Evolution of Superconducting-Transition Temperature with Superfluid Density and Conductivity in Pressurized Cuprate Superconductors

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What factors fundamentally determine the value of superconducting transition temperature T_c in high temperature superconductors has been the subject of intense debate. Following the establishment of an empirical law known as Homes' law, there is a growing consensus in the community that the T_c value of the cuprate superconductors is closely linked to the superfluid density (ρ_s) of its ground state and the conductivity (σ) of its normal state. However, all the data supporting this empirical law ($\rho_s = A\sigma T_c$) have been obtained from the ambient-pressure superconductors. In this study, we present the first high-pressure results about the connection of the quantities of ρ_s and σ with T_c, through the studies on the $Bi_{1.74}Pb_{0.38}Sr_{1.88}CuO_{6+\delta}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$, in which the value of their high-pressure resistivity ($\rho = 1/\sigma$) is achieved by adopting our newly established method, while the quantity of ρ_s is extracted using Homes' law. We highlight that the T_c values are strongly linked to the joint response factors of magnetic field and electric field, i.e., ρ_s and σ, respectively, implying that the physics determining T_c is governed by the intrinsic electromagnetic fields of the system.

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There are more than two-hundred copper-oxide (cuprate) superconductors that have been found after the discovery of superconductivity in Ba-doped La_2CuO_4,[1–3] in which their T_c values vary widely from 35 K in Ba-doped La_2CuO_4 to 134 K in $HgBa_2Ca_2Cu_3O_9$ at ambient pressure,[4] and to 164 K under the pressure of 30 GPa.[5] Although intense investigations have been carried out in the past decades, the issues about why the critical transition temperature (T_c) is high for some cuprate superconductors and low for some others, why T_c can be tuned by the parameters such as doping, pressure or magnetic field, and what the physical quantities decisively control the superconducting properties, are still on the list of mysteries of the condensed mater physics.[6–25] Previous studies found that T_c of the superconductors is associated with the superfluid density (or penetration depth) and conductivity (or effective mass),[26–28] however these empirical relations work well only for some of the high-T_c superconductors. Based on these relations, a more powerful empirical law was proposed by Homes *et al.* (known as Homes' law),[11] which states that the superfluid density (ρ_s) in the ground state and the normal-state conductivity (σ) just above T_c are directly connected to the T_c value of both conventional and high-temperature superconductors $-\rho_s = A\sigma \times T_c$ (A is a constant). Currently, Homes' law can provide reasonable description for many ambient-pressure superconducting materials, however, the validity of this law for superconductors under high pressure remains largely unknown. If we assume that Homes' law is applicable to all bulk superconductors, it should hold true for the superconductivity that can be tuned through various methods. To know the relation of the superconductivity in compressed high-T_c superconducting materials to superfluid density and normal-state conductivity, we performed high-pressure resistance measurements on the bulk cuprate superconductors that contain single and double CuO_2 planes in a unit cell. Subsequently, we utilized our newly established method to convert the measured resistance under pressure into resistivity,[29] enabling us to examine the correlation among the three physical quantities, T_c, σ, and ρ_s, with increasing pressure.

Figure 1 illustrates the temperature dependence

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of *in-plane* resistance for the three over-doped $Bi_{1.74}Pb_{0.38}Sr_{1.88}CuO_{6+\delta}$ (referred to as Bi-2201) single crystals, measured at different pressures. As shown in Figs. 1(a)–1(c), these samples display similar superconducting transition temperatures (18 K, 19 K, and 21 K, respectively) at ambient pressure. Furthermore, we observed that the normal-state resistance of these samples initially decreases until it reaches a minimum value. This minimum resistance occurs at 4.1 GPa for sample 1, at 4.0 GPa for sample 2, and at 3.8 GPa for sample 3 [Figs. 1(a)–1(c)]. However, beyond these pressure points, the normal-state resistance increases significantly with further compression. To determine the normal-state conductivity σ for the samples, we employed our newly established method.[29] to convert the measured resistance values to resistivity ρ. Subsequently, we obtained σ by taking the reciprocal of ρ ($\sigma = 1/\rho$). The obtained resistivity versus temperature are plotted in Figs. 1(d)–1(f).

