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Low Temperature Phonon Anomalies in Cuprates

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Abstract

Our inelastic neutron scattering measurement on $La_{1.85}Sr_{.15}CuO_4$ single crystals shows that the in-plane LO phonon dispersion at low temperature is incompatible with the current view on the dynamic charge stripes, which for this composition should have the periodicity of $4a$. Instead the results are consistent with the dynamic stripes with the periodicity of $2a$, half of what is expected and a quarter of the magnetic periodicity. Calculations with the two-band t-t'-J model suggest that such $2a$ stripe charge ordering may help hole pairing.

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1. Introduction

For some time the electron-lattice coupling has been considered to play a negligible role in the superconductivity of high-T_C cuprates, and most theories have focused on magnetic mechanisms. However, the observation of the static spin-charge stripes in non-superconducting $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ opened up the possibility that the role of the lattice is more important than has been previously thought [1]. Currently the prevailing thought is that the charge-spin stripes exist even in the superconducting systems, but are dynamic and short range. The well known dynamic incommensurate magnetic peaks observed by inelastic neutron scattering are supposed to be the signature of magnetic stripes, but the corresponding lattice signature of charge stripes has not been observed. Our initial aim of this work has been to observe the dynamic charge-spin stripes by studying high energy phonons, since for such phonons the stripes may look frozen in time. As it turned out we observed something related but decidedly different. Our data are NOT consistent with the current idea of the charge-spin stripes, but suggest that the periodicity of the dynamic charge stripes is a half of what is currently believed. For $La_{1.85}Sr_{0.15}CuO_4$ the periodicity of the magnetic correlation determined from the dynamic magnetic satellites is ~ $8a$. This implies the periodicity of the charge stripes ~ $4a$, while our data are more consistent with the charge periodicity of $2a$.

2. Experimental Results

We have carried out inelastic neutron scattering measurements of the high frequency bond-stretching LO phonons of $La_{1.85}Sr_{0.15}CuO_4$ (T_C = 37 K). This phonon branch is known to show strong softening with hole doping [2]. Two single crystals were co-mounted in an aluminum can filled with He exchange gas. Both samples were grown by the floating zone method and were obtained from the same batch. The

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total size of the sample is approximately 40 x 20 x 10 mm^3. Previous measurements and characterizations attest to the high quality of this sample [3].

The experiments were made on the HB-3 triple axis spectrometer at the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory. The spectrometer configuration used the beryllium (002) and pyrolitic graphite (002) reflections as the monochromator and analyzer, respectively. The analyzer angle was fixed when performing inelastic scans, giving a fixed final energy of 14.87 meV. Soller collimators of angular divergence 48'-40'-80'-240' were placed along the flight path from source to detector. To reduce higher order Bragg scattering contamination from the analyzer, a pyrolitic graphite filter was placed before the analyzer.

Some of the results of constant-**Q** energy scans taken in the (3+q_x, 0, 0) Brillouin zone, in tetragonal notation (a = 3.78 Å), are shown in Fig. 1 for q_x = 0.5, 0.35 and 0.2. Because of the large incident energy required for measurements up to 90 meV the flux was low, and the count rates were 1~5 ct/min. The measurement in the Brillouin zone around (5, 0, 0) [2,4] suffers from spurious scattering consisting of the (6, 2, 0) Bragg reflection from the sample scattering incoherently from the analyzer that obscured the main phonon branch. Thus we stayed in the (3+q_x, 0, 0) zone, even though the intensity here is significantly weaker than in the (5±q_x, 0, 0) zone. In Fig. 1 the large peak at 58 meV is due to the oxygen in-plane Cu-O bond-bending mode. The 70 meV peak is associated with the oxygen Cu-O bond-stretching mode also in the CuO_2 plane. The energy scans at various values of q_x shows that the frequency of the 70 meV branch remains constant from (3.5, 0, 0) to (3.25, 0, 0), below which its intensity diminishes rapidly. At the same time some intensity appears at 85 meV, and becomes a strong peak below (3.25, 0, 0) down to (3, 0, 0). The peak positions are shown in Fig. 2. Thus it appears that bond-stretching phonon branch has split into two nearly dispersionless sub-branches, with the intensity crossover at (3.25, 0, 0).

