

## Quadratic Electron-phonon Coupling and the Isotope Effect in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$

We develop a path-integral theory to study the angle-resolved photoemission spectra (ARPES) of high- $T_c$  superconductors based on a two-dimensional model for the  $\text{CuO}_2$  conduction plane. We find that the experimentally observed isotopic band shift in ARPES can be clarified based on a quadratically coupled electron-phonon ( $e$ -ph) model, and that the positive-negative sign change is due to the momentum dependence of the  $e$ -ph coupling.

The study of high- $T_c$  superconductivity has been one of the most active fields in solid state physics over the last two decades. Since angle-resolved photoemission spectroscopy (ARPES) directly probes the electronic occupied states, it has become an important technique for investigating the electronic properties of cuprates [1]. Recently, the oxygen isotope effect has been detected with ARPES in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  (Bi2212) by two groups [2,3]. Both groups report the common feature that the spectra are shifted with  $^{16}\text{O}/^{18}\text{O}$  substitution, providing direct evidence for electron-phonon ( $e$ -ph) coupling in this material. However, since the first report by Gweon *et al.* [2], this isotopic band shift has been a subject of considerable controversy [4], since the observed shift is as large as 40 meV, far larger than the isotopic energy change of the oxygen phonon which is  $\sim 5$  meV. Subsequently, Douglas *et al.* [3] repeated the experiment, but found a shift of only  $2\pm 3$  meV. Thus it turns out to be an interesting problem whether the large shift observed by Gweon *et al.* is possible or not in the cuprates. To answer this question, we have recently investigated the isotopic band shift from a theoretical point of view using the path-integral quantum Monte Carlo (QMC) method, and we find that off-diagonal quadratic  $e$ -ph coupling plays an essential role [5].

Since we are primarily concerned with the band shift, we start with the following model Hamiltonian ( $\hbar = 1$  and  $k_B = 1$ ),

$$H = -\frac{1}{2} \sum_{l,\sigma} \sum_{l+\delta} t(l, l+\delta) (a_{l,\sigma}^+ a_{l+\delta,\sigma} + a_{l+\delta,\sigma}^+ a_{l,\sigma}) - \mu \sum_{l,\sigma} a_{l,\sigma}^+ a_{l,\sigma} + \frac{\omega_0}{2} \sum_{\langle l,l' \rangle} \left( -\frac{1}{\lambda} \frac{\partial^2}{\partial q_{ll'}^2} + q_{ll'}^2 \right)$$

In the conduction plane of  $\text{CuO}_2$ , the electronic hopping integral can be expanded to the second order terms with respect to the phonon displacements parallel to the Cu-O bond. For example, in Fig. 1,

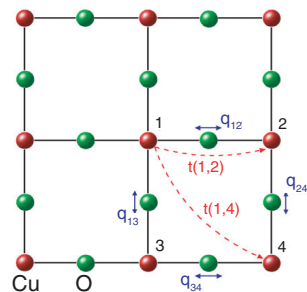


Figure 1 Schematic plot of the  $\text{CuO}_2$  conduction plane in the cuprates. The copper atoms (red) form a simple square lattice, and the oxygen atoms (green) are located between the nearest-neighbor Cu sites.

the nearest-neighbor hopping can be expanded as  $t(1,2) = t_1 + s_1 q_{12}^2$ , and the next-nearest-neighbor hopping as  $t(1,4) = t_2 + s_2 (q_{12}^2 + q_{24}^2 + q_{13}^2 + q_{34}^2)$ . Here  $t_1$  ( $t_2$ ) is the bare nearest- (next-nearest-) neighbor hopping energy,  $s_1$  ( $s_2$ ) is the off-diagonal  $e$ -ph coupling strength due to the nearest- (next-nearest-) neighbor hopping effect, and  $q_{ll'}$  is the displacement of the oxygen phonon located between the nearest-neighbor Cu sites  $l$  and  $l'$ . One should note here that by using the above Hamiltonian we have assumed that the electrons reside only on the Cu sites, and that the electronic transfer is quadratically modulated by the oscillation of the oxygen phonons, as the linear coupling contributes little to the isotopic shift [4]. To reproduce the band structure and Fermi surface of Bi2212 (see the inset of Fig. 2), we set  $t_1=0.22$  eV and make this the unit of energy for the numerical calculation. In units of  $t_1$ , we assume  $t_2=-0.2$ , and the chemical potential is  $\mu=-1$ , which is invariant with isotope substitution. The bare phonon energy is set as  $\omega_0=1.0$ , and the two  $e$ -ph coupling constants are fixed at a ratio of  $s_1:s_2=1:-1$  for simplicity.

