International Journal of Modern Physics B, Vol. 16, Nos. 11 & 12 (2002) 1691–1696


# PHOTOEMISSION STUDY OF THE INTRA-UNIT-CELL COUPLING IN A TRILAYER CUPRATE

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Received 10 December 2001

The electronic structures of the nearly optimally doped single-layer, bilayer and trilayer Bi-based cuprates are investigated by angle-resolved photoemission spectroscopy. A line-shape analysis of data taken for different photon energies indicates that the interlayer coupling within the trilayer is not stronger than its counterpart in the bilayer system. This suggests that the higher $T_c$ of the trilayer cuprate superconductors is not due to an enhancement of the coupling strength between the neighboring $CuO_2$ planes within each unit cell.

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## 1. Introduction

The high-temperature superconductors (HTSCs), based on the number of $CuO_2$ planes ($n$) in each characteristic block, are classified into single-layer materials [e.g. $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201) and $La_{2-x}Sr_xCuO_4$], bilayer materials [e.g. $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) and $YBa_2Cu_3O_{7-\delta}$], trilayer materials [e.g. $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi2223), and $HgBa_2Ca_2Cu_3O_{8+\delta}$], and so on. This structural characteristic influences the superconducting properties to such an extent that, within each family of cuprates, $T_c$ increases monotonically with the layer number up to $n \leq 3$ and then start decreasing.[1,2] For example as far as the Bi-based cuprate HTSCs are concerned, the maximum $T_c$ is approximately 34, 90, and 110 K for optimally doped Bi2201 ($n = 1$), Bi2212 ($n = 2$), and Bi2223 ($n = 3$), respectively.[2] However, despite all the experimental and theoretical efforts, a conclusive microscopic understanding of the empirical relation between $n$ and the superconducting transition has not been reached yet.

A change in $n$ affects the system in many different ways. First of all, it results in a change of the chemical environment around the $CuO_2$ planes themselves. Taking Bi2223 as an example, the two outer $CuO_2$ layers have the same chemical environment as the two planes present in Bi2212, while the inner $CuO_2$ layer of Bi2223 has no apical oxygen as in the electron doped cuprate superconductor $Nd_{2-x}Ce_xCuO_4$. Second, the presence of $CuO_2$ multilayer blocks in the unit cell and the coupling between the neighboring $CuO_2$ layers alter the electronic structure of the system resulting, for example, in the splitting of the otherwise degenerate $CuO_2$ electronic bands.[3] This and more subtle effects can be studied by angle-resolved photoemission spectroscopy (ARPES), which is considered as one of the most direct probes of the electronic structure of solids and has greatly contributed to the understanding of the HTSCs.[4] Recent ARPES results from the bilayer compound Bi2212[5,6] have demonstrated the presence of bilayer splitting in the heavily overdoped Bi2212 systems where one can distinguish two spectral features that correspond to the two split bands. To gain a systematic picture of the role of multiple $CuO_2$ planes and interlayer coupling on the electronic structure of the cuprates and, more importantly, on high-$T_c$ superconductivity, we here study the intra-trilayer coupling of the trilayer Bi2223 system. Our ARPES lineshape analyses at different photon energies indicate a weak intra-trilayer coupling (as compared to the bilayer case of Bi2212[5,6]), suggesting that the coupling strength between different $CuO_2$ layers may not directly relate to the enhancement of the superconducting phase transition temperature.

## 2. Experimental

The Bi2201, Bi2212 and Bi2223 single crystals were grown by travelling-solvent-floating-zone method. Nearly optimally doped samples were obtained by annealing the as-grown crystals. $T_c = 34$ K, $\Delta T_c(10\% - 90\%) = 1$ K, $T_c = 90$ K, $\Delta T_c(10\% - 90\%) = 1$ K, and $T_c = 108$ K, $\Delta T_c(10\% - 90\%) = 2$ K are achieved for the $n =$