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3. Discussion

The experimental results at T = 10 K shown here are in conflict with the previous measurement of the bond stretching branch in $La_{1.9}Sr_{0.1}CuO_4$ at room temperature [2]. The previous result shows a strongly dispersing, but continuous, single branch from (0, 0, 0) to (0.5, 0, 0), in contrast to the two remarkably dispersionless sub-branches shown in the data presented here. The difference in these two measurements most likely arises from the difference in temperature, according to our preliminary measurements of temperature dependence. On the other hand our results are similar to the ones observed for $YBa_2Cu_3O_7$ [2] and $Bi_{0.6}K_{0.4}BaO_3$ [5]. The two peaks seen in Fig. 1 for $q_x = 0.35$ resemble those observed for $YBa_2Cu_3O_7$ that provoked a dispute on the "extra" phonon branch [2,6]. The claim of the extra phonon branch was later withdrawn citing the possibility of compositional inhomogeneity, but such inhomogeneity is highly unlikely for the present sample that shows a very clean spin gap [3]. Thus our observation revives the controversy over the extra-branch.

The dynamic magnetic satellites in this sample were observed at $(0.5\pm\delta, 0.5, 0)$ and $(0.5, 0.5\pm\delta, 0)$, with $\delta = 0.125$, indicating the wavelength of magnetic periodicity is $8a$ [3]. Thus we expected the lattice signature of the charge-spin stripes at $(\pm 2\delta, 0, 0)$ and $(0, \pm 2\delta, 0)$. Such dynamic superstructures will create pseudo-Brillouin zone boundaries for high energy phonons at δ, 2δ, and 3δ. The observed dispersion shown in Fig. 2 is consistent with the pseudo-Brillouin zone boundary at 2δ, but not at δ and 3δ. Indeed a spring model created assuming the charge-spin stripes with the period of $4a$ shows a dispersion which is split at δ and 3δ, and is qualitatively very different from the observed one as shown in Fig. 3. On the other hand, if we assume the charge-spin stripes with the periodicity of $2a$ (Fig. 4) instead of $4a$, the calculated dispersion is in good agreement with the observation as shown in Fig. 5. It is interesting to note that in this case the charge must be on oxygen ions. The model with the charge on Cu showed poor agreement. Since we assumed a periodic structure in the simulation the branches have some dispersion. If we introduced disorder and localization the branches would have shown less dispersion. From the flatness of dispersion we estimate the correlation length of charge ordering to be about 20 Å ($5a$) along the stripes and 8 Å (2*a*) across them. These correlation lengths define an area which contains just about one hole, since the linear charge density in the stripe is ¼ per unit cell. Our earlier search for superlattice diffraction (elastic) at (0.5, 0, 0) was negative. Thus we conjecture that the charge periodicity with $\lambda = 2a$ is dynamic and short range.

Our results cast a serious doubt on the conventional picture of charge-spin stripes obtained by merely extrapolating from the static stripe structure of non-superconducting compounds. Instead our results suggest that while the dynamic charge stripes indeed exist, they have the periodicity of 2*a* and are centered on oxygen ions. Such charge stripes will interact strongly with the Cu bond-stretching half-breathing mode at (0.5, 0, 0). It is interesting to note that Harashina *et al.* reported strange phonon behavior in $YBa_2Cu_3O_{6.6}$ for this mode in the vicinity of the (0.5, 0, 0) point around 31 - 33 meV [7]. We have observed that the corresponding mode in the present sample at 29 meV also shows anomalous temperature dependence. This provides another evidence of the presence of the 2*a* charge periodicity.

