Evidence for very strong electron-phonon coupling in $YBa_2Cu_3O_6$

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From the observed oxygen-isotope shift of the midinfrared two-magnon absorption peak of $YBa_2Cu_3O_6$, we evaluate the oxygen-isotope effect on the in-plane antiferromagnetic exchange energy J. The exchange energy J in $YBa_2Cu_3O_6$ is found to decrease by about 0.9% upon replacing ^{16}O by ^{18}O, which is slightly larger than that (0.6%) in La_2CuO_4. From the oxygen-isotope effects, we determine the lower limit of the polaron binding energy, which is about 1.7 eV for $YBa_2Cu_3O_6$ and 1.5 eV for La_2CuO_4, in quantitative agreement with angle-resolved photoemission data, optical conductivity data, and the parameter-free theoretical estimate. The large polaron binding energies in the insulating parent compounds suggest that electron-phonon coupling should also be strong in doped superconducting cuprates and play an essential role in high-temperature superconductivity.

The behavior of undoped insulating compounds such as La_2CuO_4 and $YBa_2Cu_3O_6$ is of interest as the starting point for discussion of the physics of cuprates, particularly with regards to the microscopic pairing mechanism of high-temperature superconductivity in doped systems. The antiferromagnetic (AF) ordering found in the parent compounds[1–4] signals a strong electron-electron Coulomb correlation. On the other hand, there is overwhelming evidence that electron-phonon coupling is very strong in the cuprate superconductors.[5–20] In particular, various unconventional oxygen-isotope effects that Zhao and co-workers have observed since 1994 clearly indicate that the electron-phonon interactions are so strong that polarons/bipolarons are formed in doped cuprates[5–9,11,12,14–17,19] and manganites,[21,22] in agreement with a theory of high-temperature superconductivity[23] and the original motivation for the discovery of high-temperature superconductivity.[24]

Now it is well accepted that the parent compounds are charge-transfer insulators and can be described by a three-band Hubbard model. Recently, Eremin *et al.*[25] have considered strong electron-phonon coupling within the three-band Hubbard model. They show that the antiferromagnetic exchange energy J depends on the polaron binding energy E_p^O due to oxygen vibrations, on the polaron binding energy E_p^{Cu} due to copper vibrations, and on their respective vibration frequencies ω_O and ω_{Cu}. At low temperatures, J is given by[25]

$$J = J_\circ\left(1 + \frac{3E_p^O\hbar\omega_O}{\Delta_{pd}^2} + \frac{3E_p^{Cu}\hbar\omega_{Cu}}{\Delta_{pd}^2}\right), \qquad (1)$$

where Δ_{pd} is the unrenormalized charge-transfer gap. The bare superexchange interaction $J_\circ$ is obtained within the fourth-order perturbation theory and its renormalization due to electron-phonon coupling takes place in the sixth-order term.[25] That is why the effective exchange energy J is not strongly renormalized by electron-phonon coupling, in contrast to a strong renormalization of the hopping integral by electron-phonon coupling.[25]

Since the polaron binding energy is independent of the masses of nuclear ions,[26] it is apparent from Eq. (1) that there should be an observable oxygen-isotope effect on J if

the polaron binding energy is comparable with Δ_{pd}. An increase of the oxygen mass leads to a decrease of the phonon energy, which in turn results in a reduction of the exchange energy J according to Eq. (1). Quantitatively, the oxygen-isotope effect on J can be readily deduced from Eq. (1) as follows:

$$\frac{\Delta J}{J} = \left(\frac{3E_p^O\hbar\omega_O}{\Delta_{pd}^2}\right)\left(\frac{\Delta\omega_O}{\omega_O}\right). \qquad (2)$$

Zhao *et al.* initiated studies of the oxygen-isotope effect on the AF ordering temperature in several parent compounds.[5] A noticeable oxygen-isotope shift of T_N (about 1.9 K) was consistently observed in undoped La_2CuO_4 with $T_N=315$ K.[5] From the observed oxygen-isotope shift of T_N, they found[15] that the antiferromagnetic exchange energy is reduced by about 0.6% upon replacing ^{16}O by ^{18}O, i.e., $\Delta J/J=-0.6\%$. However, this isotope effect is negligible in electron-doped cuprates that do not have apical oxygen.[5] This implies that the apical oxygen, which can stabilize the Q_1-type Jahn-Teller distortion, enhances the electron-phonon interaction significantly.

