Anisotropic softening of collective charge modes in the vicinity of critical doping in a doped Mott insulator

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Momentum resolved inelastic resonant x-ray scattering is used to map the evolution of charge excitations over a large range of energies, momenta and doping levels in the electron doped Mott insulator class Nd$_{2-x}$Ce$_x$CuO$_4$. As the doping induced AFM-SC (antiferromagnetic–superconducting) transition is approached, we observe an anisotropic softening of collective charge modes over a large energy scale along the $\Gamma \to (\pi, 0)$-direction, whereas the modes exhibit broadening ($\sim 1$ eV) with relatively little softening along $\Gamma \to (\pi, \pi)$ with respect to the parent Mott insulator ($x=0$). Our study indicates a systematic collapse of the gap consistent with the scenario that the system dopes uniformly with electrons even though the softening of the modes involves an unusually large energy scale.

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The evolution of a strongly correlated material with doping—from a Mott insulator to a conducting metal—is one of the most intensively studied issues in current condensed matter physics. This fascinating evolution has proven to be full of surprises such as the appearance of high-$T_c$ superconductivity, non-Fermi liquid behavior, and nanoscale phase separation [1]. Mott insulators often exhibit phase transitions upon doping, which are signaled or hallmarked by the softening of collective charge or spin modes. The behavior of spin modes has been investigated extensively via neutron scattering [2]. Although charge excitations near the Brillouin zone (BZ) center can be accessed by optical techniques [3], their behavior with momentum over the full BZ remains largely unexplored. Here, as demonstrated in recent experimental [4, 5] and theoretical studies [6, 7], inelastic x-ray scattering indeed provides such a unique opportunity.

While previous studies have focused largely on either the undoped 1-D [8] or 2-D [4, 5] insulators, or the hole-doped superconductors [9], Mott insulators can be doped with electrons as well. In fact, it appears that with electron doping bands in the cuprates evolve in a much more straightforward and systematic manner [10, 11, 12, 13, 14] than with hole doping. However, a direct study of the doping induced changes in the collective excitation spectra of a Mott insulator is lacking. Previous x-ray scattering work [12] focused on the highly electron doped superconductor (SC) Nd$_{1.85}$Ce$_{0.15}$CuO$_4$, observed intraband and interband excitations and interpreted the excitations in a one-band model with long-range hoppings. However, Ref[15] does not report the undoped Mott insulator and the way gap modes of the Mott insulator are related to the doping where superconductivity sets in. In this Letter, we report a high-resolution study of how the collective charge excitations of the Mott insulating state ($x=0$) develop with electron doping in approaching the critical AFM-SC transition (before reaching the optimal doping) for the first time. Our finding, made possible by studying the insulating states, is that, as the electron-doping induced AFM-SC transition is approached from the $x=0$ Mott side, the system exhibits an anisotropic softening of excitations over a large energy scale ($\sim 5$ eV) along the $\Gamma \to (\pi, \pi)$-direction, whereas the modes exhibit broadening ($\sim 1$ eV) with relatively little softening along $\Gamma \to (\pi, 0)$. Our results suggest that a multi-orbital Hubbard model is essential to describe the observed evolution from the $x=0$ Mott state in contrast to the 1-band model with long-range hoppings proposed in Ref[15] based on the superconductor only. Moreover, our results show that the evolution of Mott physics of the electron-doped cuprate is dramatically different from that reported in the hole-doped cuprates [9].

The electronic structure of Nd$_{2-x}$Ce$_x$CuO$_4$ (NCCO) has been studied by angle-resolved photoemission (ARPES) and optical spectroscopies. ARPES studies [16] find that the electrons dope directly into the bottom of the upper Hubbard band and yield small Fermi (FS) pockets, with a crossover around optimal doping to a large FS. Such a scenario, where the magnetic order remains commensurate without signs of ‘stripe’ (charge inhomogeneity) or other phase separation, also describes magnetization [14, 17, 18] and optical data [19].

The experiments were performed at CMC-CAT beamline 9-ID-B at the Advanced Photon Source. Resonant inelastic x-ray scattering (RIXS) at copper K edge allows a large enough momentum transfer to cover several BZ’s. The scattered photon energy was measured by using a
the sample was mounted with incident polarization. Induced artifacts when measuring the in-plane anisotropy depend on details of the spectra. All data are taken at count.

Resolution was set to about 0.37 eV in order to diced Ge (733) crystal analyzer, and the intensity was measured at room temperature. To avoid possible polarization-induced artifacts when measuring the in-plane anisotropy, the sample was mounted with incident polarization directed along the c-axis (Ref.1(a)). The data were collected at several values of the momentum transfer vector $q = k_i - k_f$ in the 2nd BZ along the [100] direction, and in the 4th BZ along the [110] direction. The coupling to various excitations in the x-ray scattering process, near a resonance, depends in general on the energy of the incident photons. Accordingly, we first examined the detailed incident energy dependence of the loss spectra at several momenta. Representative data sets are presented in Fig. 1. It is known that the charge-transfer gap excitations resonate near the first absorption peak $0.4 eV$. Similar resonance behavior is seen in Nd$_2$CuO$_4$, which is like that in the more extensively studied La$_{2-x}$Sr$_x$CuO$_4$. $0.4 eV$. However, the scattering intensity at the low-energy branch is associated with leading edges are plotted in (b) as a function of momentum and doping. The upper branches around $2 eV$ are seen to split at $\pi$, $\pi$ and softens rapidly near the zone center as one approaches the superconducting phase ($x=0.14$). These results are in clear contrast to the behavior in hole-doped cuprates such as the La$_{2-x}$Sr$_x$CuO$_4$ or the YBa$_2$Cu$_3$O$_{7-\delta}$ series.$0$