It is still possible that our data are consistent with the stripe periodicity of 4*a*, if we assume that the 2*a* charge periodicity exists *within* the stripe. Since the linear charge density is ½ in the conventional stripe, the Peierls distortion in the spin-polarized state will produce the lattice distortion with the periodicity of 2*a*, and charges will be localized. However, our simulations for such a case failed to reproduce the strong crossover at $q_x = 0.25$, and the dispersion appeared similar to the result in Fig. 3. Furthermore this picture is inconsistent with the high conductivity of the system and the widely held assumption that spins are unpolarized within the stripes. Thus in our view this possibility is very remote.

The static stripes observed earlier were associated with the antiferromagnetic (AFM) domain boundaries [1,8]. In order for the charge periodicity of 2*a* to be compatible with the magnetic periodicity of 8*a*, the spin rotation through the charge stripe has to be $\pi/2$, rather than π as in the static stripe, and the magnetic structure has to be in a chiral AFM state with a phase slip of $\pi/2$ at every other Cu-Cu bond. The average linear charge density is about ¼ per unit cell, rather than ½ as in the static stripes. This provokes interesting thoughts about the relationship among charge, spin of the charge, the magnitude of spin rotation, and chirality. This subject deserves a very detailed study.

We have expanded the t-t'-J model of Emery and Reiter [9] with the holes on oxygen ions, and studied the effect of Cu half-breathing mode using the exact diagonalization method [10]. For a perfectly periodic CuO_2 plane the strongest hole-hole pairing occurs for oxygen ions separated by $2a$ along the Cu-O chain, followed by the pairs separated by a across the Cu_4O_4 square. This is already suggestive of the possible relation between the stripes and pairing. If a static half-breathing Cu mode was introduced by the frozen phonon approach, the $2a$ charge stripes are induced. Also the strengths of the two kinds of pairs above are exchanged, making the intra-stripe pairing (separated by a across the Cu_4O_4 square) more favored compared to the inter-stripe pairing (separated by $2a$ along the Cu-O chain). These results suggest that the presence of the $2a$ charge stripes may enhance superconductivity in the CuO_2 plane.

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Figure captions

Figure 1 - Constant-Q energy scans along the $(3+q_x, 0, 0)$ direction at 10 K at $q_x = 0.5$, 0.35 and 0.2. The dashed line is the background obtained from the counting time for each point and assuming a time-independent count rate of 1.4 counts/minute. The solid line is the fit to a sum of two or three gaussians plus the background. The strong peak at 58 meV is due to the oxygen bond bending mode which is not important here.

Figure 2 - The peak position for the upper two longitudinal optic branches along $(Q_x, 0, 0)$ as obtained from the gaussian fits shown in Fig. 1. The empty circles indicate the frequency of the weak extra branches. The shade at (3.25, 0, 0) indicates a broad peak in the intensity.

Figure 3 - Contour plot (each 15 % of the maximum) of the scattering intensity for the $(q_x, 0, 0)$ LO phonon mode calculated with the spring model assuming the charge-spin stripe structure with $\lambda = 4a$. The oxygen-Cu interaction parameter was softened by 40 %, oxygen-oxygen parameter by 20 % when the oxygen charge is present. The

phonon peaks were broadened with a gaussian with the full width 7 meV to simulate the experimental resolution. The results are qualitatively different from the observation.

Figure 4 - Schematic hole stripes in the CuO_2 plane. Small circles are Cu ions, and large circles are oxygen ions. Dark shade on oxygen ions represents low hole density, and light shade high hole density. Here for simplicity a fully periodic model is shown. In reality, however, charge ordering is dynamic and short range, over 20×8 Å.

Figure 5 - A similar plot as Fig. 3, but for the stripe pattern with $\lambda = 2a$ shown in Fig. 4. The result shows only two branches and a large gap between the two. This simulation shows much better agreement with the experiment, except that the dispersions here are stronger.

