Probing single-crystalline $YBa_2Cu_3O_7$ across the superconducting transition temperature by positron annihilation measurements

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Shape parameter S determined from the Doppler broadening of the positron annihilation radiation line-shape measurement (DBPARL), gives the fraction of suitably defined low momentum electrons among the annihilating electrons. Since BCS pairing in momentum space is accepted for the so-called conventional superconductors, such studies of possible changes in the electron momentum distribution as a function of temperature across the superconducting transition in high-temperature superconductors (HTSC) may help clarify the yet unknown mechanism of superconducting pairing in HTSC. Our S vs T data at close intervals (down to 1 K) of temperature revealed one minimum at T_c in Bi-2212 pellets and two minima in the T_c region in (Bi,Pb)-2223 pellets, in addition to the somewhat better known steplike change at T_c. Similar DBPARL measurements presently on Y-123 single crystals show the S vs T minima in these as well as in previous (Bi,Pb)-2223 pellets to be at T_c and $0.95T_c$. These observations can imply a redistribution of electron momentum similar to the better discussed charge redistribution.

I. INTRODUCTION

The pairing of electrons in momentum space is a key factor in the BCS mechanism of transition to the superconducting state. Whether the mechanism of the so-called high-temperature superconductivity (HTSC) is BCS-like or not,[1] it is of great interest to investigate experimentally the temperature dependence of the momentum distribution of electrons in various HTSC, particularly across T_c, the superconducting transition temperature. Partial but important[2–5] information on this distribution can be conveniently obtained from the Doppler-broadened positron annihilation radiation line shape (DBPARL) or similar experiments. Similarly the density of electrons at the annihilating sites can be probed from positron lifetime, τ, measurements.[6,7] Such measurements[3–6] across T_c can be justified also from a recent suggestion[8] that the vortex cores of a type-II superconductor in a mixed state may get excess charge. The mechanism of such charging on cooling has been understood[8] for materials with such electronic subsystems that one subsystem becomes superconducting. Below T_c, the charge carriers in the superconducting subsystem should have lower energy. This invites charge carriers from other subsystems to the superconducting subsystem and enhances the density of carriers at vortices below T_c. Electron density and electron momentum distribution being related, increased density leads to increased average momentum. This concept[8] has not been applied earlier to explain the interlayer charge transfer, proposed[2,4] to be activated at T_c, to explain the observation in various HTSC of steplike changes[9,10] in τ and S across T_c. But we find that it leads to charge-carriers or holes concentrating in the superconducting subsystem of CuO_2 layers on cooling below T_c.

The line-shape parameter S is defined[5,7] as the ratio of the area of a suitably selected central region of the energy spectrum $N(E)$ vs E of the annihilation radiations to the area under the full spectrum, after the background correction. Due to the Doppler shift, the two oppositely emitted gamma rays of energy $E_0=511$ keV appear to be of energy $E=[E_0 \pm pc/2]$ in the laboratory frame where p is the momentum of the annihilating electron in the direction of detection. Consequently, S gives the fraction of chosen low momentum (corresponding to 510.4 to 511.6 keV energy in the present analysis) electrons among the electrons participating in the positron-electron annihilation in the solid. One must add here that these positrons in a solid populate[6,7] preferably all available lower potential-energy regions of the lattice and vacancylike lattice defects. The latter group of positrons, trapped in vacancylike defects, have a lifetime τ_2 longer than τ_B, the lifetime in the bulk. Relative intensity I_2 for τ_2 for various HTSC samples studied by various authors has been seen to be up to 15%, and oxygen vacancies or unoccupied oxygen sites are believed to be the main trapping centers. The rest of the positrons annihilate in the bulk. It may be added that the mean positron lifetime and S parameter should have the same[6,11] behavior.

