Scaling of pressure-induced and doping-induced superconductivity
in the Ca$_{10}$(Pt$_n$As$_8$)(Fe$_2$As$_2$)$_5$ arsenides

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The Ca$_{10}$(Pt$_n$As$_8$)(Fe$_2$As$_2$)$_5$ (n=3,4) compounds are a new type of iron pnictide superconductor$^{1-7}$ whose structures consist of stacking Ca-Pt$_n$As$_8$-Ca-Fe$_2$As$_2$ layers in a unit cell. When n=3 (the “10-3-8 phase”), the undoped compound is an antiferromagnetic (AFM) semiconductor, while, when n=4 (the “10-4-8 phase”), the undoped compound is a superconductor ($T_c=26$K), a difference that has been attributed to the electronic character of the Pt$_n$As$_8$ intermediary layers. Here we report high-pressure studies on 10-3-8 and 10-4-8, using a combination of in-situ resistance, magnetic susceptibility, Hall coefficient and X-ray diffraction measurements. We find that the AFM order in undoped 10-3-8 is suppressed completely at 3.5 GPa and that superconductivity then appears in the 3.5-7 GPa pressure range with a classic dome-like behavior. In contrast, $T_c$ in the 10-4-8 phase displays a monotonic decrease with increasing pressure. Our results
allow for the establishment of a unique correspondence between pressure-induced and doping-induced superconductivity in the high-\(T_c\) iron pnictides, and also points the way to an effective strategy for finding new high-\(T_c\) superconductors.

Superconductivity at high temperatures usually develops in the proximity of suppressing antiferromagnetic (AFM) ordered phases that can be tuned by control parameters such as pressure or carrier doping\(^8\text{-}^{10}\). The recently discovered \(\text{Ca}_{10}(\text{Pt}_n\text{As}_8)(\text{Fe}_2\text{As}_2)_5\) (the “10-n-8 phases”, \(n=3\) and 4) arsenide superconductors display an alternative way to suppress AFM order; they achieve superconductivity through control of electronic dimensionality via different intermediary layers. Their structures can be described as replacing alternating \(\text{Fe}_2\text{As}_2\) layers in the \(\text{CaFe}_2\text{As}_2\) (Ca-122) unit cell with \(\text{Pt}_n\text{As}_8\) intermediary layers, \(i.e.,\) by stacking \(\text{Ca-Pt}_n\text{As}_8-\text{Ca-Fe}_2\text{As}_2\) layers. The two different types of intermediary layers have a diverse influence on the physical properties. When they are \(\text{Pt}_3\text{As}_8\), which have a semiconducting nature, the undoped 10-3-8 compound is an antiferromagnetic semiconductor\(^1\text{-}^3\); it is non-superconducting down to temperatures below 1.5 K at ambient pressure. In contrast, when they are \(\text{Pt}_4\text{As}_8\), which have a metallic nature, the 10-4-8 compound presents metallic behavior at ambient pressure and shows superconductivity at 26 K\(^1\text{-}^3\text{-}^4\). When the intermediary layer is semiconducting, the electronic system is more 2D, and, when metallic, the electronic system is more 3D. Upon chemical doping with Pt on the Fe site of the FeAs layer of the 10-3-8 compound, the system can be tuned into the superconducting state\(^1\text{-}^4\text{-}^7\).
High pressure can serve a dual role in tuning both the stability of AFM long-range order and the carrier density through adjustment of the lattice parameters. Therefore it is commonly adopted as an important tool to probe newly discovered superconductors. Here we report a systematic high-pressure study on the semiconducting undoped 10-3-8 compound and the superconducting 10-4-8 compound through a combination of in-situ resistance, ac susceptibility, Hall coefficient and X-ray diffraction measurements performed with diamond anvil cells. We find that the AFM order in the 10-3-8 phase is continuously suppressed under pressure until it disappears at ~ 3.5 GPa. At this pressure, superconductivity emerges. A superconducting dome with maximum $T_c$ of 8.5 K at 4.1 GPa lies in the pressure range of 3.5-7 GPa. In contrast, we find that $T_c$ of the compressed 10-4-8 decreases monotonically with increasing pressure. No pressure-induced structural phase transitions are observed in the 10-3-8 or 10-4-8 compounds for the pressure range investigated at room temperature. Scaling between pressure-induced and doping-induced changes in the superconductivity in these systems reveals unique similarities and differences in behavior, and leads us to propose that the 10-n-8 arsenide superconductors provide a versatile platform for testing available theories of the microscopic physics in iron pnictide superconductors.

