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Pressure effects on the structural phase transitions in $La_{2-x}Ba_xCuO_4$

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Abstract

Triple axis neutron scattering experiments with improved momentum transfer resolution were used to study the sequence of the structural phase transitions in $La_{2-x}Ba_xCuO_4$ under high pressure. For the composition x=0.12 and ordinary pressure we confirm the previously reported sequence of a high temperature tetragonal (HTT) phase (I4/mmm) which, around 220 K, continuously changes into the low temperature orthorhombic (LTO) phase (Abma) and finally transforms almost completely into a low temperature tetragonal (LTT) phase ($P4_2/ncm$). Scanning the hydrostatic pressure up to 5 Kbars at given temperature chosen in the intermediate LTO phase we show that the orthorhombicity (a-b)/(a+b) is continuously reduced at the rate of $1.310^{-4}/Kbar$ compatible with a decrease of the HTT-LTO transition temperature at the rate of $-6.5K/kbar$. At low temperature we were able to study the pressure dependence of particular Bragg reflections and find that the LTO-LTT phase transition temperature is also depressed at the rate of $-3K/kbar$. Moreover we have determined the compressibility in the LTT phase. At 10 K we have found a compressibility of the order of 1.36 $10^{-3}/kbar$ while in the LTO we have measured a significantly smaller compressibility 4 $10^{-4}/kbar$ at about 80K. The evolution of the transition lines will be discussed within the framework of the electron-lattice coupling.

INTRODUCTION

It is well known that the lattice instability associated with the CuO_6 tilt mode which transforms $La_{2-x}Sr_xCuO_4$ from the high temperature tetragonal (HTT) phase into the low temperature orthorhombic (LTO) phase, is well understood in terms of ionic forces[1–3]. The dependence of the HTT-LTO upon the dopant concentration is attributed to the screening of these forces by the mobile holes. In $La_{2-x}Ba_xCuO_4$ a second structural instability into a low temperature tetragonal phase (LTT) occurs[4] near the composition x=0.12 along with a drastic suppression[5] of the superconducting transition temperature Tc. This result as well as other modifications of the transport properties reported[6] at the LTO-LTT transition, suggest that the electronic structure is significantly modified. Although structurally, apart the difference in the rotation axis (110) in the LTO phase and (100) in the LTT phase, the amplitude of the tilt angle of the CuO_6 octahedra is practically identical[7]. As a matter of fact, as noticed by Barisic[8], the orientation of the tilt is important. The LTT tilt mode unlike the LTO tilt mode, lifts the degeneracy between the O^x and O^y oxygen atoms in the CuO_2 planes introducing different site energies which would favour the in-plane O^x-O^y charge fluctuations. These charge fluctuations specific of the LTT phase must be part of the electronic structure and then responsible of the drastic modification of the transport properties. Extensive resitivity measurements in $La_{2-x}Ba_xCuO_4$ performed[9] under high pressure show that the dip observed in Tc(x) at ordinary pressure near 0.12 is completely washed out at about 20 Kbars. This result naturally leads to the question of the simultaneous effects of the hydrostatic pressure on the structural instabilities.

In this paper we will give a detailed account of the neutron scattering studies of the HTT-LTO and LTO-LTT instabilities as a function of pressure up to 5 Kbars and for two compositions x =0.10 and 0.12 .

EXPERIMENTAL

The neutron scattering studies were performed on the 1T triple-axis spectrometer at the Orphée reactor at the Laboratoire Léon Brillouin in Saclay. Constant neutron energy of 14.7meV was selected using Cu(111) and PG(002) crystals as monochromator and analyzer.

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A pyrolytic graphite filter was used to suppress higher order contaminations. The necessary good momentum resolution was achieved using the Cu(111) monochromator combined with a tight collimation set at 40'-30'-10'-10' from reactor to detector.

The powder samples corresponding to x=0.10 and 0.12 were prepared from appropriate starting composition of La_2O_3, $BaCO_3$ and CuO following the procedure described earlier in reference (5). The quality of the samples was checked using X-ray powder diffraction. At room temperature all the observed peaks could be indexed in the body centered tetragonal unit cell with space group I4/mmm and the lattice parameters a=b=3.805 , c=13.174 Å. No evidence of impurity phases could be detected. The powder sample was then mounted in the aluminum pressure cell placed in a cryostat with appropriate temperature and helium pressure controlers.

