = 1$, the right-hand side of Eq. (1) is usually called the "bare" susceptibility χ_0. The imaginary part $\Gamma(q)$ of the renormalization can be represented as a Fermi-surface integral involving the same form factors $g^{\kappa\alpha}$ and is the essential ingredient to the phonon linewidths γ_{qj}. The latter enter the expression for the Eliashberg function in the strong-coupling theory of superconductivity[12]

$$\alpha^2F(\omega) = [hN(E_F)]^{-1}\sum_{qj} \gamma_{qj}/\omega_{qj}\delta(\omega-\omega_{qj}), \quad (2)$$

where ω_{qj} are the phonon frequencies. The e-ph coupling parameter $\lambda = 2\int d\omega \alpha^2(\omega)F(\omega)/\omega$.

The e-ph calculations have been performed using methods that are analogous to those used previously for $La_{2-x}(Ba,Sr)_xCuO_4$ (see Ref. 5). In particular, the same gradients of the NTB integrals have been used. A mesh of 243 k points in the irredicible BZ was employed for the electronic states while a mesh of 50 q points was used for the phonons in calculating the Eliashberg function. This latter mesh was probably too coarse for the finer details of the Fermi-surface nesting. However, we were limited by computational constraints. T_c was then calculated by solving the Eliashberg equations numerically. For the lattice-dynamical model, a short-range force-constant model similar to that for $La_{2-x}(Ba,Sr)_xCuO_4$ was used. We have utilized force constants that produce agreement with recent phonon measurements, both for $La_{2-x}(Ba,Sr)_xCuO_4$ and $Ba_2YCu_3O_7$. In particular, we have included the observation that the highest O stretch vibrations are about 10 meV lower in $Ba_2YCu_3O_7$ than in $La_{2-x}(Ba,Sr)_xCuO_4$.[13]

We have performed three different model calculations for the e-ph coupling in $Ba_2YCu_3O_7$. The first calculation focused on the e-ph interaction within the three $pd\sigma$ bands. Here, we have included all intra- and interband contributions of these three bands in the evaluation of D_2 and Γ, and we chose a value for $N(E_F) = 3$ states/eV cell which corresponds to the NTB value of the $pd\sigma$ partial density of states at E_F. As expected, our results exhibit only a renormalization of the longitudinal Cu–O stretch modes. However, the magnitude of D_2 and Γ that arises from these $pd\sigma$ bands is found to be rather small, as can be seen by comparing the two phonon density-of-states spectra of Figs. 1(a) and 1(b). The Eliashberg function

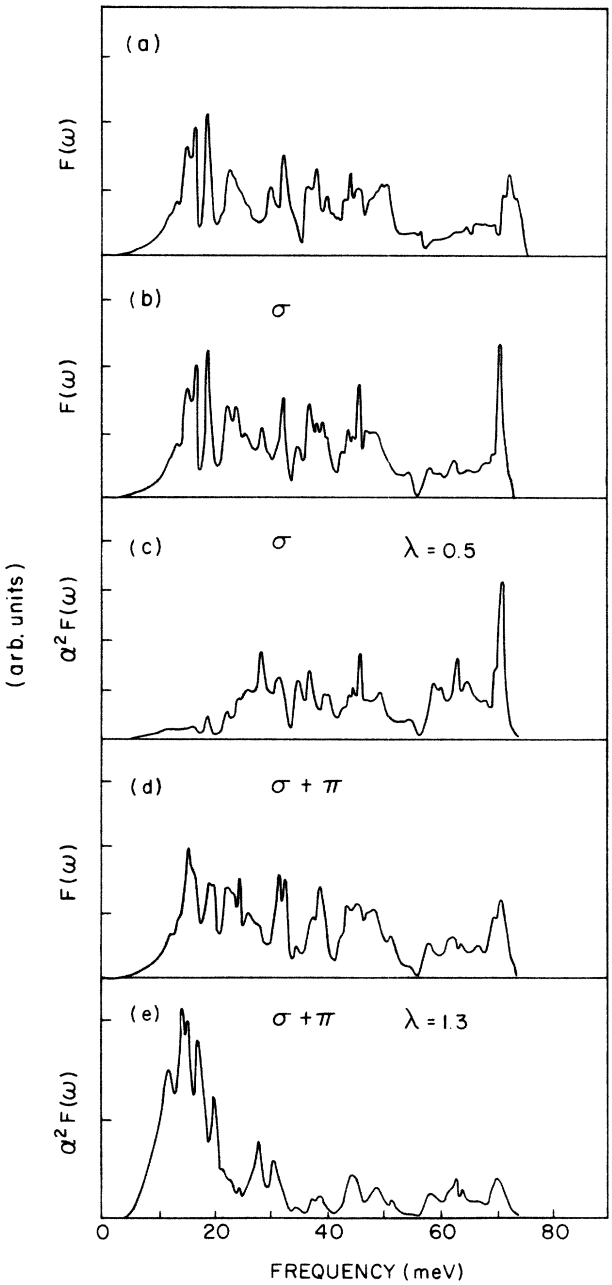

FIG. 1. Comparison of calculated phonon density-of-states curves $F(\omega)$ and the Eliashberg function $\alpha^2F(\omega)$ for $Ba_2YCu_3O_7$. Curve (a) represents the "bare" phonon results. Curves (b) and (c) include intra- and interband D_2 contributions from the three $pd\sigma$ bands, while (d) and (e) represent analogous results with the $pd\pi$ band included. Note that the weight of the high-frequency phonons in the $\alpha^2F(\omega)$ of (c) is much larger than that in (e) because of the smaller $N(E_F)$ value used for the calculation (c).

