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Temperature-dependent x-ray studies of the high-T_c superconductor $La_{1.9}Ba_{0.1}CuO_4$

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Simultaneous x-ray diffraction and four-probe electrical resistance measurements have been performed as a function of temperature on a polycrystalline sample of the high-T_c material $La_{1.9}Ba_{0.1}CuO_4$. Comparisons of the diffraction spectra between 290 and 18 K show no evidence of any gross structural distortions. Least-squares refinements of the tetragonal unit-cell parameters at several temperatures indicate smooth monotonic thermal expansions of about $0.10 \pm 0.05\%$ along the a axis and about $0.35 \pm 0.10\%$ along the c axis over this temperature range.

I. INTRODUCTION

It has recently been reported[1] and confirmed[2] that superconductivity occurs in La-Ba-Cu-oxides at temperatures (T_c) in excess of 30 K. Subsequent structural studies[3] have identified the superconducting phase as being $(LaBa)_2CuO_{4-y}$, crystallizing in the tetragonal K_2NiF_4-type structure. More recently, a magnetic and resistive anomaly was reported in $La_{1.9}Ba_{0.1}CuO_4$ near 65 K, well above T_c. We have performed a series of x-ray diffraction measurements as a function of temperature to investigate whether this anomaly is associated with a structural distortion and to obtain structural information on this material at low temperatures.

II. EXPERIMENTAL PROCEDURES

The test specimen was prepared by mixing appropriate proportions of reagent grade powders of La_2O_3, $BaCO_3$, and CuO to yield a product of nominal composition $La_{1.9}Ba_{0.1}CuO_4$. The mixture was calcined twice in air at 1000–1100 °C for approximately 12 h and pressed into a cylindrical pellet.

The pellet was mounted with thermally conducting grease on the cold stage of a Heli-tran cryogenic refrigerator. This refrigerator system is coupled to a computer controlled x-ray diffractometer in a manner similar to that previously described.[4] A calibrated Si-diode thermometer is also bonded to the low-temperature block, and coarse temperature control is achieved by regulating the flow rate of He fluid from a remote storage dewar. Fine control is accomplished with an electrical heater and feedback control circuit using either a Pt or Ge sensor, depending on the temperature range. The measured temperatures are estimated to be accurate and stable to within ±0.5 K.

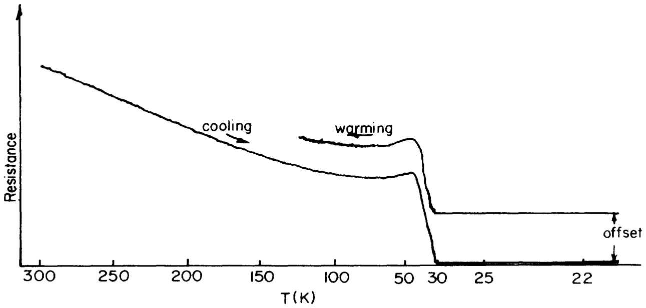

FIG. 1. Temperature dependence of the electrical resistance exhibiting a local minimum near 70 K.

Four leads were bonded to the sample with In solder and the electrical resistance was continuously monitored. A typical resistance curve is shown in Fig. 1. Although the resistive anomaly reported in Ref. 5 was not observed, possibly due to aging effects, a local minimum in the resistance near 70 K was observed to precede the onset of superconductivity which occurs at about 38 K. The superconducting transition is complete at 29 K.

Concurrent with the resistance measurements the sample was illuminated with radiation from a Cu x-ray tube operated at 30 kV and 20 ma. A 9 μm-thick Ni foil was placed in the incident beam to attenuate the Cu-K-β radiation.

III. RESULTS AND DISCUSSION

Diffraction spectra recorded at 297.5 and 24.0 K are shown in Fig. 2. All but four of the diffraction peaks can be indexed on the basis of the tetragonal K_2NiF_4-type structure. These four peaks, identified as A–D in Fig. 2, are believed to be associated with a small component of the orthorhombic phase, a distortion of the K_2NiF_4 structure,[6] present in the sample.