The 10-3-8 samples investigated are single crystals. As shown in Fig.1a, the diffraction patterns perpendicular to the crystal plates show only narrow (00l)-type reflections, indicating that the crystals are high quality and single phase. Chemical analysis (employing the WDS method) shows that the composition of the crystals
studied is \( \text{Ca}_{10}(\text{Pt}_{3}\text{As}_8)(\text{Fe}_{1.996}\text{Pt}_{0.004}\text{As}_2)_5 \). Since the amount of Pt in the Fe\(_2\)As\(_2\) layers is much too small to change either the physical or structural properties, we denote the material as undoped 10-3-8 hereafter.

The temperature \((T)\) dependent resistance \((R)\) of the 10-3-8 phase measured at ambient pressure displays three significant features (Fig.1b). The first is an obvious resistance minimum near 175 K; the resistance decreases with cooling and then increases with further cooling, demonstrating a crossover from a metallic to a semiconducting state (the crossover temperature is defined by the resistance minimum and denoted by \(T'\)). The other two features can be seen by careful inspection of the temperature derivative of electrical resistance \(dR/dT\) (inset of Fig.1b) - one is at 103 K and the other is at 90 K. Recent experimental results indicate that both structural and AFM phase transitions occur in the parent 10-3-8 compound\(^2\),\(^6\),\(^11\) and that these transition temperatures can be inferred from the dip temperatures in \(dR/dT\)\(^12\), however, no clear evidence shows which one happens first. We therefore denote these two characteristic temperatures in \(dR/dT\) by \(T_1\) and \(T_2\), respectively. The magnetic susceptibility data above 100 K presents only weak temperature dependence, nearly \(T\)-linear, indicating that the material is in a paramagnetic (PM) metallic-like state at higher temperature (Fig.1c)\(^13\)-\(^16\).

Figure 2 depicts the in-situ temperature dependence of resistance measured at different pressures for the 10-3-8 compound. It is apparent that the resistance is reduced over the entire temperature range when pressure is applied (Fig.2a) and that \(T_1\) shifts to lower temperature (Fig.2a and inset). The evolution of \(T_2\) with pressure is...
more obscure, so the PM-AFM transition temperature is estimated by assuming that $T_1$ and $T_2$ mirror each other with pressure, as is seen in the undoped FeAs-122 phase \(^{17-19}\). At a pressure of \(~3.5\) GPa, $T_1$ is invisible and a sharp drop in the resistance is seen at \(~7\) K. The resistance drop becomes more dramatic with increasing pressure (Fig. 2b), indicating the emergence of the superconducting transition. At pressures above \(7\) GPa, superconductivity is completely suppressed (Fig. 2c). From ambient pressure to \(10.8\) GPa, the normal state resistance minimum moves to lower temperature, and the upturn feature in resistance becomes less pronounced. This is similar to what is observed in Pt-doped 10-3-8 superconductors \(^{1-4,6,7}\) at ambient pressure. In the pressure range of \(10.8-24.3\) GPa, the normal state resistance minimum disappears and the temperature ($T$) dependent resistance ($R$) shows metallic behavior down to \(4\) K. $R(T)$ is almost unchanged between \(24.3 - 31.4\) GPa (Fig. 2d).

