

Doping Effect of Cu-O Bond Stretching Phonon in $La_{2-X}Sr_XCuO_4$ Studied by Inelastic X-ray Scattering

Since the discovery of superconductivity in copper oxides in 1986 many different kinds of experiments have been carried out in order to investigate the mechanism of superconductivity, and also many different mechanisms have been suggested from a theoretical point of view to explain the high transition temperatures of cuprates. However, the driving mechanism for Cooper-pair formation in cuprates remains unclarified despite such large efforts from both experimental and theoretical sides for almost two decades. Since the critical temperature (TC) observed in various cuprates, including La_{2-X}Sr_xCuO₄, is too high to explain the Cooper-pair formation by the so called BCS theory, which is based on a phonon mechanism, only limited attention has been paid to phonons for the mechanism of superconductivity in cuprates. Recently, however, angular resolved photoemission spectroscopy (ARPES) [1] and inelastic neutron scattering (INS) measurements [2] led to a renewed interest in phonons. INS measurements show a strong softening in the highest energy LO phonon branch with hole doping of La_{2-X}Sr_XCuO₄ (LSCO) and $YBa_2Cu_3O_{7-\delta}$ (YBCO). However, since such softening is observed among other nonsuperconducting oxides such as manganites and nickelates, the relationship between the softening and high-T_C superconductivity is not clear. In order to clarify a clear correlation of this phonon mode with the superconductivity, one would like to see the behavior of this phonon softening in the over-doped region where superconductivity is degraded and normal metallic conductivity dominates. For this purpose, we study by IXS the Cu-O bond stretching LO phonon of LSCO, which has a simple layered structure with single CuO₂ planes and whose doping concentration X can be controlled readily from the nondoped insulator phase to the overdoped normal metallic phase through the superconducting phase (see Fig. 1(a)). Therefore, one can expect to successfully investigate the doping dependence of the phonon properties precisely. For this purpose, we used two types of sample: one with a uniform Sr concentration in the crystal, and an other in which the concentration varies smoothly along the cylindrical axis (gradient sample) (see Fig. 1(b)). These samples enable us to investigate the doping dependence easily because IXS requires a small sample size. In this report, we use the tetragonal notation, in which the main (a,b) axes are parallel to Cu-O bonds in the CuO₂ plane. The superconducting transition was observed by measurement of the diamagnetic susceptibility, and the results are consistent with those in the published literature.



Fig. 1. (a) Schematic drawing of phase diagram of $La_{2,X}Sr_XCuO_4$ as a function of doping concentration X. (b) Doping concentration gradient of single crystal. X changes from 0.07 to 0.23 from one end to the other end of the single crystal.

46

The IXS experiments were performed at beamline BL35XU. The momentum and energy resolution in the experiment with 15.816 keV incident X-ray energy and about 100 µm spot size were about 0.076 Å⁻¹ and 6.0-6.2 meV (FWHM), respectively. The IXS measurements were performed with the sample in a reflection mode at 13 K and room temperature for X = 0.12, and at 60 K for X = 0.29. The reason for the measurements at low temperature is simply to reduce the tails of low energy phonons, and is not related to the measurement of superconducting transition. We focus on the LO high-energy Cu-O bond stretching phonons in the CuO_2 plane. Figure 2 shows the obtained dispersion of this mode for the samples with various doping concentrations together with neutron data, and Fig. 3 shows the amplitude of the softening, which is defined by the difference in energy between the maximum and minimum of the dispersion as a function of X [3], together with the results of recent theoretical calculations [4]. As can be seen, the softening shows a highly nonlinear dependence on X. Khaliullin and Horsch theoretically predicted that the softening simply depends linearly on X if no interaction between the low energy charge collective mode and the phonon is introduced [5]. Then, the anomalous softening can be defined as the deviation from the linear dependence of the softening on X, which is shown in Fig. 3 as a dashed line. The X dependence of T_C measured for the studied single crystals is also



shown in Fig. 3. What should be noticed is that anomalous softening occurs near the superconducting region, and the maximum deviation is seen at around the optimum doping concentration. The present observation of the precise X dependence of the phonon softening in the normal metallic phase will evoke further discussion and studies to elucidate the reason for the anomalous softening in hightemperature superconducting cuprates.



Fig. 3. Amplitude of softening as a function of doping concentration, X, together with results of theoretical calculations [4].

J. Mizuki*, T. Fukuda and K. Yamada

SPring-8 / JAERI

*E-mail: mizuki@spring8.or.jp

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47

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