Alternative pseudogap scenario: Spectroscopic analogies between underdoped and disordered $Bi_2Sr_2CaCu_2O_{8+x}$

I. Vobornik, H. Berger, M. Grioni, and G. Margaritondo
Institut de Physique Appliquée, Ecole Polytechnique Fédérale, CH-1015 Lausanne, Switzerland

L. Forró
Laboratoire de Physique des Solides Semicristallins, IGA, Ecole Polytechnique Fédérale, CH-1015 Lausanne, Switzerland

F. Rullier-Albenque
Laboratoire des Solides Irradiés, CEA, Ecole Polytechnique, 91128 Palaiseau, France
(Received 17 November 1999)

Disorder has a strong influence on the spectral properties of the $Bi_2Sr_2CaCu_2O_{8+x}$ high-T_c superconductor, both in the normal and in the superconducting state. High-resolution photoemission reveals a progressive quasiparticle suppression with increasing disorder in electron-irradiated optimally doped single crystals. The spectral line shapes of the disordered samples show striking analogies with those of underdoped samples specifically the widely discussed pseudogap at the chemical potential. Disorder provides therefore an alternative mechanism for spectral weight suppression near the Fermi surface, possibly coexisting and competing with other mechanisms. Since doping unavoidably introduces disorder, these results require a reconsideration of the relative importance of intrinsic effects vs disorder in high temperature superconductivity.

Superconductivity in the cuprates is remarkably sensitive to stoichiometry and the density of carriers, n. The highest transition temperature T_c identifies an optimum doping level, $n_{\rm opt}$, which separates overdoped (OD,$n>n_{\rm opt}$) and underdoped (UD,$n<n_{\rm opt}$) regions of the phase diagram. For OD samples, both T_c and the superconducting (SC) gap $\Delta_{\rm SC}$ decrease as n increases; for UD samples, lower n's give a smaller T_c but a larger $\Delta_{\rm SC}$, reflecting a stronger pairing. UD and OD samples are also different in the normal state. OD cuprates exhibit a metallike Fermi surface in angle-resolved photoemission (ARPES), and well-defined quasiparticle peaks. For UD samples the quasiparticle features are strongly suppressed indicating strong correlations, and there is evidence[1] for a normal-state pseudogap, Δ^*, for $T_c<T<T^*$. Various experiments and theoretical models suggest a relation between pseudogap and superconductivity.[2–5] We show, however, that the peculiar UD spectral signatures are also present in optimally doped samples, for which T_c is reduced by controlled disorder. This surprising result requires a careful reassessment of the role of disorder in the cuprates.

Superconductors with anisotropic pairing symmetry like the high-T_c cuprates are particularly sensitive to disorder. Even non-magnetic impurities, which have negligible effects in conventional superconductors, act as strong pair breakers and rapidly suppress superconductivity if the order parameter has nodes. This was confirmed by recent results on disordered $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212).[6] High-resolution ARPES spectra reveal a residual density of states in the SC gap, induced by the pair-breaking events.[7–9]

Disorder can also affect the normal state. In UD cuprates, due to the reduced carrier concentration and strong two-dimensional character, defects may even lead to localization. Various experiments suggest a superconductor-insulator transition at low doping.[10–12] Zn substitution in underdoped $YBa_2Cu_3O_{7-y}$ and $La_{2-x}Sr_xCuO_4$, as well as electron irradiation in $YBa_2Cu_3O_{7-y}$ and $Tl_2Ba_2CuO_{6+x}$, reduce T_c and eventually force a transition to an insulating state.[13,14] Magnetotransport in insulating and underdoped $Bi_2Sr_2Ca_{1-z}Pr_zCu_2O_{8+x}$ indicate the localization of carriers near E_F.[15] These findings are quite relevant because doping in high-T_c cuprates is obtained either by cation substitution or by varying the oxygen concentration—and both procedures introduce disorder. We identified the characteristic ARPES spectral signatures of disorder by investigating irradiated optimally doped Bi-2212.

