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Fermi-surface and low-energy excitation spectrum of $YBa_2Cu_3O_7$: Role of the Ba-O plane

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We present evidence for the existence of two close-lying, nearly parallel energy bands in the vicinity of the Fermi level in $YBa_2Cu_3O_7$. The electrons in these bands are located in the Ba-O planes and have nearly no Cu-O chain or plane components. The higher band is partially unoccupied and the corresponding Fermi surface was recently measured by the de Haas–van Alphen effect. We have calculated the extremal Fermi-surface areas and the results are in excellent agreement with experiment, when spin splitting in high magnetic field is taken into account. We show furthermore that interband transitions for in-plane polarizations are allowed between these two bands and that they result in a sharp peak at about 350 cm^{-1} in our calculated optical conductivity.

The electronic structure of the high-temperature superconductors, and in particular of $YBa_2Cu_3O_7$ has been studied intensively both experimentally and theoretically, and a better understanding of the normal-state electronic structure seems to emerge. On the experimental side there has been considerable progress in angular-resolved photoemission spectroscopy (ARPES),[1] positron annihilation,[1] infrared optical spectroscopy,[2–4] and even de Haas–van Alphen (dHvA) measurements have now appeared.[5] On the theoretical side, more accurate local-density-approximation (LDA) electronic structures have been published,[6,7] which have clarified the details of the LDA band structure close to the Fermi level. Besides an impressive overall agreement between the measured and the calculated main Fermi-surface (FS) parts, i.e., the three sheets derived from the plane and the chain bands, we shall point out in this paper that there are strong experimental indications for the existence of the theoretically predicted small hole pocket of the FS around the SR line $(\pi/a, \pi/b, k_z)$ in the Brillouin zone, shown in Fig. 1. In contrast to the other FS sheets, this sheet has nearly only O(4)(p) character, where O(4) is the oxygen atom bridging plane and chain (often called the "apex oxygen"). Since the apex oxygen has attracted much attention, especially in connection with nonadiabatic mechanisms for superconductivity (see, e.g., Ref. 8), it is of interest to study the electronic excitations from this part of the FS. In this paper we describe in detail two bands near the Fermi level, which are formed neither by plane nor by chain electrons. We shall show below that the characteristic energy scale of these excitations is of the same order of magnitude as the phonon frequencies (≈ 400 cm^{-1}) and the corresponding interband transitions can therefore be quite important in the superconducting regime. We show that the dHvA cross sections clearly indicate the existence of one of these bands, when the spin splitting due to the strong magnetic field is taken into account. Then we present calculations of the contribution to the interband optical conductivity in the infrared region from these bands, and suggest that it can and probably has been observed in experiment.

We shall first describe these two bands, which are located near the S point $(\pi/a, \pi/b, 0)$ in the Brillouin zone and are shown in the inset of Fig. 1. They were calculated recently[6] using the LDA and the full-potential linear-muffin-tin-orbital (LMTO) method. As shown in Fig. 1, there are two bands in the vicinity of the Fermi energy: the upper one has mainly O(4)(p_y) character and the lower one mainly O(4)(p_x) character. We shall denote them as the y band (the y axis is along the chains) and the x band, respectively. The O(p_x) and O(p_y) states are hybridized with the Ba(p_y) and Ba(p_x)

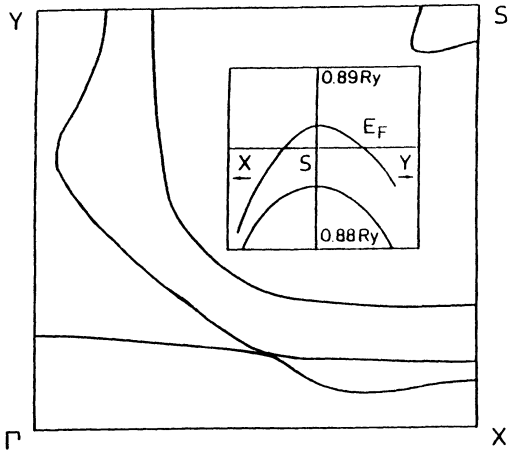

FIG. 1. The Fermi surface of $YBa_2Cu_3O_7$ in the $k_z = 0$ plane. The inset shows the band structure near the S point and close to the Fermi level, according to Ref. 6.