Since the 1950s the failure to see a steplike change of the S-parameter across T_c for the conventional superconductors like Nb_3Sn and V_3Si has been attributed[9,10] to the involvement in superconductivity of not all the conduction electrons but of only the electrons near the Fermi energy E_F. The new high-T_c oxide superconductors have favorably larger values of the energy gap 2Δ and also favorably smaller values of E_F, which together should allow a much larger fraction[9,10] of charge carriers to participate in the superconducting pairing process. Abrupt changes in τ and S at T_c were expected to be large and hence measurable in these high-T_c oxides. While an abrupt decrease or increase of S on cooling various HTSC below T_c has been strongly claimed[4,9,10] on the basis of many positron annihilation experiments since 1987, an extra feature of a minimum at T_c $(R=0)$ and another at a slightly lower temperature has been observed[5] rather recently for (Bi,Pb)-2223 pellets. The minimum at T_c implies a sharp increase in the fraction of large momentum electrons on cooling towards T_c and then a sharp decrease on further cooling below T_c. In view of the important implication of such observations to the poorly understood mechanism of high-T_c superconductivity, a need was felt to extend such detailed DBPARL measurements to an entirely different HTSC and to single crystals. Here the variation of S with T across the superconducting region is measured for $YBa_2Cu_{(3-x)}Al_xO_z$, $x\simeq 0.02$, $z\simeq 7.0$, or Y-123 single crystals[12] with $T_c=89.5$ K and compared to our recent results[5] for granular Bi-2212 and (Bi,Pb)-2223 samples in an attempt to understand the intriguing results for high-temperature superconductors in general.

Presently, only a steplike[2,4] increase or decrease of S on cooling a HTSC below T_c seems to be recognized.[5,9,10] However, a closer look, as is possible from our data[5,6] at closer intervals of temperature and some earlier experiments,[11] reveals either two minima or one minimum in the superconducting region of the S vs T graph, depending on the system studied, in addition to the steplike change. That makes the situation more complex and interesting. A minimum at $T_c(R=0)$ and another one at a slightly lower temperature has been clearly observed[5] for (Bi,Pb)-2223 pellets.

The present experiment is a DBPARL experiment on well-characterized single crystals, in contrast to our earlier work on granular[5] samples. It has been argued that in such pellets[5] consisting of HTSC grains, the superconducting transition of the network of grains can be at a slightly lower temperature than the T_c of the individual grains, to give two-step transitions[5,13,14] as observed in magnetic susceptibility measurements. This possibility of explaining the lower-temperature minimum in S vs T, observed[5] in a granular HTSC, is put to test in the present measurements on HTSC single crystals. Moreover, it is also to confirm for Y-123 HTSC the peculiarities observed for two Bi-HTSC.

II. EXPERIMENTAL OUTLINE

Samples for positron annihilation experiments have to be sufficiently large to sandwich a nickel-foil ^{22}Na source between two approximately identical samples. Also to allow positrons to annihilate within the single-crystal samples, the crystal thickness needs to be about 1 mm. Large (squares with 3 to 4 mm sides) Y-123 crystals of such thickness could be grown[12] by a slow cooling of the mixed melt with BaO-CuO self-flux in Al_2O_3 crucibles. Unavoidable dissolution of almost all crucible materials into the melt accounts for the earlier-mentioned traces of Al in our crystals. However, an *in situ* coating of the inner crucible surface by the corrosion product $YAl_2Ba_3O_{7.5}$ prior to crystal growth ensured a low Al content in our $YBa_2Cu_{(3-x)}Al_xO_z$ single crystals, as estimated in many of our samples by energy dispersive x-ray analysis (EDAX). A.C. The susceptibility measurement gave the onset of superconductivity at T_c(acs onset)= 89.5 K with a width (for 10% to 90% transition) less than 2 K. This T_c(acs onset) and $T_c(R=0)$ are seen to coincide in all well-prepared samples and hence abbreviated, in the following

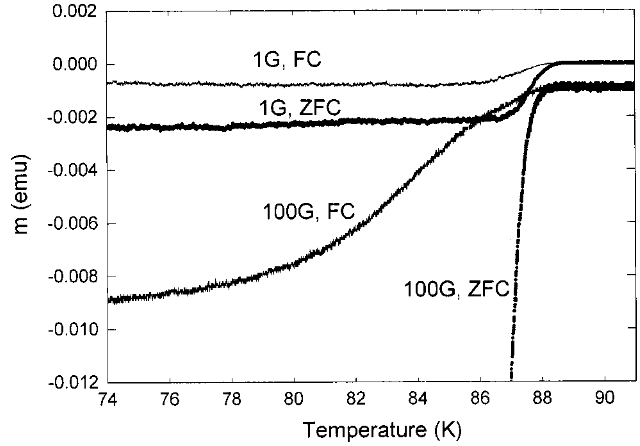

FIG. 1. Variation of the magnetization (at fields of 1 and 100 G, for field cooling and zero field cooling, in a VSM setup) with sample temperature for the Y-123 single-crystal sample.