To provide further evidence that the 10-3-8 phase in the pressure range of \(3.5-7\) GPa is superconducting, magnetic fields parallel to the \(c\) axis and currents along the \(ab\) plane are applied on the sample separately at fixed pressure of \(4.1\) GPa (Fig. 3a and Fig. 3b). A monotonic suppression of the resistance transition is observed with applied fields and currents, \(i.e.\) the resistance drop moves to lower temperature, and it is almost undetectable at \(7\) T and \(20\) mA, respectively. These are universal indications of the presence of a superconducting state. To fully characterize the superconducting state in the compressed 10-3-8 phase, we then performed alternating current (\(ac\)) susceptibility measurements at high pressure in a diamond anvil cell. As shown in Fig. 3c, no diamagnetism is detected down to \(4\) K at \(1.5\) GPa. However, at \(4.3\) GPa a
remarkable diamagnetism appears at 6.8 K, which unambiguously demonstrates that
the pressure-induced resistance drop in the 10-3-8 phase is a superconducting
transition. $T_c$ further increases to 8.2 K at 5.5 GPa but no longer exists at 10.4 GPa,
consistent with our resistance data. In-situ high pressure X-ray diffraction
measurements at room temperature found no pressure-induced structural phase
transitions under pressure up to ~14 GPa (supplementary information S1 and S2).

We also studied the effect of pressure on superconductivity in the 10-4-8 phase
(Fig.4). The 10-4-8 sample is from the same batch reported in Ref [1]; its $T_c$ at
ambient pressure is about 26 K. $T_c$ decreases with pressure at a rate of -2 K/GPa (The
temperature dependence of the resistance at different pressures for the 10-4-8 phase
can be found in Fig.S3), similar to other overdoped iron pnictide and cuprate
superconductors $^{10,20-23}$. This result is also consistent with previous measurements for
the 10-4-8 phase at pressure below 3 GPa $^3$. In-situ X-ray diffraction measurements
for the 10-4-8 at high pressure show no pressure-induced structure phase transition
under pressure up to 10.2 GPa (Fig.S4).

Employing our data, we construct the pressure-temperature phase diagram for the
10-3-8 phase, as displayed in Fig.5a. $T_J$ is very sensitive to pressure; it decreases with
increasing pressure at an initial rate of -21K/GPa. At ~ 3.5 GPa, $T_J$ is fully suppressed
and superconductivity appears. This is reminiscent of what has been found for
LaFeAsO and A-122 (A=Ca, Sr and Ba), in the sense that superconductivity arises in
the proximity of a suppressed AFM phase $^{13,14,17-19,24}$, revealing that the suppression of
AFM long-range order is also crucial for achieving superconductivity in the 10-3-8
phase. The superconducting dome, with a maximum $T_c$ of ~8.5 K at 4.1 GPa, lies in the pressure range from 3.5 GPa to 7 GPa. Furthermore, the crossover temperature ($T'$) of the semiconducting-to-metallic like state is reduced under pressure, and its extrapolated line approaches zero at ~ 10 GPa. The superconducting dome stands within the semiconducting normal state region, a characteristic that has also been reported in the 10-3-8 superconductor at ambient pressure as a function of Pt doping.