RESULTS

In figure (1) we show, for the composition x=0.10 and a pressure of about 5.2 kbars, a set of longitudinal scans through the (2,2,0) reflection as a function of temperature. Notice that the two peak structure of the scattering profile corresponds to the orthorhombic splitting. The top and bottom scans which correspond to a single peak are associated with the high and low temperature tetragonal phases. As in the previous X-ray studies at normal pressure[4] we have analyzed the intermediate scattering profiles assuming two or three peaks. We were able to determine precisely the HTT-LTO transition temperature T_d by appropriate extrapolation of the orthorhombic splitting to zero. We obtained $T_d = 235 \pm 5K$ at 5.2 kbars while at normal pressure we get $270K$. Thus the pressure dependence of the HTT-LTO transition temperature can be accounted for by dT_d/dP = -6.5 K/kbar. At low temperature below about 40K, the scattering profiles are satisfactorilly fit assuming three peaks. This reflects the first order nature of the LTO-LTT transition with a coexistence of the two phases already described at normal pressure[4]. We determine the LTO-LTT transition temperature T_d' as the temperature at which the three peak fit gives better agreement than the two peak fit.

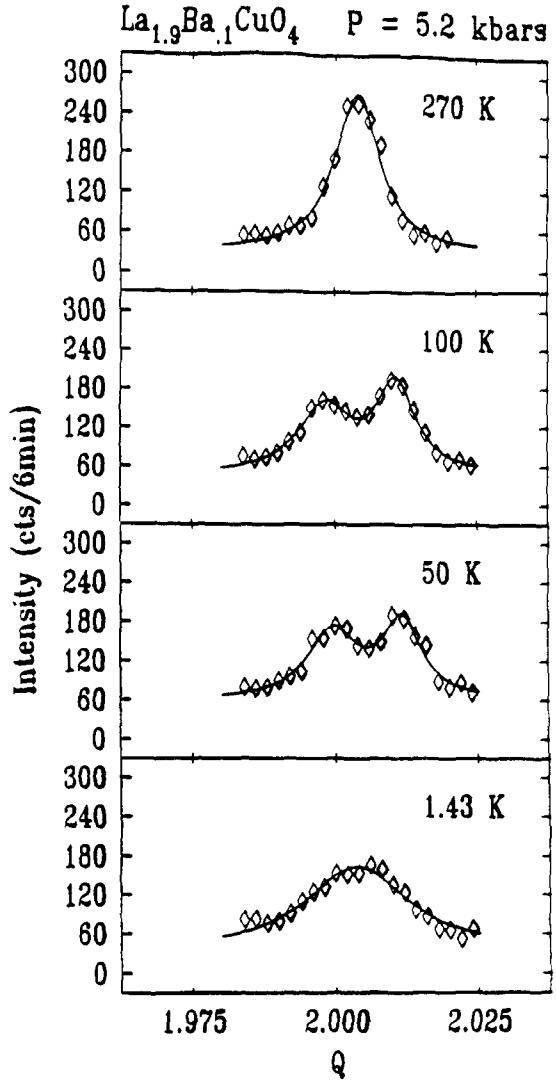

Fig-1 *Evidence of HTT-LTO-LTT phase transitions in $La_{2-x}Ba_xCuO_4$ at high pressure P = 5.2 Kbars and for x = 0.1. Solid lines are fits to Gaussian lineshapes.*

We find $T_d' = 40 \pm 5K$ at 5.2 kbars while at normal pressure it is about $55K$. Here again the hydrostatic pressure depresses the LTO-LTT transition at the rate of -3 K/kbars. We performed similar studies at the higher composition x=0.12 for which the orthorhombicity is even smaller at normal pressure. In figure(2) we show the pressure evolution of the scattering profile at 79 and 9 K. The resulting transition temperatures T_d and T_d' are summarized in the Pressure-Temperature phase diagram shown in figure(3) for the two compositions 0.1 (solid lines) and 0.12 (dashed lines).