discussion, as just T_c. This slight depression of T_c is consistent[12] with a doping concentration of $x\simeq 0.02$ Al on the Cu(1) chain sites and oxygen content of 6.99, which was confirmed by EDAX and x-ray profile refinement measurements. Neither EDAX nor x-ray diffraction (XRD) detected any impurity phase in the samples. Magnetization measurements at 1 and 100 G have been carried out as a second check on the quality of the crystals rather recently, i.e., about two years after the low-temperature cycling for positron experiments. This result in Fig. 1 shows the onset of superconductivity practically at the same temperature for FC and ZFC samples to confirm that there is no detectable granularity in our single-crystal samples. A fully oxygenated state, the absence of any detectable microcracks due the specially developed cooling rates[12] during oxygenation (taking 630 h for cooling from 873 K to 653 K, for example), and clear evidence of nongranularity from magnetization measurements confirm the high quality of the samples. Positron annihilation data on such well-characterized samples have been recommended by many earlier authors.

For mounting the delicate sample-source-sample combination between the cold head and a screwed cold plate of a Leybold 10-300 cryogenerator, with the a,b planes of the SXL samples parallel to the cold head surface, a covering support of indium sheet was found most effective for mechanical stability and thermal contact. Such mounting with thin indium padding has been checked[5] to eliminate thermal hysteresis in well-prepared and stable samples. The positron lifetime measurement at room temperature on the same sample-source-sample combination gave two lifetime components 179 ± 2 ps (91%) and 585 ± 13 ps (9%), implying $\tau_B=191$ ps for these Y-123 crystals with traces of Al in CuO chains. The lifetime values confirm that positrons annihilated essentially in the samples and not in the sample holder. The contribution of the positron to the momentum of the electron-positron pair is known[7] to be negligible on account of complete thermalization of the positrons before annihilation. A horizontally placed HPGe detector with associated electronics[5,7] was used in a fixed setting to obtain the $N(E)$ vs E annihilation radiation spectrum with about 0.8 million counts at each sample temperature, maintained by a Leybold LTC-60 temperature controller. Presently used detector-

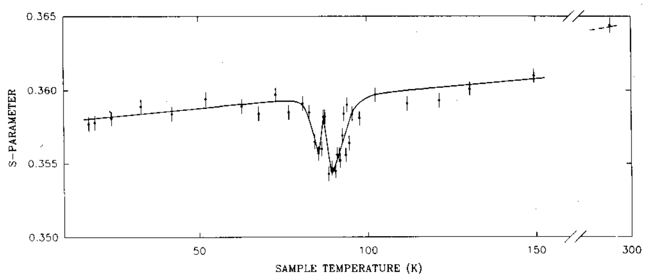

FIG. 2. Variation of the DBPAR line-shape parameter S with sample temperature T for the Y-123 single-crystal samples.

sample (Y-123 crystal) geometry allows no gamma rays other than those parallel to (a,b) planes be observed. This implies that practically only the a,b plane components, $p(a,b)=p$ of the momentum of the annihilating electrons contribute to the observed Doppler shift, $\pm pc/2$, in energy. So S is the fraction of annihilating electrons that make this energy less than or equal to 0.6 keV, as defined in the Introduction. In later sections, these are called ''low'' momentum electrons, implying that their momentum component in the (a,b) plane is less than or equal to 1.2 keV/c.

III. OBSERVATIONS AND DISCUSSION

The resulting S vs T data over the 20 to 300 K range and concentrating in the superconducting region is depicted in Fig. 2. It shows deeper minima compared to those for (Bi,Pb)-2223 pellets (as given in Fig. 1 of Ref. 5) due to the larger superconducting volume fraction in the single crystals. In the present work, one minimum is at about 89.5 K and another is at about 85.5 K. It is remarkable that the two minima are experimentally observed at the same T/T_c values, 1.00 and 0.95, in the (Bi,Pb)-2223 pellets and Y-123 single crystals.

Structures, minima or maxima, similar to those identified above in the superconducting region, are well reported[5,11] at higher temperatures and tentatively attributed to magnetic or structural changes or trapping by lattice defects. But the average slope[5] of S with respect to T over a large temperature range, 100 K or so, deliberately ignoring these ''local'' structures, should be positive in cases where the lattice contracts on cooling. Such a positive average slope for the $T>T_c$ has been observed in Y-123 repeatedly[2,11] in spite of a broad minimum[11] at around 225 K. No attempt has been made in this work to record these structures (over the range $T_c<T<2T_c$), as was done in our S-parameter[5] and lifetime[6] experiments in (Bi,Pb)-2223 or in above-mentioned Ref. 11 for Y-123 pellets. However, the room-temperature point was measured several times as a check and the same value was always reproduced within the accepted experimental error.

