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Quantum phase transition from superconducting to insulating-like state in a pressurized cuprate superconductor

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Copper oxide superconductors continue to fascinate the communities of condensed matter physics and material sciences because they host the highest ambient-pressure superconducting transition temperature and unconventional electronic behaviour that are not fully explained^{1–3}. Searching for universal links between the superconducting state and its normal metallic state is believed to be an effective approach to elucidate the underlying mechanism of superconductivity. One of the common expectations for copper oxide superconductors is that a metallic phase will appear after the superconductivity is entirely suppressed by chemical doping^{4–8} or the application of a magnetic field⁹. Here we report the first observation of a quantum phase transition from a superconducting state to an insulating-like state as a function of pressure in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) superconductors with two CuO_2 planes in a unit cell for doping below, at and above a level that achieves the highest transition temperature. We also find the same phenomenon in related compounds with a single CuO_2 plane as well as three CuO_2 planes in a unit cell. This apparently universal phenomenon poses a challenge for achieving a unified understanding of the mechanism of high-temperature superconductivity.

Although a huge body of experimental work has been reported for copper oxide (cuprate) superconductors since they were discovered more than thirty years ago^{10,11}, the correlation between the superconducting state and its normal state or the neighbouring ground state is still widely debated^{2,6,12–14}. By changing the chemical makeup of interleaved charge-reservoir layers, electrons can be added to or removed from the CuO_2 planes, resulting in the suppression of the antiferromagnetic insulating state of the parent compound². As the doping level reaches a critical value, superconductivity appears and its transition temperature (T_c) grows to a maximum on doping to an optimal one, then declines for higher doping and finally vanishes at the maximum doping level^{2,4,7,9,15}. It is important to recognize that once the superconducting state is completely suppressed by chemical doping, the material undergoes a quantum phase transition from a superconducting state to a metallic state^{16–18}. However, detailed experimental studies of the breakdown of the superconducting state in cuprates are still lacking, and they may be crucial for

understanding how the superconducting state melts into or emerges from its neighbouring ground states.

Pressure is an alternative method of tuning the superconductivity beyond the chemical doping or external magnetic field, and it can provide significant information on the evolution of superconductivity, electronic state and crystal structure without changing the chemical composition. On the other hand, it can also provide valuable assistance in the search for superconductors with higher values of T_c at ambient pressure by the substitution of smaller ions¹⁹. A notably successful application of this strategy leads to the discoveries of important cuprate- and Fe-based superconductors^{10,20,21}. Therefore, high-pressure studies on superconductivity can not only help the search for new superconductors but also provide a deeper understanding of the correlation between the superconducting state and its neighbouring normal or ground state^{22–26}. To reveal how the superconducting state or non-superconducting state develops—a central issue for understanding the high- T_c superconductivity in cuprates, we performed a series of high-pressure investigations by employing our newly developed state-of-the-art technique—combined in situ high-pressure measurements of the resistance and alternating current (a.c.) susceptibility for the same sample at the same pressure. We studied samples that have been investigated broadly by a variety of methods^{24,27–31}, namely, the under-doped (UD), optimally doped (OP) and over-doped (OD) $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) superconductors with two CuO_2 planes in a unit cell.

Figure 1 shows the results of temperature versus in-plane resistance at different pressures for the UD sample with $T_c = 74$ K (Fig. 1a,d), the OP sample with $T_c = 91$ K (Fig. 1b,e) and the OD sample with $T_c = 82$ K (Fig. 1c,f). It is found that the onset T_c of these samples exhibits the same high-pressure behaviour: a slight increase initially and then a monotonous decrease on elevating pressure until it is not detectable. Subsequently, an unexpected insulating-like state appears at pressure (p_i) of 34.3 GPa for the UD sample, 39.9 GPa for the OP sample and 42.2 GPa for the OD sample. Further, the insulating-like behaviour appears when the pressure is higher than p_i (Fig. 1a–f), which is a grand surprise because one naively expects that by increasing pressure, the bandwidth should widen and the system should become more metallic. We repeated the measurements on new samples and proved that the results are reproducible