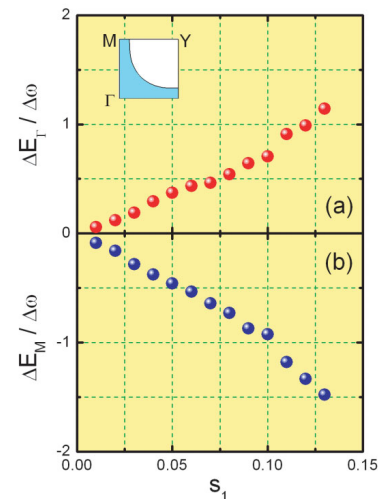


Figure 2 The positive-negative sign change effect of the isotopic band shift as a function of the  $e$ -ph coupling constant  $s_1$ , on a square lattice. The ratio  $\Delta E_i/\Delta\omega$  at the  $\Gamma$  (upper panel) and  $M$  (lower panel) points are also shown. The inset shows a quadrant of the Brillouin zone with the Fermi surface.

In Fig. 2, the QMC results of the ratio  $\Delta E_i/\Delta\omega$  at the  $\Gamma$  [ $\mathbf{k}_\Gamma=(0,0)$ ] and  $M$  [ $\mathbf{k}_M=(\pi,0)$ ] points are presented as functions of  $s_1$ , where  $\Delta E_i$  and  $\Delta\omega$  are the electronic band shift and phonon energy change due to the oxygen isotopic substitution. Our calculation shows an

increase in  $\Delta E_i/\Delta\omega$  with  $s_1$ , meaning that if the  $e$ -ph coupling is strong enough, a small change in phonon energy  $\Delta\omega$  can be amplified to a large band shift  $\Delta E_i$  observed in ARPES. The results shown in Fig. 2 also imply that the band shift has a momentum dependence, resulting in an anisotropy of the isotope effect and a positive-negative sign change of the ratio  $\Delta E_i/\Delta\omega$ . Because of the size limitations of QMC, here we do not have enough data for a continuous dispersion along the  $\Gamma M$  direction. Nevertheless, in Fig. 2, it is clearly shown that the isotope substitution yields a positive band shift at the  $\Gamma$  point and a negative one at the  $M$  point. Thus, one can see that the sign reversal of the isotope effect is a natural consequence of the momentum dependence of the  $e$ -ph coupling. In conclusion, our QMC results demonstrate that the off-diagonal quadratic  $e$ -ph coupling is responsible for the anomalous isotopic band shift.

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## A Theoretical Study of the Photoemission Spectra of Boron-doped Diamond

Path-integral theory is applied to calculate the photoemission spectra (PES) of boron-doped diamond, mainly investigating electron-phonon coupling and the impurity effect. The coupling and the increase of doping rate contribute to the emergence of a Fermi edge, indicating the electron's itinerancy - moving freely from boron atom to boron atom through the intermediate carbon atoms. Simultaneously, the coupling results in multi-phonon side structure, indicating electron localization by Einstein phonons. These qualitatively clarify the coexistence of a Fermi edge and the step-like structure observed in PES experiments.

In the photoemission spectra (PES) of boron-doped diamond (BDD), the emergence of a clear Fermi edge and a step-like structure slightly below the Fermi level are observed with increasing boron doping rate [1]. Notably, this step-like structure is in normal states, which is the basis of the superconductivity. Further, this side structure is distributed over nearly equal distances, around 0.150 eV. This reminds us of its similarity to the spectrum of the localized Einstein model. Combined with the emergence of the Fermi edge, which indicates the free movement of electrons, the coexistence of localization and itinerancy - the electron's two intrinsic characteristics - seems obvious, although its observation is unique. In this work, we use a path-integral theory to calculate the PES of BDD. To clarify the coexistence of localization and itinerancy theoretically, an impurity Holstein model is employed, focusing on the electron-phonon (e-ph) interaction and the impurity effect. The Hamiltonian is given as

