Excitation of a bosonic mode by electron tunneling into a cuprate superconductor $NdBa_2Cu_3O_{7-\delta}$

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We performed scanning tunneling spectroscopic experiments on hole-doped $NdBa_2Cu_3O_{7-\delta}$. The dI/dV curves obtained at 4.2 K are asymmetric with clear peak, dip, and hump structures. Energy derivatives of these curves show peaks at energies beyond the dip features. Highly precise full potential band-structure calculations confirm a featureless electronic density of states in that energy region. Our results indicate that tunneling electrons couple to a collective mode in the CuO_2 plane.

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The identification of the microscopic mechanism for pair formation in the unconventional superconductors is still a challenge. In conventional superconductors, apart from the isotope effect, tunneling experiments provided the most direct evidence of electron-phonon interaction mediating the Cooper-pair formation.[1,2] In case of high-T_c superconductors (HTSC), inelastic neutron-scattering (INS) experiments and high-resolution angle-resolved photoemission spectroscopy (ARPES) experiments were useful to find collective modes in the range of 30–70 meV.[3–9] Collective spin excitations have been suggested as the bosonic mode which interacts with electrons to produce a resonance peak at a wave vector (π,π) in INS spectra and a kink along the $(\pi,0)$ direction of the Brillouin zone in ARPES spectra.[3–6,10] On the other hand, several ARPES results also show evidence of phonons as the collective modes.[7–9] Thus the assignment of the boson-mediating Cooper-pair formation in HTSC is far from being settled.

Due to the microscopic electronic inhomogeneity in these superconductors,[11] tunneling experiments using scanning tunneling microscopy (STM) are expected to play an important role in resolving such issues being related to pairing. While tunneling, electrons can excite a collective mode of a certain energy if the energy of the electrons is equal to that of the collective mode. Due to the inelastic interaction of the electrons with the mode, a new scattering channel is induced which results in a steplike feature in the dI/dV curves.[12] Ideally, these steplike features can be observed more easily in the form of a delta function in the energy derivative of the dI/dV curves. In experiments, the peak heights and widths are dependent on the energy resolution, temperature, and the modulation voltage.[12] For the superconducting state of *d*-wave superconductors, Balatsky *et al.*[13] predicted that features due to inelastic scattering of tunneling electrons would be observed in the d^2I/dV^2 curves as satellite peaks at $\Delta+\Omega$ in addition to a very weak peak at Ω, where Δ is the energy gap and Ω is the excitation energy. Indeed, Lee *et al.*[14] observed peaks in the second derivatives, which carry the signature of a bosonic mode in the hole-doped cuprate $Bi_2Sr_2CaCu_2O_{8-\delta}$ (Bi-2212). The tunneling spectra of the cuprates such as $YBa_2Cu_3O_{7-\delta}$ (Y-123), which contain an additional Cu-O layer in the unit cell in the form of quasi-one-dimensional chains, often exhibited features which were not observed for Bi-2212.[15,16] These additional features in the spectra impeded the general understanding of the electronic structure of the superconducting state of cuprates and also masked the relevant features due to bosonic mode from the tunneling spectra of these cuprates. Very recently, Niestemski *et al.*[17] observed a bosonic mode from the tunneling spectra on $Pr_{0.88}LaCe_{0.12}CuO_4$ (PLCCO), which is an electron-doped superconductor. In this work, we present evidence of a bosonic mode, which appears to be originated from the CuO_2 plane, from scanning tunneling spectroscopy (STS) data obtained on a twinned single crystal of $NdBa_2Cu_3O_{7-\delta}$ (Nd-123). Nd-123 belongs to the Y-123 family. They have isostructural unit cells. The ionic size of Nd^{3+} ions, which are paramagnetic at 4.2 K,[18] is bigger than that of Y^{3+}.

The single crystal used for the measurements was grown from a BaO/CuO flux. The details of the growth process are according to those mentioned in Ref. 18. After the growth, the crystal was properly oxidized to achieve a T_c onset of 93.5 K. The transition width of $\Delta T_c \sim 3.5$ K was determined from ac-susceptibility measurements. For the STS experiments, the as-grown crystal surface was cleaned with absolute ethanol and dried by pure helium gas. No special surface preparation was performed.[19] The STS measurements were carried out at 4.2 K in He gas atmosphere using a homemade STM. A mechanically cut Pt-Ir tip was used for the measurements. The tunneling parameters for the measurements were set as −0.1 V and 0.2 nA giving a tunneling resistance of 500 MΩ. The bias voltage was applied to the sample, so that a negative (positive) voltage refers to a filled (empty) sample energy state. Spectroscopic data were obtained as $[dI/dV](V)$ curves using the standard lock-in technique where a modulation voltage of 2 mV (rms) was used. Data were taken at many different locations on flat areas of the sample surface. The spectra show that the surface is electronically inhomogeneous. However, at some locations of the sample, we obtained curves which are nearly homogeneous in the length scale of 20–25 nm. Here we discuss only those spectra which contain coherence peaks (P), dips (D), and humps (H). These features are hallmarks of the superconducting state in cuprates as was independently observed in STS and ARPES experiments on Bi-2212.[20–24] In Fig. 1, a set of dI/dV spectra obtained along a line cut of 20 nm is shown. These spectra were observed while taking a dI/dV curve at every 0.5 nm of a 32×32 nm^2 area. It is remarkable that the PDH features are observed on the as-grown sample