The Bi-2212 single crystals were heat-treated in an O_2 atmosphere, to obtain an identical oxygen stoichiometry and T_c of 90 K. Overdoped samples went through an additional annealing at 4 kbar O_2 pressure, resulting in T_c values between 57 and 62 K. Underdoping was performed by substituting divalent Ca by trivalent Pr. The details of the electron irradiation of optimally-doped Bi-2212 were described elsewhere.[6] The resulting defect concentrations in the CuO_2 planes were in the 10^{-3} dpa (displacements per atom) range. By varying the irradiation fluences we obtained T_c's of 82, 72, and 62 K. ARPES experiments were performed by a Scienta electrostatic hemispherical analyzer with an energy and momentum resolution $\Delta E=10$ meV and $\Delta k=0.04$ Å^{-1}. The samples were oriented by Laue x-ray diffraction, then transferred into the UHV system and cleaved *in situ* at a base pressure better than 1×10^{-10} torr. The SC state measurements were performed at 25 K and the normal state data were taken at $T=1.1$-$1.4T_c$.

Figure 1 shows the resistivity curves for different doping levels from highly OD ($T_c=62$ K) to nearly optimally doped ($T_c=90$ K) and to UD ($T_c=84$, 75, and 56 K). The resistivity increases with decreasing carrier concentration.

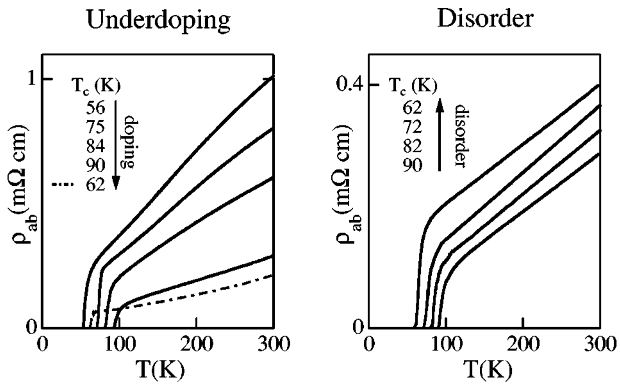

FIG. 1. Resistivity vs temperature; left: for different carrier concentrations [$T_c=62$ K (OD), 90 K (optimal doping), and 84, 75, 56 K (UD)]; right: for the pristine ($T_c=90$ K) and irradiated ($T_c=82$, 72, and 62 K) optimally doped samples. UD samples were obtained by substituting divalent Ca with trivalent Pr, and OD samples by annealing in a 4 kbar oxygen atmosphere. The disorder-inducing electron irradiation of optimally-doped Bi-2212 was described in Ref. 6, and produced defect concentrations in the CuO_2 planes in the 10^{-3} dpa (displacements per atom) range.

The temperature dependence is approximately linear at optimum doping, and exhibits the usual *S*-shape anomalies in the UD regime. The extrapolation of the high-T resistivity to $T=0$ K yields residual resistivity values which grow with underdoping. Figure 1 also illustrates the data for the electron-irradiated samples.[6] All curves exhibit a linear dependence with similar slope, indicating that irradiation did not modify the carrier concentration, as required to distinguish the effects of disorder from those of doping.

The ARPES spectra of Fig. 2 illustrate the effects of doping and disorder in the normal state. As T_c decreases, one sees a progressive reduction of the coherent spectral weight near E_F indicating a pseudogap.[1,3] This is true for both UD and irradiated samples. The suppression of the quasiparticle peaks does not eliminate the Fermi surface crossing along ΓY (not shown). Following the standard practice,[1] we derived the pseudogap magnitude from the midpoint of the spectral leading edge. There is a striking similarity between the two sets of data, with the pseudogap size tracking the decrease of T_c.

The SC-phase ARPES line shape also strongly varies with T_c. Figure 3 shows the dependence on doping, from strongly OD ($T_c=57$ K), to strongly UD ($T_c=56$ K) Bi-2212. All spectra were measured near the M point where the SC gap size is maximum. The SC condensate peak, which tracks the low-energy scale Δ_{SC}, is largest for the most overdoped sample and it decreases with decreasing doping. It also progressively moves away from E_F, reflecting an increasing pairing strength.

The two extreme spectra of Fig. 3(a) are compared in Fig. 3(b) with a silver reference measured at the same temperature. The leading edge of the 57 K (OD) spectrum is resolution limited. By contrast, in the UD sample it is much broader, and extends into the gap all the way to E_F. Figure 3(c) shows that the SC condensate peak is also progressively reduced by disorder.[6] Nevertheless, the peak position remains constant indicating a constant pairing strength. At the higher irradiation doses, a weak feature appears at the spec-

