High-$T_c$ superconductivity up to 55 K under high pressure in a heavily electron doped Li$_{0.36}$(NH$_3$)$_x$Fe$_2$Se$_2$ single crystal

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We report a high-pressure study on the heavily electron doped Li$_{0.36}$(NH$_3$)$_x$Fe$_2$Se$_2$ single crystal by using a cubic anvil cell apparatus. The superconducting transition temperature $T_c \approx 44$ K at ambient pressure is first suppressed to below 20 K upon increasing pressure to $P \approx 2$ GPa, above which the pressure dependence of $T_c(P)$ reverses and $T_c$ increases steadily to ca. 55 K at 11 GPa. These results thus evidence a pressure-induced second high-$T_c$ superconducting (SC-II) phase in Li$_{0.36}$(NH$_3$)$_x$Fe$_2$Se$_2$ with the highest $T_c^{\text{max}} \approx 55$ K among the FeSe-based bulk materials. Hall data confirms that in the emergent SC-II phase the dominant electron-type carrier density undergoes a fourfold enhancement and tracks the same trend as $T_c(P)$. Interestingly, we find a nearly parallel scaling behavior between $T_c$ and the inverse Hall coefficient for the SC-II phases of both Li$_{0.36}$(NH$_3$)$_x$Fe$_2$Se$_2$ and (Li,Fe)OHFeSe. The present study demonstrates that high pressure offers a distinctive means to further raise the maximum $T_c$ of heavily electron doped FeSe-based materials by increasing the effective charge-carrier concentration via a possible Fermi-surface reconstruction at $P_c$.

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To find out the approaches to raise the critical temperature $T_c$ of unconventional superconductors is one of the most enduring problems in contemporary condensed-matter physics. The great tunability of $T_c$ for bulk FeSe from 8 K to over 40 K [1–7] and the possible high $T_c$ exceeding 100 K in the monolayer FeSe/SrTiO$_3$ [8,9] have spurred tremendous interest recently. The principal route to raise the $T_c$ of FeSe is to dope electron, which has been successfully achieved via the interlayer intercalations [A$_x$Fe$_{2-y}$Se$_2$ (A = K, Rb, ...), A$_x$(NH$_3$)$_x$Fe$_2$Se$_2$, and (Li,Fe)OHFeSe] [2,3,7,10,11], interface charge transfer [8], surface K dosing [12,13], and gate-voltage regulation [6,14]. A common Fermi-surface topology consisting of electron pockets only has been confirmed by the angle-resolved photoemission spectroscopy (ARPES) measurements on these heavily electron doped (HED) FeSe derivatives [15–17]. Based on the bulk resistivity measurements on these HED FeSe bulk materials, the highest $T_c$ at ambient pressure, i.e., $T_c^{\text{onset}} = 46.6$ K and $T_c^{\text{zero}} = 44.8$ K, was achieved in the FeSe flake in a field-effect transistor device based on a solid ion conductor [14]. Further enhancement of $T_c$ via adding more electrons seems to be plagued by the observed insulating state in the overdoped regime [14,18,19]. Whether the $T_c$ of HED FeSe bulk materials can reach well above 50 K or even approach that of monolayer FeSe/SrTiO$_3$ remains an open issue.

Given the limitations of electron doping, it is imperative to explore other routes to further enhance $T_c$ of these HED FeSe materials. The application of high pressure can provide an alternative means. It was reported that pressurization on A$_x$Fe$_{2-y}$Se$_2$ [20,21] and Cs$_{0.4}$(NH$_3$)$_x$FeSe [22] can first reduce $T_c$ and then above a critical pressure $P_c$ induce a second high-$T_c$ superconducting phase (denoted as SC-II to distinguish from the ambient pressure SC-I phase). The observed $T_c$ of SC-II is ca. 10 K higher than that of SC-I. Our recent high-pressure study on (Li,Fe)OHFeSe also evidenced such an SC-II phase above $P_c \approx 5$ GPa, reaching a record high $T_c^{\text{onset}} = 51$ K and $T_c^{\text{zero}} = 46.5$ K at 12.5 GPa [23]. More intriguingly, we observed a sharp transition of the normal state from Fermi liquid for SC-I to non-Fermi liquid for SC-II in (Li,Fe)OHFeSe. In addition, the emergence of SC-II is accompanied by a concurrent enhancement of electron carrier density. These observations demonstrated that high pressure can play a very distinctive role to tune the normal and superconducting properties of these HED FeSe materials.