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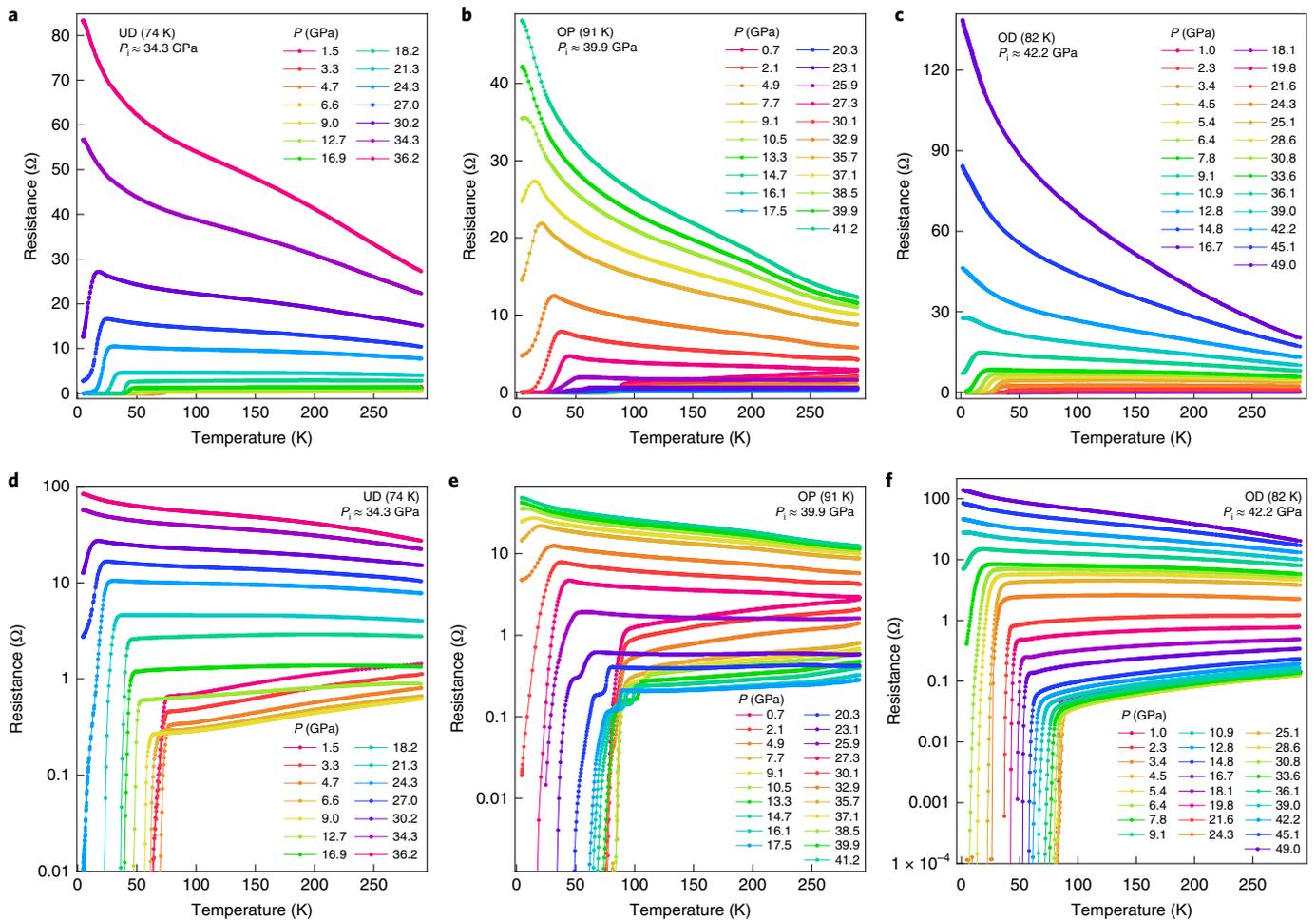


