Evidence for a Common High-Temperature Superconducting Effect in $La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_7$

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We report the positron-annihilation lifetime and Doppler-broadening energy spectra in $La_{1.85}Sr_{0.15}CuO_4$ superconductors for $10 < T < 297$ K. A strong temperature dependence in positron lifetime and momentum distribution parameters is observed for positron-electron annihilation data while the sample is in the superconducting state, but not while it is in the normal state. We show experimental evidence that the superconducting mechanisms in $La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_7$ are similar and suggest that this is a consequence of a common physics involving the delicate balance between localized- and itinerant-electron behavior.

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We have recently observed[1] that positron annihilation in $YBa_2Cu_3O_7$ is affected by the degree of crystal perfection. Oxygen defects in the CuO(1) chain, arising from oxygen substoichiometry with respect to $YBa_2Cu_3O_7$, have been shown to act as positron-trapping sites.[2-4] It is the intent of the present investigation to study another nearly defect-free high-T_c material, $La_{1.85}Sr_{0.15}CuO_4$. Without positron trapping into vacancy sites we expect to observe results similar to those seen for $YBa_2Cu_3O_7$.[1]

Two samples of $La_{1.85}Sr_{0.15}CuO_{4-d}$ ($d < 0.04$) were synthesized by a direct solid-state reaction from high-purity oxides as described elsewhere.[5] The samples were consolidated by sintering yielding a polycrystalline material with a $\simeq 10^{-6}$-m grain size and a $\simeq 10\%$ porosity. The resistivity and ac magnetic susceptibility show that $T_c = 33$ K. A nonsuperconducting material was also produced for comparative experimentation by heating of the superconducting material at $T = 950$ K under vacuum. A 24-μCi ^{22}NaCl positron source was directly deposited on the surface of one of two identical samples, each of which was 2 mm thick $\times 5 \times 4$ mm^2. The two samples were arranged as a sandwich with the deposit between and placed in an Al cell under a high-purity He atmosphere. The cell was attached to a He refrigerator which could control the temperature to ± 0.1 K. Both positron-lifetime and Doppler-broadening experiments were performed as a function of temperature in three temperature cycles with no temperature hysteresis effects being observed.

Approximately 10^6 counts were accumulated in each of the lifetime and Doppler-broadening spectra. The lifetime spectra were analyzed by our fitting a sum of exponential functions using the program PATFIT.[6] All the results show only two lifetime components. A long-lived component is identified in all the lifetime spectra with a lifetime of $\simeq 0.5$ ns and an intensity of $\simeq 0.4\%$ We attribute this part of the spectrum to surface and source annihilation of Ps and positrons. No more than one remaining lifetime could be resolved from the present data at all temperatures studied. Our attempts to fit two lifetimes in excess of the "source term" resulted in unstable or unphysical results. We conclude that only a single positron bulk lifetime (175–190 ps) exists in $La_{1.85}Sr_{0.15}CuO_4$. The Doppler-broadening spectra are reported in terms of a shape parameter (S) which is tak-

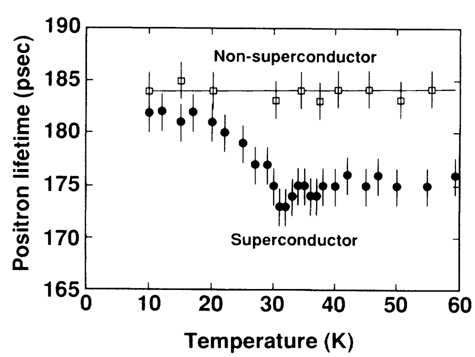

FIG. 1. Positron lifetime vs temperature in superconducting and nonsuperconducting $La_{1.85}Sr_{0.15}CuO_4$. The line for the nonsuperconductor is the linear fit to the data points.

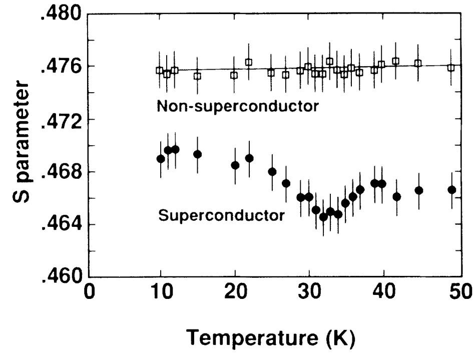

FIG. 2. S parameter of the Doppler-broadening energy spectra vs temperature in superconducting and nonsuperconducting $La_{1.85}Sr_{0.15}CuO_4$. The line for the nonsuperconductor is a linear fit to the data points.

