# Gap Anisotropy in the Layered High Temperature Superconductors

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We present a new strategy for constructing semiphenomenological models which deal with the electronic structure of the normal and superconducting states on equal footing. We demonstrate that it can reproduce all the striking features of the high-resolution photoemission experiments on the layered high $T_c$ superconductors.

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Recently, high-resolution photoemission experiments have been providing fairly convincing evidence that the gap in the excitation spectrum of the layered high $T_c$ superconductors is strongly anisotropic [1,2]. While this may be decisive information, the interpretation of the data is elusive due to the lack of credible models which deal with the normal and superconducting states on equal footing [3,4]. In this Letter we present a new strategy for constructing such models and *illustrate* their use and power by interpreting complex features of experimental data.

To make progress we combine the eight-band model advocated by Andersen *et al.* [5] for describing the electronic structure of these materials, near the Fermi energy, in the normal state and the semiphenomenological density functional approach to superconductivity of Suvasini *et al.* [6]. The former is an effective, nearest neighbor, tight-binding model Hamiltonian for a Cu-O bilayer, and it features four $\sigma$ orbitals: $\varphi_1^s$(Cu $d_{x^2-y^2}$), $\varphi_2^s$(Cu $s$), $\varphi_3^s$(O $2p_x$), $\varphi_4^s$(O $3p_y$), and four $\pi$ orbitals: $\varphi_5^s$(O $2p_z$), $\varphi_6^s$(O $3p_z$), $\varphi_7^s$(Cu $d_{zx}$), $\varphi_8^s$(Cu $d_{xy}$), for the bottom ($s = 0$) and top ($s = 1$) layers of bilayer material. It is the result of a down-folding procedure based on first-principles local density approximation (LDA) calculations [5], and it gives an accurate account of all the interesting bands near the Fermi level [5]. In particular, it describes the bifurcated saddle point which is often regarded as essential for the occurrence of superconductivity [3–5]. For a physical interpretation of the model it is worthwhile to add that the Cu $s$ orbital contains some $\mathrm{Cu}_{3d_{x^2-1}}$ and apex oxygen $p_z$ characters. The description of pairing [6] is based on the density functional theory (DFT) which, under a very general condition, describes superconductivity by a self-consistent Kohn-Sham Bogoliubov–de Gennes equation with normal potential $v(\mathbf{r};[n,\chi])$ and pairing potential $\Delta(\mathbf{r},\mathbf{r}';[n,\chi])$ which are functionals of the charge density, $n(\mathbf{r})$, and the pairing amplitude, $\chi(\mathbf{r},\mathbf{r}')$, which is the proper order parameter of the problem [7]. In this Letter we consider *singlet* pairing only.

For the normal potential $v(\mathbf{r};[n,\chi])$ we shall take the usual LDA functional of $n(\mathbf{r})$. Clearly, to make a similar approximation for the pairing potential functional would imply knowing the mechanism of pairing. Instead of assuming a specific model we adopt a semiphenomenological approach. Namely, in the general functional relation between $\Delta(\mathbf{r},\mathbf{r}')$ and $\chi(\mathbf{r},\mathbf{r}')$,

$$\Delta(\mathbf{r},\mathbf{r}') = \int \mathbf{d^3}\mathbf{r}_1 \int \mathbf{d^3}\mathbf{r}_1' \lambda(\mathbf{r},\mathbf{r}';\mathbf{r}_1,\mathbf{r}_1')\chi(\mathbf{r}_1,\mathbf{r}_1'), \quad (1)$$

we parametrize the kernel $\lambda(\mathbf{r},\mathbf{r}';\mathbf{r}_1,\mathbf{r}_1')$ (which, as $\Delta$, in a complete theory would be additionally a functional of both $n$ and $\chi$, and would be determined by the pairing mechanism) by a set of adjustable parameters (orbital and site dependent coupling constants) to be chosen so that a realistic $T_c$ would be obtained, and the anisotropy of the observed gap could be reproduced.