Fig. 1 | Temperature dependence of in-plane resistance for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi2212) at different pressures. a,d, Plots of temperature versus resistance with linear (a) and log (d) scales for the UD superconductor in the pressure range of 1.5–36.2 GPa. **b,e**, OP sample with linear (b) and log (e) scales in the pressure range of 0.7–41.2 GPa; the two-step superconducting transition demonstrates a crossover from two-dimensional to three-dimensional superconducting phases in the pressure range of 4.9–23.1 GPa, which is in good agreement with previous studies²⁶. **c,f**, OD sample with linear (c) and log (f) scales in the pressure range of 1.0–49.0 GPa. The three kinds of samples display the same behaviour of an insulating-like state above pressure p_i .

(Supplementary Fig. 2). Moreover, we found that the transition from the superconducting state to an insulating-like state is reversible when the pressure is released. As shown in Supplementary Fig. 5, the superconductivity with a zero-resistance state reappears in the sample released from the insulating-like state and a metallic normal state appears. In addition, our careful inspection on the sample recovered from 41.6 GPa, by a scanning electronic microscope, demonstrates that no crack is found (Supplementary Fig. 4). These results not only exclude the possibility that the insulating-like state is associated with some pressure-induced cracks but also provides a reasonable explanation on the reversible superconducting transition with a zero-resistance state.

The combined high-pressure measurements of a.c. susceptibility and in-plane resistance were performed for the three kinds of samples mentioned above. The superconducting transitions of the samples detected by a.c. susceptibility can be clearly identified by the onset signal of the deviation from the almost-constant background on the high-temperature side (Fig. 2, blue plots) and the plunge of resistance to zero (Fig. 2, red plots). On compression to 34.3 GPa for the UD sample, 39.9 GPa for the OP sample and 42.2 GPa for the OD sample, all their resistances show an insulating-like behaviour (Fig. 2d,h,i, red plots) and also no diamagnetic signal is captured by

the a.c. susceptibility measurements (Fig. 2d,h,i, blue plots). These results reveal that the pressure induces a quantum phase transition from a superconducting state to an insulating-like state in all these superconductors.

We summarize the experimental results in the normalized pressure– T_c phase diagram (Fig. 3, left), which is established on the basis of the pressure– T_c phase diagrams of the UD, OP and OD Bi2212 samples (Fig. 3, right). The phase diagram for the three kinds of samples shows two distinct regions: the superconducting state (SC) and the insulating-like state (I), and demonstrates a universal quantum phase transition from the superconducting to the insulating-like states. It is shown that T_c displays a slight increase initially within a small pressure range, and then a continuous decrease with elevating pressure, in good agreement with the results reported previously³². At a critical pressure P_c and above (determination of P_c is shown in Fig. 3, inset), an insulating-like state emerges (Fig. 3, left) (details of the normalizing analysis are provided in Supplementary Fig. 11). It is worth noting that P_c for the superconductor–insulator transition increases with the increase in the doping level but does not follow the change in T_c (Fig. 3, top). These results demonstrate that the factors determining thermal stability (T_c) and quantum stability (P_c) of the superconducting ground state are not the same. These results