$\alpha^2F(\omega)$ exhibits considerable weight in the high-frequency region because of the coupling of the O stretch modes [see Fig. 1(c)]. Calculated values of $\lambda \sim 0.5$ were obtained, and thus $T_c \sim 3$ K. Since the numerical evaluation of the nesting in the 1D band was probably insufficient, we estimate that λ may in fact be as high as 1, resulting in $T_c \sim 10$ K. This span of T_c values is actually in good agreement with the previous calculations for $La_{2-x}(Ba,Sr)_xCuO_4$ with $x=0.3$ for which the formal Cu valence of +2.3 is the same as that in $Ba_2YCu_3O_7$.

The second calculation incorporated the intra- and interband contributions of the 4th band with $pd\pi$ character. In this case, we have used the full value of $N(E_F)=8$ states/eV cell. We now find a large increase of $\lambda=1.3$, and with an estimated error margin $\Delta\lambda \sim 1$, λ could reach values up to 2.3, thereby producing T_c values between 19 and ~ 30 K. However, even if λ were much larger than 2, we would not expect any further significant increase in T_c because of saturation effects in the T_c versus λ relation. This increase in the e-ph coupling is caused by interactions between the σ bands and the π band. The results exhibit a large softening of certain *transverse* phonons with wave-vectors near the BZ boundary. These vibrations involve mainly the Cu(1) and O(4) atoms along the chains with displacements along the c direction. The strong renormalization of these bond-bending modes is accompanied by a large e-ph coupling, which leads to an increase in λ [see Figs. 1(d) and 1(e)]. A real-space analysis of the electron-ion form factors $g^{\kappa\alpha}$ shows that interactions between Cu(1) $d(zy)$ and O(4) $p(x)$ orbitals as well as between Cu(1) $d(z^2-y^2)$ and O(4) $p(z)$ orbitals are involved. Although the interaction between these orbitals is zero when the atoms are in equilibrium positions, large NTB gradients can occur when the symmetry is reduced by bond-bending displacements. Interactions of this type have been found to be important in the refractory and $A15$ compounds, where they have been called "dormant" interactions.[11]

Finally, a third calculation was performed assuming a formal Cu valence of +2 in which E_F was raised to accommodate one additional electron per unit cell (corresponding to $x=0$ for $La_{2-x}(Ba,Sr)_xCuO_4$, the half-filled band case). For this value of E_F, the $pd\pi$ bands are completely occupied and do not contribute to the e-ph interaction. This calculation yielded approximately the same modest values of λ and T_c as the first model calculation.

There appear to be two reasons why the e-ph coupling in the $pd\sigma$ bands of $Ba_2YCu_3O_7$ is small compared to $La_{2-x}(Ba,Sr)_xCuO_4$. One is the fact that the pair of 2D $pd\sigma$ bands of the CuO_2 layers interact and the resulting splittings are sufficient to spoil the almost perfect Fermi-surface nesting features of $La_{2-x}(Ba,Sr)_xCuO_4$. Instead of a very strong coupling (and an equally strong renormalization) of relatively few phonons near the BZ boundary that become unstable as x approaches 0 in $La_{2-x}(Ba,Sr)_xCuO_4$, we now find structural stability and a relatively small coupling (and renormalization) of basically all Cu-O stretch modes in the CuO_2 layers. Secondly, the fortunate coincidence in $La_{2-x}(Ba,Sr)_xCuO_4$ that the bare susceptibility and the e-ph matrix elements of O breathing-type displacements both have their maxima at the same wave vector is no longer the case here. This effect is especially important for the quasi 1D linear-chain band, where the nesting vectors are $\sim \frac{1}{2} q_{ZB}$ while the strongest breathing interactions are expected at q_{ZB} so that the two do not coincide. Both effects combine to yield small values of λ in $Ba_2YCu_3O_7$.

From an energy-band point of view, a significant difference between the two cuprate superconductors is that a $pd\pi$ band is present at E_F in $Ba_2YCu_3O_7$. This introduces bond-bending–type interactions as a new feature in the e-ph coupling, and one may argue that anharmonic effects in the e-ph coupling become even more important here. This view is supported by observations of large Debye-Waller factors for some of the O atoms in $Ba_2YCu_3O_7$.[9] Phillips[14] has argued along such lines in his proposed model of giant defect-enhanced e-ph interactions.

However, the top of the $pd\pi$ bands is only ~ 0.2 eV above E_F. Considering the uncertainties of the energy band results for these cuprates, especially the probability of strong electronic correlations, it is also possible that the $pd\pi$ bands are pushed below E_F and are therefore unimportant for the superconducting properties of these materials.

In summary, we have presented calculations of the electron-phonon interaction for $Ba_2YCu_3O_7$ using a realistic tight-binding theory based on first-principles energy-band results. We find that, in the harmonic approximation, the calculated values of T_c are much smaller than observed. Thus, it is concluded that conventional electron-phonon interaction cannot account for the high superconducting transition temperatures in $Ba_2YCu_3O_7$. An additional or alternative mechanism for electronic pairing is required.

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