Tc-dependence of energy gap and asymmetry of coherence peaks in NdBa2Cu3O7−δ superconductors

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Abstract – We report here the results of scanning tunneling spectroscopy experiments performed on hole-doped NdBa2Cu3O7−δ single crystals of Tc values of 76 K, 93.5 K and 95.5 K. The energy gaps are observed to be increasing with decreasing Tc values. The coherence peaks are asymmetric with the peaks at the filled states being larger than those at the empty ones. The asymmetry increases with decreasing Tc. The observed asymmetry and its Tc-dependence can be explained by considering the Mott insulating nature of the material at the undoped state.

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Tunneling spectroscopy played an important role in the understanding of superconductivity in conventional superconductors. In case of high-temperature cuprate superconductors (HTSC), scanning tunneling spectroscopy (STS) and angle-resolved photoemission spectroscopy (ARPES) experiments have turned out to be very important in providing detailed information on the electronic structure. The cuprates being Mott insulators in the undoped state show strongly doping-dependent characteristics. From the tunneling and ARPES studies, a monotonic dependence of the energy gap has been observed on Bi2Sr2CaCu2O8+δ (Bi-2212) [1–3]. However, Yeh et al. reported that the energy gap of YBa2Cu3O7−δ (Y-123) does not vary monotonically with doping as it does for Bi-2212 [4]. In addition to the information on the energy gap, STS and ARPES experiments also have revealed dip and hump structures beyond the coherence peak in the spectroscopic data [5–7].

Apart from these features, STS data often revealed an asymmetry of the conductance peak heights at both empty and filled states [5,6]. The peak height at the filled state is usually higher than that at the empty ones. In some cases, where conductance peaks are not very clear, the asymmetry is still clearly visible in the background of the tunneling conductance [8,9]. The origin of this asymmetry so far remains unclear. Yusof et al. have shown that the asymmetric peaks in the local density of states (LDOS) are related to the symmetry of the order parameter [10]. Using tight-binding band structure calculations for Bi-2212 and with the inclusion of tunneling directionality and group velocity, their calculations suggested that for the dxy symmetry the LDOS should be asymmetric. The important factor giving rise to the asymmetry in their calculations seems to be the directionality. The calculations were performed for tunneling almost along the (π,0)-direction in order to match the asymmetry observed in experiments. However, in case of STS, the tunneling tip positioned above the ab-plane is unlikely to have the directional preference that, according to Yusof et al., is necessary to produce the observed asymmetry [10]. On the other hand, it was shown that if the energy gap function in a superconductor depends on the wave vector k through the band energy ϵk (i.e. ∆ ≡ ∆(ϵk)), and if the slope of ∆ at the Fermi energy is positive, asymmetric peaks with the peak at the occupied state higher than that at the unoccupied one will result [11]. Recently, Randeria et al. and Anderson et al. have suggested that the observed asymmetry in strongly correlated systems like cuprates is due to hole doping [12,13]. As the cuprate superconductors are lightly hole-doped systems, where strong Coulomb repulsion prohibits double occupancy at a site, they considered the role of electron and hole occupancies in their calculations, which produced an asymmetry of

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the coherence peaks. This theory also predicts that the asymmetry would increase with underdoping. Experimentally, the respective features were mostly observed for Bi-2212, where very reproducible results could be obtained [5,14,15]. This is due to the fact that the surface of Bi-based systems can be easily prepared for STM/STS and ARPES studies by cleaving. However, in order to gain a general understanding it is necessary to study the electronic structures of different cuprate systems.

In a very recent work Kohsaka et al. observed that the asymmetry decreases with doping in Ca$_{2-x}$Na$_x$Cu$_2$Cl$_2$ [9]. Y-123 defines another class of HTSC, where in addition to CuO$_2$ layers, there is another one-dimensional CuO chain layer. Due to the surface-related complexities, so far there is too little reproducible STS and ARPES data on this cuprate system to suggest that the features which are observed on chainless Bi-2212 could be generalized for the cuprates with chain layers.

In this work we present STS data obtained on NdBa$_2$Cu$_3$O$_{7-\delta}$ (Nd-123) which is isostuctural to Y-123. STS studies were performed on the ab-plane of Nd-123 single crystals with $T_c$ values of 76 K (underdoped, sample UD76) and 93.5 K (slightly underdoped, sample UD93). Data obtained on the bc-plane of an optimally doped single crystal ($T_c = 95.5$ K, sample OP95) will also be presented.