1, 2, and 3 material respectively. Magnetic susceptibility measurements excluded the presence of any second phase. X-ray and low-energy electron diffraction show well ordered bulk and surface structures. ARPES experiments were performed at the Stanford Synchrotron Radiation Laboratory (SSRL) beamline 5-4, which is equipped with a Scienta SES200 electron analyzer. ARPES spectra were acquired in a narrow window of about $0.3^\circ \times 0.5^\circ$. The overall energy resolution is about $10 \sim 17$ meV depending on different photon energies. The samples were aligned by Laue diffraction, and cleaved *in-situ* at a pressure better than $5 \times 10^{-11}$ torr $10 \sim 20$ K above $T_c$. The flatness of the cleaved surfaces was confirmed by the small laser reflection from the samples. Data were collected within 12 hours after cleaving and aging effects were negligible. Particularly, since we are focused on large energy scale features, small differences in temperature, momentum window and energy resolution will not affect the conclusion drawn here.

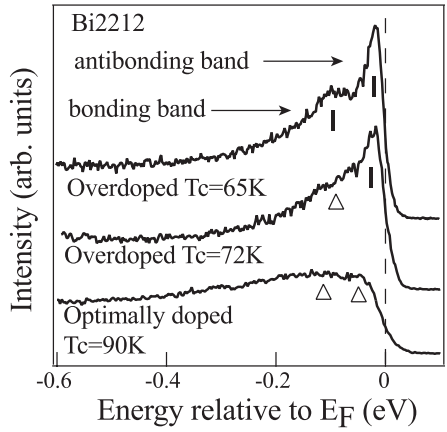


Fig. 1. Normal state photoemission spectra of Bi2212 taken at $(\pi, 0)$ for three different doping levels (from top to bottom, carrier doping level decreases). Data were taken with $h\nu = 22.7$ eV photon. Bars indicate identifiable feature positions, and triangles indicate possible feature positions. Note the seeming feature near the Fermi energy could be due to the Fermi cutoff.

## 3. Results and Discussion

The doping range of the existing Bi2223 samples is limited, unlike Bi2212, heavily overdoped regime of Bi2223 has not been reached. This conspires the immediate experimental difficulty in the study of the intra-trilayer coupling of Bi2223. For optimally doped Bi2223, ARPES spectra and the measured Fermi surface appear to only contain one component,[7] instead of three separate features corresponding to the bonding, non-bonding and anti-bonding bands as expected from the intra-trilayer coupling. However, this does not necessarily mean that intra-trilayer coupling is negligible: similar behavior has also been observed in optimally doped Bi2212, although there exist sizable intra-bilayer coupling.[8] Due to the anisotropic

nature of the intra-bilayer coupling, the amplitude of the bilayer splitting is also anisotropic with zero splitting in the nodal region and maximum splitting at $(\pi, 0)$. As demonstrated in Fig. 1, the normal state $(\pi, 0)$ photoemission spectrum of heavily overdoped system ($T_c = 65$ K) clearly shows two features that exhibit a normal state peak-dip-hump structure, and are assigned to the two bilayer split bands.[5,6] This was not observed in previous measurements on overdoped samples, mainly due to extrinsic factors such as energy and angular resolution. With a slight decrease of the doping (overdoped, $T_c = 72$ K), the two components of the $(\pi, 0)$ spectrum are barely distinguishable. Compared with the $T_c = 65$ K system, the two features become broader and their intensities smaller. For optimally doped system ($T_c = 90$ K), the spectrum is *intrinsically* too broad to distinguish the two split features, which makes the identification of the bilayer splitting very difficult in this manner. Therefore, one needs to look for other signs of bilayer splitting. In a recent study,[8] by comparing the spectra of Bi2212 with those of single layer Bi2201, it has been shown that the presence of incipient bilayer splitting can be addressed also on the basis of more subtle effects: even when only one broad envelope (instead of two clear peaks) is detected, its centroid and overall lineshape would change significantly with photon energy. This is because the split bands have different symmetry along the *c*-axis, and thus different dependence of the matrix elements on the final-state electron wavelength [see Fig. 2(b)].[8] On the basis of this behavior , the signature of bilayer splitting in optimally doped and even moderately underdoped Bi2212 was recognized,[8] which also naturally explains the broad linewidth of the Bi2212 spectra near $(\pi, 0)$, as it contains two features [see Fig. 2(d)].