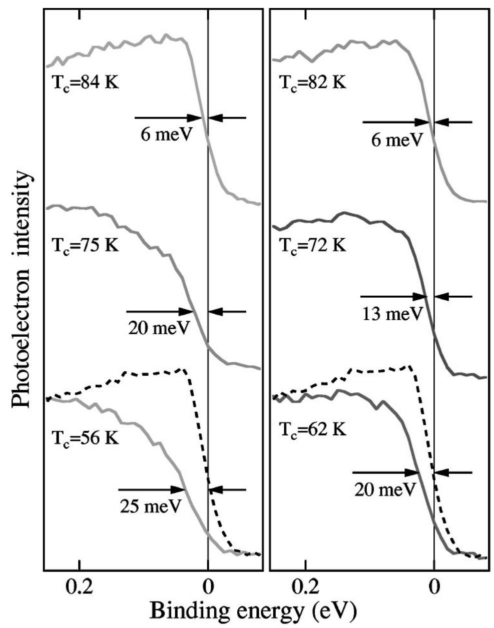

FIG. 2. Doping (left) and disorder (right) dependence of the ARPES spectra for samples of Fig. 1. The spectra were taken at the Fermi surface crossing along the M-Y direction, where the d-wave gap is maximum, in the normal state, at $T=77$ K ($T_c=56$, 75 K) or $T=94$ K ($T_c=62$, 72, 82, 90 K). Dashed line: spectrum of optimally doped ($T_c=90$ K), pristine Bi-2212.

tral onset near E_F. This reflects normal carriers induced by pairbreaking scattering on defects, in agreement with theoretical models of resonant scattering in disordered superconductors.[9] A similar spectral feature was recently reported for scanning tunneling measurements of Bi-22121.[16,17]

The data reveal strong analogies between doping and disorder: (i) the appearance of a pseudogap; (ii) the suppression of quasiparticles in the normal state; (iii) the reduction of the condensate peak in the SC state. The main difference is the invariance of the SC peak energy in the irradiated crystals, reflecting a constant carrier concentration. The experimental facts are summarized in Fig. 4. For both disordered and underdoped samples, the pseudogap size Δ^* [Fig. 4(a)] exhibits a similar monotonic increase as T_c decreases. The T_c dependence of the SC gap Δ_{SC} is illustrated in Fig. 4(b). The gap size was estimated from the binding energy of the condensate peak, the procedure which yields the best agreement

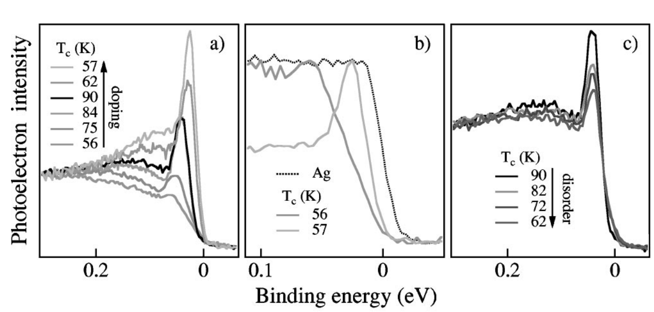

FIG. 3. Evolution of the superconducting state spectra with doping (a) and disorder (c); (b) the OD (57 K) and UD (56 K) spectra, normalized to the peak intensity to enhance line shape differences, are compared to an Ag metal reference.

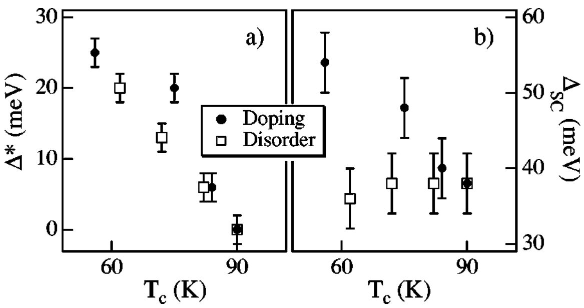

FIG. 4. Pseudogap (a) and superconducting gap (b) values vs T_c for underdoped and disordered samples.

with tunneling measurements. In the irradiated samples Δ_{SC} is nearly constant, as suggested by the raw spectra of Fig. 3(c). For nonirradiated samples, Δ_{SC} increases with underdoping.

The crucial question is: do the spectral features of UD and disordered samples have the same origin? Impurities act as resonant scatterers in the cuprates,[9,18,19] and it is known that resonant scattering can induce a pseudogap at E_F.[20] By contrast, in complementary experiments we found that electron irradiation of a normal metal reduces the quasiparticle peaks but does not open a pseudogap. As to the SC state, the momentum dependence of Δ_{SC} in strongly UD samples deviates from pure *d*-wave behavior near the nodes[1,21] as predicted for disordered samples (dirty *d*-wave scenario).[9] Also consistent with the dirty *d*-wave scenario is the fact that the leading edge of the UD samples extends up to E_F reflecting states in the gap as in disordered samples.