To combine this phenomenology of superconductivity with the description of the normal state by the eight-band model [5], we expand $\chi(\mathbf{r},\mathbf{r}')$ and $\Delta(\mathbf{r},\mathbf{r}')$ in terms of the orbitals of the model

$$\chi(\mathbf{r},\mathbf{r}') = \sum_{i,j}\sum_{\mu,\mu'}\sum_{\mathbf{s},\mathbf{s}'} \varphi_\mu^s(\mathbf{r}-\mathbf{R}_i)\chi_{\mu,\mu'}^{s,s'}(i,j)\varphi_{\mu'}^{s'}(\mathbf{r}'-\mathbf{R}_j),$$
$$\Delta(\mathbf{r},\mathbf{r}') = \sum_{i,j}\sum_{\mu,\mu'}\sum_{\mathbf{s},\mathbf{s}'} \varphi_\mu^s(\mathbf{r}-\mathbf{R}_i)\Delta_{\mu,\mu'}^{s,s'}(i,j)\varphi_{\mu'}^{s'}(\mathbf{r}'-\mathbf{R}_j),$$

where $\varphi_\mu^s$'s are the orbitals mentioned earlier, $\mathbf{R}_i$'s are the positions of the atoms, $i, j$ are the site indices, and $\mu, \mu'$ the basis function indices. Clearly, which atom contributes which orbital at which site is governed by the coefficients $\chi_{\mu,\mu'}^{s,s'}(i,j)$ and $\Delta_{\mu,\mu'}^{s,s'}(i,j)$, and Eq. (1) becomes

$$\Delta_{\mu,\mu'}^{s,s'}(i,j) = \sum_{k,l}\sum_{\mu_1,\mu_1'}\sum_{s_1,s_1'} \lambda_{\mu,\mu';\mu_1,\mu_1'}^{s,s';s_1,s_1'}(i,j;k,l)\chi_{\mu_1,\mu_1'}^{s_1,s_1'}(k,l), \quad (2)$$

where

  

$$\lambda^{s,s';s_1,s_1'}_{\mu,\mu';\mu_1,\mu_1'}(i,j;k,l) = \int d^3\mathbf{r} \int d^3\mathbf{r}' \int d^3\mathbf{r}_1 \int d^3\mathbf{r}_1' \times \varphi^{s}_{\mu}(\mathbf{r}-\mathbf{R}_i)\varphi^{s'}_{\mu'}(\mathbf{r}'-\mathbf{R}_j)\lambda(\mathbf{r},\mathbf{r}';\mathbf{r}_1,\mathbf{r}_1')\varphi^{s_1}_{\mu_1}(\mathbf{r}_1-\mathbf{R}_k)\varphi^{s_1'}_{\mu_1'}(\mathbf{r}_1'-\mathbf{R}_l)\,.$$

The central point of our approach is that we shall regard the matrix elements $\lambda^{s,s';s_1,s_1'}_{\mu,\mu';\mu_1,\mu_1'}(i,j;k,l)$ as adjustable parameters describing an effective electron-electron attraction of unknown origin. The power of the method lies in being able to attribute the rich variety of consequences, predictions for various experiments, of the theory to interactions between electrons at specific sites and in specific orbitals. Our hope is that guided by the data from such searching probes as high-resolution photoemission experiments [1,2] we shall be able to identify the sites and orbitals relevant to superconductivity, and hence provide new bases for speculations about the nature of pairing. In what follows we *illustrate* the power of this approach by analyzing the gap anisotropy reported in [1] and [2], and identifying the site and orbital dependence of the electron-electron attraction with specific features of the data.

The calculations are much simpler if we make the necessary but reasonable approximation,

$$\lambda^{s,s';s_1,s_1'}_{\mu,\mu';\mu_1,\mu_1'}(i,j;k,l) = \lambda^{s,s'}_{\mu,\mu'}(i,j)\delta_{\mu,\mu_1}\delta_{\mu',\mu_1'} \times \delta_{s,s_1}\delta_{s',s_1'}\delta_{i,k}\delta_{j,l}\,,$$

which reduces (2) to *one* term, and base our theory on the fundamental self-consistency condition,

$$\Delta^{s,s'}_{\mu,\mu'}(i,j) = \lambda^{s,s'}_{\mu,\mu'}(i,j)\chi^{s,s'}_{\mu,\mu'}(i,j)\,.$$

