ISSN 1330–0008
CODEN FIZAE4

LETTER TO THE EDITOR

DEFECT-INDUCED CHANGES IN THE SPECTRAL PROPERTIES OF HIGH-T_c CUPRATES

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Dedicated to Professor Boran Leontić on the occasion of his 70th birthday

Received 9 November 1999; revised manuscript received 1 February 2000
Accepted 14 February 2000

Superconductivity in high-T_c cuprates is particularly sensitive to disorder due to the unconventional d-wave pairing symmetry. We investigated effects of disorder on the spectral properties of $Bi_2Sr_2CaCu_2O_{8+x}$ high-T_c superconductor. We found that already small defect densities suppress the characteristic spectral signature of the superconducting state. The spectral line shape clearly reflects new excitations within the gap, as expected for defect-induced pair breaking. At the lowest defect concentrations the normal state remains unaffected, while increased disorder leads to suppression of the normal quasiparticle peaks.

PACS numbers: 74.25.-q, 74.62.Dh, 74.72.Hs UDC 538.945

Keywords: superconductivity, high-T_c cuprates, effects of disorder, spectral properties, $Bi_2Sr_2CaCu_2O_{8+x}$, defect-induced pair breaking, suppression of the normal quasiparticle peaks

The superconducting (SC) order parameter in high-T_c cuprates is highly anisotropic. A number of experimental results indicate that the carriers in the

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SC state pair with the $d_{x^2-y^2}$ symmetry. The unconventional pairing stimulates interest in finding how disorder affects superconductivity in these materials. In low-T_c superconductors, with isotropic pairing symmetry, magnetic defects lead to a rapid suppression of T_c and ultimately destruction of superconductivity [1,2]. If the order parameter is anisotropic, both magnetic and nonmagnetic defects are harmful for superconductivity [3–7].

These theoretical predictions can be tested in experiments on samples with controlled amounts of introduced disorder [8,9]. Chemical substitution in the CuO_2 planes may perturb the local antiferromagnetic order, resulting in magnetic scattering centers [10–12]. Electron irradiation, on the contrary, introduces homogeneously distributed Frenkel-type point defects [8,9], most likely of nonmagnetic character [9].

We report here the results of a systematic angle-resolved photoemission (ARPES) investigation of the effects of disorder on the spectral properties of electron-irradiated $Bi_2Sr_2CaCu_2O_{8+x}$. In the SC state, a residual density of states at the Fermi level, due to the defect-induced pair breaking, is clearly observed [13]. At the lowest defect concentrations, only the superconducting state is affected. At increased concentrations, however, the observed changes in the normal state quasiparticle features are similar to the ones found in highly underdoped regime [14].

Our samples were grown by the flux method [15]. The electron irradiation was performed on a van de Graaff accelerator with the irradiation flux fixed to 2×10^{14} e/(cm^{2}s) and the electron energy of 2.5 MeV. The electron irradiation in these conditions displaces mainly Cu and O atoms, resulting in defect concentrations confined to the low 10^{-3} dpa (displacements-per-atom) range at constant carrier concentration [8,9,13]. Depending on the irradiation time, we obtained critical temperatures of 82, 72 and 62 K, as opposed to 90 K before the irradiation.

ARPES experiments were performed with a Scienta 300 electrostatic hemispherical analyzer whose energy resolution is better than 10 meV and the angular acceptance is $\pm 1^\circ$. Photons of 21.2 eV were used, which gives a total k-space window of $0.4\,\mathrm{nm}^{-1}$. The high symmetry directions were determined by the Laue X-ray diffraction. The samples were cleaved in-situ at the base pressure better than 2.7×10^{-8} Pa (2×10^{-10} torr). The SC-state measurements were performed at 25 K and normal state data were taken at 95 K.

Figure 1 illustrates the SC-state spectra for the three irradiated samples and for the pristine sample (each specimen is identified by its T_c). All data were taken close to the $(0,\pi)$ point in the Brillouin zone where a $d_{x^2-y^2}$ gap exhibits a maximum. The signature of the SC state in ARPES is a coherent peak at a binding energy approximately corresponding to the half-magnitude of the SC gap, followed by a dip and a hump features at larger binding energies [16–18]. The leading edge of the spectra is limited only by the experimental resolution.