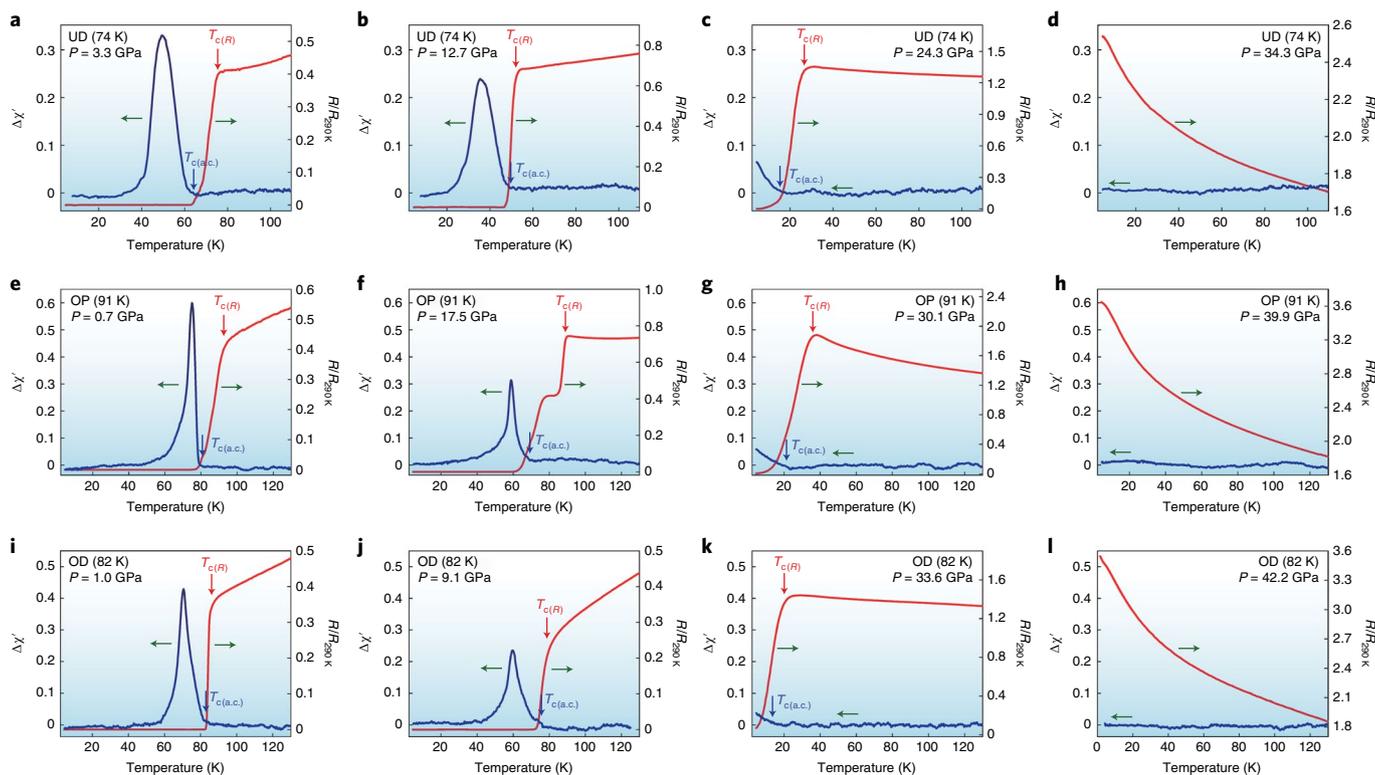


Fig. 2 | In-plane resistance (R) and a.c. susceptibility ($\Delta\chi'$) as a function of temperature (T). **a-d**, Plots of the UD superconductor. **e-h**, Plots of the OP superconductor. **i-l**, Plots of the OD superconductor. The blue lines in the figures are the data for $\Delta\chi'(T)$, whereas the red lines are the data for $R(T)$. The red and blue arrows indicate the temperatures of the onset of superconducting transition detected by the resistance ($T_{c(R)}$) and a.c. susceptibility ($T_{c(a.c.)}$) measurements, respectively.