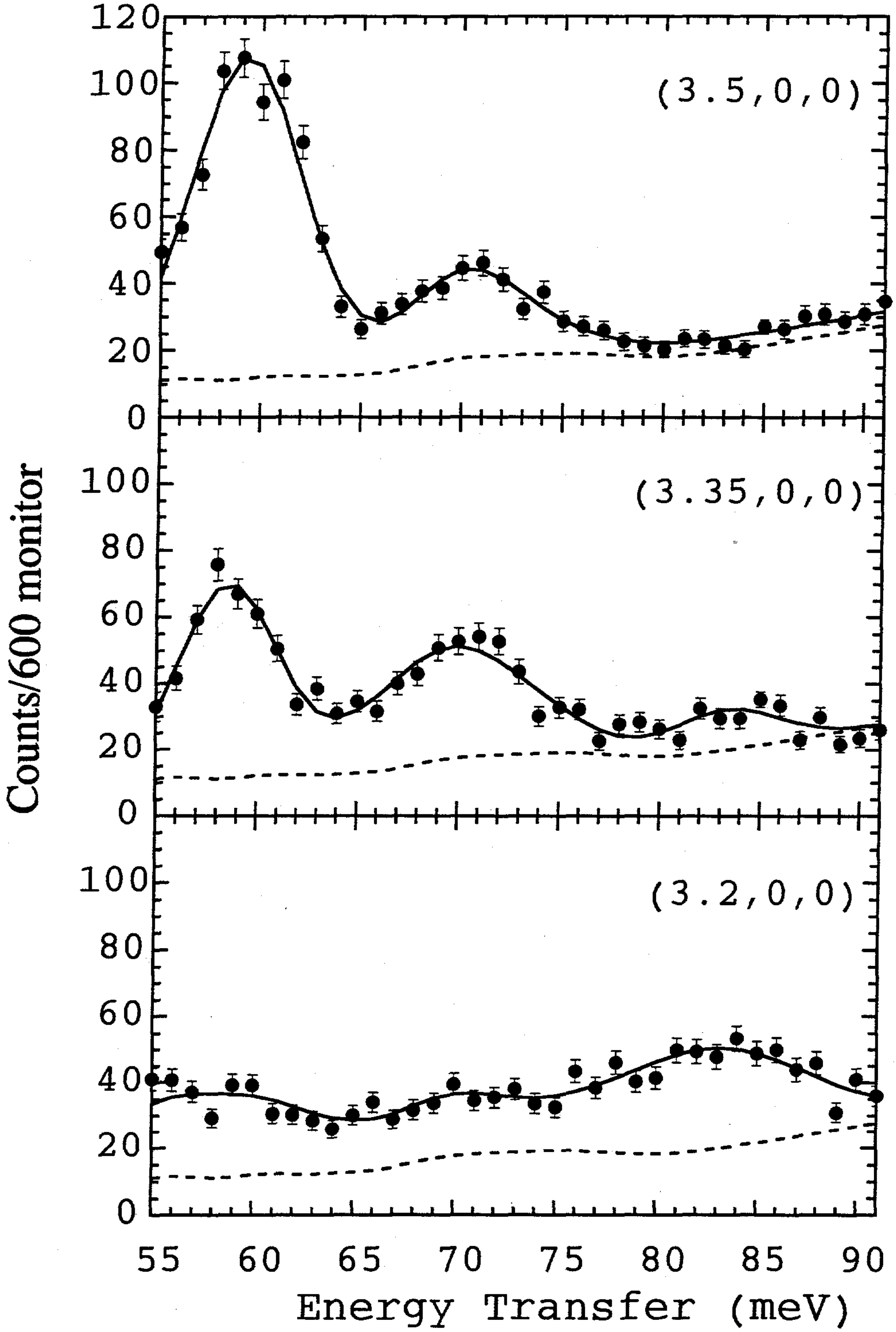

Fig. 1

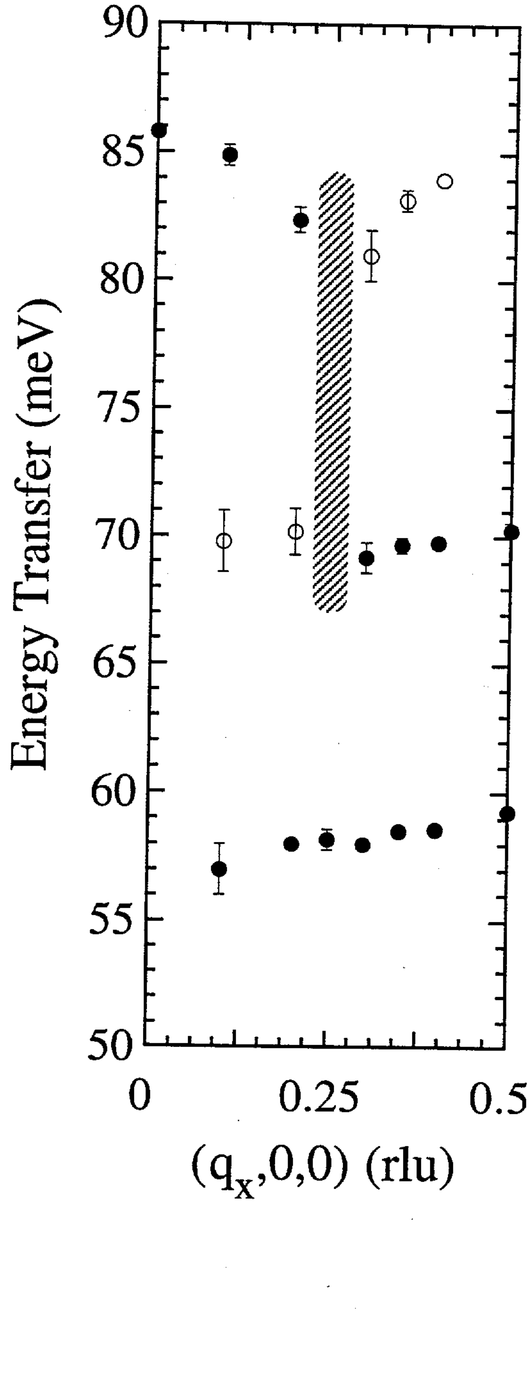