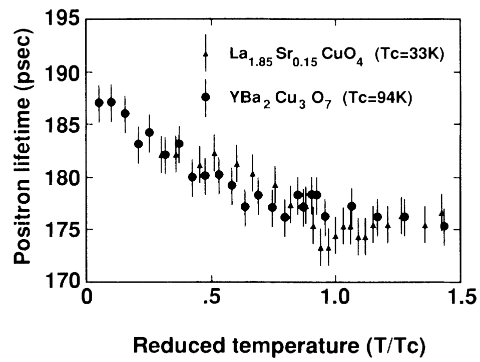

FIG. 3. Positron lifetime vs reduced temperature (T/T_c) for $La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_7$ (data from Ref. 1).

en as a ratio of the sum of the counts in the central region (± 0.9 eV about the centroid) to the total counts in the 511-keV peak.

In Figs. 1 and 2 the positron lifetime and S parameter are shown to increase below T_c. The lifetime above T_c ($=175$ ps) is similar to the lifetime for positron annihilation in single-crystal $YBa_2Cu_3O_7$ ($=176$ ps). Although the dependence is similar to that previously reported[1] in single-crystal $YBa_2Cu_3O_7$ it is opposite to the results in sintered powders[2-4] where a significant amount of vacancy trapping exists. Positrons annihilating from oxygen-vacancy–trapped states in sintered $YBa_2Cu_3O_{6.8}$ exhibit a lifetime of $\simeq 220$ ps which decreases to $\simeq 200$ ps below T_c. From the existing results in high-T_c oxide superconductors we conclude that the evidence points to a general synergism which requires that there be a relatively high degree of crystal perfection to observe the increasing lifetime for $T < T_c$.

To compare more directly the present data and those obtained in $YBa_2Cu_3O_7$,[1] we plot the positron lifetime data versus the reduced temperature (T/T_c) in Fig. 3. Although the data were collected with two different spectrometers, the results in $La_{1.85}Sr_{0.15}CuO_4$ and in $YBa_2Cu_3O_7$ agree very well. We note that in comparison the $La_{1.85}Sr_{0.15}CuO_4$ lifetime exhibits a "saturation" at the lowest temperatures when compared to the $YBa_2Cu_3O_7$, and further that at $T/T_c \simeq 1$ the $La_{1.85}Sr_{0.15}CuO_4$ lifetime data show a dip or kink, which is possibly indicative of the early states of a phase transformation, but which, with decreasing temperature, appears to be overcome by the superconducting transition itself.

Since the positron-annihilation rate is an overlap integral between the electron density and positron density at the same sites, we need to obtain the positron density distribution in these materials. We have extended a variational method developed by Chiba,[7] to calculate the positron probability distribution in these perovskite materials.[8] In Fig. 4 we show the positron densities in two orthogonal planes representative of the crystal La_2CuO_4. Similar results are also obtained for $YBa_2Cu_3O_7$.[8] The similarity in the nature of the spatial distributions of the positron wave function in the two perovskite materials can be described as envisioning the positron wrapped about the Cu(1)O(1) spine at the interstices of the bonds. From this point of view the La_2CuO_4 and $YBa_2Cu_3O_7$ (Ref. 8) materials look the same to the positron.

Because of the particular ability of the positron to probe the copper-oxygen bonding in $La_{2-x}Sr_xCuO_4$, as it was shown in Fig. 4, one can obtain an appropriate description of the electron-positron annihilation process by considering the electronic properties in the cluster so defined, i.e., the distorted octahedron of oxygens cen-

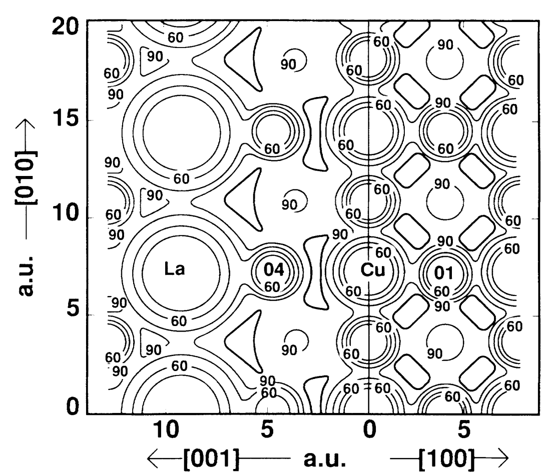

FIG. 4. Planar isodensity plot of the square of the positron wave function for La_2CuO_4, calculated by the method of Ref. 7. The right-hand side shows isodensity information in the basal plane; to its left, and joined along a common boundary, is the corresponding information for the orthogonal direction. The principal atomic sites, La, O(1), and O(4), have been labeled. The small numbers refer to percentages of the maximum probability density; highlighted levels enclose the regions of highest probability.

