## **Tunneling spectroscopy of the quasiparticle Andreev bound state** in ion-irradiated  $YBa_2Cu_3O_{7-\delta}/Pb$  junctions

M. Aprili,\* M. Covington,† E. Paraoanu, B. Niedermeier, and L. H. Greene

*Loomis Laboratory of Physics, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801*

(Received 18 November 1997)

Effects of disorder on the in-plane density of states of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> are investigated by planar tunneling spectroscopy. Low-fluence 1-MeV He<sup>+</sup> irradiation reduces the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta T_c$ </sub> and mean-free path without altering the junction quality. As the dirty limit is approached, the zero-bias conductance peak (ZBCP) disappears, as expected for a quasiparticle Andreev bound state. Thus, the quasiparticle scattering rate may be determined by the ZBCP magnitude. This rate is a direct measure of the pair-breaking strength and accounts for the  $T_c$  suppression. [S0163-1829(98)51814-0]

The growing evidence for unconventional pairing in hightemperature superconducting cuprates has made the issue of gap suppression around impurities and interfaces a topic of increasing interest. Recent theoretical work showed that the local quasiparticle (QP) density of states (DOS) at the surface<sup>1</sup> or around an impurity<sup>2</sup> is phase sensitive and might provide a powerful tool for studying details of the order parameter symmetry. In particular, an Andreev bound state forms at a (110)-oriented surface of a  $d_{x^2-y^2}$  symmetry superconductor.<sup>3,4</sup> This bound state produces a peak in QP DOS at the Fermi energy (defined to be zero). Since the QP DOS is directly probed by tunneling, this bound state is manifested as a zero-bias conductance peak (ZBCP) in the conductance of a tunnel junction.

The node of a  $d_{x^2-y^2}$  symmetry order parameter (OP) is perpendicular to the  $(110)$ -oriented surface allowing specularly reflected QP's to suffer strong Andreev reflections due to the sign change of the order parameter for any QP trajectory. Andreev scattering is a process in which QP refection results from spatial variations of the amplitude or phase of the order parameter and induces branch conversion of the electronlike excitations into holelike excitations and vice versa. As a result Buchholtz *et al.* found<sup>1</sup> that in the vicinity of the (110)-oriented surface of a  $d_{x^2-y^2}$ -symmetry superconductor, the order parameter is reduced within a few coherence lengths producing a pair potential well. Hence the quasiparticles are in turn bound to the interface by Andreev scattering and the coherent superposition of electronlike and holelike excitations generates zero energy bound states. Although Andreev bound states in a  $d_{x^2-y^2}$ -symmetry superconductor has been predicted only for any  $(x,y,0)$ -oriented surface which is not purely  $(100)$  or  $(010)$  oriented, it has recently been shown that surface nanofaceting on the scale of the coherence length  $\xi_{ab}$  ~ 20 Å allows the ZBCP to be observed in all  $(x,y,0)$ -oriented surfaces with comparable spectral weight.<sup>5</sup>

Similarly, zero energy bound states develop in the center of a vortex core for a type-II superconductor.<sup>6</sup> The superconducting order parameter is completely suppressed within the vortex core and the QP excitations undergo Andreev reflections at the vortex edges. A  $\pi$ -phase shift is produced by the circulating supercurrents around the vortex core, and bound states at zero energy are generated. These states have been theoretically predicted by early work of Caroli, de Gennes, and Matricon<sup>'</sup> and recently observed by scanning tunnel microscopy (STM).<sup>6</sup> In a  $d_{x^2-y^2}$ -symmetry superconductor, the  $\pi$ -phase shift is intrinsic to the superconducting order parameter, so specular reflection at an *ab*-plane interface generates a bound state at the Fermi energy.