In order to avoid the complications from the magnetism of intercalated (Li,Fe)OH layer in (Li,Fe)OHFeSe [3], we turn our attention to the recently synthesized Li$_{0.36}$(NH$_3$)$_x$Fe$_2$Se$_2$ single crystal [24], which can reach an optimal $T_c^{\text{onset}} \approx 44.3$ K at ambient pressure. By performing magnetotransport measurements up to 12 GPa, we find that the SC-I phase is quickly suppressed under a low $P_c \approx 2$ GPa, above which an SC-II phase emerges and the highest $T_c^{\text{onset}}$ reaches ca. 55 K at $P \geq 10$ GPa. Similar to (Li,Fe)OHFeSe [23], the reemergence of SC-II is also accompanied with a concurrent enhancement of electron carrier density. Importantly, we obtain a linear relationship between $T_c$ and the inverse Hall coefficient for the SC-II phases of both systems. In this Rapid Communication,
we demonstrate a way for high pressure to further raise $T_c$ of these HED FeSe materials via increasing effective electron carrier density.

Details about the crystal growth and characterizations of Li$_{0.36}$(NH$_3$)$_3$Fe$_2$Se$_2$ single crystal at ambient pressure can be found elsewhere [24]. Each unit cell consists of two FeSe layers with lattice parameters $a = 3.7704$ and $c = 16.973$ Å in the space-group $I4/mmm$ (No. 139). The chemical composition was determined via inductively coupled plasma atomic emission spectroscopy and energy-dispersive x-ray spectroscopy measurements [24]. Because the NH$_3$ molecules are neutral and will not transfer electron carriers into FeSe layers, they should not affect the physical properties of the Li$_{0.36}$(NH$_3$)$_3$Fe$_2$Se$_2$ single crystal.

A palm cubic anvil cell (CAC) apparatus was employed for the accurate measurements of magnetotransport and ac magnetic susceptibility under hydrostatic pressures up to 12 GPa [25]. The standard four-probe method was used for the resistivity measurement, and the current is applied within the $ab$ plane with the magnetic field along the $c$ axis. An antisymmetrized (symmetrized) method was performed to get the $\rho_{xx}(H)$ and $\rho_{xy}(H)$ data. The mutual induction method was employed for the ac magnetic susceptibility measurements with an excitation current of 1 mA and 317 Hz. The superconducting shielding volume fraction was estimated by comparing with the superconducting signal of Pb. Glycerol was used as the pressure transmitting medium, and the pressure values inside the CAC were calibrated at room temperature by measuring the characteristic transitions of bismuth and lead from resistivity.

Figure 1(a) shows the resistivity $\rho(T)$ under various pressures up to 12 GPa in the whole temperature range for Li$_{0.36}$(NH$_3$)$_3$Fe$_2$Se$_2$. Here, we determine $T^\text{zero}$ as the zero-resistivity temperature and define the onset $T^\text{onset}$ as the temperature where $\rho(T)$ above and below intersect with each other. At ambient pressure, $\rho(T)$ displays a broad hump centered around 220 K and shows a sharp superconducting transition with $T^\text{onset} = 44.3$ K and $T^\text{zero} = 42$ K, in agreement with the previous report [24]. The normal-state $\rho(T)$ decreases considerably, and the hump feature fades away gradually with increasing pressure to 3 GPa, above which a quasilinear behavior is restored in a broad temperature range. Similar behavior has also been observed in (Li,Fe)OHFeSe [23]. As illustrated in Fig. 1(a), $\rho(T)$ at $P > 10$ GPa exhibits an anomalous bendover in the normal state. As discussed below, such a behavior should be attributed to the partial conversion of Li$_{0.36}$(NH$_3$)$_3$Fe$_2$Se$_2$ to pristine FeSe, which transforms to three-dimensional MnP-type structure with a semiconducting behavior above 10 GPa [26].

At low temperatures, the superconducting transition displays a nonmonotonic variation with pressure, which can be seen more clearly in Fig. 1(b) from the vertically shifted $\rho(T)$ curves below 100 K. Pressure first reduces $T_c$ quickly to $T^\text{onset} = 25$ K and $T^\text{zero} = 15$ K at 2 GPa. Interestingly, the pressure dependence of $T_c(P)$ suddenly reverses at $P > 2$ GPa and $T^\text{onset}$ ($T^\text{zero}$) increases to 37 K (26 K) at 3 GPa, thus evidencing the emergence of SC-II as seen in (Li,Fe)OHFeSe [23]. But the critical pressure of $P_c \approx 2$ GPa is lower than that of (Li,Fe)OHFeSe. As illustrated in Fig. 1(b), $T^\text{onset}$ and $T^\text{zero}$ exhibit distinct pressure dependences at $P > 2$ GPa: $T^\text{onset}$ first increases quickly with pressure to $\sim 50$ K at 6 GPa and then levels off reaching the highest 55 K at 11 GPa and finally decreases slightly with pressure; in contrast, $T^\text{zero}$ first tracks $T^\text{onset}$ and reaches the maximum value of $\sim 40$ K at 6 GPa and then decreases quickly with the difference between $T^\text{onset}$ and $T^\text{zero}$ enlarged considerably at $P > 6$ GPa. Eventually, $T^\text{zero}$ can be barely reached at 11 and 12 GPa despite a high $T^\text{onset}$ of $\approx 55$ K. Since our previous studies have demonstrated an excellent hydrostatic pressure condition up to 15 GPa for CAC [25], the observed discrepancy between $T^\text{onset}$ and $T^\text{zero}$ above 6 GPa reflects an intrinsic pressure response of Li$_{0.36}$(NH$_3$)$_3$Fe$_2$Se$_2$, implying that the superconducting transition either consists of a distribution of different $T_c$’s or is not bulk in nature.