Using the pressure dependence of the lattice parameter we were able to determine compressibilities. In the LTT phase at about 10 K and x=0.12 we find $\chi = -\frac{dV}{VdP} \sim 1.3 \pm 0.1\ 10^{-3}/kbar$ whereas in the LTO phase for the same composition at about 80 K we find a smaller value $\chi \sim 4. \pm 0.5\ 10^{-4}/kbar$.

DISCUSSION

We have shown that when applying a hydrostatic pressure on $La_{2-x}Ba_xCuO_4$, both T_d and T_d' are depressed at the rates of -6.5 and $-3K/kbars$ respectively. In the undoped La_2CuO_4 , the modification of the HTT-LTO transition has been investigated theoretically by B Piveteau et al [3]. They have clearly shown that in this limit the coulomb interactions favour the orthorhombic phase but as the hydrostatic pressure is increased the short range Born-Mayer forces are increasingly limiting the tilt angle. In the present Ba doped compounds the LTO phase is destabilized with respect to both the HTT and LTT phases. The fact that the charge carriers interact strongly with the LTT instability as shown in the transport properties[6], is necessary to include the effects of the mobile holes to account at least partly for the pressure effects on T_d and T_d'. For a qualitative discussion of the electronic effects, we chose to follow L. Vasiliu-Doloc et al[11] modeling of the successive transitions HTT-LTO-LTT. These authors have used the polaron theory of superconductivity[12] based on the coupling of the charge carriers with local optical-type vibrations of the oxygen atoms. Considering the tilt angle of the octahedra to be time independent parameter and assuming it to enter the coupling of the in-plane charge carriers to the sheet bending mode they were able to derive the dependence of the tilt angle θ on the carrier concentration $\theta = \theta_o e^{-ni/x_{cr}}$, where x_{cr} is the upper critical concentration above which the HTT phase is stable. At zero temperature the average occupation number n_i is identified with the hole concentration and x_{cr} is identified with the transition concentration. Taking into account the experimental fact that the carrier concentration changes with temperature, a two level model of the electronic structure has been introduced with appropriate thermal distribution of the carriers. A comparison of the average occupation

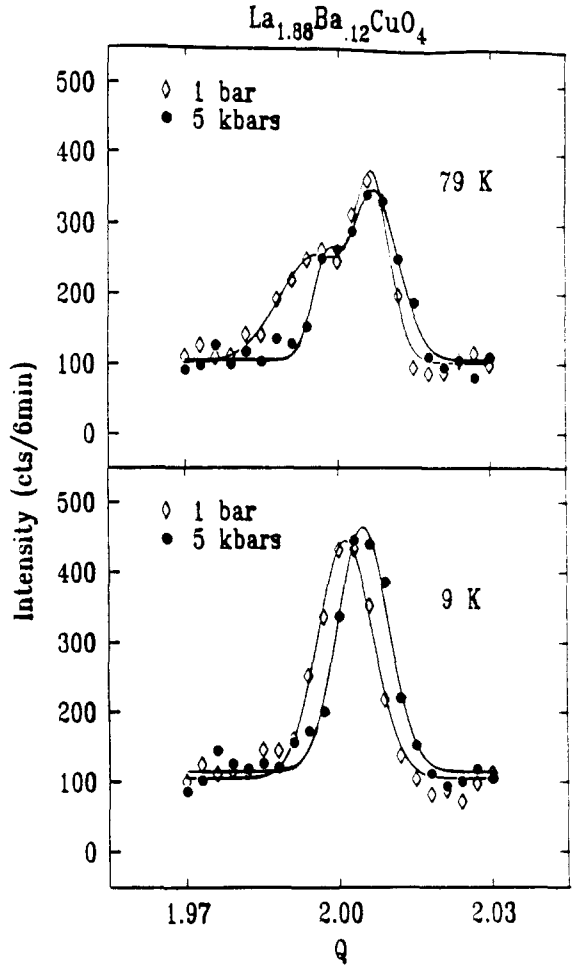

Fig-2 *Pressure dependence of the (2,2,0) reflection in the LTO and LTT Phases for x=0.12. Solid lines are fits to Gaussian lineshapes.*