Zero-bias conductance peaks (ZBCP's) have been seen in the in-plane tunneling spectra of high- $T_c$  superconductors by planar tunneling<sup>8,9</sup> break and grain-boundary junctions<sup>10,11</sup> and STM.<sup>12</sup> Even though ZBCP's have earlier been explained by energy-dependent tunneling via spin-flip scattering at the interface,<sup>13</sup> previous data from Lesueur *et al.*<sup>9</sup> show that the magnetic-field dependence of this anomaly is inconsistent with a spin-flip scattering mechanism. Moreover, it was recently observed by STM that for very smooth surfaces, the ZBCP appears on  $(110)$ - but not on  $(100)$ oriented films.<sup>14</sup> This behavior, which also challenges the spin-flip scenario, supports the origin of the ZBCP as an Andreev bound state at the surface of a  $d_{x^2-y^2}$ -symmetry superconductor. Furthermore the ZBCP is absent in  $Nd_{2-x}Cu_{x}O_{4}$ -based tunnel junctions.<sup>11</sup> Finally, recent inplane tunneling results using normal counter electrodes on planar YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>/Cu junctions show a zero-field splitting of the ZBCP at temperatures below  $\sim 8$  K.<sup>15</sup> Both the field evolution and the zero-field splitting are remarkably consistent with predictions by Fogelström et al.<sup>5</sup> for an Andreev bound state and surface-induced broken time-reversal symmetry.

Here we report effects of ion-induced disorder upon the ZBCP in  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-\delta</sub>$  (YBCO) tunnel junctions. With increased damage, the ZBCP is found to be quenched when the mean-free path,  $\ell_0$ , is reduced to the order of the in-plane coherence length,  $\xi_{ab}$ , or  $\ell_0 \sim \xi_{ab}$ . It is argued that, as the mean-free path is reduced, the constructive interference of particlelike and holelike excitations is lost, which leads to a suppression of the Andreev bound state. Therefore the ZBCP amplitude provides a sensitive measure of the QP scattering time. An analogy with the results of Renner *et al.*<sup>16</sup> for bound states in the vortex core of a dirty type-II superconductor is presented.

Planar tunneling spectroscopy is carried out on  $He<sup>+</sup>$ -irradiated YBCO/Pb junctions fabricated on  $(103)$ - and

 $(110)$ -oriented thin films grown by off-axis magnetron sputtering. The films are typically between 1500 and 2000 Å thick and consistently show midpoint  $T_c$ 's of 89–90 K. Furthermore, x-ray diffraction shows no evidence for misoriented phases. Tunnel junctions are fabricated *ex situ* using Pb as the counter electrode. Details of film growth and junction fabrication are given elsewhere.<sup>17,18</sup> The junctions are irradiated at room temperature in a Van de Graff accelerator using  $1$ -MeV He<sup>+</sup>. The beam energy is chosen to produce a homogeneous defect profile throughout the film thickness. Heating is minimized during the irradiation by using a low beam current ( $\sim$  400 nA/cm<sup>2</sup>) and by keeping the film in good thermal contact with a room temperature Cu stage. By monitoring the film resistance rise during irradiation, the temperature is estimated to increase by approximately 10 K. Only low fluences are employed which correspond to doses that are less than  $1 \times 10^{16}$  He<sup>+</sup>/cm<sup>2</sup>; the dose being measured with an uncertainty of 30%. A Monte Carlo simulation  $(TRIM92)$  (Ref. 19) shows that the He<sup>+</sup> energy loss is as low as  $0.07$  eV/Å, minimizing the formation of extended defects.<sup>20</sup> The He<sup>+</sup> mean-free path is  $\sim$ 3  $\mu$ m, which is much larger than the combined thickness of the YBCO film and Pb electrode ( $\sim$  5000 Å). Therefore, the He<sup>+</sup> ions stop in the substrate, far from the YBCO film. The junctions studied exhibit between 8 and 25 % nontunneling current at  $T=4.2$ K, and the spectroscopic features discussed in this paper are independent of the amount of this leakage current. It is found that the ion damage does not affect the tunnel junction leakage for doses up to  $3.0\times10^{15}$  He <sup>+</sup>/cm<sup>2</sup>.