Now a question arises: Does this isotope effect also exist in other parent cuprates with apical oxygen? Recent midinfrared spectra of $YBa_2Cu_3O_6$ crystals show that the two-magnon absorption peak is shifted down by 3.5 meV upon replacing ^{16}O by ^{18}O.[27] This isotope shift was explained in terms of the shift in the frequency of a high-energy oxygen vibration mode that assists the two-magnon absorption process.[27] However, in order to reproduce the experimentally observed high-frequency spectral weight within this scenario, one requires a very large coupling constant that is 1 order of magnitude larger than an expected value.[27] Moreover, the deduced exchange energy[27] (99.5 meV) is significantly lower than those (110–118 meV) inferred from neutron-scattering and Raman-scattering data[29,30] (see below).

Here we demonstrate that the phonon modes that assist the two-magnon absorption process are not the high-energy oxygen-related vibration modes, but the low-energy phonon modes (20–30 meV) which are mainly associated with cop-

per vibrations and very strongly coupled to electrons.[17,31] Within this scenario, the observed oxygen-isotope shift of the two-magnon absorption peak in $YBa_2Cu_3O_6$ is actually consistent with a significant oxygen-isotope effect on J, that is, J decreases by about 0.9% upon replacing ^{16}O by ^{18}O, which is slightly larger than that (0.6%) in La_2CuO_4. From the oxygen-isotope effects, we determine the lower limit of the polaron binding energy, which is about 1.7 eV for $YBa_2Cu_3O_6$ and 1.5 eV for La_2CuO_4, in quantitative agreement with angle-resolved photoemission data, optical conductivity data, and the parameter-free theoretical estimate. Such quantitative agreement strongly supports the model where the low-energy phonon modes mainly assist the two-magnon absorption process. The large polaron binding energies in the insulating parent compounds suggest that electron-phonon coupling should also be strong in doped superconducting cuprates and play an essential role in high-temperature superconductivity.

It is known that Raman spectra can probe two-magnon scattering in antiferromagnets. In contrast, two-magnon absorption in infrared (IR) spectra is expected to be inactive. However, the excitation becomes IR active when a phonon is simultaneously created.[28] A photon with an energy of $\hbar\omega_{IR} = 2.73J + \hbar\omega_{ph}$ is required for this process in single-layer systems,[28,32] where $\hbar\omega_{ph}$ is the phonon energy. For double-layer systems such as $YBa_2Cu_3O_6$, the two-magnon absorption peak shifts up by about $1.6J_\perp$,[27] that is,

$$\hbar\omega_{IR} = 2.73J + 1.6J_\perp + \hbar\omega_{ph}, \tag{3}$$

where $J_\perp$ is the interlayer exchange energy within the bilayers.

From Eq. (3), we can determine the value of J from the energy position of the two-magnon absorption peak if we are able to independently determine the phonon energy. The phonon energy can be determined by the temperature dependence of the linewidth of the peak. Choi $et\ al.$[32] have shown that the temperature dependence of the linewidth of the absorption peak in $Sr_2CuO_2Cl_2$ is in quantitative agreement with the two-magnon absorption process that involves scattering by phonon modes centered at about 25 meV. This is consistent with inelastic neutron scattering, which shows a broad maximum at about 27 meV in the phonon density of states of single-layer $La_{2-x}Sr_xCuO_4$.[33,34] This is also in quantitative agreement with tunneling spectra in both optimally doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ and $YBa_2Cu_3O_{7-\delta}$, which indicate a very large electron-phonon coupling constant (about 2.6) for the 20 meV phonon modes.[17] Substituting $\hbar\omega_{IR} = 363$ meV (Ref. 32) and $\hbar\omega_{ph} = 25$ meV into Eq. (3), we find that $J = 124$ meV for $Sr_2CuO_2Cl_2$. The deduced $J = 124$ meV from the IR spectrum is in perfect agreement with that (125 meV) evaluated precisely from the measured temperature dependence of the spin-correlation length.[35] This quantitative agreement provides additional evidence that the 25 meV phonon modes contribute to the phonon-assisted two-magnon excitation in this single-layer compound.