Based on least-squares analyses of the measured Bragg angles of 15 diffraction peaks between 40° and 80°, the $[a;c]$-tetragonal unit cell parameters are found to be (3.7873 ± 0.0012 Å; 13.235 ± 0.019 Å) at 297.5 K and (3.7820 ± 0.0025 Å; 13.168 ± 0.037 Å) at 24.0 K. To provide an estimate of the linear thermal expansivities and possibly detect anomalous behavior in the vicinity of the electrical or magnetic anomalies, measurements of five diffraction peaks (2,1,7), (2,0,8), (3,0,3), (3,1,0), and (1,1,10), were performed at several different temperatures. The temperature dependence of the fractional change in the unit cell parameters derived from the data

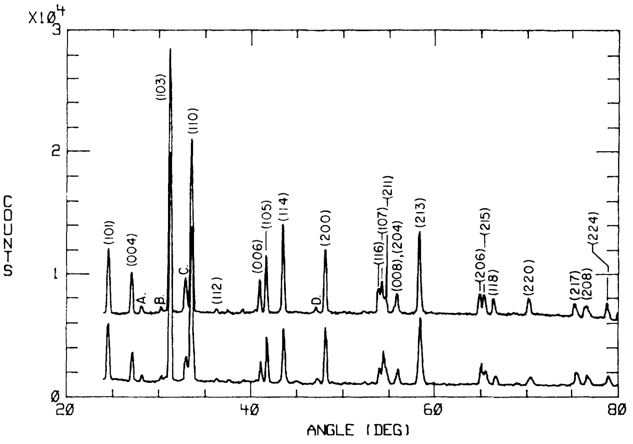

FIG. 2. X-ray diffraction spectra as recorded at 297.5 K (upper curve) and 24.0 K (lower curve).

are plotted in Fig. 3. There is an overall expansion of about 0.10 ± 0.05% and 0.35 ± 0.10% in the *a* and *c* axes, respectively. To within the uncertainty of the measurements, there is no indication of any unusual behavior in these linear expansivities.

Most recently, neutron powder diffraction measurements have been reported on $La_{1.85}Ba_{0.15}CuO_4$ at 295 and 10 K.[7] The observations reported here are in agreement with the results of the neutron measurements. In summary, the results of these experiments do not indicate any gross low-temperature lattice distortions in $La_{1.9}Ba_{0.1}CuO_4$ over the temperature range from 18 to 300 K. It should be noted that this does not rule out the possibility of a charge density wave transition or other less demonstrative effect occurring in these materials. Single-crystal samples would be helpful in searching for these possibilities.

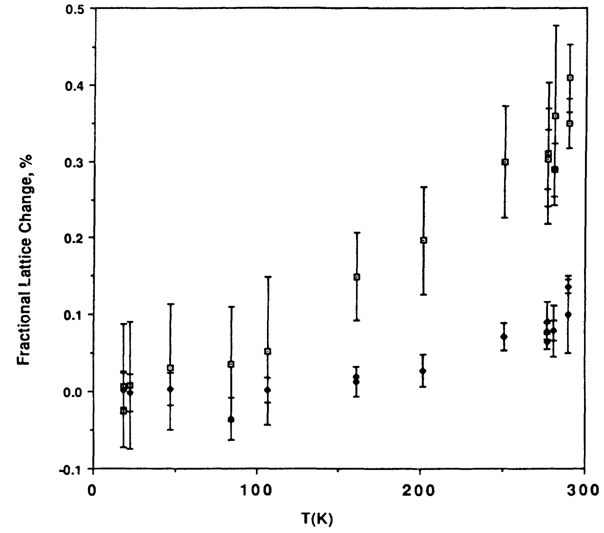

FIG. 3. Fractional change in the tetragonal *a* and *c* lattice parameters on increasing temperature from 18 K.

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