Fig. 2

Cu->O<-Cu (π/2,0) mode

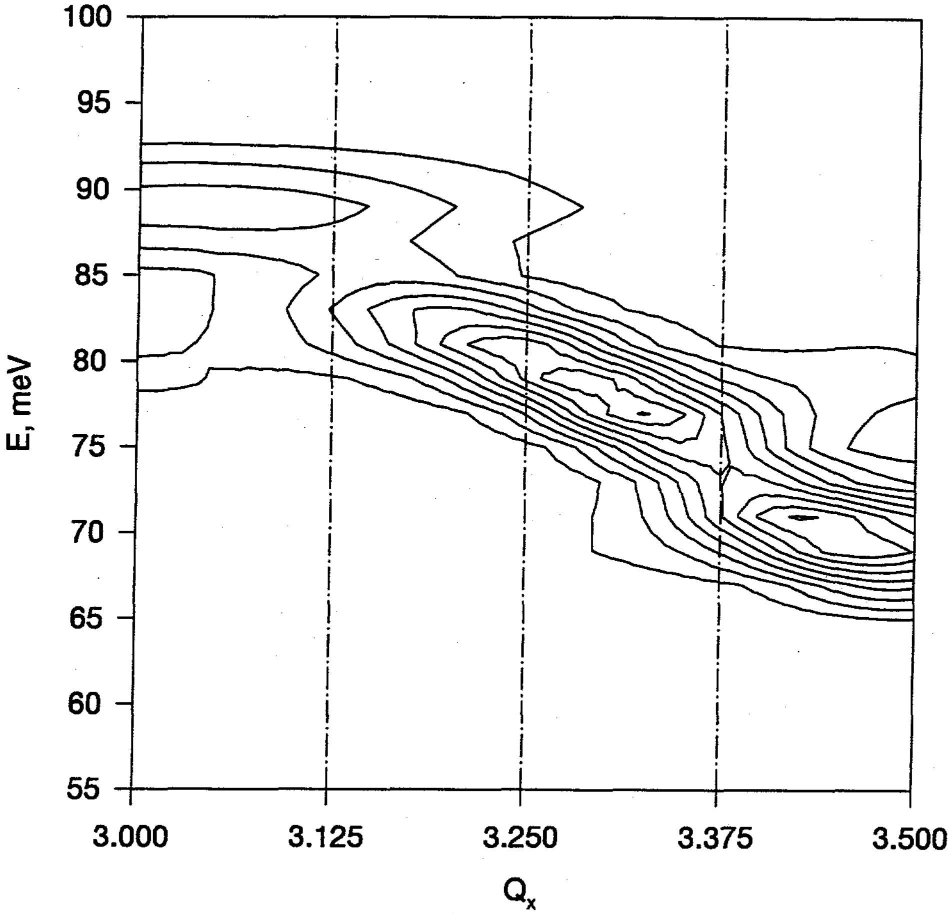

Fig. 3