All spectra in Fig. 1a exhibit the superconductivity-related features. The coherent peak is at the same binding energy for all samples, consistent with the constant and optimal carrier concentration [13]. The peak intensity decreases as T_c decreases, i.e., as the amount of disorder increases. The leading edge is no longer resolution-limited. It extends instead beyond the Fermi energy, suggesting new states within the gap. This is emphasized in Fig. 1b, which illustrates the near-edge portion of the spectra compared to a reference 90 K spectrum. The shaded area in Fig. 1(b) corresponds to the gap states and clearly increases with the induced disorder beyond the experimental uncertainty.

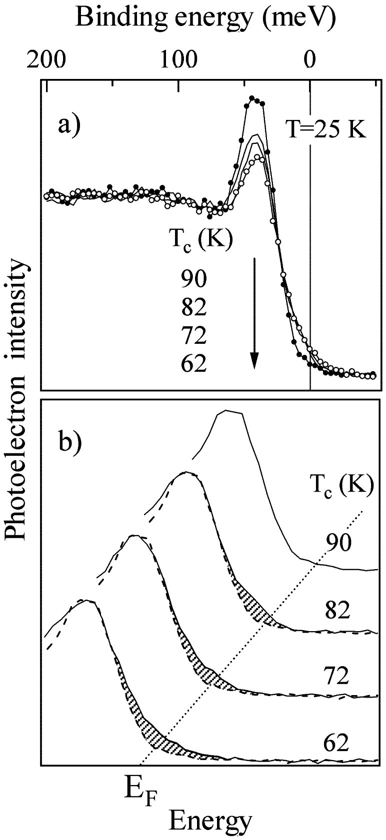

Fig. 1. (a) Superconducting state spectra for the pristine (T_c = 90 K) and irradiated (T_c = 82 K, 72 K and 62 K) samples, normalized at the highest binding energy; (b) Near-edge portion of the same spectra normalized to the coherent peak and compared to a reference pristine T_c = 90 K) spectrum. The shaded area corresponds to the new states within the SC gap, created through the defect-induced pair breaking.

The spectral feature near E_F reflects normal quasiparticle (QP) states, which obey the Fermi-Dirac distribution. They coexist with the SC condensate below T_c, as expected for the defect-induced pair-breaking. A similar spectral feature was recently reported in the scanning tunneling measurements of $Bi_2Sr_2CaCu_2O_{8+x}$, where it was attributed to the quasiparticle scattering resonances [19]. There is a good agreement between our results and the theoretical predictions of Ref. [7], which analyzed the effects of nonmagnetic defects on the SC state spectral function. At our low defect concentrations ($< 10^{-3}$ dpa), Ref. [7] argues that the observed spectral changes are due to the resonant scattering, previously investigated in heavy-fermion superconductors [20–23]. The concept of the resonant scattering is important for the cuprates, since it rules out pairing without nodes in the gap

[7,24].

Figures 2 and 3 illustrate the normal state dispersion along the Γ-M (0, π) and Γ-X (π, $-\pi$) high-symmetry directions. The pristine 90 K sample exhibits all features found in previous ARPES studies [16,18]. Along Γ-X, a fast dispersing state crosses the Fermi surface. Along Γ-M-Z, no crossing occurs; the quasiparticle states remain just below the chemical potential, forming a flat band or an extended van Hove singularity.

