Spectrum of elementary excitations and interplanar tunneling in layered cuprates

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(Submitted 14 June 1994)
Pis'ma Zh. Eksp. Teor. Fiz. **60**, No. 2, 118–122 (25 July 1994)

Data on photoelectron emission obtained recently with a resolution better than 10 meV can be described well by the model of a singlet-correlated motion of oxygen holes in CuO_2 planes. This model predicts the existence of a peak in the density of states near the bottom of the band. Experimental points corresponding to an energy of 120 meV near the center of the Brillouin zone in $YBa_2Cu_4O_8$ are explained on the basis of an interplanar tunneling of singlet-correlated oxygen holes.

The spectrum of elementary excitations in layered cuprates such as $YBa_2Cu_4O_8$ has yet to be explained. Recent studies of photoelectron emission with a resolution better than 10 meV revealed an interesting aspect of this spectrum: so-called extended saddle regions near the coordinate axes of the Brillouin zone. These extended regions lead to a peak in the density of states, which in turn leads to an explanation of several features of high-T_c superconductors, in particular, the weak isotope effect.[1,2]

In this letter we show that the features of the dispersion relation observed near the Fermi surface in Refs. 1 and 3 can be explained on the basis of a model of a singlet-correlated motion of oxygen holes in CuO_2 planes.[4,5] We also predict several important new features of the spectrum, which can be tested experimentally.

We first consider a single CuO_2 plane. In the hole representation, the energy operator is[5]

$$H=\epsilon_d\sum_{i\sigma} d_{i\sigma}^{+}d_{i\sigma}+\epsilon_p\sum_{i\sigma} p_{i\sigma}^{+}p_{i\sigma}+\sum_{ij} t_{ij}^{pd}(d_{ij}^{+}p_{j\sigma}+p_{j\sigma}^{+}d_{i\sigma})+\sum_{jj'} t_{jj'}^{pp}p_{j\sigma}^{+}p_{j'\sigma}$$

$$+J_{pd}\sum_{i}\psi_i^{+}\psi_i, \qquad (1)$$

where the operators $d_{i\sigma}^{+}$ and $p_{j\sigma}^{+}$ create a hole at a copper site and an oxygen site, respectively, and the operator ψ_i^{+} creates a Zhang–Rice singlet[4,5] centered at copper site i. All the parameter values of the Hamiltonian are well known: $\epsilon_p-\epsilon_d\approx 2-3$ eV, $t^{pd}=1$ eV, $t^{pd}=0.6$ and 0.4 eV for nearest and second-nearest neighbors, respectively, and $J_{pd}\approx -1.4$ eV.

Under the standard approximations of the Hubbard model, the spectrum of elementary excitations is determined by a secular equation[5]

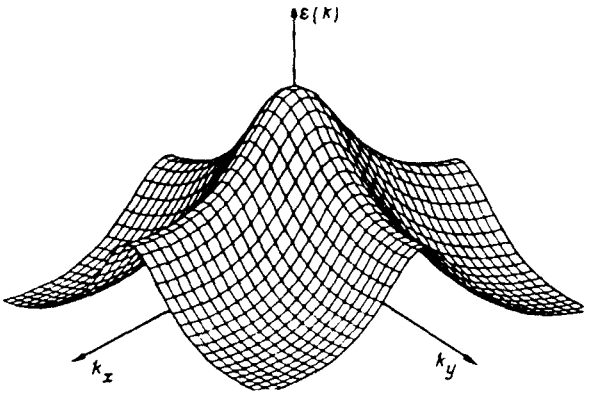

FIG. 1. Theoretical values of $\epsilon(k)$ of the singlet-correlated band of oxygen holes in a CuO_2 plane.

$$\begin{vmatrix} a_{11}-\epsilon & a_{12} & a_{13} & a_{14} \\ a_{21} & a_{22}-\epsilon & a_{23} & a_{24} \\ a_{31} & a_{32} & a_{33}-\epsilon & a_{34} \\ a_{41} & a_{42} & a_{43} & a_{44}-\epsilon \end{vmatrix} = 0. \tag{2}$$

It leads to four bands. The lowest, the "copper" Hubbard band, is completely filled. The two "oxygen" bands are empty. The band of singlet-correlated motion of an oxygen hole against the background of singly occupied copper sites is partially filled. The creation operator for this quasiparticle excitation can be described approximately by

$$S_{k\sigma}^{+} \approx \frac{1}{\sqrt{N}} \sum_i \psi_i^{+} d_{i\sigma} e^{i\vec{k}\vec{R}_i}. \tag{3}$$