To further track the evolutions of $T_c(P)$ and to investigate the nature of broad superconducting transitions above 6 GPa, we measured ac magnetic susceptibility $4\pi \chi(T)$ up to 11 GPa. As shown in Fig. 2(a) for $P < 6$ GPa, a single superconducting diamagnetic drop can be clearly observed below $T^\text{d}$, which increases with pressure in perfect agreement with the $\rho(T)$ data. In addition, the superconducting shielding volume fraction increases with pressure and reaches $\sim 90\%$ at 5 GPa, signaling a bulk nature for the observed SC-II at $P \lesssim 6$ GPa. In contrast, the $\chi(T)$ curves at 7 and 9 GPa, Fig. 2(b), evidence two superconducting transitions as indicated by the two successive drops, which correspond to the $T^\text{onset}$ and $T^\text{zero}$ determined from $\rho(T)$. The superconducting phase with a higher $T_c \approx 50$ K can attain a volume fraction of ca. 30% and induces the sudden drop in resistivity at $T^\text{onset}$, but the sample can reach zero resistivity only when the lower-$T_c$ phase also enters the superconducting state below $\sim 33$ K. Although the higher $T_c$ phase remains nearly unchanged with pressure, both $T_c$ and

![FIG. 1. High-pressure resistivity $\rho(T)$ for a Li$_{0.36}$(NH$_3$)$_3$Fe$_2$Se$_2$ single crystal. (a) $\rho(T)$ curves in the whole temperature range illustrating the overall behaviors under pressure up to 12 GPa. At 11 and 12 GPa, the $\rho(T)$ curves were scaled by a factor of 0.5. (b) $\rho(T)$ curves below 100 K illustrating the variation of the superconducting transition temperatures with pressure. Except for data at 0 GPa, all other curves in (b) have been vertically shifted for clarity. The onset $T^\text{onset}$ (up-pointing arrow) was determined as the temperature where $\rho(T)$ curves above and below intersect with each other, whereas the $T^\text{zero}$ (down-pointing arrow) was determined as the zero-resistivity temperature.](020508-2)
the volume fraction of the lower-\(T_c\) phase decrease and nearly vanish at 11 GPa. As such, \(\rho(T)\) at 11 GPa can hardly reach zero until very low temperatures.

From these above characterizations, we can conclude that the SC-II phase is bulk in nature for \(P \leq 6\) GPa, whereas the sample contains two superconducting phases with different \(T_c\)’s above 6 GPa: The high-\(T_c\) (\(\geq 50\) K) phase has a small but nearly constant volume fraction \(\sim 30\%\) to 11 GPa, whereas the low-\(T_c\) (\(\leq 33\) K) phase shrinks and vanishes completely above 11 GPa. Figure 3 summarizes the pressure dependences of \(T_{c_{\text{onset}}}, T_{c_{\text{zero}}}, T_c\) for \(\text{Li}_{0.36}(\text{NH}_3),\text{Fe}_2\text{Se}_2\) together with the \(T_{c_{\text{zero}}}^{\text{FeSe}}\) of FeSe for comparison [26]. Since the obtained \(T_c\)’s of the low-\(T_c\) phase and the \(T_{c_{\text{zero}}}\) from \(\rho(T)\) at \(P \geq 6\) GPa match perfectly with the \(T_{c_{\text{zero}}}^{\text{FeSe}}\) of FeSe, it is likely that \(\text{Li}_{0.36}(\text{NH}_3),\text{Fe}_2\text{Se}_2\) has been partially converted to the pristine FeSe due to the extrusion of Li\(^+\) and ammonia, which might be associated with the solidification of the pressure transmitting medium, glycerol, at about 6 GPa at room temperature [27].