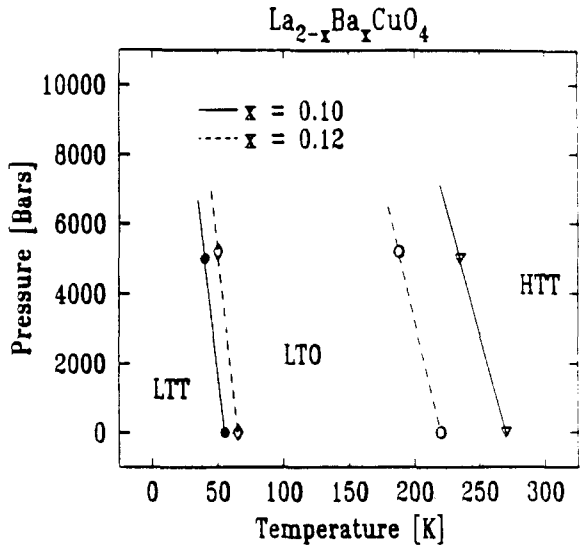

Fig-3 *Pressure-Temperature phase diagram for the two compositions x=0.1 and x=0.12.*

number with x_{cr} leads to the structural HTT-LTO transition line $Td(x)$ given by

$$T_d = \frac{\delta}{2}\frac{1}{ln(\frac{1-x_{cr}}{x_{cr}-x})}$$

where δ measures the two levels separation, it can be considered as a variational parameter of the electronic structure. To take into account the modification of the electronic structure in the LTT phase, these authors introduced a second critical concentration x'_{cr} above which we suggest that the effective number of holes per CuO_2 layer is reduced by a quantity $2\times(x'_{cr}-x)$. This also means that for some higher composition the charge carriers may change sign consistent with Hall effect studies[13,14] in the Sr doped compounds. Assuming that the average effective number of free carriers in the LTT phase must be smaller than the critical composition x'_{cr}, similar expression for the LTO-LTT transition line is derived

$$T'_d(x) = \frac{\delta}{2}\frac{1}{ln(\frac{1-x'_{cr}}{x-x'_{cr}})}$$

using the same parameter δ . The pressure derivative of T_d and T'_d reads

$$\frac{dlnT_d}{dP} = \frac{dln\delta}{dP} + \frac{1-x}{(1-x_{cr})(x_{cr}-x)ln(\frac{1-x_{cr}}{x_{cr}-x})}\frac{dx_{cr}}{dP}$$

$$\frac{dlnT'_d}{dP} = \frac{dln\delta}{dP} - \frac{1-x}{(1-x'_{cr})(x-x'_{cr})ln(\frac{1-x'_{cr}}{x-x'_{cr}})}\frac{dx'_{cr}}{dP}.$$

The numerical evaluation of this system at the composition $x = 0.12$ and for the observed values $x_{cr} \sim 0.24$ and $x'_{cr} \sim 0.04$ leads to

$$\frac{dlnT_d}{dP} = \frac{dln\delta}{dP} + 1.26\frac{dlnx_{cr}}{dP}$$

$$\frac{dlnT'_d}{dP} = \frac{dln\delta}{dP} - 0.18\frac{dlnx'_{cr}}{dP}.$$

At this level it is natural to make use of the pressure dependence of x_{cr} as related to the in plane compressibility by $\frac{dlnx_{cr}}{dP} = -\chi_p \sim -2/3\chi$. With our experimental values of the depression rates for T_d and T'_d we can reasonably say that the electronic parameter δ decreases at the rate of $2.2meV/kbar$. Moreover our results indicate that, while the critical concentration x_{cr} which controles the amplitude of the tilt angle slightly decreases with pressure, the critical concentration x'_{cr} which controles the relative number of carriers in LTT with respect to LTO phase, strongly increases at the rate of $\frac{dlnx'_{cr}}{dP} \sim 0.11/kbar$. This result is rather consistent with the fact that even though the Sr-doped La_2CuO_4 has no LTT phase, its critical concentration x_{cr} is very close to that of the Ba doped compound. Thus it is reasonable to conclude from the present pressure studies that the modification of the electronic structure within the range of $2meV$ is sufficient to induce the LTT phase and from the structural point of view the Ba system under 16 kbars hydrostatic pressure is equivalent to the Sr doped system. To get more quantitative understanding of x'_{cr} other studies at different compositions and higher pressure are planned. *We would like to thank Drs. J.D. Axe, M. Apostol for interesting discussions and R. Leprovost for technical assistance.*

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