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ION-CHANNELING OBSERVATION OF CORRELATED ATOMIC DISPLACEMENTS BELOW T_c IN $YBa_2Cu_3O_{7-x}$ AND Pb-DOPED $Bi_2Sr_2CaCu_2O_x$*

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Temperature dependent studies of ion channeling in high-quality, high-T_c single-crystals are summarized. The measurements revealed an abrupt change across T_c in displacements in the a-b plane of the Cu(1 and 2) and O(4) atoms; normal "Debye-like" vibrations were found for the Y and Ba atoms. The anomalous atomic displacements were found for both proton and He channeling, and manifested themselves as an abrupt increase in the critical angle and a simultaneous decrease in the minimum yield. The anomalous change in Cu-O displacements shifted directly with stoichiometry-induced changes in T_c, implying a causal link between the observed phonon anomaly and the superconducting state. An apparently identical anomaly was found in $(Bi_{1.7}Pb_{0.3})Sr_2CaCu_2O_x$, indicating that it is a general feature of high-T_c superconductivity. A comparison with other experimental measurements in $YBa_2Cu_3O_{7-x}$, including a detailed neutron diffraction study, indicates that the anomaly is not due to an overall reduction in average vibrational amplitude, but arises instead from a strongly correlated sequence of Cu(1 and 2) and O4 displacements that appears with the onset of superconductivity. These strongly correlated displacements are either dynamic, or they are static distortions that fail to preserve the overall crystal symmetry.

1. Introduction

Ion channeling[1] results from a sequence of small-angle collisions produced when an energetic ion-beam becomes closely aligned with a major crystallographic axis of a single-crystal. The resultant steering of the ions between the atomic rows and planes of the crystal causes a large reduction in ion-atom collisions, which produces a strong reduction in Rutherford Backscattering (RBS) and characteristic X-ray yields. The critical angle ($\leq 1^{\circ}$) of incidence for ion-channeling to occur is determined by the ion energy, the atomic numbers of the projectile and target, the interatomic spacings, and most important for the present discussion, any displacements (static or thermal) of the target atoms from their regular lattice sites.

Stoeffel et al.[2] first demonstrated the excellent ion-channeling properties of $YBa_2Cu_3O_{7-x}$ single crystals along the [001] axis, obtaining room temperature minimum yields as low as a few per cent, and only a small amount (<1 monolayer) of equivalent surface disorder (for Y, Ba, and Cu) even in as-annealed specimens. The excellent ion-channeling properties make it possible to determine changes in average atomic displacements smaller than 1 pm in high-quality single crystals of this material. Channeling along other directions is degraded by the presence of twins, as well as by the existence of small periodic lattice distortions. The presence of an incommensurate phase modulation causes similar degradation of channeling scans from the Tl and Bi high-T_c compounds.

2. Summary of Results

A summary of the measurements of the critical angle (FWHM) for He-ion channeling in $YBa_2Cu_3O_{7-x}$ along the [001] axis as a function of temperature by Sharma et al.[3] is shown in Fig. 1. As the temperature is lowered to approximately 100K, the FWHM of the channeling scan increases by approximately 25%. The FWHM increases (the channeling effect becomes stronger) because the atomic rows which provide the steering become smoother as the thermal vibration amplitude of individual atoms decreases by about 0.03 Å between 295 and 100K. As the specimen temperature is lowered further, through T_c (= 92K), an abrupt additional increase (~7-8%) in the FWHM is clearly evident . No additional change occurs with further cooling to ~25K. The anomalous jump in the FWHM has been shown[4] to shift directly with stoichiometry induced changes in T_c, indicating a direct link between the anomaly and the superconducting transition.