The samples are prepared as described in ref. [16]. X-ray diffraction patterns for the three samples show that the samples are of single phase. The unit cells have the usual orthorhombic structure and the samples are twinned. STM/STS experiments were performed on the as-grown samples. These were cleaned in absolute ethanol and dried by pure He gas before mounting into the STM. The STS data were obtained as $dI/dV$ curves with a lock-in amplifier using a modulation voltage of 2 mV (rms). All data were obtained at a tunnel junction resistance of 500 MΩ. A negative (positive) bias addresses the filled (empty) states of the sample. Platinum (80%)-iridium (20%) tips were used for the measurements. All measurements were carried out at $T = 4.2$ K and in He gas environment.

STS data were obtained at different locations on the samples. $dI/dV$ curves show a strong variation of electronic structures. Curves with different energy gaps and sometimes without any signature of a clear energy gap were observed on the samples. In the following, we discuss the spectra with clear energy gap features. Such curves (raw data) which are representative for the superconducting state for the three samples are shown in fig. 1. As is evident from the figure, the energy gap with asymmetric peaks is very reproducibly observed for all the three samples. On the optimally doped sample, where the STS experiments were performed on the bc-plane, curves with energy gap features were not observed as frequently as in the case of the other two samples. This is most likely due to the fact that the probability of probing a superconducting CuO$_2$-plane, while tunneling to or from the ab-plane, is higher than that on the bc-plane, where all the superconducting and nonsuperconducting layers of the unit cell terminate.
As can be seen from fig. 1, the curves have three important features: a peak (close to the Fermi energy), a dip (next to the peak) and a hump (marked as P, D and H for the UD76 and UD93 samples) at both filled and empty states [17]. The dips and humps are present on top of a nearly linear background. These features are very clear in the data obtained on the UD93 sample. Apart from the PDH features, the peak heights are asymmetric with filled state heights being larger than those at the empty ones.

The PDH features have been often observed in STS and ARPES data obtained at the superconducting state of Bi-2212 [6,18]. Although, the physics behind the origin of the features is still unknown, the prior experiments suggest that these fine structures are intrinsic to the CuO$_2$-plane [19,20]. Whether the features are due to any band structure or strong-coupling effect similar to the reduced gap values $2\Delta$ the above values suggest a qualitatively similar dependence of $\Delta$ on doping [4]. The highest $T_c$ obtained at an oxygen content of $O_T$ (i.e., $\delta = 0$) [22]. Thus, there is no overdoped region in the $T_c$-vs.-doping phase diagram for this material. Since the isotherms are believed to depend on the Nd/Ba ratio of the Nd-123 phase, the determination of the exact oxygen content was not possible [23]. However, since the $T_c$-dependence on hole doping is almost linear, the above values suggest a qualitatively similar dependence of $\Delta$ on hole-doping values. From the observed data, the reduced gap values $2\Delta/k_BT_c$ are found to be increasing from 6.9 for OP95 to as high as 18 for UD76. These values are in the range of values reported for both Bi-2212 and Y-123 [24]. In general, underdoped cuprates which yield smaller $T_c$ values have been found to have large $\Delta$ values [24]. A very high value (as high as 20) for underdoped Bi-2212 has been observed by Miyakawa et al. [25]. For Bi-2212, $\Delta$ decreases monotonically with an increase of doping and hence with increasing $T_c$ up to optimal doping [1,2,26]. The present data show that for Nd-123 the $\Delta$ values similarly decrease with increase of $T_c$ (hence doping) up to the highest $T_c$ value (i.e., optimal doping). This contradicts the data on Y-123 reported by Yeh et al. who observed no monotonic dependence of $\Delta$ on doping [4].

The observation of identical features in different cuprate materials containing an additional CuO chain layer in the unit cell implies that these are general features of cuprates. The strong dependence of $\Delta$ on $T_c$ and hence on hole doping observed here indicates a similar pairing mechanism for cuprate materials with or without chain layers. Although, most of the STS and ARPES spectra at the underdoped region showed clear peaks as in the present data on Nd-123, in a very recent study on underdoped Bi-2212, Gomes et al. observed ill-defined large gaps without sharp peaks in the $dI/dV$ spectra [15]. Similar features have also been observed on underdoped La$_{2-x}$Ba$_x$CuO$_4$ with $x = 1/8$ where superconductivity is strongly suppressed [27]. Here, the ill-defined gaps, marked by humps across the Fermi level, have been suggested to be pseudogaps. In a recent ARPES experiments on deeply underdoped Bi-2212, Tanaka et al. observed two distinct gap features: superconducting gap features with coherence peaks close to the nodal direction and pseudogap features without sharp peaks along the antinodal directions of the Fermi surface [28]. Considering these two gap scenarios in a underdoped cuprate, it is tempting to suggest that the tunneling data which averages over the entire k-space might indeed give a resultant of both pseudogap and pairing gap. This would then lead to big gap values [15].