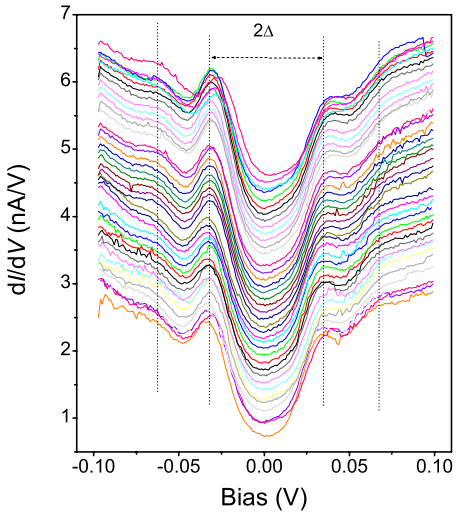

FIG. 1. (Color online) dI/dV curves (shifted along y axis for clarity) obtained at 4.2 K along a 20 nm line. The peaks and humps are indicated by dotted vertical lines.

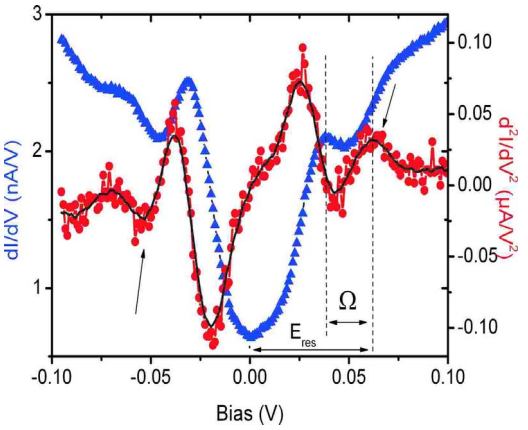

FIG. 2. (Color online) Typical dI/dV curve (triangles) containing peak-dip-hump structures together with the energy derivative (circles). The smooth dark line is a guide to the eyes. The arrows show the inelastic features at both empty and filled states. The boson mode energy (Ω) at empty state is shown.

surface. On the as-prepared surface of NdBCO, Ting *et al.*[25] found the CuO chain layer as the surface terminating layer. The spectra shown in Fig. 1 do not have any similarity to the ones observed on the well-characterized CuO chain layer.[26] It is possible that the topmost CuO chain layer in the present case is an insulating layer due to surface degradation. In that case, at the small bias voltage range of ± 100 mV, the tunneling would take place between the metal tip and the CuO_2 plane layer, which is situated within ~ 4.2 Å of the CuO chain layer, thus revealing the clear PDH features. Apart from the PDH features, the asymmetry of the coherence peaks is evident from the data. The peak height is found to be always larger at the filled sample states compared to the empty ones. Similar asymmetric curves were also observed for Bi-2212 (Refs. 11, 20, and 27) and Nd-123 (Ref. 28) earlier. The asymmetry is most likely related to the probability of electron extraction and injection from or to the material.[29] Beyond the peaks, there is an asymmetric V-shaped background. On top of the background, humps are evident at both empty and filled states. The humps are observed at $\sim 2\Delta$ and there are dips at $\sim 1.4\Delta$. The respective features are broader for the empty states than for the filled ones. This becomes clear from the representative curve shown in Fig. 2. The average energy gap measured from peak to peak is 70 meV which leads to $2\Delta/k_BT_c = 8.68$.

Recently, Ngai *et al.*[16] reported subgap features in addition to the coherence peaks in the dI/dV curves obtained on $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films. Similar features were also reported earlier on Y-123 single crystals.[15] On the other hand, subgap features were not observed in the ARPES data on cleaved single crystal and STS data on thin film of Y-123.[30,31] However, the PDH features are very clearly present in those data. The subgap features were also not observed by Wu *et al.*[28] on Nd-123. In cuprate systems with an additional copper-oxygen chain layer, the subgap features were suggested to be due to multiband superconductivity.[16] In our data, we do not find any indication of such an influence of the metallic chain layer on the electronic structure representing superconducting state (see Fig. 1). Interestingly, Derro *et al.*[26] observed strong subgap resonances in the energy scale of ~ 5 meV while tunneling to the chain layer of Y-123 single crystals. These subgap features were suggested to be possibly due to the oxygen vacancies in the chains. It is most likely that the metallic CuO chain layers acquire superconductivity due to the proximity effect.[26,32] Within this scenario, the spectra with subgap features possibly reflect the local density of states (DOS) (LDOS) of CuO chains and those with clear PDH features reflect the LDOS of CuO_2 plane layers as these features are also observed on chainless Bi-2212. If the superconductivity in 123 systems would be due to multiband effects, the signature of this should also be seen in the electronic spectra of Nd-123. Although from the present data it is not clear if the subgap states would be observed in the LDOS of CuO_2 plane that exists in the bulk, however, it gives another indication that the PDH features are the generic features in the CuO_2-plane-derived DOS of cuprate superconductors.