For granular superconductors, an intrinsic T_c of the grains and a lower T_c for the weakly coupled network of grains have been reported by various authors[13,14] as discussed above. Had the two minima for non-single-crystal or granular samples[4] been due to granularity, there would have been just a single minimum for the single crystals. Here in Fig. 2 two minima have been seen for the single crystals also. This proves that the lower temperature minimum is not linked to granular superconductivity.

Positron density has been shown[2,3,15] to be significant in the neighborhood of CuO chains and very small in the CuO_2 planes. This appears to imply that positrons are practically unable to probe the superconductivity in CuO_2 planes. Whether positron probing of the superconducting gap, possible in CuO chains according to an independent particle model, will be seriously affected by the electron-positron correlation has not been understood even theoretically. There is, however, no conceptual difficulty in positrons probing some of the effects of the superconducting transition or superconductivity in CuO_2 planes. One possibility is a previously presumed[2,4] transfer of electrons from CuO_2 planes to CuO chains at the on-set of superconductivity on cooling the sample, a qualitative explanation already outlined in our Introduction. So positrons distributed in chain regions are already believed[2] to probe ''superconductivity in CuO_2 planes'' via this gain of electrons in the chains on cooling below T_c. The second such effect now known to be associated with the superconducting transition is the set anomolous changes[16] in interatomic separations at or in the vicinity of T_c. Interestingly, it roughly explains the origin of at least one minimum in S, as detailed below, and also supports[16] the charge transfer conjecture.

HTSC ''structures in the vicinity of T_c'' have not yet been as widely and as closely investigated as would permit a full interpretation of our positron S parameter minima, these structural investigations being difficult and time consuming. But still the XRD data[16] at just eight chosen temperatures for ''superconducting single crystals'' of Tl-2212 and Y-124 can be used to partly explain our positron result on Y-123. Positron distribution[17] in Y-124 is mostly in the region between the CuO chains involving Cu1 atoms, from where most of the annihilations (with electrons) should take place. Figure 5 of Ref. 16 shows a minimum, at or near T_c, of the Cu1-O4a interatomic separation. This minimum implies a minute compression of the electron cloud around the above-mentioned annihilating sites occurring over a short range of temperature. This should lead to a minute minimum in S spanning a short range of temperature. It is the increase in the average momentum of the electrons due to an increase of the density of the electron gas. In this explanation, one need not consider the change with temperature of the separation of such atom pairs that are away from positron regions. However, structural data for even Y-124 is inadequate for any quantitative or perfect explanation. It is worth checking whether there are two minima in the XRD data in the T_c region of some HTSC to support two S vs T minima in these compounds, or only one minimum in the XRD result to motivate an alternative explanation of the second minimum in S. We[18] plan a detailed investigation in the next available beam times of these interatomic separations[19] in Y-123 as the sample is cooled across T_c.

Critical fluctuation is active only over a few Kelvin above and below T_c covering the width of the S vs T minimum. This similarity alone may not be sufficient to explain[5] the sharp decrease of the fraction of lower momentum electrons on approaching T_c from either side or a minimum in S at T_c. Kresin *et al.*[10] showed that two conducting subsystems in Y-123 (the CuO_2 planes and CuO chains) can lead to a two gap structure in agreement with the NMR observations.[20] Kresin *et al.* considered intrinsic superconductivity in CuO_2