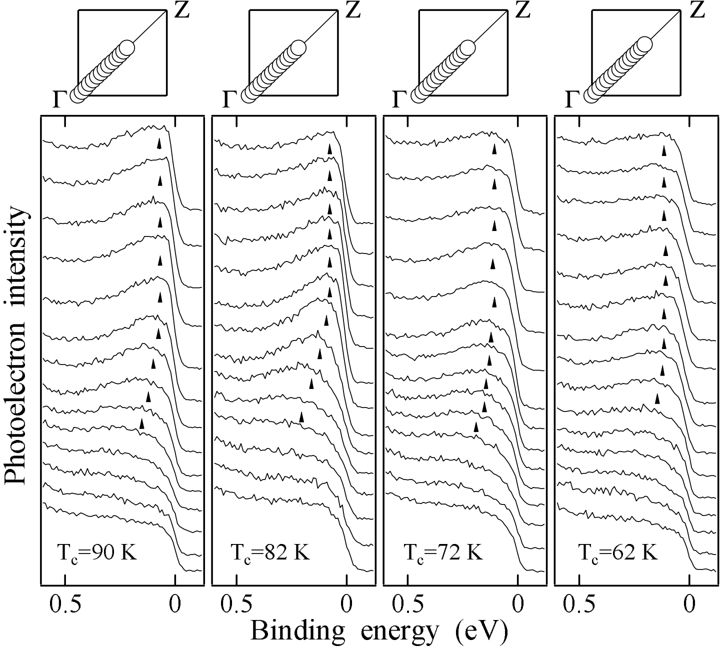

Fig. 2. Normal state spectra along the Γ*-M (0,* π*) high-symmetry direction for the pristine* $T_c = 90$ K *sample and the irradiated* $T_c = 82$ K*, 72* K*, 62* K *samples. The insets indicate the locations in the Brillouin zone where the spectra were taken.*

These normal-state characteristics remain unchanged for the 82 K sample - in contrast to the significant difference in the SC state (Fig. 1). On going from the 82 K to the 72 K and to the 62 K samples, the normal-state properties change dramatically. The most pronounced is the loss of coherent spectral intensity. The spectral features become broader and their dispersion cannot be easily traced.

We found similar behaviour in the normal state of neutron-irradiated $Bi_2Sr_2CaCu_2O_{8+x}$, where the quasiparticle features were completely suppressed at the highest disorder levels [25]. The normal state behaviour [14] of underdoped $Bi_2Sr_2CaCu_2O_{8+x}$ also resembles that of the irradiated samples. The suppression of the quasiparticle peaks in the underdoped regime was initially attributed uniquely to reduced carrier concentration and increased electronic correlations. Our data suggest instead that disorder introduced through doping could be, at least in part, responsible for the peculiar spectral properties of underdoped cuprates. In fact, doping is usually achieved by chemical substitution or oxygen reduction, both affecting the CuO_2 planes and likely to increase disorder.

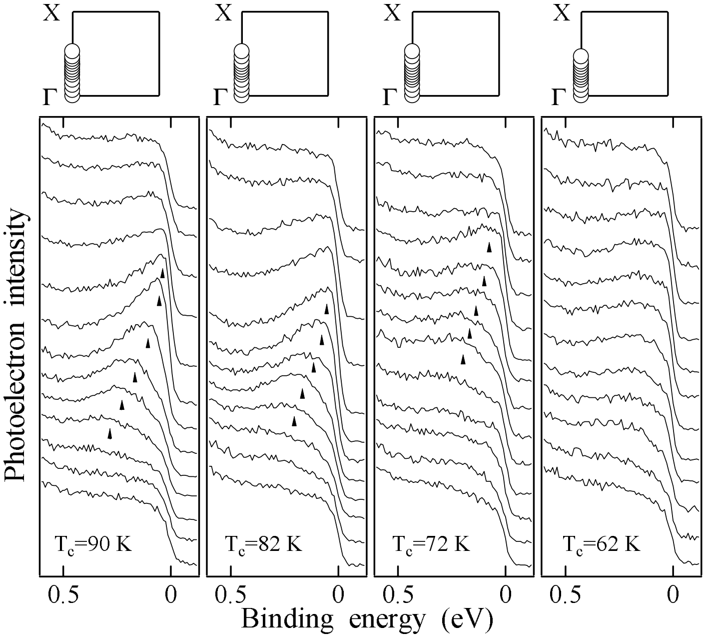

Fig. 3. Same as Fig. 2, but for the Γ-*X* $(\pi, -\pi)$ *direction.*

In summary, our ARPES data provide a first direct evidence for defect-induced pair-breaking in a d-wave superconductor. The loss of the coherent-peak intensity in the SC-state spectra is accompanied by an increase of the residual density of states at the Fermi level, as expected for coexisting normal and superconducting carriers due to the pair-breaking events. In the normal state, disorder causes a dramatic suppression of the quasiparticles features, which qualitatively resembles the spectral evolution in highly-underdoped cuprates. These findings call for a reconsideration of the effects of disorder in the cuprates, in particular at low carrier concentrations.

Acknowledgements

This work was supported by the Fond National Suisse de la Recherche Scientifique and by the Ecole Polytechnique Fédérale de Lausanne.