To better understand the superconductivity in the 10-n-8 iron arsenide superconductors, we have added to the same figures the temperature-composition electronic phase diagram for the Pt-doped 10-3-8 phase (Fig. 5a) and the $T_c$-composition relationship for the 10-4-8 phase (Fig. 4). For the 10-3-8 phase, the doping composition is scaled by 0.018 Pt/2.24 GP also that the two $T_1$ phase lines match each other, i.e, the rate of suppression of $T_1$ is the same for both systems. Similarly, the doping composition for the 10-4-8 phase is scaled by 0.056Pt/4GPa in figure 4 so that the initial rate of suppression of $T_c$ under pressure and doping match; the figure shows that the scaling is excellent to pressures up to 4GPa and doping levels up to ~0.056. For the 10-3-8 phase, superconductivity emerges with the same $T_c$ in the phase diagrams for both pressurized and doped systems, at the point where the AFM phase is completely suppressed. This demonstrates that until 3.5 GPa, the tuning effect of applied pressure is equivalent to 1GPa per 0.008Pt doping, leading to the strong similarity of the pressure-composition-temperature electronic phase diagrams. This can be explained by the fact that applied pressure increases the bandwidths in the
10-3-8 phase, as has been seen in other materials\textsuperscript{25,26}, the decreasing semiconducting behavior of the 10-3-8 phase with increasing applied pressure implies that the gap in the Pt\textsubscript{3}As\textsubscript{8} layer is closing due to the increase in bandwidth. As a result of the increased bandwidth in the Pt\textsubscript{3}As\textsubscript{8} layer under pressure, some formerly full states are pushed above $E_F$ and charge transfer occurs between the Pt\textsubscript{3}As\textsubscript{8} layer and the Fe\textsubscript{2}As\textsubscript{2} layer, leading to the electronic doping of the Fe\textsubscript{2}As\textsubscript{2} layers. This hypothesis is proven by our Hall measurements for the undoped 10-3-8 phase under pressure. The Hall coefficient ($R_H$) obtained at different pressures, shown in Fig.5b, is negative at all pressure points investigated, indicating that the pressure makes the sample more $n$ type. Within a single band, the carrier density $n$ can be estimated as $-1/eR_H$. $n$(at 20 K) against pressure is plotted in inset of Fig.5b, together with $n$ against Pt doping. It is seen that $n$ increases from $3.07 \times 10^{19}$ C/cm$^3$ at 0.2 GPa to $1.13 \times 10^{20}$ C/cm$^3$ at 3.5 GPa. The pressure-induced change in carrier concentration in the pressure region <4 GPa is similar and comparable to that seen for Pt-doping\textsuperscript{7}, undisputedly confirming that pressure plays the same role as electron doping. Therefore we can propose that the critical carrier concentration ($n_c$) for pressure-induced superconductivity in the undoped 10-3-8 phase is about $11 \times 10^{19}$ C/m$^3$.

Increasing pressure above 4 GPa decreases $T_c$ (Fig.5a), indicating that pressurized 10-3-8 is “overdoped” from 4 GPa to 7GPa. In contrast, the $T_c$ values at the corresponding scaled Pt doping levels continue to increase, a signature that at these compositions Pt doped 10-3-8 remains in the “underdoped” regime. This, and the fact that superconductivity disappears at low pressures relative to the doping level,
shows a breakdown of the pressure-temperature scaling in this regime, and implies that pressure is a much weaker factor in stabilizing superconductivity in the 10-3-8 phase than Pt doping is. This suggests in turn that high pressure (P>4 GPa) and heavy Pt doping (x>0.05) have distinctly different impacts on the Fermi surface topology, which plays a key role in determining $T_c$ for the arsenide.

Our results also demonstrate the different roles that pressure plays in tuning the properties of the 10-3-8 parent compound and the 10-4-8 superconductor. The electronic phase diagrams in the electron-doped and pressurized 10-3-8 phase are remarkably similar in the moderate pressure and doping range, and a simple scaling law reveals their equivalence. This implies that pressure is equivalent to electronic doping in the 10-3-8 parent phase due to a bandwidth-controlled semiconductor to metal transition in the Pt$_3$As$_8$ intermediary layer and thus a charge transfer to the Fe$_2$As$_2$ layer. Similarly, there is also a simple scaling law of $T_c$ as a function of pressure and composition in the moderate doping/pressure regime in the superconducting 10-4-8 phase. This again implies that transfer of electrons between the Pt$_4$As$_8$ and Fe$_2$As$_2$ layers is occurring under pressure. In this case, pressure and chemical doping appear to be largely equivalent in their impact on the stability of the superconductivity, while at large relative doping and pressures, pressure appears to be less favorable for the stability of superconductivity than doping is.