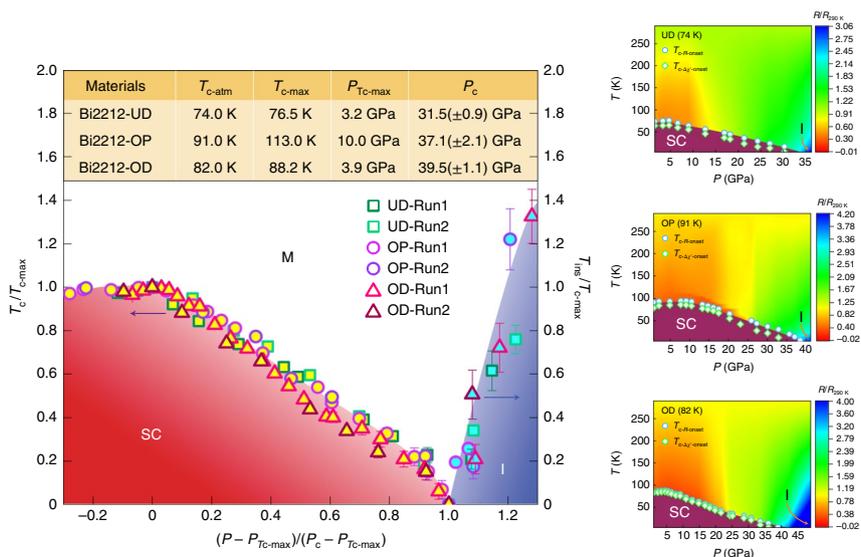


Fig. 3 | Pressure- T_c phase diagrams for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ superconductors. Left, a normalized phase diagram that is built on the basis of the experimental phase diagrams (right). Right: phase diagrams established by the experimental results from the UD, OP and OD samples, together with the mapping information of temperature- and pressure-dependent R (shown in the colour scale). P_{Tc-max} and P_c stand for the pressures where T_c reaches the maximum and zero, respectively. T_{c-atm} stands for the superconducting transition temperature at ambient pressure. In the normalizing analysis, we define the pressure as P_{Tc-max} when $(P - P_{Tc-max}) / (P_c - P_{Tc-max}) = 0$ and define the pressure as P_c when $(P - P_{Tc-max}) / (P_c - P_{Tc-max}) = 1$. The results of the normalizing analysis for T_c / T_{c-max} versus $(P - P_{Tc-max}) / (P_c - P_{Tc-max})$ and T_{ins} / T_{c-max} versus $(P - P_{Tc-max}) / (P_c - P_{Tc-max})$ show that the three kinds of samples display a universal quantum phase transition from the superconducting state to an insulating-like state. SC, M and I stand for superconducting state, metallic state and insulating-like state, respectively. The region of the M phase is determined by the critical value of R/R_{290K} where the quantum phase transition occurs. $T_{c-R-onset}$ and $T_{c-\Delta\chi'-onset}$ denote the onset temperatures of the superconducting transition detected by the resistance and a.c. susceptibility measurements, respectively. T_{c-max} and T_{ins} are the maximum value of T_c and the characteristic temperature of the insulating-like transition, respectively. P_c represents the critical pressure from the superconducting state to insulating-like state (methods for determining T_{ins} and P_c are shown in Supplementary Fig. 11). The details about determining error bars can be found in SI.