states, respectively. Exactly at the S point they belong to different irreducible representations and therefore cannot interact. In a tetragonal structure these bands would be degenerate, but due to a small chain oxygen admixture into the y band they split apart. The simplest tight-binding model with only p states yields isotropic parabolic hole bands. However, hybridization with Ba(d_{xy}) states, which is allowed away from the S point, makes the mass of the x band in the x direction larger than in the y direction and vice versa for the y band. The last factor determining the dispersion is the interaction between the two bands, which is forbidden along the SX and the SY lines, but allowed otherwise. This explaines why the y band is less dispersive and the x band is more dispersive in the S-Γ direction (not shown) than in the directions shown in the figure (see Ref. 6). In the nearest-neighbor approximation the band structure is determined by three effective hopping integrals: $t_1 = t(p_x^{O(4)} - p_y^{Ba} - p_x^{O(4)}) = t(p_y^{O(4)} - p_x^{Ba} - p_y^{O(4)})$, $t_2 = t(p_x^{O(4)} - d_{xy}^{Ba} - p_x^{O(4)}) = t(p_y^{O(4)} - d_{xy}^{Ba} - p_y^{O(4)})$, and $t_3 = t(p_x^{O(4)} - d_{xy}^{Ba} - p_y^{O(4)})$.

$$E_x = E_{x0} + t_1 \cos^2 x \cos^2 y - t_2 \cos^2 x \sin^2 y - t_3 \sin^2 x \sin^2 y,$$

$$E_y = E_{y0} + t_1 \cos^2 y \cos^2 x - t_2 \cos^2 y \sin^2 x + t_3 \sin^2 x \sin^2 y,$$

where $x = k_x a/2$ and $y = k_y b/2$ and the origin is at the S point. There is no dispersion in the z direction. We have discussed this model in such detail because the square oxygen lattice with metal ions in the center of the squares is a structural element of essentially all superconducting cuprates. In $YBa_2Cu_3O_7$ because of the orthorhombicity the parameters t_1 and t_2 are not exactly the same for the x and the y band. Only the upper band crosses the Fermi level, while the lower, separated by about 3 mRy from it, is occupied.

We shall now indicate some experimental evidences for these bands. First of all, the occupied part of the y band seems to have been seen at least along the S-X (Y) direction in ARPES.[1] Secondly, the dHvA measurements[5] show two small extremal orbits perpendicular to the c axis with areas $A \approx 0.014$ and 0.021 a.u. and masses $m \approx (7.0 \pm 2.5)m_0$ and $(7.2 \pm 2.5)m_0$. It was suggested[5] that these orbits correspond to the cross sections of the y-band FS sheet by the $k_z = 0$ and $k_z = \pi/c$ planes. However, since the band-structure calculations[6,7] predict negligible z dispersion for this band, it is difficult to explain the existence of two considerably different extremal orbits.

We suggest that this contradiction can be resolved by taking into account the spin splitting of the bands due to the extremely strong magnetic field (80–100 T). In Fig. 2 we show our calculated extremal areas and the corresponding masses as function of the position of the Fermi level. The broken lines correspond to the spin-up and spin-down bands at a field of 100 T. The two extremal areas are 0.013 and 0.020 a.u. in excellent agreement with experiment and the corresponding masses are

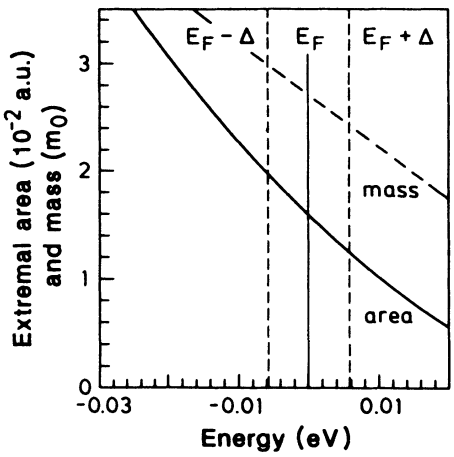

FIG. 2. Areas and masses of the de Haas–van Alphen extremal orbits for the Fermi-surface pocket near the S point as a function of the Fermi energy. Broken lines indicate the positions of the Fermi level for a magnetic field of $H = \pm 100$ T.

$2.6m_0$ and $3.0m_0$. This gives the mass renormalization parameters $\lambda \simeq 1.7 \pm 1.0$ and 1.4 ± 0.8, respectively. Another interesting point is that the dHvA measurements were performed at temperatures $T << T_c$, and it has been argued that in a field of 80–100 T, which is probably less than H_{c2} for the Cu-O planes, the samples are superconducting and the outcome of the dHvA measurements is unclear. However, since the electronic states of the y band have no component in the Cu-O planes, it is possible that at 80–100 T they are not superconducting. In this case the dHvA measurements would have their usual interpretation for this part of the FS.