Finally, from a materials design perspective, we highlight that some intermediary layers are very effective in decreasing the stability of AFM long-range order in parent compounds, lowering the $T_I$ of the system. For example, Pt doping in the 10-3-8 phase
suppresses the AFM state and induces superconductivity. This is in contrast to the situation in the Ca-122 phase, where Pt doping on the Fe site cannot lead to superconductivity\textsuperscript{17}, even though the Ca-122 and the 10-3-8 phases have the same solubility limit for Pt\textsuperscript{3}. The absence of superconductivity in Pt-doped Ca-122 appears to be due to the fact that Pt doping only weakly suppresses the AFM order. The replacement of one Fe\textsubscript{2}As\textsubscript{2} layer with an Pt\textsubscript{3}As\textsubscript{8} layer, however, appears to be more effective in suppressing the AFM order of the Fe\textsubscript{2}As\textsubscript{2} layers; recent NMR studies\textsuperscript{11} on the undoped 10-3-8 phase found that its AFM transition temperature ($T_2$) is dramatically lower than is usually found in iron pnictide parent compounds ($T_2 \sim$170K in Ca-122 and $\sim$100K in 10-3-8), a result of the larger distance between Fe\textsubscript{2}As\textsubscript{2} layers. Therefore, in the 10-3-8 phase, by decreasing the coupling between active superconducting layers, superconductivity may more easily emerge under modest external pressure or light doping. Thus, our results imply that increasing the complexity of the intermediary layers in materials whose magnetic transitions are otherwise too stable, may, in new materials, more easily result in superconductivity through doping or under applied pressure.

**Methods**

Single crystals of the 10-3-8 were grown using the flux method as described in Ref.12. Ambient-pressure electrical resistance and magnetic susceptibility as a function of temperature were performed in a Quantum Design Physical Property Measurement System (PPMS) and a Quantum Design Magnetic Property
Measurement System, respectively. Pressure was generated by a diamond anvil cell with two diamond anvils sitting on a Be-Cu supporting plate oppositely. The anvil diameter is about 300µm. The nonmagnetic rhenium gasket was preindented down to 50µm thickness for different runs. The 10-n-8 single crystal with dimensions of around 80×80×20µm was loaded into the gasket hole. High-pressure electrical resistance experiments were carried out using a standard four-probe technique. High-pressure alternating current (ac) susceptibility measurements were conducted using home-made coils around a diamond anvil\textsuperscript{22,28}. High-pressure Hall coefficient measurements with a four wire symmetry were carried out for the undoped 10-3-8 phase, with a magnetic field perpendicular to the \textit{ab} plane of the sample to 0.2 T by sweeping the magnetic field at fixed temperature for each given pressure. High-pressure angle dispersive x-ray diffraction (XRD) measurements were carried out at beamline 15U at the Shanghai Synchrotron Radiation Facility (SSRF). Silicon oil was used as a pressure-transmitting medium to ensure the sample in a hydrostatic pressure environment. Pressure was determined by ruby fluorescence\textsuperscript{29}.

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Figure 1 X-ray diffraction patterns of the 10-3-8 single crystal and its temperature dependence of resistance and susceptibility at ambient pressure. (a) X-ray diffraction pattern collected at room temperature and ambient pressure for the
sample with actual composition of $\text{Ca}_{10}(\text{Pt}_{3}\text{As}_8)(\text{Fe}_{1.996}\text{Pt}_{0.004}\text{As}_2)_{5.5}$. (b) Electrical resistance as a function of temperature for the same single crystal. The $T'$ represents the crossover temperature of metallic-to-semiconducting like state. The Inset displays the temperature derivative of electrical resistance $dR/dT$. The dip at 103 K is related to a structural phase transition ($T_1$) and the dip at 90 K ($T_2$) is associated with the AFM transition. (c) Temperature dependence of magnetic susceptibility for the sample cut from the same batch for the resistance measurement.

Figure 2 Temperature dependence of electrical resistance for the 10-3-8 at
**different pressures.** (a) Resistance-temperature curves in the semiconducting-like state under pressure to 2.5 GPa. The inset demonstrates the temperature derivative of electrical resistance $dR/dT$ obtained at different pressures. $T_1$ and $T_2$ shift to lower temperatures. (b) A resistance drop is observed in the pressure range between 3.5 GPa - 7 GPa. (c) The semiconducting-like state at lower temperatures is suppressed upon increasing pressure. (d) The resistance-temperature curve exhibits metallic behavior at pressure above 24.3 GPa.

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**Figure 3** Electrical resistance as a function of temperature measured at different magnetic fields and currents at a fixed pressure of 4.1 GPa, and high-pressure alternating current (ac) susceptibility measurements at different pressures for the 10-3-8 single crystal. (a) The resistance-temperature curve of the sample at different magnetic fields. The resistance drop is suppressed with increasing field. (b)
Evolution of the resistance-temperature curve with current. The resistance drop is almost invisible when current reaches 20 mA. (c) Temperature dependence of the real part of the ac susceptibility at different pressures. Diamagnetism is clearly seen at 4.3 GPa, demonstrating the presence of a superconducting state.