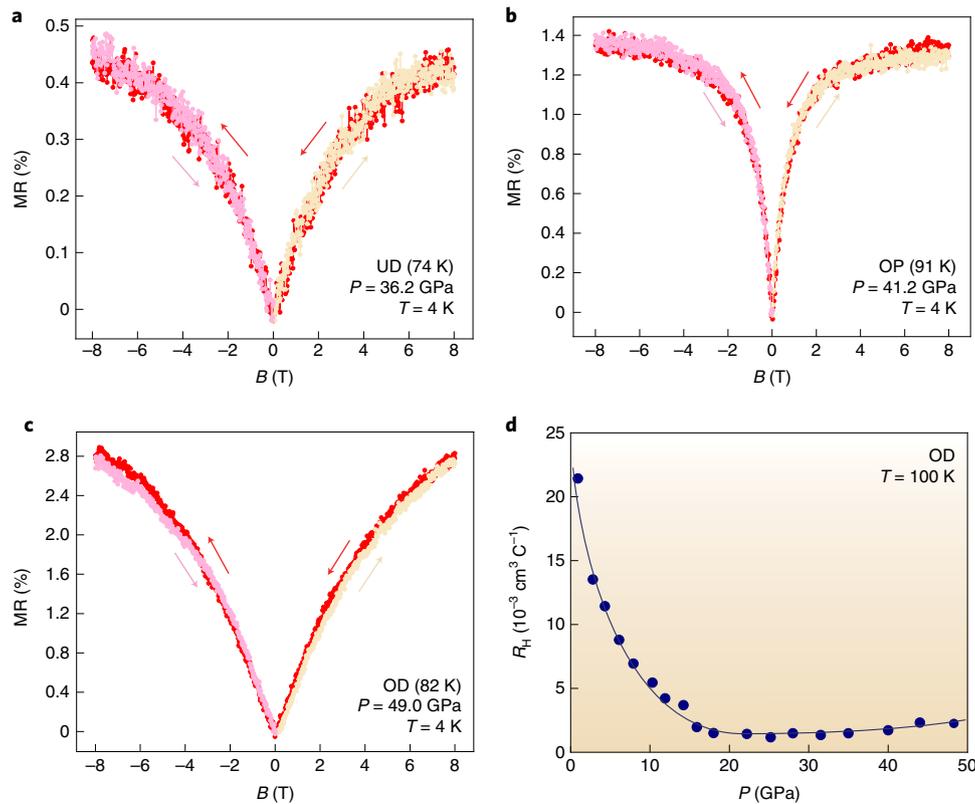


Fig. 4 | Magnetoresistance (MR) of the UD, OP and OD $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ superconductors, and Hall coefficient of the OP sample under pressure.

a–c, Plots of MR versus B for the UD (**a**), OP (**b**) and OD (**c**) samples measured at 4 K and 36.2 GPa for the UD sample, 41.2 GPa for the OP sample and 49 GPa for the OD sample. It is seen that all of them exhibit a positive magnetic effect. The red and beige arrows represent the directions of increasing and decreasing magnetic field, respectively. **d**, Pressure dependence of the Hall coefficient (R_H) for the OD superconductor measured at 100 K.

raise two important questions: what is the factor that determines the value of T_c and what is the factor that controls the P_c value of the superconducting ground state? Further, how do they connect with each other? These questions are still open and call for further investigations from a broader perspective.

To know whether the quantum phase transition discovered in this study is a common phenomenon beyond the Bi2212 superconductors investigated, we conducted the same measurements on the $\text{Bi}_2\text{Sr}_{1.63}\text{La}_{0.37}\text{CuO}_{6+\delta}$ (Bi2201) superconductor with one CuO_2 plane as well as the $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ (Bi2223) superconductor with three CuO_2 planes in a unit cell. The same phenomenon is also found in these superconductors (Supplementary Fig. 3), indicating that the observed quantum phase transition is universal in bismuth-bearing cuprate superconductors, regardless of the doping level and number of CuO_2 planes in a unit cell.

These results impact our knowledge about cuprate superconductors that after the superconducting state is destroyed, the sample should show a metallic state because pressure generally increases the bandwidth. To clarify the possible origin that leads to the destruction of the superconducting state and the emergence of an insulating-like state under pressure, we carried out more experiments.

First, we conducted the high-pressure synchrotron X-ray diffraction measurements at 50 K for the OD sample on 4W2 beamline at the Beijing Synchrotron Radiation Facility. Our results indicated that there is no structural phase transition in the range of pressures up to 43.1 GPa, except that the volume of the lattice is apparently compressed (Supplementary Fig. 9). These results ruled out the possibility that the quantum phase transition from superconducting to insulating-like states connects with a pressure-induced structural phase transition.

Second, we measured the magnetoresistance (MR) at 4 K for the compressed UD, OP and OD samples that host the insulating-like state. The magnetic field was applied perpendicular to the a – b plane of these samples. As shown in Fig. 4a–c, the MR values of all the samples exhibit a positive effect. These results are reminiscent of what has been seen in disordered superconducting films. When the film samples move into an insulating state (driven by the magnetic field) from a superconducting state, a positive MR value indicating a superconducting fluctuation state exists on increasing the field^{33–35}. These results lead us to consider that the positive MR effect may be a universal feature manifesting the existence of a superconducting fluctuation in the insulating state induced either by a magnetic field in the film samples or pressure in the bulk cuprates. As only a small MR value (of the order of 1%) is observed in the insulating-like state of our bulk cuprate superconductor, it implies that the superconducting fluctuation—if it indeed exists in these samples—should be weak.