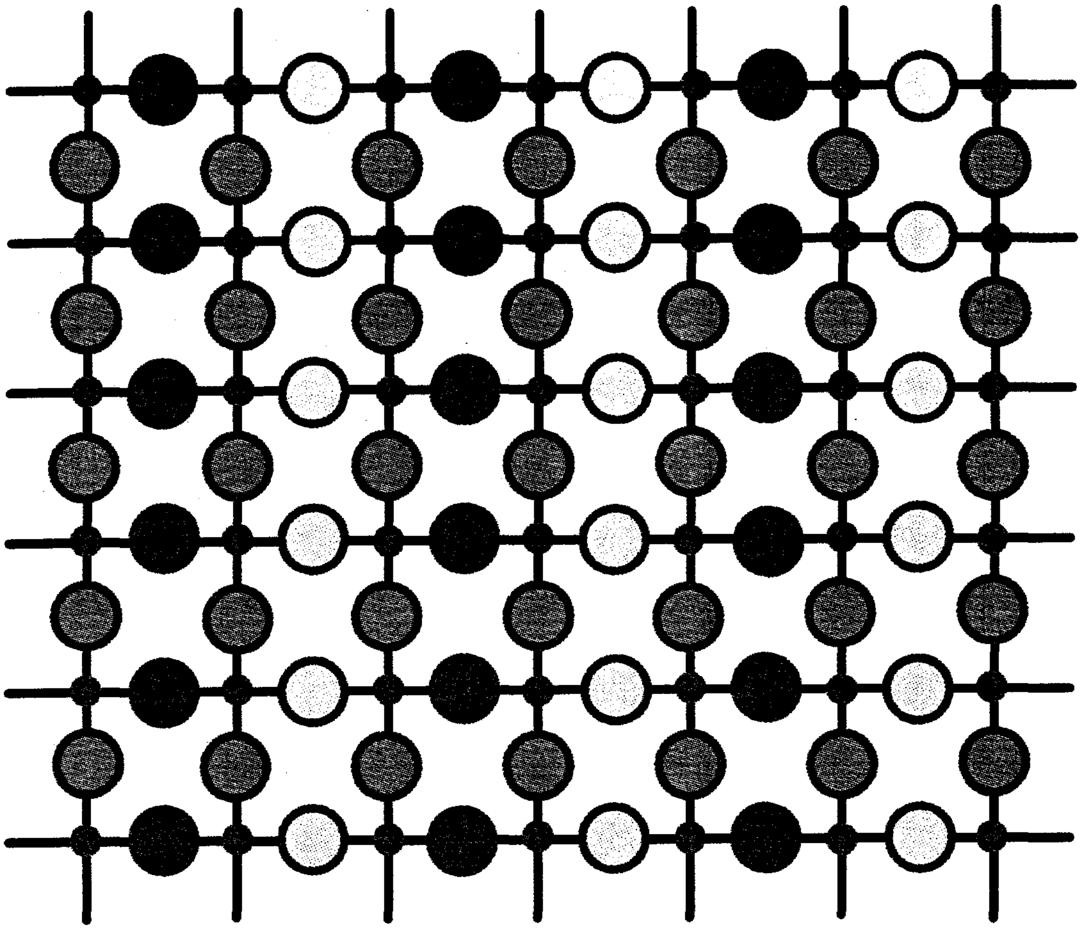

Fig. 4

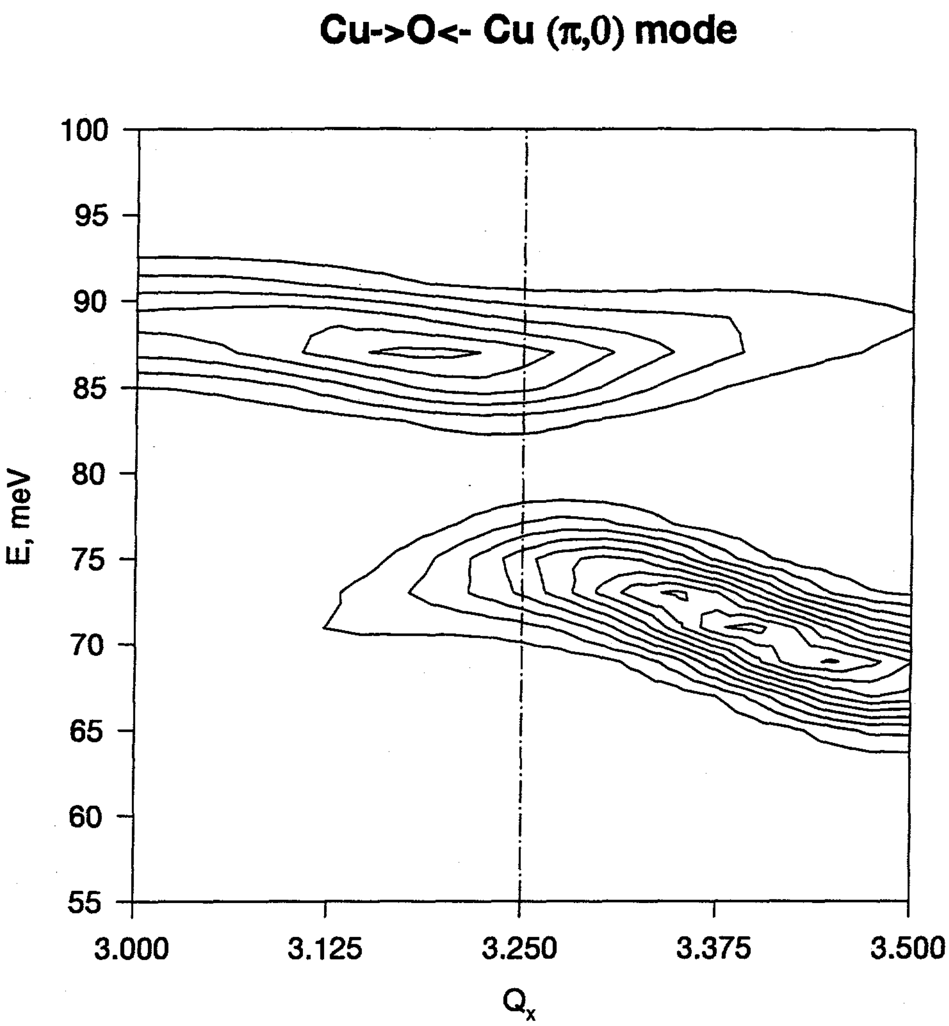
Cu->O<- Cu (π,0) mode
100
95
90
85
80
75
70
65
60
55
E, meV
3.000
3.125
3.250
3.375
3.500
Q_x

Fig. 5