It is reminiscent of the quasiparticle creation operator corresponding to the upper "copper–oxygen" Hubbard band. Since the insulating gap of the spectrum, $\epsilon_p - \epsilon_d$, is comparable to t^{pd}, ordinary perturbation theory is not suitable for calculating the roots of Eq. (2). For reliability below, as in Ref. 5, we use numerical solution methods. Explicit expressions for the matrix elements a_{ij} are given in Ref. 5. Here we would simply like to call attention to two refinements in them:

$$a_{41} = \frac{t^{pd}}{4\sqrt{2}}[4n_p + n_d(2-n_p)(\cos k_x a + \cos k_y a)], \tag{4}$$

$$a_{44} = \epsilon_p + J_{pd} + (-2t^{xy} + t^{xx})\left(1 - \frac{n_p}{2}\right) - \frac{1}{16}(2-n_p)n_d J_{pd}(\cos k_x a + \cos k_y a). \tag{5}$$

The last terms in (5) stem from the coherent natural motion of singlet-correlated oxygen holes; that motion was not considered in Ref. 5.

Figure 1 shows the dispersion of the singlet-correlated band calculated from Eq. (2). Note the saddle points near the coordinate axes and the "wings" at the bottom of the band. Figure 2, a and b, compares the results with existing experimental points in two cross sections. For convenience, we are using the electron representation in this figure, as

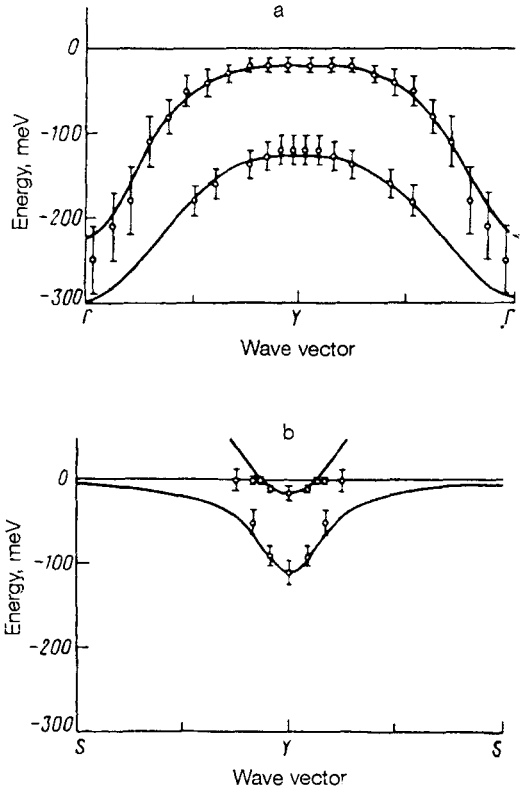

FIG. 2. Curves of $\epsilon(k)$ measured in Refs. 1 and 3 and calculated in the present study along two directions in the Brillouin zone. a—The Γ–Y–Γ line; b—the S–Y–S line.

in Refs. 1 and 3. We see that the upper theoretical curve agrees remarkably well with data from photoelectron emission.[1,3] The results of our calculations are stable with respect to possible changes in the parameters of Hamiltonian (1). In particular, at both $\epsilon_p - \epsilon_d = 2$ eV and $\epsilon_p - \epsilon_d = 3$ eV, the theoretical curve conforms to the experimental data within the errors. The picture found for the density of states is also stable; it is shown in Fig. 3. The small peak corresponds to the saddle points described in Ref. 1. The large peak near the bottom of the band corresponds to the wings and has not been mentioned previously. This is a prediction of the model of this letter. At low doping levels, the Fermi energy is evidently near this peak. If this peak exists, then it should be responsible for several unusual properties of cuprates of the $La_2CuO_{4+\delta}$ type. This question obviously requires further experimental study.

Let us discuss the origin of the lower band in Fig. 2a, i.e., the origin of the experimental points near the center of the Brillouin zone at energies of 120 meV (Ref. 3). We will show that these points can be described satisfactorily under the assumption that a tunneling of singlet-correlated oxygen holes occurs between neighboring planes in $YBa_2Cu_4O_8$ or $YBa_2Cu_3O_7$. Here we mean the planes that are separated from each other by a distance on the order of 3.3 Å . There are no such planes in $La_2CuO_{4+\delta}$. The idea of a possible interplanar tunneling of singlet pairs has been raised previously by Hsu and Anderson[6] and other investigators, but this tunneling has not been linked with Zhang–Rice singlets.