Since a ZBCP is not observed when tunneling in the *c* direction (as expected for a *d*-wave Andreev bound state), only in-plane tunneling spectroscopy is addressed here. Tunneling spectra obtained on  $(103)$ - and  $(110)$ -oriented films do not exhibit any measurable difference, so they will be considered equivalent. Analysis by scanning electron micros $copy$  (SEM) reveals that  $(103)$ -oriented films show a nanofaceting along the  $(010)$  direction on the order of  $100-200$ Å, which is larger than the *ab* plane coherence length. Therefore, the equivalence of junctions fabricated on  $(103)$ and  $(110)$ -oriented films can be explained by a mixing of different crystallographic orientations.<sup>5,15</sup>

The effects of disorder on tunnel junctions are summarized in Table I. The radiation-induced increase in the normal-state junction resistance,  $R_nA$ , cannot be due to a simple mixing of materials. In the ballistic limit, $^{21}$  appropri-

TABLE I. Materials parameters as a function of irradiation. The surface resistivity,  $R_nA$ , increases by a factor of 15 while the bulk  $T_c$  reduces by nearly 20 K. The in-plane residual resistivity is measured by extrapolation of the linear behavior in the normal state. The mean free path,  $\ell_0$ , is calculated using a Drude model for a 2D system

Fluence $(He^{+}/cm^{2})$	R <sub>n</sub> A $(\Omega \text{ cm}^2)$	$T_c$ (K)	$\rho_0$ $(\mu \Omega \text{ cm})$	∩ گ Ă)
$0.0 \times 10^{15}$	0.02	90	< 20	>285
$1.0 \times 10^{15}$	0.20	82	125	48
$1.8 \times 10^{15}$	0.30	78	180	33
$2.2 \times 10^{15}$	0.23	74	225	27
$3.0 \times 10^{15}$	0.28	65	340	18

ate at this low He $^+$  energy loss and fluence, the thickness of the mixing layer is estimated to be less than 0.5 Å. Since YBCO and Pb naturally interdiffuse over at least  $\sim$  10 Å,<sup>22</sup> the very small increase in the thickness of the interdiffusion layer cannot explain the strong enhancement of  $R_nA$ . Instead, we propose that the cause for the increase in junction resistance is a damage-induced metal-insulator transition in the oxygen deficient region in YBCO within a few Å of the YBCO/Pb interface. After irradiation, the critical temperature,  $T_c$ , is obtained from the temperature dependence of the zero-bias conductance  $G(V=0)$ : Since the four-probe geometry employed in these tunneling measurements can detect film resistance when it is comparable to the junction resistance, which is the case for these damaged *ab*-oriented films, the superconducting transition and  $T_c$  are easily determined. The residual resistivity,  $\rho_0$ , determined by the extrapolation of the normal-state linear resistivity to zero temperature, is measured along the  $(010)$  direction of patterned, crystallographically aligned  $(103)$ -oriented films.  $(103)$ -oriented films are irradiated so that the  $T_c$  is suppressed to the value measured for an irradiated junction. The irradiation has been carried out using the same ion  $He<sup>+</sup>$  and the same energy used for the YBCO/Pb junctions. Finally, the mean-free paths in Table I have been calculated using a Drude model for a two-dimensional (2D) system  $\ell_0 = hc/2e^2k_F\rho_0$  where  $k_F$  $\sim$  2.7/*a* as found by recent measurement of the Fermi surface,<sup>23</sup> *a* being the in-plane unity cell. The film residual resistivity is reduced by a factor of 3 to get rid of disorderinduced changes in transport through grain boundaries, as discussed below.

Normalized tunneling conductance curves taken on the same junction before and after irradiation are presented in Fig. 1. Note that the energy of the Pb tunneling features, including the energy gap and phonon structure, are unchanged by irradiation. The amount of nontunneling current also remains unchanged, verifying that elastic tunneling is the dominant transport mechanism through the junction, even after irradiation. Moreover we can exclude any ''ag-



FIG. 1. Effects of disorder on the low-temperature in-plane YBCO DOS is shown. The tunneling spectra correspond to the same junction before and after irradiation damage. An applied 0.2 T magnetic field is used to quench the Pb superconductivity. Note that the Pb  $T_c$  and QP DOS are unchanged by the low-fluence irradiation. The unirradiated data are shifted vertically for clarity.