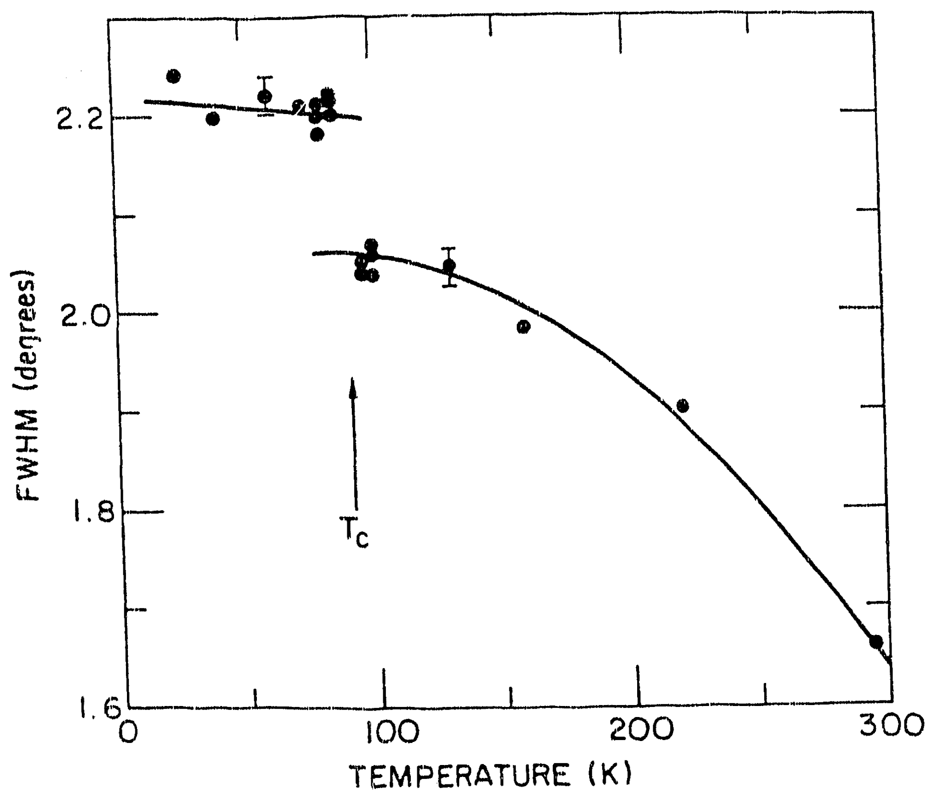

Fig. 1. Measured temperature dependence of the FWHM of [001] channeling scans obtained with the Y, Ba, and Cu RBS signal from 1.5 MeV ^{4}He.

Channeling along the [001] axis in $YBa_2Cu_3O_{7-x}$ results primarily from the atomic rows of Y and Ba, and of Cu and O, which occur along this axis. Because of their relatively low atomic numbers and large interatomic spacings, the contributions to channeling from the two different O-only [001] rows can be neglected. An $ErBa_2Cu_3O_{7-x}$ single-crystal, which increases substantially the net RBS yield from the rare-earth element, was employed by these same authors[5] to extract the contributions from the individual atomic rows to the observed anomaly at T_c. A sample of these results is given in Fig. 2, where the [001] axial scans containing the combined information from the Cu-O and Er-Ba rows exhibits a significant (~8%) change in FWHM across T_c, but the scans containing only the Er-Ba contribution do not. This demonstrates that the observed anomaly is confined to the Cu(1 and 2) and O(4) atoms. Similar effects, i.e., anomalous jumps in the channeling scans from the Cu-O row but not from the Y-Ba row, were observed using characteristic x-ray production in $YBa_2Cu_3O_{7-x}$.

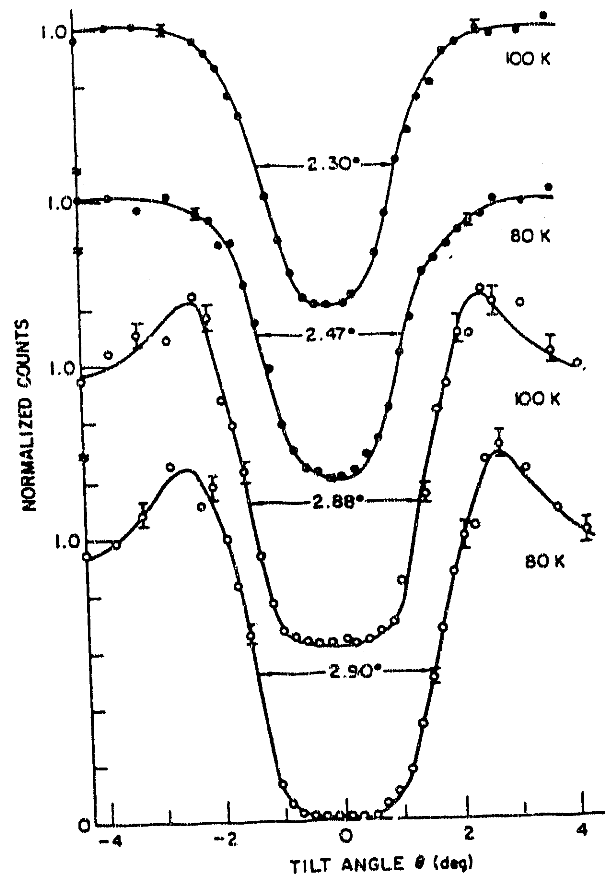

Fig. 2. [001] axial channeling scans obtained with the combined Er-Ba-Cu (closed symbols) and with the Er-Ba (open symbols) RBS signals at 80 and 100K.