For another single-layer compound La_2CuO_4, the two-magnon absorption peak is at 410 meV.[36] Substituting $\hbar\omega_{IR} = 410$ meV and $\hbar\omega_{ph} = 25$ meV into Eq. (3) yields $J = 141$ meV, in quantitative agreement with that

(138±4 meV) deduced from the measured temperature dependence of the spin-correlation length.[37] From the measured long-wave spin velocity[38] ($\hbar c = 0.85 \pm 0.03$ eV Å) and using a renormalization factor $Z_c = 1.14$,[39] we obtain $J = 139 \pm 5$ meV, in excellent agreement with the above values. Therefore, the average energy of the phonon modes assisting the two-magnon absorption process is also about 25 meV in La_2CuO_4.

We can also extract the energy of the phonon modes assisting the two-magnon absorption process if we can reliably obtain the J value from other independent experiments. Two-magnon Raman scattering and resonant two-magnon Raman-scattering experiments[30] can independently determine the J value. For two-magnon scattering, the peak position $\hbar\omega_R$ is related to J and $J_\perp$ as[30]

$$\hbar\omega_R = 2.8J + J_\perp. \tag{4}$$

For resonant two-magnon Raman scattering, the first resonance peak occurs at[30]

$$\hbar\omega_{res}^1 = \Delta_{pd} + 2.9J. \tag{5}$$

The Raman spectrum of $Sr_2CuO_2Cl_2$ shows a two-magnon scattering peak at $\hbar\omega_R = 355$ meV.[40] Substituting $\hbar\omega_R = 355$ meV and $J_\perp = 0$ into Eq. (4) yields $J = 126.8$ meV. The measured temperature dependence of the spin-correlation length indicates $J = 125$ meV.[35] Hence, the J value determined from the two independent experiments is 126 ± 1 meV. Substituting $\hbar\omega_{IR} = 363$ meV and $J = 126$ meV into Eq. (3) yields $\hbar\omega_{ph} = 19$ meV, slightly lower than that (25 meV) extracted from the temperature dependence of linewidth of the absorption peak.

From Eqs. (4) and (5), we can extract the exchange energy J for $YBa_2Cu_3O_6$. Substituting $J_\perp = 11$ meV (Ref. 29) and $\hbar\omega_R = 342.7$ meV (Ref. 30) into Eq. (4) yields $J = 118$ meV. Resonant two-magnon Raman scattering[30] shows a resonance peak at $\Delta_{pd} + 328$ meV, that is, $\hbar\omega_{res}^1 - \Delta_{pd} = 328$ meV. Then from Eq. (5), we find $J = 113$ meV. Neutron data[29] imply that $Z_cJ = 125 \pm 5$ meV, leading to $J = 110 \pm 5$ meV with $Z_c = 1.14$.[39] Thus, the J value deduced from the three independent experiments is 114 ± 4 meV.