Similar to the case of the bilayer splitting, one can also study the intra-trilayer coupling or trilayer splitting in optimally doped Bi2223 based on the photon energy dependence data and lineshape analysis. In Fig. 2(a)–(c) the normal state $(\pi, 0)$ spectra from optimally doped Bi2201, Bi2212 and Bi2223 are presented. Contrary to the case of Bi2212 discussed above, on Bi2201 the feature position is independent

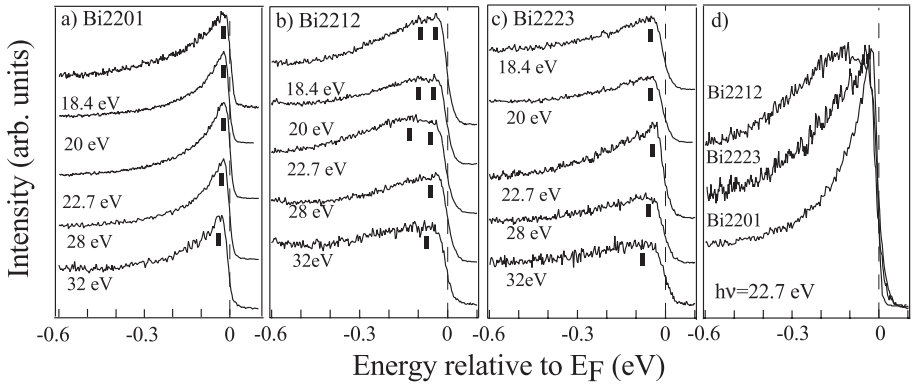


Fig. 2. Photon energy dependence of the normal state $(\pi, 0)$ ARPES spectra for optimally doped: (a) Bi2201 at 40 K ($T_c = 33$ K); (b) Bi2212 at 110 K ($T_c = 90$ K); (c) Bi2223 at 125 K ($T_c = 108$ K). (d) Direct comparison of the $(\pi, 0)$ ARPES spectra taken with 22.7 eV photons (from panels a-c).

of the photon energy. The slow change in the overall spectral lineshape may be due to photon energy dependence of the in-plane photoemission matrix element and/or background. This is in agreement with the absence of multi-band structure in Bi2201. Furthermore on Bi2223, even though multi-layer splitting is expected, the spectral centroid position does not show as significant changes with photon energy as on Bi2212. This is further confirmed by the direct comparison of the $(\pi, 0)$ spectra from the three systems taken with 22.7 eV photons, where the spectra are sharpest among other photon energies, and backgrounds are relatively the weakest. It is found that [see Fig. 2(d)] the lineshape for Bi2223 is intermediate between those of Bi2201 (sharpest) and Bi2212 (broadest). Therefore, these results suggest that the trilayer splitting effects in Bi2223, though not totally absent, are at least weaker than the bilayer splitting effects in Bi2212.[9]

The weakness of the trilayer effects in Bi2223 could be understood by considering the differences in chemical environment among the three $CuO_2$ planes. Measurements of the $^{17}O$ Knight shift by nuclear magnetic resonance showed that the inner and outer layers of Bi2223 have different hole concentrations[10]: it was argued that the inner-layer hole density is 15% to 25% less than that of the outer layers (similar conclusions were also drawn for other $n \geq 3$ HTSC's.[11]) Also ionic-model calculations of the charge distribution among the three $CuO_2$ layers indicate that the inner layer is underdoped[12] or even depleted of holes.[1] Empirically, it was shown that it is energetically unfavorable to hole-dope a $CuO_2$ plane with no apical oxygen.[13] Because the inner layer is underdoped, electronic correlations are strong and will severely reduce the *effective* hopping between the outer and inner layers.[5,14] In other words, the inner layer serves as a separator between the two outer layers, and trilayer splitting effects are thereby weakened.

If the coupling between neighboring $CuO_2$ planes within each unit cell is not responsible for the $T_c$ enhancement with $n$, one might need to search for other factors that can alter $T_c$ such as chemical and/or structural changes near the $CuO_2$ planes. Recent material study[15] shows that the different ionic radius of impurities in cuprates alters $T_c$ dramatically. It is also intriguing to note that among the single-layer systems the highest $T_c$ (95 K) is found in the single-layer Hg-based cuprate, which is also the material characterized by the flattest $CuO_2$ plane.