All this does suggest that disorder contributes to the reduction of the critical temperature from T^* to T_c in the UD cuprates. The effects of disorder become stronger away from optimal doping. An irradiation defect density $\sim 10^{-3}$ dpa in the CuO_2 planes reduces T_c by 30% for optimal doping, whereas underdoping at the levels considered here involves a density of substitutional impurities at least 10 times larger. These impurities are probably not as effective as the in-plane defects produced by irradiation, but they do yield a higher residual resistivity (Fig. 1). Moreover, the low carrier density and the two-dimensional character reduce the screening of the impurity potential, thus making strongly UD materials more sensitive to defects.[22]

In summary, the spectral properties of optimally doped irradiated Bi-2212 are markedly similar to those of underdoped samples. Our results show that disorder can significantly contribute to the spectral properties in the UD regime and this cannot be ignored when analyzing the pseudogap and its role in high-T_c superconductivity. More generally, our data show that defect-induced pair breaking could play a significant but so far neglected role in the reduction of the critical temperature from T^* to T_c in the UD regime. It would be extremely interesting to perform a further quantitative evaluation of this effect in epitaxial thin films, where the carrier density can be tuned electrically, without introducing structural defects.[23]

We acknowledge useful discussions with Dr. D. Ariosa. This work was supported by the Swiss National Science Foundation and by the Ecole Polytechnique Fdrale de Lausanne.

[1] H. Ding *et al.*, Nature (London) **382**, 21 (1996).
[2] N. Miyakawa, P. Guptasarma, J.F. Zasadzinski, D.G. Hinks, and K.E. Gray, Phys. Rev. Lett. **80**, 157 (1998); C. Renner, B. Revaz, J.-Y. Genoud, K. Kadowaki, and O. Fischer, *ibid.* **80**, 149 (1998).
[3] A.G. Loeser *et al.*, Science **273**, 325 (1996).
[4] M.R. Norman *et al.*, Nature (London) **392**, 157 (1998).
[5] S. Kivelson and V. Emery, Nature (London) **374**, 434 (1995).
[6] I. Vobornik *et al.*, Phys. Rev. Lett. **82**, 3128 (1999).
[7] Y. Sun and K. Maki, Phys. Rev. B **51**, 6059 (1995).
[8] P.J. Hirschfeld, W.O. Putikka, and D.J. Scalapino, Phys. Rev. B **50**, 10 250 (1994).
[9] R. Fehrenbacher, Phys. Rev. B **54**, 6632 (1996).
[10] G.S. Boebinger *et al.*, Phys. Rev. Lett. **77**, 5417 (1996).
[11] D.N. Basov, B. Dabrowski, and T. Timusk, Phys. Rev. Lett. **81**, 2132 (1998).
[12] G.A. Levin *et al.*, Phys. Rev. Lett. **80**, 841 (1998).
[13] Y. Fukuzumi, K. Mizuhashi, K. Takenaka, and S. Uchida, Phys. Rev. Lett. **76**, 684 (1996).
[14] F. Rullier-Albenque, P.A. Vieillefond, P. Lejay, and A.W. Tyler, cond-mat/9908405 (unpublished).
[15] B. Beschoten, S. Sadewasser, G. Guntherodt, and C. Quitmann, Phys. Rev. Lett. **77**, 1837 (1996).
[16] A. Yazdani, C.M. Howald, C.P. Lutz, A. Kapitulnik, and D.M. Eigler, Phys. Rev. Lett. **83**, 176 (1999).
[17] E.W. Hudson, S.H. Pan, A.K. Gupta, K.-W. Ng, and J.C. Davis, Science **285**, 88 (1999).
[18] P.J. Hirschfeld and N. Goldenfeld, Phys. Rev. B **48**, R4219 (1993).
[19] D. Poilblanc, D.J. Scalapino, and W. Hanke, Phys. Rev. Lett. **72**, 884 (1994).
[20] B. Janko, J. Maly, and K. Levin, Phys. Rev. B **56**, R11 407 (1997).
[21] J.M. Harris *et al.*, Phys. Rev. B **54**, R15 665 (1996).
[22] J. Maly, K. Levin, and D.Z. Liu, Phys. Rev. B **54**, R15 657 (1996).
[23] C.H. Ahn *et al.*, Science **284**, 1152 (1999).