Ion channeling was also performed on single crystals of $(Bi_{1.7}Pb_{0.3})Sr_2CaCu_2O_x$ between 35 and 295K[6]. Substantial effects of the incommensurate phase modulation in this material on the channeling properties were observed. The relatively large (≤0.03 nm) static atomic displacements from this modulation narrowed the critical angle for channeling by more than 50%, sharply reduced its normal temperature dependence, and increased the minimum yield to ≥7%. Despite these complications, measurements of the critical angle as a function of temperature revealed an anomaly at T_c in Cu-atom displacements perpendicular to the [001] axis that appeared identical to that found in $YBa_2Cu_3O_{7-x}$. This suggests that the observed anomalous lattice displacements are a general feature of high-T_c materials.

3. Discussion

As explained in detail elsewhere[3,5,7], one simple interpretation of the observed increase in the FWHM of the channeling scans across T_c is that the average vibrational amplitude in the a-b plane of the Cu and O atoms decreases abruptly by ~30% across T_c; i.e., an abrupt stiffening of the lattice occurs. However, this interpretation is clearly inconsistent with several other measurements of Debye properties across T_c in these materials, which demonstrate that any such change is smaller than one part in 10^4. An alternative explanation for the substantial changes observed in channeling behavior across T_c is that the displacements of the Cu(1 and 2) and O4 atoms in the a-b plane become strongly correlated in the superconducting state. Qualitatively, the effect of such correlations on the observed channeling scans is easy to understand. Correlations can smooth the atomic rows in the same sense as a decrease in thermal vibration amplitudes would, enhancing the channeling effect. Quantitative analysis of correlation effects on ion channeling would require sophisticated computer codes, which unfortunately are not currently available for multicomponent targets such as the high-T_c materials.

In an attempt[8] to more accurately characterize the structural and dynamic response of the lattice at T_c, we performed high-resolution neutron powder diffraction measurements as a function of temperature between 10 and 300K on a large, single-phase specimen of $YBa_2Cu_3O_{7-x}$. The structural and vibrational properties were obtained using Rietveld refinement. No evidence was found for an anomalous change in any of the structural parameters in the vicinity of T_c. A comparison of the thermal vibration amplitudes extracted for the Cu and O4 atoms from both the channeling and neutron diffraction data following the procedures described elsewhere is shown in Fig. 3. The general agreement between the two techniques regarding the temperature dependence of the vibrational amplitudes is clearly quite good. The total decrease of u for the Cu and O4 atoms between 295 and 30 K as seen in the channeling measurements is somewhat larger than that seen by neutron diffraction, but only by about the magnitude of the abrupt drop seen at T_c in the channeling results.

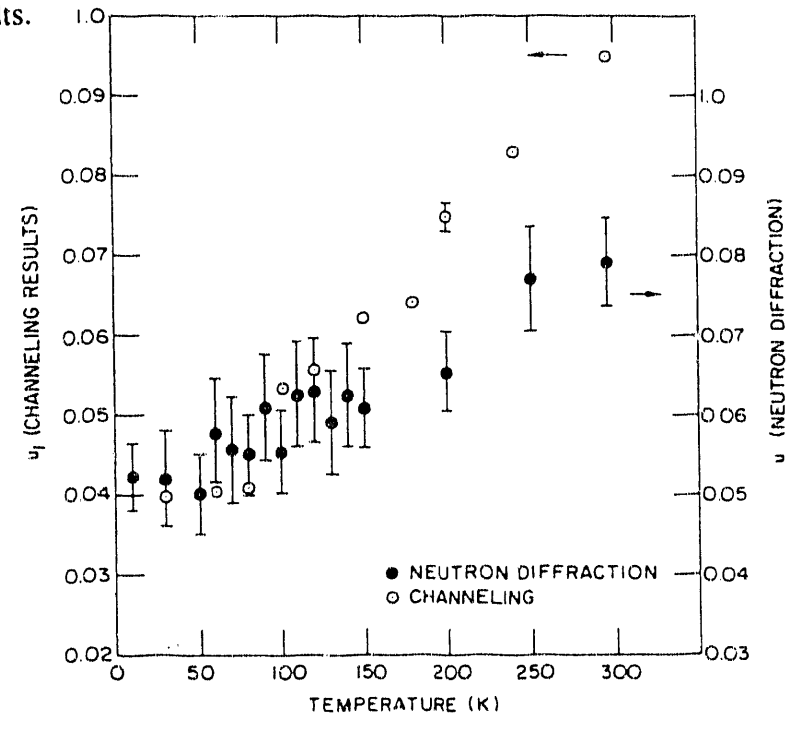

Fig. 3. Comparison of the temperature dependence of the average displacement amplitude, u, of the Cu and O4 atoms extracted from the ion channeling (open symbols) and neutron diffraction (closed symbols) results.