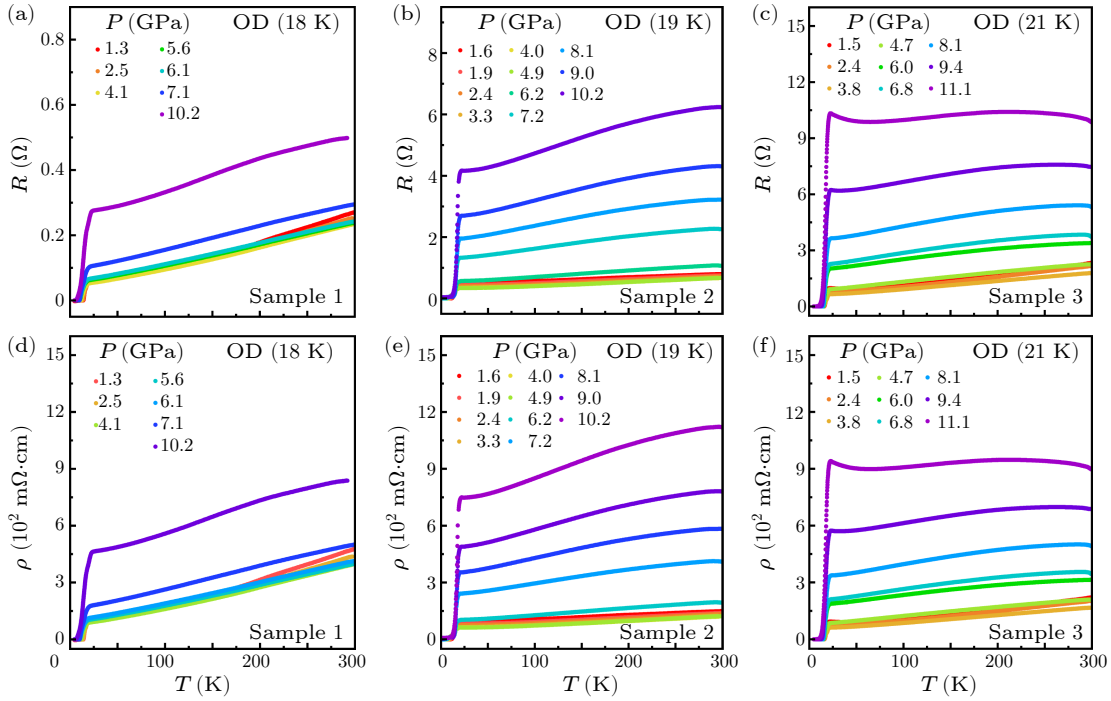

Fig. 1. Transport properties of the over-doped $Bi_{1.74}Pb_{0.38}Sr_{1.88}CuO_{6+\delta}$ (Bi-2201) superconductors. (a)–(c) Temperature dependence of the electrical resistance at different pressures for the three samples with superconducting transition temperature (T_c) of 18 K, 19 K, and 21 K, respectively. (d)–(f) Resistivity versus temperature for the same three samples. OD: over-doped samples.