We shall now present some predictions following from the band structure in Fig. 1. An obvious consequence of this band structure is the existence of interband electronic transitions with extremely small energies ($\simeq$ 40 meV). The simplest property in which such transitions can manifest themselves is the optical conductivity, whose interband part is given by

$$\sigma_{\text{inter}}(\omega) = (\pi/3\Omega\omega) \sum |p_{if}(k)|^2 \delta(E_f(k) - E_i(k) - \hbar\omega) \times [\Theta(E_i(k)) - \Theta(E_f(k))]. \quad (1)$$

Here i and f refer to the initial (x-band) and the final (y-band) states, $p_{if}(k)$ are optical matrix elements, and Ω is the cell volume. Different susceptibilities are defined by analogous expressions with different matrix elements. For constant matrix elements, $p_{if} = 1$, the sum in Eq. (1) is called the joint density of states (JDOS). We have calculated[11] the JDOS with the band structure from Ref. 6, and found a sharp very narrow peak at about 350 cm^{-1}. From the description of the tight-binding band structure it may be seen that nonzero optical matrix elements exist, due to the Ba states, namely $\langle Ba_y^i | \hat{p}_x | Ba_{xy}^f \rangle$ and $\langle Ba_{xy}^i | \hat{p}_y | Ba_x^f \rangle$, and $(p_{if})_x \propto y$ and $(p_{if})_y \propto x$. The corresponding proportionality factors can be obtained from standard LMTO calculations, and the k dependence of the matrix elements can be calculated analytically. The optical conductivity obtained in

this way is shown in Fig. 3, after convolution with a Lorenzian of width 80 cm^{-1} to account for interband relaxation. It is extremely difficult to observe this feature in an experiment because it is superposed on the strong intraband component. The situation below T_c is even more complicated because the superconducting gap may well be in the same energy region. However, a feature in the optical conductivity has been observed at about 350 cm^{-1} for $T > T_c$.[2-4] In Fig. 3 we also show the *interband* conductivity extracted from the measured reflectivity by Kamaràs *et al.*[2] It is noteworthy that there is a clear feature exactly at the position of the calculated peak, which even has nearly the same intensity. One should of course be cautious of this interpretation because of the difficulties in separating the interband and the intraband conductivity, and the contribution from optical phonons.

Another physical effect in which such interband transitions may show up is the interband electronic Raman scattering. This is described by the same sum as the optical absorption,[9] except that the momentum matrix elements now have to be summed over available intermediate states. Some Raman experiments[10] show that besides the constant background, there is a relatively smooth maximum centered at 300–400 cm^{-1} and with a width of several hundred cm^{-1}. One possible interpretation of this maximum is that it is due to the interband scattering of electrons in the two apex oxygen bands. It could also explain the extraordinary strong interaction of the Ba phonon with the electronic background.[12] It is also interesting that, according to the calculated band structure, available intermediate states are missing between 1.9 and 2.8 eV below the Fermi level. Most Raman experiments use the laser energy $\hbar\omega \approx 2.4$ eV, and it would therefore be interesting to look for this effect using smaller or larger laser energy, for which it should show up more clearly.

It should be mentioned that there are other low-energy transitions possible, namely for k points close the $k_z = \pi/c$ plane where the two plane bands nearly touch, and close to the $k_z = 0$ plane, where the chain and the upper plane band cross at the Fermi level. In both cases broad features are expected in the optical conductivity in z polarization. These are, however, even more difficult to observe than the sharp feature we have focused on.

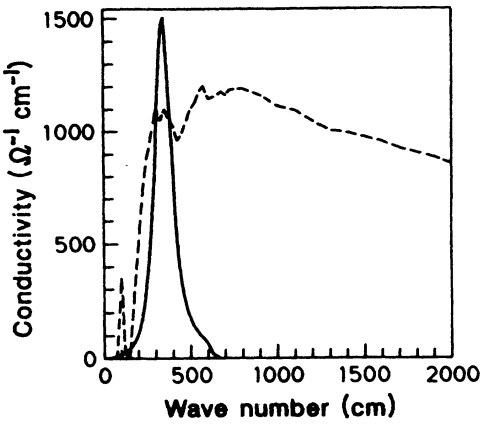

FIG. 3. Experimental optical conductivity (without the Drude part) from Ref. 2 (broken line), compared with the calculated interband optical conductivity (full line).

In summary, we have presented theoretical and pointed out experimental evidence for the existence of two close-lying and nearly parallel energy bands in $YBa_2Cu_3O_7$, one of which crosses the Fermi level. Both bands have mostly apex oxygen character. These *nonplane nonchain* electronic states near the Fermi level should be taken into account in the discussions of the electronic properties of the normal and superconducting state of $YBa_2Cu_3O_7$. Furthermore, the fact that there are interband electronic transitions with energies comparable to the superconducting gap should be taken into account as well.

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