FIG. 2. The evolution of the conductance with increasing disorder for *ab*-oriented YBCO/Pb tunnel junctions is shown. As the ion fluence is increased from 0 to  $3.0\times10^{15}$  He <sup>+</sup>/cm<sup>2</sup>, the strength of both the ZBCP and GLF is reduced. All the data shown are normalized by the voltage-dependent conductance measured in the respective junction at higher temperature ( $\sim$ 40 K). The data for irradiated junctions are shifted vertically for clarity. The surface resistivity for each junction is listed in Table I.

ing'' effects of the junction during irradiation. First, unirradiated junctions held at room temperature for a typical irradiation time show only a slight increase in junction resistance, and the strength of the YBCO conductance features remain unchanged. Second, unirradiated junctions prepared next to irradiated junctions, *on the same film*, exhibit the same YBCO conductance features before and after irradiation.

From Fig. 1, one can see that when the Pb electrode is driven normal under application of a 0.2 T magnetic field, the undamaged film exhibits all the usual conductance features observed when tunneling into the *ab*-plane direction of YBCO. These are a gaplike feature (GLF) at energies  $\sim$  17 meV and a well defined ZBCP, whose magnitude is  $\sim$ 30% change in the conductance. The ZBCP is only observed when the GLF is present, giving further confirmation that the ZBCP is associated with the YBCO superconductivity, as expected for an Andreev bound state. The data from the damaged film show that disorder produces three main effects:  $(1)$  a steeper background conductance;  $(2)$  a broader GLF; and (3) a reduced ZBCP. The steeper background conductance is most likely due to changes in the tunnel barrier. The reduction in the strength of the GLF and the ZBCP are intimately related to the amount of damage produced in the YBCO film and will be the focus for the remainder of this article.

The effect of increasing irradiation dose on the tunneling conductance of *ab*-plane oriented YBCO/Pb junctions is shown in Fig. 2. The tunneling conductance data taken at *T*=7.5 K (lead  $T_c$ =7.2 K) for each junction are normalized by data from the same junction taken at higher temperature  $(\sim 40 \text{ K})$ , where the ZBCP is absent. This normalization eliminates the background conductance and highlights the GLF and ZBCP. The curves, each taken for a different junction, have been shifted for clarity. Several junctions irradiated at the same doses have been investigated, verifying the



FIG. 3. (a) Disorder-reduced ZBCP amplitude vs  $\xi_{ab}/\ell_0$  ratio is presented. (b)  $T_c$  vs impurity scattering rate expressed in units of  $\xi_{ab}/\ell_0$ , is shown. The linear behavior reported in Ref. 27 for ionirradiated and Pr-doped thin films is plotted.

reproducibility of these data. We observe that the increasing amount of disorder strongly reduces the ZBCP amplitude, while the ZBCP completely disappears at doses higher than  $\sim$  2.5 $\times$  10<sup>15</sup> He <sup>+</sup>/cm<sup>2</sup>. No evidence for ZBCP restoring has been observed for doses up to  $\sim$  9.0 $\times$  10<sup>15</sup> He <sup>+</sup>/cm<sup>2</sup>, corresponding to three times the highest dose presented in Fig. 2. Moreover, spectra taken at lower temperature in a 0.2 T magnetic field, show only negligible variations of the ZBCP magnitude. This behavior which cannot be explained in the framework of the Appelbaum model<sup>13</sup> without evoking an unlikely damage-induced reduction of the magnetic impurity density, has a natural explanation if the ZBCP is an Andreev bound state. As a result of the increasing amount of scattering, the mean-free path is reduced and when the dirty limit is approached, the constructive interference of particlelike and hole- like excitations breaks down. Then the zero energy bound state disappears. This is shown in Fig. 3, where the ZBCP height vs the  $\xi_{ab}/\ell_0$  ratio is presented; the  $\xi_{ab}/\ell_0$ ratio is calculated from the estimation of the mean-free path listed in Table I, while the YBCO coherence length is set to be 20 Å. Finally the ZBCP height is determined from a background obtained by a linear extrapolation of the conductance within the GLF. The reduction of the ZBCP amplitude follows a linear behavior and the extrapolation to zero height shows that the ZBCP does disappear for a mean-free path shorter than the *ab*-plane coherence length. Thus the zerobias conductance provides a sensitive measure of the QP scattering time. A similar behavior has been recently observed for increasing doping concentration in Pr-doped *ab*-oriented YBCO/Pb tunnel junctions.<sup>24</sup> We also note from Fig. 2 that the ZBCP width as a function of disorder is almost unchanged. Such a width cannot be accounted for by simple thermal broadening due to the convolution of the ZBCP by the derivative of the Fermi function, which at *T*  $=7.5$  K is small (0.6 meV) compared to the measured ZBCP width  $(5–6 \text{ meV})$ .