In order to gain further information from the spectra, we focus on the part of the curves beyond the coherence peaks. By numerically differentiating the curves we observe a peak in the empty state at an energy $E_{\rm res}$ as shown in Fig. 2. The above mentioned peaks in the d^2I/dV^2 curves correspond to very weak steplike features in the dI/dV curves. The observed fine structure is consistent with the predicted inelastic electron tunnel spectroscopic (IETS) features in d-wave cuprates.[13,33] It also has considerable similarity to the IETS features observed for hole-doped Bi-2212 (Ref. 14) and electron-doped PLCCO superconductors.[17] In order to check if the fine structure in the STS spectra is due to the specific band structure of the superconductor, full potential density-functional calculations were performed. We applied the full potential local-orbital (FPLO) minimal basis scheme[34] (version 5.00-19) within the local spin-density approximation (LSDA), where the exchange-correlation potential of Perdew and Wang[35] was chosen for the calculations. The strong Coulomb repulsion in the localized Nd 4f shell has been modeled in a mean-field way within the LSDA+U approxima-

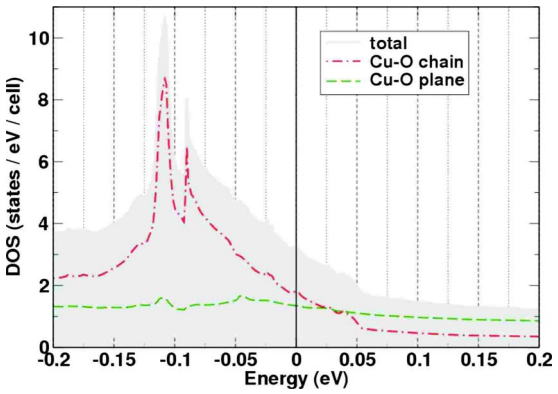

FIG. 3. (Color online) Calculated total electronic density of states (shaded gray) in the vicinity of the Fermi level (zero energy) for NdBCO. The sharp features at about 85 and 110 meV binding energy can be assigned to Van Hove singularities originating from the Cu-O chains [(red) dot-dashed line]. The contribution from the plane [(green) dashed line] is rather flat in the shown energy window.

tion. A typical value of U_{4f}=8 eV has been applied throughout the calculations. Our results are basically independent of the choice of U within physically reasonable limits. As basis set, Nd $4f5s5p/6s6p5d$, Ba $5s5p/6s6p5d$, Cu $3s3p/4s4p3d+4d$, and O $2s2p3d+3s$ states (notation: semicore/valence+polarization states) have been chosen. The extent of the valence states is optimized with respect to the total energy.[36] The inclusion of the semicore states is necessary to account for their non-negligible overlap. The polarization states provide a more complete basis set to ensure highly accurate band structure and DOS results. A very fine k mesh of 16 200 points in the Brillouin zone (2560 in the irreducible wedge) was used to resolve the singularities in the DOS near the Fermi level accurately. Convergency with respect to basis set and k mesh was carefully checked. The structural data of Ref. 37 were used.

We find a total valence bandwidth of about 9 eV, which is typical for the quasi-two-dimensional cuprates. Three bands cross the Fermi level, two of them originating from the CuO_2 layer and one originating from the CuO_3 chains along the y direction. In Fig. 3, we show the calculated total DOS of Nd-123 in the vicinity of the Fermi level. In this energy region, only the states originating from the CuO_2 planes and the CuO_3 chains exhibit significant contributions. Whereas the CuO_2-plane-related DOS is almost constant close to the Fermi level, two distinct Van Hove singularities related to the CuO_3 chain show up at about 85 and 110 meV. Otherwise, no significant features in the DOS near the Fermi level are obtained. The slight underdoping of the present sample can be modeled by a small rigid shift of the Fermi level toward negative energies. For an oxygen deficiency δ=0.01, this shift is estimated to be ~ -18 meV. There is no sharp structure present in the DOS of the CuO_2 plane at energies close to the region where peaks are observed in the experimental d^2I/dV^2 spectra. Thus, these features cannot be related to the DOS. The calculations also show that the bilayer splitting is not responsible for the PDH features.