Now we analyze the observed oxygen-isotope shift of the two-magnon absorption peak for $YBa_2Cu_3O_6$.[27] Figure 1 shows the midinfrared optical conductivity for the ^{16}O and ^{18}O samples of $YBa_2Cu_3O_6$ at $T = 4$ K. The figure is reproduced from Ref. 27. The intersection point of the two straight lines defines the peak position. Upon replacing ^{16}O by ^{18}O, the peak position shifts down by about 26.8 cm^{-1}. The energy of the peak for the ^{16}O sample is about 352 meV. Substituting $\hbar\omega_{IR} = 352$ meV and $J = 114$ meV into Eq. (3) yields $\hbar\omega_{ph} = 23$ meV, close to the values for both $Sr_2CuO_2Cl_2$ and La_2CuO_4. It is interesting to note that the optical conductivity data of $YBa_2Cu_3O_{6.95}$ are consistent with very strong coupling to a bosonic mode whose energy is lower than 32 meV.[41] If one takes the realistic value of the superconducting gap to be 31 meV (Ref. 42) rather than 25 meV used in Ref. 41, one should obtain the mode energy of about 24 meV, in quantitative agreement with $\hbar\omega_{ph} = 23$ meV. Moreover, the coupling strength of this bosonic mode is found to be independent of magnetic field, suggesting that
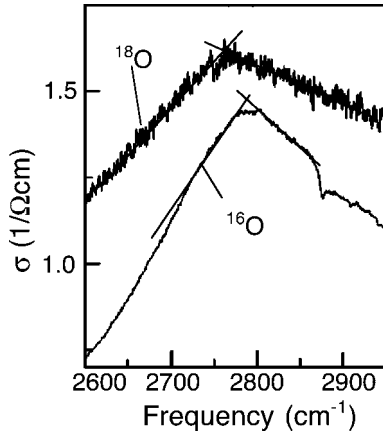
FIG. 1. The midinfrared optical conductivity for the ^{16}O and ^{18}O samples of $YBa_2Cu_3O_6$ at $T=4$ K. The figure is reproduced from Ref. 27. Upon replacing ^{16}O by ^{18}O, the peak position shifts down by about 26.8 cm^{-1}.

this mode is not associated with the magnetic resonance mode.[43]

The average energy of these low-energy phonon modes should not have a significant oxygen-isotope shift since the weight of oxygen vibrations for these modes is less than 30%.[44] We expect that, upon replacing ^{16}O by ^{18}O, $\hbar\omega_{ph}$ should shift down by ~ 0.5 meV. Then from the oxygen-isotope shift (3.38 meV) of $\hbar\omega_{IR}$, we readily find that $\Delta J/J \simeq -0.9\%$. The magnitude of the isotope effect on J for $YBa_2Cu_3O_6$ is slightly larger than that for La_2CuO_4 ($\Delta J/J \simeq -0.6\%$).

Now let us use Eq. (2) to deduce the value of E_p^O for both La_2CuO_4 and $YBa_2Cu_3O_6$. The charge-transfer gaps Δ_{pd} have been measured for both systems,[45] that is, $\Delta_{pd} = 1.81$ eV for La_2CuO_4 and $\Delta_{pd} = 1.60$ eV for $YBa_2Cu_3O_6$. If we take $\hbar\omega_O = 0.075$ eV and substitute the above parameters into Eq. (2), we obtain $E_p^O = 1.5$ eV for La_2CuO_4 and $E_p^O = 1.7$ eV for $YBa_2Cu_3O_6$. Although the E_p^O values for the two systems are similar, the oxygen-isotope effect on J is significantly larger in $YBa_2Cu_3O_6$ due to a smaller Δ_{pd}. Since $E_p^{Cu} \neq 0$, the deduced E_p^O values should be the lower limit of the total polaron binding energy.

Very recently, angle-resolved photoemission spectroscopy (ARPES) data of undoped La_2CuO_4 have been explained in terms of polaronic coupling between phonons and charge carriers.[46] From the width of the phonon sideband in the ARPES spectra, the authors find the polaron binding energy to be about 1.9 eV, in good agreement with their theoretical calculation based on a shell model.[46] On the other hand, the observed binding energy of the sideband should be consistent with a polaron binding energy of about 1.0 eV.[46] This should be the lower limit because the binding energy of the sideband decreases rapidly with doping and because the sample may be lightly doped.[46] Therefore, the ARPES data suggest that 1.0 eV $< E_p < 1.9$ eV, which is in quantitative agreement with the value deduced from the isotope effect on the exchange energy.

The parameter-free estimate of the polaron binding energy due to the long-range Fröhlich-type electron-phonon interaction has been made for many oxides including cuprates and manganites.[47] The polaron binding energy due to the long-range Fröhlich-type electron-phonon interaction is estimated to be about 0.65 eV for La_2CuO_4.[47] The polaron binding energy due to the Q_1-type Jahn-Teller distortion is about 1.2 eV for La_2CuO_4.[48] The total polaron binding energy should be about 1.85 eV, in excellent agreement with the value deduced from the isotope effect on J and the ARPES data.