For a periodic system it is useful to look for solutions such that $\Delta^{s,s'}_{\mu,\mu'}(i,j)$, $\lambda^{s,s'}_{\mu,\mu'}(i,j)$, and $\chi^{s,s'}_{\mu,\mu'}(i,j)$ depend only on the differences $\mathbf{R}_i - \mathbf{R}_j$, and hence only their lattice Fourier transforms $\Delta^{s,s'}_{\mu,\mu'}(\mathbf{k})$, $\lambda^{s,s'}_{\mu,\mu'}(\mathbf{k})$, and $\chi^{s,s'}_{\mu,\mu'}(\mathbf{k})$ enter the theory. Under this assumption, the eight-band model Bogoliubov–de Gennes equation, at a particular $\mathbf{k}$ point in the Brillouin zone, reads as follows:

$$\sum_{\mu'=1}^{8}\sum_{s'=0}^{1}\begin{pmatrix}(\epsilon^{s}_{\mu}-\mu_e)\delta_{\mu,\mu'}\delta_{s,s'}+t^{s,s'}_{\mu,\mu'}(\mathbf{k}) & \Delta^{s,s'}_{\mu,\mu'}(\mathbf{k}) \\ \Delta^{s,s'*}_{\mu,\mu'}(\mathbf{k}) & -(\epsilon^{s}_{\mu}-\mu_e)\delta_{\mu,\mu'}\delta_{s,s'}+t^{s,s'}_{\mu,\mu'}(\mathbf{k})\end{pmatrix}\begin{pmatrix}u^{\nu}_{\mathbf{k},\mu',\mathbf{s}'} \\ v^{\nu}_{\mathbf{k},\mu',\mathbf{s}'}\end{pmatrix} = E_{\mathbf{k},\nu}\begin{pmatrix}u^{\nu}_{\mathbf{k},\mu,\mathbf{s}} \\ v^{\nu}_{\mathbf{k},\mu,\mathbf{s}}\end{pmatrix},$$

where the site energies $\epsilon^{s}_{\mu}$ and the hopping integrals $t^{s,s'}_{\mu,\mu'}(\mathbf{k})$ are those of the eight-band model [5] for the Cu-O bilayer.

For each solution labeled by band index $\nu$ of Eq. (3), we construct the two-component Bloch wave solution of the Bogoliubov–de Gennes equation in the usual way;

$$\Psi_{\mathbf{k},\nu}(\mathbf{r}) = \sum_{i,\mu,s}\begin{pmatrix}u^{\nu}_{\mathbf{k},\mu,\mathbf{s}} \\ v^{\nu}_{\mathbf{k},\mu,\mathbf{s}}\end{pmatrix}\phi^{s}_{\mu}(\mathbf{r}-\mathbf{R}_i)\exp(i\mathbf{k}\cdot\mathbf{R}_i)\,,$$

and calculate the density $n(\mathbf{r})$ and the pairing amplitude $\chi(\mathbf{r},\mathbf{r}')$ using the standard relations [see Eq. (4) in [6]]. We then recalculate the pairing potentials $\Delta^{s,s'}_{\mu,\mu'}(i,j)$ and repeat the process, determining the chemical potential $\mu_e$ at each stage, until a prescribed level of convergence is reached. All the calculations we shall discuss in the remainder of this Letter have been performed in this way, making a variety of choices for the phenomenological particle-particle interaction parameters $\lambda^{s,s'}_{\mu,\mu'}(i,j)$.

The quasiparticle energies $E_{\nu}(\mathbf{k})$ come in pairs, $E^{+}_{\nu}(\mathbf{k})$ and $E^{-}_{\nu}(\mathbf{k})$, above and below the chemical potential $\mu_e$, respectively. We define the superconducting gap in the quasiparticle spectrum as the minimum energy separation $2\overline{\Delta}_{\nu}(\mathbf{k}_{F,s}) = E^{+}_{\nu}(\mathbf{k}_{F,s}) - E^{-}_{\nu}(\mathbf{k}_{F,s})$, where $\mathbf{k}_{F,s}$ is the wave vector for which such a minimum occurs. Note that $\mathbf{k}_{F,s}$ defines a surface in the superconducting state, but this need not be the same as the Fermi surface in the normal state due to the change in the chemical potential on condensation [6]. This is why we do not use the BCS formula $E_{\nu}(\mathbf{k}) = \sqrt{(\epsilon_{\mathbf{k},\nu}-\mu_e)^2 + \overline{\Delta}^2_{\nu}(\mathbf{k})}$ to define the gap.