The same evolution of the density of states with increasing disorder has been also observed for ZBCP's in the vortex core of a type-II superconductor. Renner *et al.*<sup>16</sup> showed that the Caroli–de Gennes–Matricon bound state at the center of a vortex core<sup> $\prime$ </sup> is strongly affected by impurities. In particu-

lar, for a single crystal of  $2H-Nb_{1-x}Ta_{x}Se_{2}$ , the ZBCP is reported to vanish when the concentration of Ta reduces the mean-free path to values lower than the BCS coherence length. As also seen in our data, the width of the ZBCP measured by Renner *et al.* is unchanged with increasing disorder.

Although the nature of such defects produced by ion damage is not clearly established, evidence exists that the depression of  $T_c$  is mainly due to in-plane disorder<sup>25</sup> while the carrier concentration is unchanged.<sup>26</sup> Thus the damageinduced  $T_c$  suppression is explained as a result of pair breaking due to increasing QP scattering rate (we remind that even nonmagnetic impurities are pair breaking for a *d*-wave-symmetry superconductor). This is shown in Fig. 3, where we present the  $T_c$  dependence on the impurity scattering rate, expressed in unity of  $\xi_{ab}/\ell_0$ , being  $\xi_{ab} \sim 20$  Å. Our measures are consistent with previous data of Sun *et al.*<sup>27</sup> in ion-irradiated and Pr-doped thin films. For both sets of data, the mean-free path has been calculated considering a reduced residual resistivity to reproduce the  $T_c$  vs  $\xi_{ab}/\ell_0$  dependence found in electron-irradiated and Prdoped single crystals.<sup>28</sup> This enables us to get rid of damageinduced enhancement of the grain-boundary resistivity. The linear decrease of  $T_c$  is in qualitative agreement with the Abrikosov-Gor'kov theory<sup>29</sup> in the limit of a small pairbreaking rate. However, as reported by Tolpygo *et al.*<sup>28</sup> the

- \*Present address: C.S.N.S.M. Baˆt. 108, 91405 Orsay Cedex, France.
- † Present address: NIST, Boulder, CO.
- <sup>1</sup>L. J. Buchholtz, M. Palumbo, D. Rainer, and J. A. Sauls, J. Low Temp. Phys. 101, 1099 (1995).
- $2^2$ M. I. Salkola, A. V. Balatsky, and D. J. Scalapino, Phys. Rev. Lett. 77, 1841 (1996).
- ${}^{3}$ C.-R. Hu, Phys. Rev. Lett. **72**, 1526 (1994).
- $4$ Y. Tanaka and S. Kashiwaya, Phys. Rev. Lett.  $74$ ,  $3451$  (1995).
- <sup>5</sup>M. Fogelström, D. Rainer, and J. A. Sauls, Phys. Rev. Lett. **79**, 281 (1997).
- <sup>6</sup>H. F. Hess *et al.*, Phys. Rev. Lett. **62**, 214 (1989).
- 7C. Caroli, P. G. de Gennes, and J. Matricon, Phys. Lett. **9**, 308  $(1964).$
- <sup>8</sup> J. Geerk, X. X. Xi, and G. Linker, Z. Phys. B **73**, 329 (1988).
- <sup>9</sup> J. Lesueur, L. H. Greene, W. L. Feldmann, and A. Inam, Physica C 191, 325 (1992).
- <sup>10</sup>R. Gross, IEEE Trans. Appl. Supercond. **7**, 2929 (1997).
- $11$  D. Mandrus, L. Forro, D. Koller, and L. Mihaly, Nature (London) 351, 460 (1991).
- <sup>12</sup>S. Kashiwaya et al., Phys. Rev. B 51, 1350 (1995).
- $^{13}$  J. A. Appelbaum, Phys. Rev. **154**, 633 (1967).