units and induced superconductivity in CuO units. It is possible to extend this idea of two conducting Cu-O subsystems to (Bi,Pb)-2223 on considering the central CuO_2 plane to be different[5] from the other two CuO_2 planes. Further work is needed to see whether such considerations[10] can explain two minima in Y-123 and (Bi,Pb)-2223 (Ref. 5) in contrast to one minimum in Bi-2212.[5] An alternative explanation of a minimum in *S* can be from a crystallographic transition accompanied by some soft modes, leading to a change of the positron *S* parameter for a narrow range of temperature around the transition point. Increasing lattice instability on cooling towards T_c and its arrest due to the onset of superconductivity have been well known even for A15 superconductors. Such a situation can give rise to at least one minimum at T_c in the HTSC materials. It is difficult, particularly in the absence of more detailed structural studies, to conceive lattice distortions at T_c and $0.95T_c$ to give rise to the observed double minima in *S*. Still some possibilities are outlined, although the present note limits itself mostly to the experimental observations. Indeed a number of neutron PDF (pair distribution function) experiments have indicated[19] a local structural response to a change in the electronic state, like the appearance of superconductivity and the opening of the pseudogap. Particular mention may be made of the complex maximum (Fig. 20 of Ref. 19) observed in the T_c region in Tl-2212 HTSC of the PDF peak height $\Delta\rho$, at 3.4 Å . It concerns the pair correlation between in-plane, O(1), oxygen and apical, O(2), oxygen. The striking similarity of this variation to our double minima in *S* lies in the fact that the complex maximum in $\Delta\rho$ (of a Ti-HTSC) appears to consist of maxima at about 110 and 80 K. In Y-123 also, the tunneling frequency of apical oxygen between two possible sites shows a maximum (Fig. 23 of Ref. 19) at T_c. Explanations based on such considerations need to be worked out more precisely to understand the changes observed in the positron annihilation parameters across T_c.

The two minima in the *S* vs *T* graph at 99 and 104 K in (Bi,Pb)-2223 and at 85.5 K and 89.5 K for Y-123 (Fig. 2) can best be attributed to nonequivalence of the central CuO_2 layer to the noncentral ones in (Bi,Pb)-2223 and the nonequivalence of Cu-O chains and planes in Y-123. This can be tested from the fact that for Bi-2212, which has two equivalent CuO_2 layers, a single minimum in the *S*-vs-*T* graph is expected. Within the discussed[5] limitation of a broad transition, it appears to be a single minimum at its T_c. An interpretation[21] of resistivity vs temperature data in Y-123 presumed physical processes in chains and planes to be activated at different temperatures. Planar and pyramidal Cu-O units in central and noncentral CuO_2 layers in (Bi,Pb)-2223 are not equivalent, leading to different types of temperature dependence of the resulting NMR lines.[20] So the *S*-parameter minimum may show up at one temperature for the central CuO_2 layer and at another for the noncentral ones, or at different temperatures for the Cu-O chains and CuO_2 planes.

IV. CONCLUSION

The combined conclusions of this work and our cited earlier papers[4–6] can be summarized. A sharp minimum of the DBPAR line shape, *S*, indicating a sharp minimum of the fraction of the low momentum electrons, has been observed at the bulk superconducting transition temperature for three high-T_c oxides: Bi-2212, (Bi,Pb)-2223, and Y-123. The temperature-wise location of this minimum in *S* at T_c of the HTSC confirms its link to superconductivity. A larger superconducting volume fraction (in the crystals) leads to a larger magnitude of this as well as the second minimum, if observed, further proving the role of superconductivity. Finding the second minimum for the single crystals confirms it to be an intrinsic effect of the HTSC material. The second minimum, observed at $0.95T_c$ for granular as well as single-crystal samples with nonequivalent Cu sites, cannot be attributed to any intergranular effect. It is interesting that *S* or the fraction of low momentum electrons show one minimum at T_c for Bi-2212 (having identical Cu-O units), and two minima at T_c and at about $0.95T_c$ for Y-123 and (Bi,Pb)-2223 (having two different Cu-O units in the structure).

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[1] J. Ruvalds, Supercond. Sci. **9**, 905 (1996); N. M. Plakida, *High-Temperature Superconductivity* (Springer-Verlag, Berlin, 1995).

[2] Y. C. Jean, S. J. Wang, H. Nakanishi, W. N. Hardy, M. Y. Hayden, R. F. Kiefl, R. L. Meng, P. H. Hor, Z. J. Huang, and C. W. Chu, Phys. Rev. B **36**, 3994 (1987); Y. C. Jean, C. S. Sundar, A. Bharathi, J. Kyle, H. Nakanishi, P. K. Tseng, P. H. Hor, R. L. Meng, Z. J. Huang, C. W. Chu, Z. Z. Wang, P. E. A. Turchi, R. H. Howell, A. L. Wachs, and M. J. Fluss, Phys. Rev. Lett. **64**, 1593 (1990).