Another general point of interest is that, unlike the textbook examples of single-band theories, in the present multiband case the pairing interaction is not the gap but is related to it in a complicated fashion. This is due to the matrix character of the pairing potential, $\Delta^{s,s'}_{\mu,\mu'}(\mathbf{k})$, and is of significance when we discuss their symmetry classification [8]. Evidently, we should distinguish between intraorbital and interorbital pairing potentials corresponding, respectively, to the diagonal and off-diagonal elements of $\Delta^{s,s'}_{\mu,\mu'}(\mathbf{k})$. While in the case of the former, $\Delta^{s,s'}_{\mu,\mu'}(\mathbf{k}_{F,s})$ for $\mu = \mu'$, is usually a good approximation to the gap, and its $\mathbf{k}$ dependence is governed by the interaction. In the case of the latter, namely $\mu \neq \mu'$, surprisingly, the $\mathbf{k}$ dependence of the hybridization $t^{s,s'}_{\mu,\mu'}(\mathbf{k})$ plays a dominant role in determining the variation of the gap, $\overline{\Delta}_{\nu}(\mathbf{k}_{F,s})$, over the Fermi surface. This is readily seen by noting the presence of terms such as, e.g., $\Delta_{ds}(\mathbf{k})$, $t_{sp}(\mathbf{k})$, $t_{pd}(\mathbf{k})$ in the determinant of the eigenvalue problem in Eq. (3). Unexpectedly, as discussed below, also diagonal pairing potentials, $\Delta^{s,s}_{\mu,\mu}$, for $\mu$ whose orbital character does not occur in any of the bands at the Fermi level can give rise to a superconducting gap with a complex $\mathbf{k}$ dependence involving off-diagonal hopping integrals. In what follows, these observations

will be central to our description of the gap anisotropy as seen by the high-resolution photoemission experiments [1,2].

We have applied the methodology described above to study the quasiparticle spectra for the Cu-O bilayer of $YBa_2Cu_3O_7$ (YBCO) [5] for a variety of site- and orbital-diagonal and off-diagonal couplings both within and between the Cu-O layers. Although an exhaustive study of all possibilities was not feasible, we have investigated a large number of cases fairly systematically. Our results may be summarized as follows. (a) For site- and orbital-diagonal interactions we found a more or less isotropic gap, except for $\lambda^{s,s}_{2,2}$, which gave rise to an anisotropic gap with a cusp and zero value along the $(\pi, \pi)$ direction. Although the corresponding $\lambda^{s,s}_{2,2} = 7.0$ eV is too large to be seriously considered, this result demonstrates dramatically that in the multiband model the symmetry of the gap need not be the same as the symmetry of the pairing interaction or the order parameter. By contrast, in the conventional one-band model such a gap would imply an order parameter of *d*-wave symmetry, but here it is associated with an isotropic pairing potential $\Delta^{s,s}_{2,2}$ and the $\mathbf{k}$ dependence involving a product of the off-diagonal hopping integrals $t_{d,p}(\mathbf{k})t_{p,s}(\mathbf{k})\Delta^{s,s}_{2,2}t_{s,p}(\mathbf{k})t_{p,d}(\mathbf{k})$. (b) Of single site, but orbital off-diagonal interactions, only $\lambda^{s,s}_{2,1}$ gave an anisotropic gap. This was also of the above mentioned *d*-wave symmetry. (c) Generally, intersite interactions in the Cu-O plane led to anisotropic gaps. Usually these were also of *d*-wave symmetry of the above kind, except in the case of $\lambda^{s,s}_{2,1}$, which resulted in an anisotropic gap with a minimum, as opposed to a cusp, at $(\pi, \pi)$. In the case of the one-band model the latter circumstance would correspond to the extended *s*-wave order parameter. (d) Interlayer couplings had similar effects to that of the on-site interactions, and only $\lambda^{s,s'}_{2,2}$ and $\lambda^{s,s'}_{2,1}$ resulted in anisotropic gaps. (e) No single interaction gave a minimum at an intermediate point on the Fermi surface between $(0, \pi)$ and $(\pi, \pi)$. (f) Examining the case where we assumed more than one coupling constant to be nonzero, we discovered that the intersite in-plane *s*-*d* interaction $\lambda^{s,s}_{2,1}$ and *d*-*p* interaction of $\lambda^{s,s}_{1,3}, \lambda^{s,s}_{1,4}, \lambda^{s,s}_{1,5}, \lambda^{s,s}_{1,6}$ combination produced the anisotropic gaps over the odd and even sheets of the Fermi surface displayed in Fig. 1. To illustrate the power of our approach, in our calculation we have adjusted the two independent interaction parameters ($\lambda^{s,s}_{2,1} = 2.7$ eV and $\lambda^{s,s}_{1,3} = \lambda^{s,s}_{1,4} = \lambda^{s,s}_{1,5} = \lambda^{s,s}_{1,6} = 0.45$ eV) so that the transition temperature agreed with experiments on YBCO, and the minima in the gap were located at the points on the Fermi surface as was first reported by Ding *et al.* [1] (Fig. 2). Although the characteristic "hump" in their original interpretation of the data has recently been withdrawn, it is illuminating to elaborate on the way the above result comes about.