Figure 4  Pressure-concentration-superconductivity transition temperature (Tc) diagram of 10-4-8 arsenide superconductors. The blue solid circles represent high pressure data obtained from this study, and the open squares represent the Pt-doped data taken from Ref. 5. The region in red color represents the superconducting region of the compressed material and the region with dashed line represents superconducting region of the Pt-doped material. The scaling of pressure and
chemical doping diverges at a pressure above 4 GPa, implying that the topology of Fermi surface at higher pressure is different from that of chemical doping.

Figure 5a Temperature-pressure electronic phase diagram for the 10-3-8 phase, and scaled temperature-doping phase diagram, showing their equivalence. The blue and green solid circles represent the $T_c$ values obtained from resistance ($T_c(R)$) and $ac$ susceptibility ($T_c(ac)$) measurements. The light green solid circle shows the AFM transition temperature determined by the temperature derivative of electrical resistance $dR/dT$. SC represents the superconducting region, which starts at 3.5 GPa and disappears above 7 GPa. AFM represents the antiferromagnetic phase region. PM represents paramagnetic phase. These data and regions represent the high pressure
measurements. Open symbols—corresponding data from the temperature-doping phase diagram of Pt-doped 10-3-8 [Ref.12]. Superconductivity emerges in both systems where the AFM disappears.

Figure 5b Hall coefficient as a function of temperature obtained at different pressures (main figure) and pressure dependent of carrier concentration for the undoped 10-3-8 phase (inset). Negative Hall coefficient demonstrates that the carriers of pressure-induced superconductivity in the un-doped 10-3-8 are electron dominated. The evolutions of carrier concentration with pressure and Pt doping follow the same trend. The data of $n$ (Pt doping) shown in inset are taken from Ref. [7]. The critical carrier concentration for occurrence of superconductivity in undoped 10-3-8 is given to be $-11 \times 10^{19}$ C/cm$^3$. 

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Supplementary Information for “Scaling of pressure-induced and
doping-induced superconductivity in the
Ca_{10}(Pt_nAs_8)(Fe_2As_2)_5 arsenides”

Figure S1 and Figure 4S show the X-ray diffraction patterns for powder
Ca_{10}(Pt_3As_8)(Fe_{1.996}Pt_{0.004}As_2)_5 (referred to as the 10-3-8 phase) and
Ca_{10}(Pt_4As_8)((Fe_{0.97}Pt_{0.3})_2As_2)_5 (refers as 10-4-8 phase) samples at various pressures
at room temperature. The single crystals were carefully ground to fine powders for
\textit{in-situ} high-pressure diffraction experiments. Angle-dispersive X-ray powder
diffraction experiments for these two systems were performed with a focused
monochromatic X-ray beam with wavelength of 0.6888 Å at BL15U at Shanghai
Synchrotron Radiation Facilities. All the patterns at different pressures can be refined
to the triclinic structure for the 10-3-8 phase and tetragonal structure for the 10-4-8
phase, respectively.
Figure S1  X-ray diffraction patterns of Ca$_{10}$(Pt$_3$As$_8$)(Fe$_{1.996}$Pt$_{0.004}$As$_2$)$_5$ at different pressures up to 14.3 GPa.
Figure S2 Pressure dependence of the spacing of neighboring FeAs layers for the 10-3-8 sample. The data is extracted from X-ray diffraction patterns.
Figure S3 Electrical resistance of Ca$_{10}$(Pt$_4$As$_8$)((Fe$_{0.97}$Pt$_{0.03}$)$_2$As$_2$)$_5$ superconductor as a function of temperature at different pressures.
Figure S4 X-ray diffraction patterns of Ca$_{10}$(Pt$_4$As$_8$)((Fe$_{0.97}$Pt$_{0.03}$)$_2$As$_2$)$_5$ at different pressures up to 10.2 GPa.