In accordance with Eliashberg's[38] classical theory for strong-coupling superconductors, where the electron-boson interaction was proposed to be observed at an energy of $E=\Delta+\Omega$, the Ω values were determined for both empty and filled states. The empty and filled states Ω are experimentally determined by $\Omega(r)=E_{\mathrm{res}}(r)-\Delta(r)$ from the entire set of curves obtained at different locations (r) on the sample. The mean values of $\Omega(r)$ are found to be 22.9 ± 1.8 and 23.7 ± 1.6 meV for empty and filled states, respectively. Thus, the peaks at E_{res} in the d^2I/dV^2 curves are found to be almost symmetric. This strongly supports the assumption that the observed features are signatures of inelastic tunneling. They result from an additional tunneling channel introduced due to the excitation of a collective mode at $\sim$23 meV. The mode energy is surprisingly low compared with the ones observed for Y-123 for similar dopings.[4] *A priori*, it is not clear if the mode is excited in the tunneling barrier or in the CuO_2 plane of the sample. Thus, it is instructive to plot $\Omega(r)$ against $2\Delta(r)$. As shown in Fig. 4, Ω and Δ are intimately related. A similar, but comparatively stronger, correlation was also found for Bi-2212 and PLCCO.[14,17] Moreover, this mode energy for Nd-123 was observed for only those spectra which exhibit clear PDH features. Thus, the collective mode appears to be intrinsic to the superconducting CuO_2 plane. To determine the nature of the observed collective mode, we compare the mode energy with the known phonon energies of Nd-123. The former is rather low compared to the in-phase or out-of-phase O(2)-O(3) vibrations which have energies of more than 35 meV as observed from Raman spectra.[39–41] In case of Bi-2212, Pilgram *et al.*[42] suggested that the peaks in the second derivative are due to the excitation of an apical oxygen vibration mode. However, the corresponding energy for Nd-123 as detected from Raman spectra is $\sim$68 meV.[39] Also, this is not intrinsic to the CuO_2 plane. The only low-energy modes for Nd-123 are due to the metallic Ba (14 meV) and Cu (19 meV) ion vibration modes. The latter one seems to be the only possible mode that could be excited during tunneling in the present case. However, it is not clear why in that case the same mode energy is not observed in case of Bi-2212 or PLCCO.[14,17] Thus, although the mode does not appear to be a phonon, we cannot definitely rule out the possibility of a low-energy excitation of metal ions.

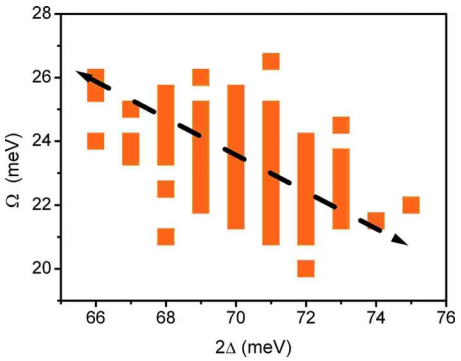

FIG. 4. (Color online) Mode energy $\Omega(r)$ vs energy gap $2\Delta(r)$ obtained from the dI/dV curves like in Fig. 2. The dashed line emphasizes the tentative correlation. The uncertainties in the evaluation of Δ and Ω are $\pm(0.5-1)$ meV and $\pm(1-1.5)$ meV, respectively.

For an underdoped Y-123 sample, incommensurate spin fluctuations were observed at 24 meV in INS experiments.[43] Niestemski *et al.*[17] suggested the 10.5 meV mode as observed for PLCCO to be a magnetic-resonance mode. At this point, it remains unclear what the origin of the observed mode for this rare-earth-based cuprate is. Since the features observed in the spectra and the correlation between Δ and Ω are qualitatively similar to those observed for Bi-2212, the identification of the nature of this low-energy mode in the rare-earth-based cuprate would be important.

In summary, our STS data on as-grown slightly underdoped Nd-123 single crystal do not show any evidence of multiband superconductivity. However, it would be important to probe the CuO_2 plane layer in the bulk in order to completely rule out the role of CuO chain band toward superconductivity in these systems. The peak-dip-hump features in the spectra indicate that these are most likely the generic features of the CuO_2-plane-derived LDOS in cuprates. Furthermore, a bosonic mode has been detected with a characteristic mode energy of about 23 meV. The mode energy is rather low compared to the other bosonic modes observed for the cuprates. The observed mode energy has certain correlation with the energy gap and thus appears to be intrinsic to CuO_2 plane. However, the correlation is weak compared to the ones reported for other cuprates. Thus, the origin of the low-energy mode due to a tunneling pathway, which is extrinsic to the CuO_2 plane layer, cannot be completely ruled out.

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