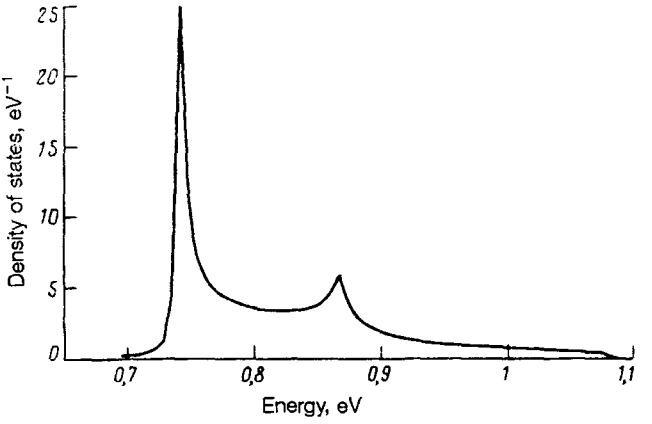

FIG. 3. Density of states of the singlet-correlated band. The energy is reckoned from the top of the lower copper Hubbard band.

We construct a tunneling operator in third-order perturbation theory, using the following procedure: An oxygen hole from plane 1 jumps to the upper copper Hubbard band, then tunnels into the upper copper Hubbard band of the neighboring plane, and finally tunnels to one of the four oxygen sites nearest a copper in plane 2. The operator representing the biplanar sandwich, which is a sum of two operators of the type in (1) (for the first and second planes), is thus supplemented by an operator

$$8\sum_i \left[\frac{t^{pd}}{U_{dd}-(\epsilon_p-\epsilon_d)-2V_{pd}}\right]^2 t^{dd}_{i_1 i_2}\psi^+_{i_1} d_{i_1\sigma} d^+_{i_2\sigma}\psi_{i_2}. \tag{6}$$

The integral t^{dd}_{12}, representing the jump of a hole between $|x^2-y^2\rangle$copper states from different planes, can be evaluated in the following way. We know that this integral is part of the operator representing the interplanar coupling of spins, $J^{dd}_\perp(S^d_1 S^d_2)$. Its parameter is given by the expression

$$J^{dd}_\perp = 4\frac{(t^{dd}_{12})^2}{U_{dd}}, \tag{7}$$

where U_{dd} is the hole repulsion energy at a copper site. The energy U_{dd} is known reliably; it is about 9 eV. Using the experimental estimate $J^{dd}_\perp = 15$ meV, as in Ref. 7, we find $t^{dd}_{12}\approx 0.2$ eV. The parameter of the interplanar tunneling of singlet-correlated oxygen holes is thus

$$\tilde{t}^{pp}_{12} = 8\left[\frac{t^{pd}}{U_{dd}-(\epsilon_p-\epsilon_d)-2V_{pd}}\right]^2 t^{dd}_{12}\approx 0.1 \text{ eV}. \tag{8}$$

In order to explain the identical behavior of the dispersion curves in Fig. 2 we must assume $\tilde{t}^{pp}_{12} = 0.13$ eV. Another important circumstance speaking in favor of our tunneling model is the following: It can be seen easily from (6) that when we go over to the k representation, the tunneling parameter is independent of k. In other words, the magni-

tude of the tunneling-induced splitting of the bands is the same for all points in the Brillouin zone. The dispersion curves of the bands are similar. This similarity can apparently be seen clearly on the basis of the experimental points[3] in Fig. 2, although it would obviously be desirable to have more experimental points in order to reach a definitive conclusion. Our model leads to one more important prediction. It can be seen from Fig. 2b that the wings of the upper hole band lie near the Fermi energy. The saddle peak in the density of states of one band is thus intensified by the peak of the wings of the other band. This circumstance may be of decisive importance for explaining the increase in T_c in biplanar cuprates, which has been mentioned in the literature.[8]

We wish to thank professor Teplov for a discussion of questions related to the increase in T_c in biplanar high-T_c superconductors and for the opportunity to read Ref. 8 before its publication. This study was carried out with support from the Russian Scientific and Technological Program "High-Temperature Superconductivity" (Project 91001).

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Translated by D. Parsons