Most importantly, and as expected from our interpretation (c.f. above) of the channeling anomaly in terms of correlated displacements, there is no convincing indication in the neutron diffraction results for any anomalous reduction in Debye properties at T_c. Additional information can be gleaned from the failure to observe any significant anomalies in structural parameters via neutron diffraction. If the anomalous displacements observed by channeling are static, and preserve the overall crystal symmetry, anomalies would have been expected in the neutron diffraction determination of the appropriate bond lengths. Since these were not observed, we can deduce that the anomalous displacements seen by ion channeling are either dynamic, or they are static distortions which fail to preserve the overall crystal symmetry.

Haga et al.[9] have also studied the temperature dependence of ion-channeling in $YBa_2Cu_3O_{7-x}$ single crystals using deuterons (d). They reported an abrupt increase in the [001] minimum RBS yield across T_c. Their characteristic x-ray measurements indicated anomalous behavior for the minimum yield of all four (Y, Ba, Cu and O) elements in the vicinity of T_c. Although both their and our studies provide strong evidence of anomalous behavior of the $YBa_2Cu_3O_{7-x}$ lattice in the vicinity of T_c, there are inconsistencies between the two studies. The He work shows an improvement in channeling at T_c as a result of displacements of only the Cu and O4 atoms, while the deuteron results indicate a degradation due to displacements of all four types of atoms. To address these inconsistencies, we performed[10] simultaneous measurements of both the FWHM and the minimum yield during channeling with both He and with protons. Although d would have provided a direct comparison, the high neutron production with such beams is not compatible with our accelerator arrangement. The results of these studies are summarized in Fig. 4.

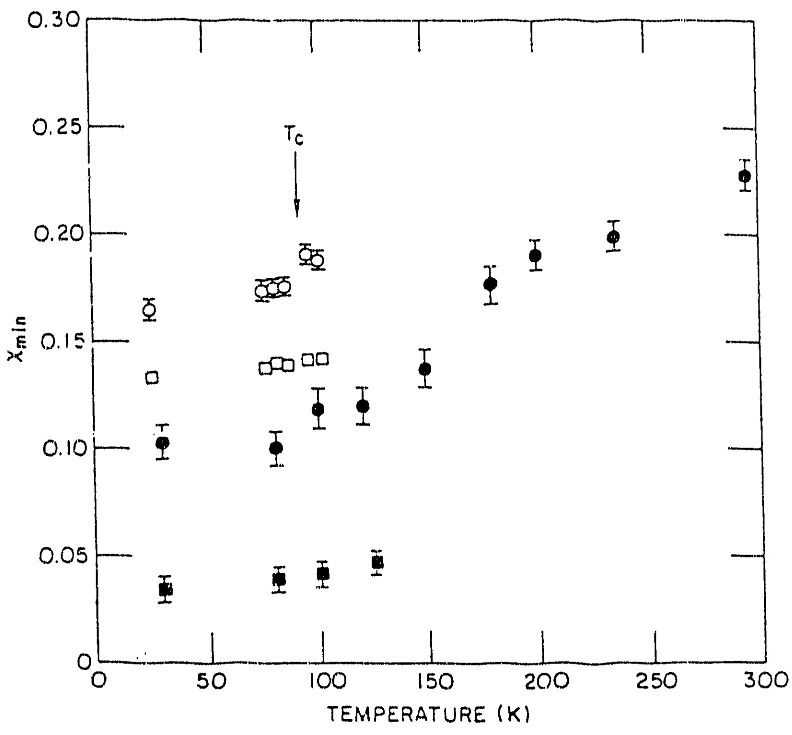

Fig. 4. Cu Kα x-ray minimum yields for 6-MeV ^4He (solid circles) and 1.3-MeV p (open circles) as a function of temperature in $YBa_2Cu_3O_{7-x}$. Y and Ba L x-ray minimum yields obtained with 1.5-MeV ^4He (solid squares) and 1.3-MeV p (open squares).

A clear decrease is seen in the Cu-minimum yield for both p and He ions as the temperature drops through T_c. Also for both ions, no indication is seen for any significant change in the vicinity of T_c of the L x-ray minimum yield for either the Y or Ba. During these same experimental runs, 6-8% increases were observed in the FWHM for the RBS (Y-Ba-Cu) yields from both He and p. In summary, we find increases in the FWHM, and decreases in the minimum yield, for both He and p channeling along the [001] Cu-O4 atomic rows, and no significant changes in either quantity for the [001] Y-Ba rows. These results therefore provide additional evidence that a correlated sequence of Cu-O4 displacements in the a-b plane appears with the onset of superconductivity in high-T_c materials.

4. Acknowledgements

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