Alternatively, another subtle scenario was proposed recently to explain the $T_c$ enhancement in Bi2223.[16] Based on the proximity effect, Cooper pairs in Bi2223 may hop to the inner layer to obtain stronger pairing and the outer layer to obtain stronger phase coherence, although the interlayer hopping probability might not be as strong as in the bilayer system. This agrees with recent experimental findings,[7] where both pairing and coherence strengths in Bi2223 are found to be enhanced, consistent with its higher $T_c$.

## 4. Conclusion

In summary, based on photon energy dependence of the ARPES spectra taken on Bi2201, Bi2212, and Bi2223, we have shown that the coupling within the multi-layer

seems to be weaker in Bi2223 than in Bi2212. The opposite trend between the intra-multi-layer coupling and $T_c$ in going from Bi2223 to Bi2212 suggests that other factors such as detailed structural difference between these materials or more complicated schemes should be examined to understand the large $T_c$ difference in different cuprates.

## Acknowledgments

SSRL is operated by the US DOE Office of Basic Energy Science Divisions of Chemical Sciences and Material Sciences. The Material Sciences Division also provided support for the work. The Stanford ARPES experiments are also supported by the NSF grant DMR-0071897 and ONR grant N00014-98-1-0195-A00002. The crystal growth work at Stanford was supported by DOE under Contract Nos. DE-FG03-99ER45773-A001 and DE-AC03-76SF00515. MG is also supported by the A. P. Sloan Foundation. GDG is supported by DOE under contract No. DE-AC02-98CH10886 DLF, HE and ZXS wish to thank S. Maekawa, T. Geballe and S. Kivelson for helpful discussions.

## References

1. M. Di Stasio, K. A. Muller and L. Pietronero, *Phys. Rev. Lett.* **64**, 2827 (1990), and references therein.
2. J. M. Tarascon *et al.*, *Phys. Rev.* **B38**, 8885 (1992).
3. O. K. Anderson *et al.*, *J. Phys. Chem. Solids* **12**, 1573 (1995); A. I. Lichtenstein *et al.*, *Phys. Rev.* **B54**, 12505 (1996).
4. Z.-X. Shen and D. S. Dessau, *Phys. Rep.* **253**, 1 (1995); A. Damascelli, D. H. Lu and Z.-X. Shen, *J. Electron Spectr. Relat. Phenom.* **117–118**, 165 (2001); D. W. Lynch and C. G. Olson, *Photoemission Studies of High-Temperature Superconductors* (Cambridge University Press, 1999).
5. D. L. Feng *et al.*, *Phys. Rev. Lett.* **86**, 5550 (2001).
6. Y.-D. Chung *et al.*, *Phys. Rev. Lett.* **87**, 117002 (2002).
7. D. L. Feng *et al.*, *Phys. Rev. Lett.* **88**, 107001 (2002).
8. D. L. Feng *et al.*, *Phys. Rev. B* (rapid communication, in press 2002).
9. We note that photemission matrix element effects are very complicated in real materials, and there might be other unexpected complications, such as those due to the finite probing depth of ARPES (see M. Mori *et al.* preprint cond-matt/0110050). To undoubtedly prove this point, further experiments on heavily overdoped Bi2223 are needed, where split bands could presumably be separated from each other.
10. A. Trokiner *et al.*, *Phys. Rev.* **B44**, 2426 (1991).
11. Y. V. Piskunov *et al.*, *Physica* **C300**, 225 (1998); Y. Tokunaga *et al.*, *Phys. Rev.* **B61**, 9707 (2000).
12. R. P. Gupta and M. Gupta, *Phys. Rev.* **B49**, 13154 (1994).
13. Y. Tokura *et al.*, *Phys. Rev.* **B39**, 9704 (1989); *Nature* **337**, 345 (1989).
14. R. Eder, Y. Otha and S. Maekawa, *Phys. Rev.* **B51**, 3265 (1995).
15. H. Eisaki *et al.*, to be published.
16. S. Kivelson *et al.*, to be published.