Recent theoretical analyses$2,15$ of data on $x=0.15$ samples suggest two different scenarios. The analysis of the data using a 1-band model Ref[15] suggests a no gap collapse scenario whereas the analysis of high doping data in Ref.[7] suggests a gap collapse scenario. Which of these two opposite viewpoints is correct cannot be ascertained from the data of Ref.[15] alone. Therefore it is crucial to study the evolution of the undoped compound of this series with high resolution so that the low-energy gap.
excitations can be accessed and modeled the evolution of the spectra from the $x=0$ Mott side. To interpret the present data in terms of RIXS spectra computed within the framework of a three-band Hubbard Hamiltonian of NCCO, based on Cu $d_{x^2-y^2}$ and two O $p_z$ orbitals, we extend the theoretical framework described in [7] by incorporating the doping evolution data made available here for the first time. The specific values of the parameters used in this work to fit the spectra are: $t_{CuO}=0.85$ eV, $t_{OO}=-0.6$ eV, $\Delta_0=-0.3$ eV, $U_p=5.0$ eV; where $t_{CuO}$ and $t_{OO}$ are the Cu-O and O-O nearest neighbor (NN) hopping parameters, $n_d$ [$n_p$] is the average electron density on Cu [O], $m_d$ the average electron magnetization on Cu, and $U$ [$U_p$] is the Cu [O] on-site Coulomb repulsion. The remaining parameter, crucial for fitting the doping evolution of the excitations, the Hubbard $U$, is taken to be 7.45 eV at $x=0$ with weak doping dependence: $U=6.69$ eV at $x=0.09$ and $U=6.27$ eV at $x=0.14$, so that the effective $U$ decreases by about 16% over this doping range, reflecting presumably the effects of screening. We note that before proceeding with RIXS computations, we determined the chemical potential and the magnetization $m_d$ on Cu sites self-consistently at each doping level. $m_d$ values so found are: 0.32 at $x=0$; 0.19 at $x=0.09$; and 0.12 at $x=0.14$.

Figure 4 (top) shows the calculated RIXS intensity maps. The positions of various experimentally observed spectral peaks (filled circles) and the leading edge of the low energy feature (filled diamonds) are superposed for ease of comparison. The high energy peaks involve transitions from the nonbonding O and bonding Cu-O bands to unoccupied states in the antibonding band, and fall in the same energy range as transitions involving other Cu and O orbitals not included in the present 3-band model. Therefore, it is appropriate to concentrate on the behavior of RIXS peaks within a few electron volts, which are associated with the antibonding Cu-O band, split by AFM ordering. In this energy region the theoretically predicted changes in the energies of various features as a function of momentum and doping are in reasonable accord with experimental observations highlighted in the discussion of Figs.2 and 3 above. In particular, the softening of low energy peaks is well reproduced and the collapse of the gap occurs only near $\Gamma$ in the computations. The latter effect can be readily understood. The RIXS transitions involve both intraband and interband contributions. In the absence of a gap, only intraband transitions are possible near $\Gamma$ which can only exist near zero energy transfer close to the Fermi level. Away from $\Gamma$, intraband transitions can take place at finite energies, but when a gap opens up, interband transitions become allowed, even at $\Gamma$.

Although the computed maps in the top row of Fig.4 are shown unbroadened to highlight spectral features, the theoretical spectra shown in the bottom panels of Fig.4 (blue lines) have been smoothed to mimic the broadening of experimental lineshapes. The relative intensities of the computed peaks are seen to be in agreement with our experimental results in lower energies, excepting...
In summary, we have utilized the unique momentum resolution of x-ray scattering to map the evolution of particle-hole excitations in the electron doped cuprate from its parent Mott state (x=0) to the doping on-set for superconductivity. We observe a nearly degenerate charge excitation mode near the (π, π) point around 5 eV in the Mott insulator, which splits on doping away from half-filling, with its lower energy branch softening anisotropically in approaching the AFM-SC critical doping. In contrast, the response near the (π,0) wavevector exhibits damping with relatively little softening. Our results indicate a systematic collapse of the gap consistent with the scenario of uniform doping when electrons are added into the parent Mott state which is in clear contrast to what is reported in hole-doped Mott insulators where charge inhomogeneity is commonly observed. The unusual softening of the modes from the Mott state likely contributes to many unusual behaviors of cuprates.

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[22] The spectra were first smoothed with a Gaussian of 0.37 eV width to account for the resolution. The spectra were further smoothed via folding with a Lorentzian of 0.30 eV width to mimic what appears to be an intrinsic linewidth.

[23] The branch near 4 eV is weak in the experimental data. Since this branch has a different symmetry than the 2 eV branch it resonates at a different photon energy (near 8.894 eV) as evident from the raw data profile in Fig-1(f).

[24] The experimental spectra for energy transfers beyond about 6 eV contain significant contributions from bands beyond the 3-band model we considered for this work.

[25] K. Penc and W. Stephan, Phys. Rev. B 62, 12707 (2000).

[26] In the one-band model, $U$ has a different definition, and the magnetic gap is $\Delta_m = 2U_m d$. Also, at $x=0$, ARPES does not see the upper band giving a lower limit to $\Delta_m$. 

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