From the curves shown in fig. 1, the asymmetry has been calculated as

$$A = \frac{g(p) - g(p^+)}{(g(p) + g(p^+))/2},$$

where $g(p^-)$ and $g(p^+)$ are the $dI/dV$ values at the peaks of the filled and empty states, respectively. The obtained values for the three representative curves for samples UD76 (violet curve), UD93 (black curve) and OP95 (yellow curve) are $A = 0.29, 0.14$ and 0.05, respectively. In case of conventional s-wave superconductors, the coherence peaks are always symmetric. In case of HTSC, the asymmetry has been mostly observed in STS data on Bi-2212.

The surface of Nd-123 has been reported to be very stable against oxygen loss unlike that of Y-123 [29]. In STS experiments on Nd-123 single crystals, Wu et al. have observed asymmetric peaks [30]. However, the energy gap and the degree of asymmetry observed in their STS experiments, where the tip and the sample were at two different temperatures, are very different than the present data. Nishiyama et al. observed energy gaps on Nd-123 single crystals, but the other features were absent [31].

In cuprates, which are Mott insulators in the undoped state, doping of holes changes the conductivity of the material. If $x$ is the average hole-doping value, there are $1-x$ occupied sites per unit cell. Since superconductivity is obtained only for a small range of $x$ (typically 5% to ~25%), there is always a higher percentage of occupied (electron) states from which the electrons can be extracted than of empty (hole) states to which the electrons could be injected. This implies that as $x$ increases the ratio $1/x - 1$ of occupied and unoccupied states decreases. In the absence of an overdoping region in the phase diagram of Nd-123, as $x$ increases, eventually $T_c$ increases. This qualitatively suggests that the observed asymmetry is related
to hole doping in the cuprates in general. The present data on Nd-123 samples obviously confirm the aforementioned predictions by Randeria et al. [12] and Anderson et al. [13] with respect to the relationship of asymmetry and under-doping. To further verify this hypothesis, the ratio of energy-integrated differential conductance is considered for the spectra shown in fig. 1. The ratio is defined as

\[ R = \frac{\int_{V_0}^{0} gdV}{\int_{0}^{V_0} gdV} \]

where \( g = dI/dV \) is the measured differential conductance. The integrations are carried out over the experimental bias range of \( V_0 = 0.1 \text{ V} \). The limits are chosen in order to be able to compare the integrated values within a fixed energy range for the three samples. The respective ratios for the three representative curves for samples UD76, UD93 and OP95 are \( R = 1.175, 1.013, \) and 1.012, respectively.

The \( R \) values show that the energy-integrated conductance at the filled sample state also decreases with respect to the empty state part with a change of \( T_c \). This is very clear in the case of the two samples with a large difference of \( T_c \) (and thus doping). It is, however, not straightforward to compare the energy-integrated value in case of tunneling on \( ab \) and \( bc \) planes. The \( bc \)-plane, being the termination of the superconducting CuO2 and other nonsuperconducting layers of the unit cell, is \( \sim 12 \text{ Å} \) along the c-axis. The probability, that the \( dI/dV \) curves obtained a few ångströms above this plane are affected by other nonsuperconducting layers is high. This would then lead to broadened features in the STS curves. Thus, for a very small doping difference, the energy-integrated value of the curves might be difficult to compare as in the present case.

In summary, STS studies have been performed on doped Nd-123 single-crystal samples with \( T_c \) values of 76 K, 93.5 K and 95.5 K. In the \( dI/dV \) curves with features like conductance peak, dip and hump, energy gap values are found to be decreasing with increasing \( T_c \) from underdoped to optimally doped samples. This is in agreement with the variation of the gap with \( T_c \) in Bi-2212 samples. Furthermore, the coherence peak heights are asymmetric. The peaks at the filled states are found to be higher compared to the ones corresponding to the empty state. The asymmetry increases with decreasing \( T_c \) which can be explained as being due to approaching the Mott insulator state.

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