It is generally believed that the stability of superconductivity is usually related to the crystal structure. To investigate the structure stability of the compressed sample, we conducted the high-pressure synchrotron x-ray diffraction measurements on the $Bi_{1.74}Pb_{0.38}Sr_{1.88}CuO_{6+\delta}$ sample at 300 K at 4W2 beamline of the Beijing Synchrotron Radiation Facility. As shown in Fig. 2(a), all the peaks observed in the x-ray diffraction measurements can be precisely indexed by the orthorhombic structure in space group *Pnan*. Notably, no new peaks were detected within the pressure range of up to 20.5 GPa. Based on the analysis results, we obtained the lattice parameters and volume versus pressure [Figs. 2(b)–2(d)]. It is seen that the lattice parameters and volume consistently decrease as the pressure increases, indicating that no structural phase transition occurs within the pressure range investigated. This lack of structural phase transition is essential for the successful implementation of our method to convert resistance to resistivity.[29]

We summarize our high-pressure results of the coevolution of T_c, σ, and the value of ρ_s calculated through Homes' law for the over-doped Bi-2201 in the pressure–T_c phase diagram [Figs. 3(a)–3(c)], together with the results achieved from the over-doped Bi-2212 sample [Figs. 3(d)–3(f)] for comparison. To assess the validity of the ρ_s value obtained using our method, we performed extrapolation of our high-pressure data [$\rho_s(P)$] to ambient pressure and compared the extrapolated ambient-pressure ρ_s values (1.5×10^7 cm^{-2} for Bi-2201 and 9.4×10^7 cm^{-2} for Bi-2212) with the ambient-pressure ρ_s values (1.2×10^7 cm^{-2} for Bi-2201 and 9.3×10^7 cm^{-2} for Bi-2212) reported for these two types of materials[30,31] [see red diamond and circle in Figs. 3(c) and 3(f)]. Encouragingly, we found that they exhibit good agreement, indicating the reliability of the $\rho_s(P)$ obtained from our high-pressure investigation.

From the results observed at ambient pressure, we note that the average T_c value of the three Bi-2201 samples is about 19.3 K [Fig. 3(a)], which is significantly lower than that of the Bi-2212 sample [Fig. 3(d)]. The reason behind the higher T_c value in Bi-2212 compared to Bi-2201 still

lacks a comprehensive explanation based on experimental data, although some theoretical investigations proposed that this difference is primarily attributed to the coupling between the layers.[32–34] In order to unravel this puzzle, we conducted estimations of the pressure dependence of conductivity σ and superfluid density ρ_s for these two different kinds of materials [Figs. 3(b)–3(c), Figs. 3(e)–3(f), and Fig. S1 in the Supplementary Information]. Interestingly, we found that although the ambient-pressure σ values at T_c for both Bi-2201 and Bi-2212 are basically at the same level [Figs. 3(b) and 3(e)], but the ρ_s value of the Bi-2201 samples is nearly 10 times lower than that of Bi-2212 [Figs. 3(c) and 3(f)], suggesting that the superfluid density ρ_s plays a crucial role in determining the T_c values of these two high-T_c superconductors.

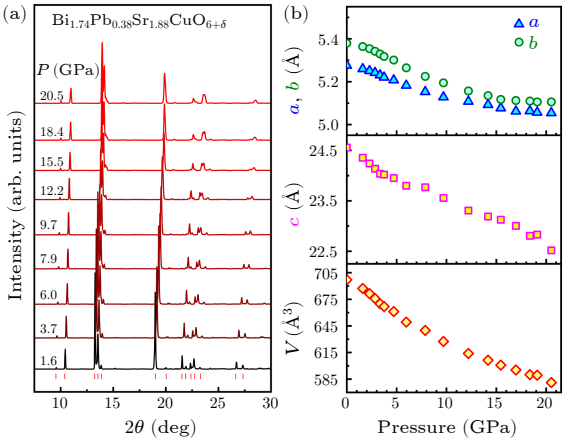

Fig. 2. High-pressure structural information of the $Bi_{1.74}Pb_{0.38}Sr_{1.88}CuO_{6+\delta}$ (Bi-2201) superconductor at room temperature. (a) X-ray diffraction patterns collected at different pressures. All peaks can be indexed well by the orthorhombic structure in space group *Pnan*. [(b), (c)] Pressure dependence of lattice parameters a, b, and c. (d) Cell volume V as a function of pressure. All the data indicates that no structural phase transition occurs in the pressure range investigated.