QP scattering rate calculated as the impurity scattering rate is a factor of 3 bigger than the pair-breaking rate predicted by the Abrikosov-Gor'kov theory in a  $d_{x^2-y^2}$ -symmetry superconductor.

In summary, a study of disorder effects on the Andreev bound state in ion-irradiated YBCO/Pb junctions is reported. Low dose irradiation does not alter the quality of the tunnel junctions, while the critical temperature and the elastic mean-free path is reduced. As observed in the vortex core of a type-II superconductor, the ZBCP disappears when the mean free-path approaches the *ab*-plane coherence length. Radiation-induced disorder destroys the constructive interference of particlelike and holelike excitations, causing the loss of the Andreev bound state. Therefore the QP scattering time may be directly measured by the ZBCP amplitude. Moreover, the  $T_c$  decrease as a function of the QP scattering rate follows the pair-breaking law observed for ion-irradiated and Pr-doped thin films.

We are grateful for many helpful conversations with M. Fogelström, J.A. Sauls, S.-K. Yip, J. Lesueur, and J. Giapintzakis. We also wish to thank J. Byers and M. Flatté for valuable discussions and B. Clymer for his assistance with the irradiations. This research was supported by the NSF-STCS (Grant No. NSF-DMR 91-20000). L.H.G. and E.P. also acknowledge support from NSF (Grant No. DMR 94-21957) and ONR (Grant No. N00014-95-1-0831).

- $^{14}$ L. Alff *et al.*, Phys. Rev. B 55, 14 757 (1997).
- <sup>15</sup>M. Covington *et al.*, Phys. Rev. Lett. **79**, 277 (1997).
- $16$ Ch. Renner, A. D. Kent, Ph. Niedermann, and  $\Phi$ . Fisher, Phys. Rev. Lett. **67**, 1650 (1991).
- <sup>17</sup>L. H. Greene *et al.*, Appl. Phys. Lett. **59**, 1629 (1991).
- <sup>18</sup>M. Covington, R. Scheuerer, K. Bloom, and L. H. Greene, Appl. Phys. Lett. 68, 1717 (1996).
- <sup>19</sup> J. F. Ziegler, J. P. Biersack, and V. Littmark, *The Stopping and Range of Ions in Solids* 1-2 (Pergamon Press, New York, 1996).
- $^{20}$ H. Bernas and A. Traverse, Appl. Phys. Lett. 41, 245 (1982).
- 21P. Sigmund and A. Gras-Marti, Nucl. Instrum. Methods **182/183**,  $25$  (1981).
- <sup>22</sup> S.-W. Han *et al.*, Physica B **221**, 235 (1996).
- $^{23}$  Rong Liu *et al.*, Phys. Rev. B 46, 11 056 (1992).
- $24$ M. Covington, E. Paraoanu, and L. H. Greene (unpublished).
- <sup>25</sup>E. M. Jackson *et al.*, Phys. Rev. Lett. **74**, 3033 (1995).
- <sup>26</sup> J. M. Valles *et al.*, Phys. Rev. B **39**, 11 599 (1989).
- $^{27}$  A. G. Sun *et al.*, Phys. Rev. B **50**, 3266 (1994).
- <sup>28</sup> S. K. Tolpygo *et al.*, Phys. Rev. B **53**, 12 454 (1996).
- 29A. A. Abrikosov and L. P. Gor'kov, Sov. Phys. JETP **12**, 1243  $(1961).$