If there are very small amounts of charged carriers in these nearly undoped compounds, the optical conductivity will show a broad peak at $E_m = 2\gamma E_p$,[47] where γ is 0.2–0.3 for layered cuprates.[47] The γ value will be further reduced in the case of $\hbar\omega/t \ll 1$, where t is the bare hopping integral. There appears to exist a third broad peak at 0.7–0.8 eV in the optical conductivity of $Sr_2CuO_2Cl_2$, La_2CuO_4, and $YBa_2Cu_3O_6$. This peak should be caused by the polaronic effect because the energy scale for the peak is similar to that predicted from the polaron theory assuming $\gamma \sim 0.2$. Hole doping will reduce the value of E_p and thus E_m due to screening of charged carriers. Indeed, E_m was found to be about 0.6 eV for $La_{1.98}Sr_{0.02}CuO_4$ and 0.44 eV for $La_{1.94}Sr_{0.06}CuO_4$.[49]

From the inferred polaron binding energy in the parent compounds, we can estimate a dimensionless coupling constant λ for the high-energy phonon modes using $\lambda = E_p/zt$, where z is the number of the nearest neighbors.[26] With $E_p = 2$ eV, $t = 0.4$ eV, and $z = 4$, we find $\lambda = 1.25$. The coupling constant is not large enough to lead to a structural instability. In fact, it has been shown that[50] there is no structural instability even at a very large electron-phonon coupling. This is because when small polarons are formed, the phonon frequency renormalization is negligible at any carrier density.[50] Moreover, a static long-range charge ordering is unlikely to occur in doped systems due to this intermediate electron-phonon coupling and the quasi-two-dimensional electronic structure.

Now we discuss the isotope effect on the antiferromagnetic ordering temperature T_N in hole-doped La_2CuO_{4+y} and $YBa_2Cu_3O_{6+y}$. It is known that the antiferromagnetic properties of La_2CuO_{4+y} can be well understood within mean-field theory, which leads to a T_N formula:[51]

$$k_B T_N \sim J'[\xi(T_N)/a]^2, \qquad (6)$$

where J' is the interlayer coupling energy and $\xi(T_N)$ is the in-plane AF correlation length at T_N, which is given by $\xi(T_N) \propto \exp(J/T_N)$ for undoped compounds with the maximum T_N. When T_N is reduced to about 80% of the maximum T_N by doping, a mesoscopic phase separation has taken place so that $\xi(T_N) = L$,[52] where L is the size of the antiferromagnetically correlated clusters, and depends only on the carrier density. In this case, we have $T_N \sim J'(L/a)^2$, which is independent of J. This can naturally explain a negligible oxygen-isotope effect on T_N in La_2CuO_{4+y} with $T_N \simeq 250$ K.[5] Because the maximum T_N is about 500 K in $YBa_2Cu_3O_{6+y}$,[2–4] the negligible oxygen-isotope effect on T_N should also be expected for $YBa_2Cu_3O_{6+y}$ with $T_N \leqslant 400$ K.

In summary, we have deduced the oxygen-isotope effect on the in-plane antiferromagnetic exchange energy J for $YBa_2Cu_3O_6$ from the observed oxygen-isotope effect on the midinfrared two-magnon absorption peak.[27] The exchange energy J in $YBa_2Cu_3O_6$ is found to decrease by about 0.9% upon replacing ^{16}O by ^{18}O, which is slightly larger than that (0.6%) in La_2CuO_4. From the isotope effect, we quantitatively estimate the lower limit of the polaron binding energy, which is about 1.7 eV for $YBa_2Cu_3O_6$ and 1.5 eV for La_2CuO_4. The results are in quantitative agreement with the recent ARPES data, optical conductivity data, and the parameter-free theoretical estimate. The large polaron binding energy in the insulating parent compounds suggests that electron-phonon coupling should also be strong in doped superconducting cuprates and play an essential role in high-temperature superconductivity.

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