$$H \equiv -T \sum_{\langle l,l' \rangle} \sum_{\sigma} (a_{l\sigma}^{\dagger} a_{l'\sigma} + h.c.) - \mu \sum_{l,\sigma} n_{l\sigma} + \Delta_c \sum_{l_0} \sum_{\sigma} n_{l_0\sigma} + \frac{\omega_0}{2} \sum_l \left( -\frac{\partial^2}{\partial Q_l^2} + Q_l^2 \right) - S \sum_{l,\sigma} Q_l (n_{l\sigma} - \bar{n}_l / 2)$$

$$n_{l\sigma} = a_{l\sigma}^{\dagger} a_{l\sigma}, \quad \sigma = \alpha \text{ or } \beta.$$

where  $T$  is the transfer energy,  $a_{l\sigma}^{\dagger}$  ( $a_{l\sigma}$ ) is the creation (annihilation) operator of a conduction electron with spin  $\sigma$  at site  $l$ . The electrons hop between the nearest neighboring sites expressed by  $\langle l,l' \rangle$ .  $\mu$  is the chemical potential of the electrons and  $\Delta_c$  is the potential difference between the original carbon atoms and the doped boron atoms.  $S$  is the e-ph coupling constant, and  $Q_l$  stands for the dimensionless coordinate operator for the phonon at site  $l$  with a frequency  $\omega_0$ .

In the simulation, a simple cubic lattice is chosen to simplify the problem, as we are just interested in the area around the Fermi level. At the same time, only the e-ph coupling at the doped boron atoms is taken into account, since the phonon effect in pure diamond has not yet been observed. A doped sites spectrum is also given to emphasize the phonon effect, which is not obvious in the total system spectrum after averaging over all sites. In all calculations, the doping is simulated by randomly replacing carbon atoms with boron atoms according to the doping rate.

In Fig. 1, the spectra for different doping rates are shown for a constant  $S/T$  of 0.6. From the total system spectra (red curves), the emergence of a clear Fermi edge is seen with increasing doping rate. The impurity band extends up to the top of the valence band and gradually fills the semiconductor gap. A semiconductor-

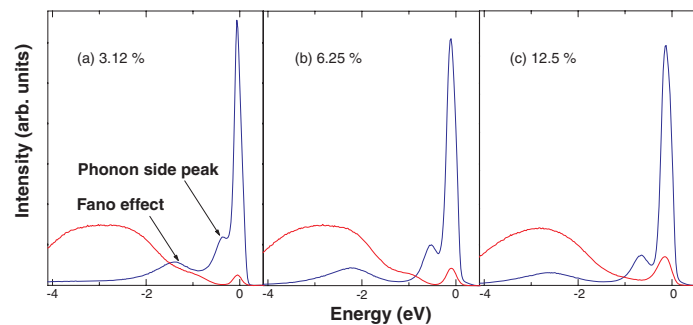


Figure 1  
Spectra of the simple cubic lattice for different doping rates. The energy zero is the position of the Fermi level. Red (blue) curves are the total system (doped sites) spectra.

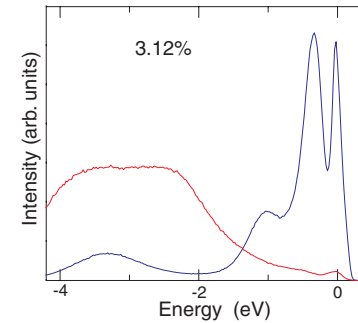


Figure 2  
Spectra of the simple cubic lattice with a larger coupling constant.

metal transition takes place, allowing electrons to move freely from boron atoms to other boron atoms through the intermediate carbon atoms. Along with a Fano tail, a phonon side structure [2] is also seen from the doped sites spectrum (blue) in each case, which reflects the phonon quantum effect. In Fig. 2, we give the spectra

for a larger coupling constant ( $S/T=1.2$ ) and a low doping rate. Comparing these spectra with those of Fig. 1(a), we conclude that the e-ph interaction can also contribute to the expansion of the impurity band and the phase transition. In addition, the multi-phonon scattering process is induced [2].

In conclusion, we have demonstrated the coexistence of the localization and itinerancy of electrons in BDD by investigating the PES calculated by applying a path-integral theory to a impurity Holstein model. We find that the e-ph coupling plays an important role in the coexistence.

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