Third, we performed the high-pressure Hall coefficient (R_H) measurements for the OD sample (Fig. 4d; measurement details can be found in Supplementary Fig. 1) and find that $R_H(P)$ decreases remarkably with increasing pressure up to approximately 18 GPa. Because the Hall resistance versus magnetic field displays a linear behaviour in the pressure range investigated (Supplementary Fig. 6), a typical feature of the single-band system—the decrease in $R_H(P)$ below 18 GPa—ought to be associated with the enhancement in carrier density. However, R_H remains almost unchanged for pressures ranging from approximately 18.0 to 35.0 GPa and then shows a slow increase from approximately 35.0 to 48.3 GPa. No apparent change in $R_H(P)$ at $P_c = 39.5$ GPa implies that the total density of charge carriers seems to remain in a steady state with a very slow change when

crossing quantum criticality. The reproducible result is also obtained in the Bi2201 superconductor (Supplementary Information).

It is noted that unlike the usual insulator, the low-temperature resistance in the insulating-like state rises way too slowly to be exponential. We attempted to fit the low-temperature resistance with exponential dependence and power law, but they failed (Supplementary Fig. 12). Slow rises of this kind have been found in low-temperature orthorhombic $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ cuprates and $\text{La}_{1-x}\text{M}_x\text{OBi}_2$ ($M=\text{Ti}$ or Th), which are perceived as fairly mysterious^{36–38}.

There is, in fact, no precedence anywhere else for such a transition from a superconducting state to an insulating-like state, to the best of our knowledge. Therefore, some questions are naturally raised: why do itinerant superconducting electrons become localized after the quantum phase transition and what is the exotic pathway that results in the quantum phase transition? All the above are attractive issues in searching for new physics behind the pressure-induced quantum phase transition from a superconducting state to an insulating-like state, which deserves further investigation with other advanced experimental probes and sophisticated theoretical studies.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41567-022-01513-2>.

Received: 22 May 2021; Accepted: 14 January 2022;

Published online: 17 February 2022

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Data Availability

Source data are provided with this paper. Other data that support the findings of this study are available from the corresponding author upon reasonable request.

Acknowledgements

We thank J. Zaanen, C.-W. Chu, L. Deng, X. Chen and Y. Zhou for helpful discussions on this work. The work in China was supported by the National Key Research and Development Program of China (grant nos. 2017YFA0302900 and 2017YFA0303103), the NSF of China (grant nos. U2032214, 12122414, 11888101 and 12004419) and the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (grant no. XDB25000000). We appreciate support from the Users with Excellence Program of Hefei Science Center CAS (2020HSC-UE015). Part of the work is supported by the Synergic Extreme Condition User System. J.G. is grateful for support from the Youth Innovation Promotion Association of the CAS (2019008). The work in BNL was supported by the US Department of Energy, Office of Basic Energy Sciences (contract no. desc0012704).

Author contributions

L.S., T.X. and Q.W. designed the study and supervised the project. G.G. grew the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{6+\delta}$ single crystals. H.Y. and X.Z. grew the $\text{Bi}_2\text{Sr}_{1.63}\text{La}_{0.37}\text{CuO}_{6+\delta}$ single

crystals. C.T.L. grew the $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ single crystals. Y.Z., J.G., S.C., J.Z. and L.S. performed the high-pressure resistance, a.c. susceptibility and magnetoresistance measurements. J.G., Y.Z., Y.G., Y.L., X.L. and C.Y. performed the high-pressure X-ray diffraction measurements. J.G., C.H. and S.L. performed the high-pressure Hall measurements. L.S., T.X., Q.W., J.H. and Y.Z. wrote the manuscript in consultation with all the authors.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41567-022-01513-2>.

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Peer review information *Nature Physics* thanks Nandini Trivedi and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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