Furthermore, we also found that the superconducting transition temperatures (T_c) of these two kinds of superconductors exhibit low sensitivity to the pressure applied in the experimental range investigated [Figs. 3(a) and 3(d)]. In contrast, both the conductivity σ and superfluid density ρ_s display significant changes as the pressure increases, exhibiting a dome-like shape [Figs. 3(b)–3(c) and Figs. 3(e)–3(f)]. These results notably highlight that the T_c value of the given material is influenced not only by its superfluid density ρ_s but also by the conductivity σ at T_c. In other words, the T_c value of a given superconductor is closely linked to the ratio of ρ_s/σ, which serves as a combined response factor that reflects the intrinsic electromagnetic state of the superconducting system to external electromagnetic fields.[12,35] Regarding the reason behind the change in slope of ρ_s and σ at a critical pressure P_c, we tentatively suggest that it could be linked to the interplay between the hole-doping effect and the pressure effect. However, this hypothesis requires further investigation to be substantiated. To provide a clearer representation of the relationship between the relevant physical quantities governing T_c, we reformulate Homes' law as $T_c = (1/A) \times (\rho_s/\sigma)$, where T_c is proportional to the ratio of the response coefficients of magnetic field (ρ_s) to electric field (σ). This modified formulation may provide a plausible explanation to why T_c remains insensitive to external pressure while presents dramatic changes in $\rho_s(P)$ and $\sigma(P)$.

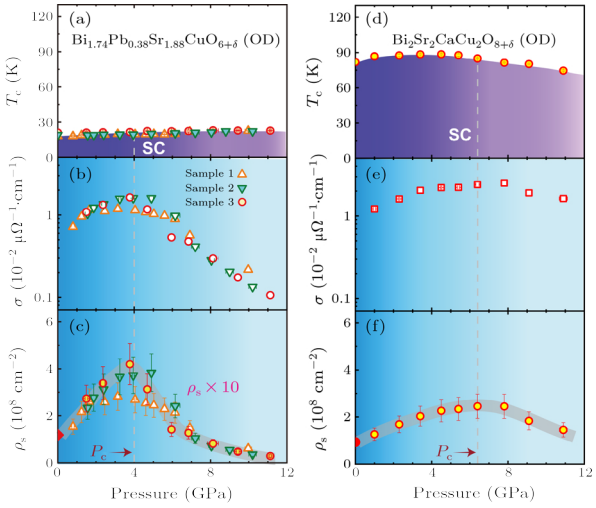

Fig. 3. Summary of the superconducting transition temperature T_c, conductivity σ, and superfluid density ρ_s versus pressure for the over-doped $Bi_{1.74}Pb_{0.38}Sr_{1.88}CuO_{6+\delta}$ (Bi-2201) and slightly over-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) superconductors. [(a), (d)] Pressure–T_c phase diagram for the Bi-2201 and Bi-2212 superconductors. The data displayed in (d) are taken from Ref. [36]. [(b), (e)] Pressure dependence of σ for the Bi-2201 and Bi-2212 superconductors. The data displayed in (e) are extracted from the results reported in Ref. [36]. [(c), (f)] Plots of ρ_s (red circle, orange, and green triangles) versus pressure. The red diamond and solid dot represent the ambient-pressure superfluid density taken from Refs. [30,31], respectively, the values are consistent with the results extrapolated from our high-pressure data.

Finally, we propose that the dome-like evolution of ρ_s and σ with pressure may be attributed to the interplay between the effects of hole-doping and pressure on the metallization process and the subsequent development of superconductivity. It is expected that our results obtained from the compressed high-T_c superconductors and the reformulated Homes' law [$T_c = (1/A) \times (\rho_s/\sigma)$] may provide valuable insights for a better understanding on the mechanism of the high-T_c superconductors, a longstanding challenge of condensed matter physics and material sciences.

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