This $k$ dependence of $\overline{\Delta}_\nu(\mathbf{k})$ is a linear combination of an extended *s*-wave pairing favored by the intersite *s*-*d* interaction and a $\cos(k_x a)\cos(k_y b)$ contribution due to a relatively small *d*-*p* interaction. The former is a surprising consequence of the interplay between the $k$ dependence coming from the lattice Fourier transform of $\lambda^{s,s}_{2,1}(i, j)$, $\lambda^{s,s}_{2,1}(\mathbf{k})$, and another induced by hybridization between *s* and *d* bands via $t_{s,p}$ and $t_{p,d}$ hopping. The latter brings in the lattice Fourier transform $\lambda^{s,s}_{p,d}(k)$ which features $\sin(k_x a/2)$ for the $\sigma$ orbitals, instead of $\cos(k_x a)$ in the case of $\lambda^{s,s}_{s,d}(\mathbf{k})$, and $\cos(k_x a/2)$ for $\pi$ orbitals. Also noteworthy is the fact that each of the $\sigma$ and $\pi$

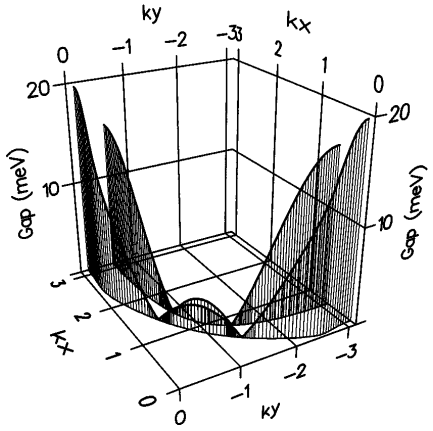


FIG. 1. The calculated superconducting gap along the even (inner curve) and odd (outer curve) sheets of the Fermi surface for Cu-O bilayer of $YBa_2Cu_3O_7$ corresponding to the $\Gamma X$ irreducible quadrant. The $x$ and $y$ axes are in units of $k$, and the length of each of them is $\pi$.

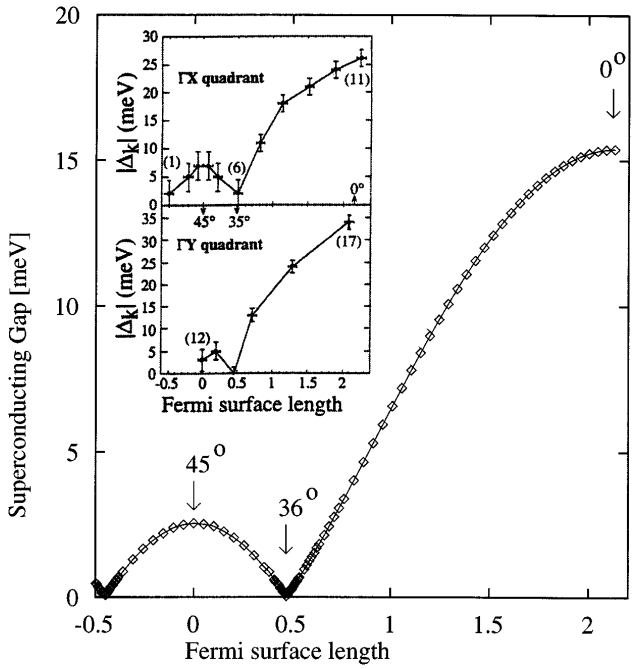


FIG. 2. The calculated superconducting gap as a function of the Fermi surface length for the even sheet of the Fermi surface of Cu-O bilayer of $YBa_2Cu_3O_7$ corresponding to the $\Gamma X$ irreducible quadrant. The $x$ axis is in units of $k$. In the inset we show the results of Ding *et al.* [1] for $Bi_2Sr_2CaCu_2O_8$.

*d-p* coupling constants separately produces minima away from $(\pi, \pi)$ with slight changes in the position of the minima and the maximum amplitudes in $(0, \pi)$ and $(\pi, \pi)$ directions. Clearly, for $\lambda^{s,s}_{d,p}(\mathbf{k}) = 0$ the hump would disappear. By contrast, in the case of the experiments suggesting that the gap is zero along the $(\pi, \pi)$ direction [2] with a cusp, our calculations strongly favor Cu $d-$Cu $d$ nearest neighbor interaction in the Cu-O plane with a conveniently small $\lambda^{s,s}_{d,d}(\mathbf{k}) = 0.67$ eV.