[3] E. C. von Stetten, S. Berko, S. S. Li, R. R. Lee, J. Brynestad, D. Singh, H. Krakauer, W. E. Pickett, and R. E. Cohen, Phys. Rev. Lett. **60**, 2198 (1988); B. Barbiellini, P. Genoud, J. Y. Henry, L. Hoffmann, T. Jarlborg, A. A. Manuel, S. Massidda, M. Peter, W. Sadowski, H. J. Scheel, A. Shukla, A. K. Singh, and E. Walker, Phys. Rev. B **43**, 7810 (1991).

[4] P. K. Pujari, T. Datta, Udayan De, and B. Ghosh, Phys. Rev. B **50**, 3438 (1994) and references therein.

[5] Udayan De, P. M. G. Nambissan, D. Sanyal, and D. Banerjee, Phys. Lett. A **222**, 119 (1996); Udayan De, P. M. G. Nambissan, S. Chaudhuri, D. Sanyal, and D. Banerjee, Physica B **230-232**, 856 (1997); D. Sanyal, P. M. G. Nambissan, Th Wolf, and Udayan De, Physica B **281-282**, 928 (2000).

[6] D. Sanyal, D. Banerjee, and Udayan De, Phys. Rev. B **58**, 15226 (1998); D. Sanyal, S. Chaudhuri, R. Bhattacharya, D. Banerjee, and Udayan De, J. Low Temp. Phys. **105**, 1665 (1996).

[7] P. Hautojarvi, *Positron in Solids* (Springer-Verlag, Berlin, 1979).

[8] D. I. Khomskii and A. Freimuth, Phys. Rev. Lett. **75**, 1384

(1995).
[9] V. L. Sedov and S. N. Kunetsov, Phys. Lett. A **193**, 413 (1994) and references therein.
[10] Vladimir Z. Kresin and H. Morawitz, J. Supercond. **3**, 227 (1990); Vladimir Z. Kresin, H. Morawitz, and S. A. Wolf, *Mechanisms of Conventional and High T_c Superconductivity* (Oxford University Press, New York, 1993).
[11] S. J. Wang, S. V. Naidu, S. C. Sharma, D. K. De, D. Y. Jeong, and T. D. Black, Phys. Rev. B **37**, 603 (1988); K. Jung, J. G. Byrne, M. C. de Andrade, M. B. Maple, S. Krichene, J. R. Reynolds, and J. M. Owens, Physica B **217**, 23 (1996).
[12] Th. Wolf, W. Goldacker, B. Obst, G. Roth, and R. Flükiger, J. Cryst. Growth **96**, 1010 (1996); V. I. Voronkova *et al.*, *ibid.* **149**, 74 (1995); E. Brecht, W. W. Schmahl, G. Miehe, H. Fuess, N. H. Andersen, and Th. Wolf, Physica C **235**, 471 (1994).
[13] T. Ishida, T. Sakuma, T. Sasaki, and Y. Kawada, Jpn. J. Appl. Phys., Part 1 **28**, L559 (1989).
[14] A. K. Bandyopadhyay, E. Gmelin, B. V. Kumaraswamy, V. P. S. Awana, Deepak Varandani, Nirupa Sen, and A. V. Narlikar, Phys. Rev. B **48**, 6470 (1994).
[15] S. Massidda, Physica C **168**, 137 (1990).
[16] V. N. Molchanov and V. I. Simonov, Acta Crystallogr., Sect. A: Found. Crystallogr. **54**, 905 (1998).
[17] C. S. Sundar, A. Bharathi, Y. C. Jean, P. H. Hor, R. L. Meng, Y. Y. Xue, Z. J. Huang, and C. W. Chu, Phys. Rev. B **41**, 11 685 (1990).
[18] S. Venkatesh, H. S. Chauhan, Th. Wolf, and Udayan De (unpublished).
[19] T. Egami and S. J. L. Billinge, in *Physical Properties of High Temperature Superconductors V*, edited by Donald M. Ginsberg (World Scientific, Singapore, 1996), p. 265, and references therein.
[20] A. P. Howes, R. Dupree, D. McK. Paul, and S. Male, Physica C **185-189**, 1137 (1991); R. Dupree, Z. P. Han, A. P. Howes, D. McK Paul, M. E. Smith, and S. Male, *ibid.* **175**, 269 (1991).
[21] Z. X. Gao, E. Osquiguil, M. Maenhoudt, B. Wuyts, S. Libbrecht, and Y. Bruynseraede, Phys. Rev. Lett. **71**, 3210 (1993).