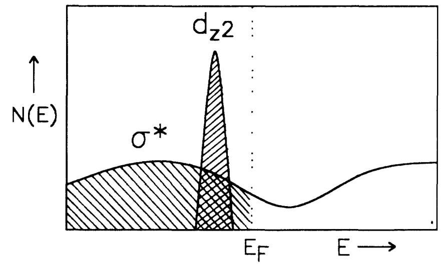

FIG. 5. Schematic density of states for high-T_c perovskite superconductors. The delocalized σ^* basal plane and localized d_{z^2} Cu-O chain contributions have been labeled. The critical role of the location of E_F is indicated.

tered around a copper site. Following the arguments developed by Goodenough and Ramasesha[9] and Singh, Ganguly, and Goodenough,[10] the interactions between the Cu-O-O-Cu connecting the perovskite layers are expected to be weak and therefore the d_{z^2} type of orbitals are fairly localized, a fact which is supported by the geometry of the orbital-overlap integrals. In contrast the $\sigma^*_{x^2-y^2}$ band, which results from a covalent mixing of Cu $d_{x^2-y^2}$ orbitals with the nearest O^{2-}-ion $2p$ and $2s$ orbitals in the basal plane, has a strong itinerant-electron character. Moreover, the inherent strong correlation between d_{z^2} electrons, enhanced by the localized d_{z^2}-electron states, will give rise to a splitting of a σ^* band. The resulting schematic density of states for the last occupied states is represented in Fig. 5. This description is supported by the following points.

(1) Itinerant $\sigma^*_{x^2-y^2}$ electrons and localized d_{z^2} electrons enhance any shortening of the Cu—O bonds in the basal plane relative to the Cu—O bond length along the c axis which is believed to account for the tetragonal $c/a > 1$ distortion of the octahedral environment of Cu.

(2) Lattice instabilities occur where the energies of a localized-electron and an itinerant-electron configuration are nearly the same.

Whether the copper-ion $3d$ hole will order into the $\sigma^*_{x^2-y^2}$ or d_{z^2} state depends on the ratio of formal Cu^{III} over Cu^{II} states which is controlled by the content of Sr.[11] As a consequence, because the positron is sensitive to any electron momentum redistribution, our lifetime results suggest that below T_c, the pairing mechanism which leads to superconductivity magnifies the ability for the electron to exhibit in nature localized or itinerant character, depending on the location of E_F. In conventional superconductors no change in the positron-annihilation rate below T_c has been observed, although on physical grounds[12] the mean lifetime of the positron is expected to be larger when the material is in the superconducting state. In the perovskite-related oxides it appears that the positron is able to probe the contribution of the electron momentum distribution responsible for superconductivity.

This reasoning can easily be extended to $YBa_2Cu_3O_7$. In this case the positron probes the Cu(1)O(1) and O(4) bonding, while the significant cluster to consider is the rhombuslike square of oxygen O(1) and O(4) centered about the Cu(1). The role of the Sr in adjusting the number of Cu-ion $3d$ holes is now played by the O(1) vacant lattice sites. The strong similarity in the temperature-dependent lifetime data for these two systems appears to be consistent with a localized-ion scheme. The present results provide a unique insight into the common nature of the superconducting mechanism. In its simplest interpretation the positron results are indicating a reduction in the sampled electron density. However, it is only in the case of the $YBa_2Cu_3O_{6.8}$ that we are able to observe a corresponding electron density increase below T_c at vacant lattice sites O(1) in the Cu(1)O(1) chain.

In summary we have measured the temperature dependence of the positron lifetime and the Doppler-broadening shape parameter S for polycrystalline samples of $La_{1.85}Sr_{0.15}CuO_4$. We have found that only a single lifetime can be associated with the experimental data over the temperature range 10–297 K and that below $T_c = 33$ K the lifetime and the S parameter increase. It is noted that this behavior is opposite to the essential characteristics of positron annihilation in sintered $YBa_3Cu_3O_{6.8}$ and remarkably similar to the behavior of positrons in single-crystal $YBa_2Cu_3O_7$. We conclude, on the basis of experimental evidence, that the superconducting mechanisms in $La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_7$ are similar, and suggest that this is a consequence of a common physics involving the delicate balance between localized and itinerant electron behavior.

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