References

1) A. A. Abrikosov and L. P. Gor'kov, Soviet Phys. JETP **12** (1961) 1243;

2) P. W. Anderson, J. Phys. Chem. Solids **11** (1959) 26;

3) K. Ueda and T. M. Rice, in *Theory of Heavy Fermions and Valence Fluctuations*, eds. T. Kasuya and T. Saso, Springer Verlag, Berlin (1985), p. 267;

4) R. J. Radtke, K. Levin, H.-B. Schttler and M. R. Norman, Phys. Rev. B **48** (1993) R653;

5) K. Ziegler, M. H. Hettler and P. J. Hirschfeld, Phys. Rev. Lett. **77** (1996) 3013;

6) K. Ziegler, M. H. Hettler and P. J. Hirschfeld, Phys. Rev. B **57** (1998) 10825;

7) R. Fehrenbacher, Phys. Rev. B **54** (1996) 6632;

8) F. Rullier-Albenque, A. Legris, H. Berger and L. Forró, Physica **254** (1995) 88;

9) J. Giapintzakis, D. M. Ginsberg, M. A. Kirk and S. Ockers, Phys. Rev. B **50** (1994) 15967;

10) H. Alloul, P. Mendels, H. Casalta, J. F. Marucco and J. Arabski, Phys. Rev. Lett. **67** (1991) 3140;

11) A. V. Mahajan, H. Alloul, G. Collin and J. F. Marucco, Phys. Rev. Lett. **72** (1994) 3100;

12) T. R. Chien, Z. Z. Wang and N. P. Ong, Phys. Rev. Lett. **67** (1991) 2088;

13) I. Vobornik, H. Berger, D. Pavuna, M. Onellion, G. Margaritondo, F. Rullier-Albenque, L. Forró and M. Grioni, Phys. Rev. Lett. **82** (1999) 3128;

14) H. Ding, M. R. Norman, T. Yokoya, T. Takeuchi, M. Randeria, J. C. Campuzano, T. Takahashi, T. Mochiku and K. Kadowaki, Phys. Rev. Lett. **78** (1997) 2628;

15) D. Mandrus, L. Forr, C. Kendziora and L. Mihaly, Phys. Rev. B **45** (1992) 12640;

16) Z.-X. Shen and D. S. Dessau, Physics Reports **253** (1995) 1;

17) H. Ding, M. R. Norman, J. C. Campuzano, M. Randeria, A. F. Bellman, T. Yokoya, T. Takahashi, T. Mochiku and K. Kadowaki, Phys. Rev. B **54** (1996) R9678;

18) M. Randeria and J. C. Campuzano, *Varenna Lectures 1997*, Report No. LANL cond-mat/9709107, and references therein;

19) E. W. Hudson, S. H. Pan, A. K. Gupta, K.-W. Ng and J. C. Davis, Science **285** (1999) 88;

20) C. J. Pethick and D. Pines, Phys. Rev. Lett. **57** (1986) 118;

21) S. Schmitt-Rink, K. Miyake and C. M. Varma, Phys. Rev. Lett. **57** (1986) 2575;

22) P. J. Hirschfeld, P. Wölfle and D. Einzel, Phys. Rev. B **37** 1988) 83;

23) H. Monien, K. Scharnberg and D. Walker, Solid State Commun. **63** (1987) 263;

24) R. Fehrenbacher, Phys. Rev. Lett. **77** (1996) 1849;

25) I. Vobornik, C. Quitmann, M. Zacchigna, F. Zwick, M. Grioni, A. Karkin, R. J. Kelley, M. Onellion and G. Margaritondo, Surf. Sci. **402-404** (1998) 761.

PROMJENE SPEKTRALNIH SVOJSTAVA KUPRATA VISOKOG T_c IZAZVANE DEFEKTIMA

Zbog nekonvencionalne d-valne simetrije, supravodljivost u visokotemperaturnim kupratima je posebno osjetljiva na neuređenost. Ispitivali smo utjecaj neuređenosti na spektralna svojstva $\mathrm{Bi_2Sr_2CaCu_2O_{8+x}}$ supravodiča primjenom ARPES metode. Već mala gustoća defekata smanjuje karakteristični spektralni odziv supravodljivog stanja. Oblik spektralne linije očito reflektira nova pobuđenja unutar zabranjene vrpce, kako se i očekuje u slučaju razbijanja Cooperovih parova izazvanog defektima. Za male koncentracije defekata, normalno stanje ostaje nepromijenjeno, dok viši stupanj neuređenosti smanjuje intenzitet spektralnih linija kvazičestica.