It is reassuring to observe that these results probe the essential features of the eight-band model [5]. The hybridization of O $p_z$ with Cu $d_{x^2-y^2}$ is responsible for the bifurcated saddle point. Its $k^4$ rather than $k^2$ dispersion in one of the directions enhances the phase space effects and, therefore, the size of the gap for a given set of coupling constants.

Of course, according to our strategy, having chosen a specific set of interaction constants which reproduces $T_c$ and the required gap anisotropy is only the beginning. Given the set, we can calculate without further adjustable parameters the full solution of the Bogoliubov–de Gennes equation depicted in Eq. (3) and, therefore, a rich variety of observables such as the Knight shifts, spin lattice relaxation rates, etc. [6]. A quantity of such general interest is the quasiparticle density of states $N(E) = \sum_k \delta(E - E_k)$. Our calculated quasiparticle density of states corresponding to the gap in Fig. 1 is shown in Fig. 3. Interestingly, the maximum gap of 20 meV at $(\pi, 0)$ marks the position of a well defined pseudogap, below which there is only a very shallow tail. Obviously, using this figure and our calculated $T_c$ of 91 K we could deduce a BCS ratio $2\Delta/k_BT$ anywhere between 3.4 and 5.1.

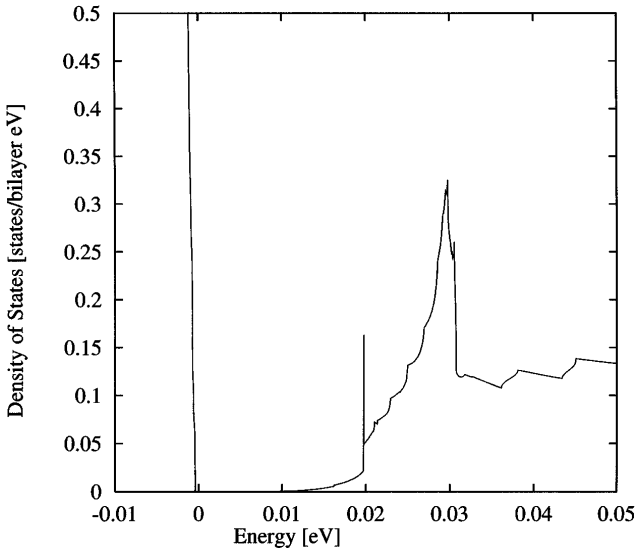


FIG. 3. The density of states for $T = 0$ K.

Given our site and orbital specific, but otherwise phenomenological, proposal for the pairing interaction we now wish to comment on the somewhat different conclusions of Fehrenbacher *et al.* [4] whose aspirations are rather similar to ours. While it is not easy to draw comparisons between their one-band model and our much more elaborate description of the electronic structure near the Fermi energy, it is interesting to note that they identify the pairing interaction between next-nearest-neighbor sites as the principal cause of the gap anisotropy shown in Fig. 2. Thus, our suggestion may be viewed as a discovery of an alternative mechanism of gap anisotropy, involving a richer variety of orbitals. The significant difference notwithstanding, it may also be worthwhile to note that the strength of their interaction in units of the hopping integrals which characterize the band structure in the normal state is rather similar to our $\lambda_{sd}/t_{sp} \sim 1$. This is a very strong interaction which, if real, should have a number of important implications. For instance, it is unlikely to be compatible with the mechanism of pairing with harmonic phonons.

Summarizing, the principal result of this Letter is the illustration of how the interpretation of sufficiently complex experimental data can lead to an identification of the orbital and site dependence of the pairing interaction. In particular, the three possible scenarios of hump, cusp, and minimum along the $(\pi, \pi)$ direction could each be interpreted as arising from a very specific nearest neighbor pairing interaction: $\lambda^{s,s}_{s,d}$ (2.7 eV) combined with $\lambda^{s,s}_{d,p}$ (0.45 eV) imply a hump, $\lambda^{s,s}_{d,d}$ (0.67 eV) gives a gap that is zero along the $(\pi, \pi)$ direction with a cusp, and, finally, $\lambda^{s,s}_{s,d}$ (2.3 eV) results in a gap which is zero and a minimum along the $(\pi, \pi)$ direction.

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