Such a speculation is supported by the disappearance of low-\(T_c\) phase at 11 GPa when the pristine layered FeSe transforms to the MnP-type structure with a semiconducting behavior [26]. The presence of semiconducting FeSe above 10 GPa can also explain the observed bendover in the \(\rho(T)\) curves of 11 and 12 GPa, Fig. 1(a). Nevertheless, the smooth evolution of \(T_{c_{\text{onset}}}, T_{c_{\text{zero}}}^{\text{FeSe}}\) and \(T_c\) of the high-\(T_c\) phase at \(P > 6\) GPa should reflect the intrinsic pressure responses for the remaining pressure-induced SC-II phase. Below we thus focus on the variations of \(T_{c_{\text{onset}}}, T_{c_{\text{zero}}}^{\text{FeSe}}\) and \(T_c\) as a function of pressure for \(\text{Li}_{0.36}(\text{NH}_3),\text{Fe}_2\text{Se}_2\).

The temperature-pressure phase diagram shown in Fig. 3 depicts explicitly the evolution of the superconducting phases of \(\text{Li}_{0.36}(\text{NH}_3),\text{Fe}_2\text{Se}_2\) under pressure. The high-\(T_c\) SC-I phase, initially achieved at ambient pressure via doping electrons through inserting Li\(^+\) and ammonia in between the FeSe layers, is quickly suppressed by pressure, and the SC-II phase emerges above \(P_c \approx 2\) GPa and exists in a broad pressure range. The pressure-induced SC-II in \(\text{Li}_{0.36}(\text{NH}_3),\text{Fe}_2\text{Se}_2\) resembles those observed in \(\text{Fe}_2\text{Se}_2\) [20,21], \(\text{Cs}_0.4(\text{NH}_3),\text{FeSe}\) [22], and \(\text{Li}_{1-x}\text{Fe}_x\text{OHOFeSe}\) [23], pointing to a universal phenomenon for these HED FeSe materials under pressure. But, some specific features for \(\text{Li}_{0.36}(\text{NH}_3),\text{Fe}_2\text{Se}_2\) are noteworthy; i.e., the \(P_c \approx 2\) GPa for the emergence of SC-II is the lowest whereas the maximum \(T_{c_{\text{onset}}}^{\text{FeSe}} \approx 55\) K is the highest among the studied HED FeSe-derived materials. For comparison, the \(P_c\) and the maximum \(T_{c_{\text{onset}}}\) are 5 GPa, 51 K for \((\text{Li}_{1-x}\text{Fe}_x,\text{OHOFeSe})[23]\) and 10 GPa, 49 K for \(\text{Fe}_2\text{Se}_2\) [20], respectively. It seems that \(P_c\) depends on the bonding strength between FeSe and the intercalated layer; the weakest bonding in \(\text{Li}_{0.36}(\text{NH}_3),\text{Fe}_2\text{Se}_2\) gives rise to the lowest \(P_c\). On the other hand, the maximum \(T_c\) achievable in the SC-II phase seems to be proportional to the initial \(T_c\) or the electron doping level at ambient pressure. The maximum \(T_{c_{\text{onset}}} \approx 55\) K in \(\text{Li}_{0.36}(\text{NH}_3),\text{Fe}_2\text{Se}_2\) is very close to the highest \(T_c\) achieved in the FeAs-based materials [28].

To further characterize the SC-II phase, we tentatively probe the information about Fermi surface under pressure by measuring the Hall effect in the normal state just above \(T_c\). Figure 4(a) shows the in-plane Hall resistivity \(\rho_{xy}(H)\) at 50 K for different pressures up to 6 GPa. All \(\rho_{xy}(H)\) exhibit a linear behavior with a negative slope, which confirms that the electron-type carriers dominate transport properties of the SC-II phase. In addition, the slope of \(\rho_{xy}(H)\) decreases gradually with pressure. To quantify this change, we obtained the Hall coefficient \(R_H \equiv d\rho_{xy}/dH\) from the linear fitting to \(\rho_{xy}(H)\) and plotted the pressure dependence of \(R_H\) at 50 K in Fig. 4(b). As can be seen, the magnitude of \(R_H\) decreases quickly and tends to level off above 6 GPa, in line with the variation of \(T_{c_{\text{onset}}}(P)\) of SC-II shown in Fig. 3. By assuming a simple one-band contribution, the electron-type carrier concentration...
can be estimated as \( n_e = -1/(R_{H0}) \). As shown in Fig. 4(b), \( n_e \) takes a value of \( \sim 0.39 \times 10^2 \text{ m}^{-3} \) at 2 GPa and experiences fourfold enhancements to \( \sim 1.5 \times 10^2 \text{ m}^{-3} \) at 6 GPa, tracking nicely the variation of \( T_c(\rho_{xy}) \). It should be noted that the carrier density at 6 GPa is slightly higher than that of \( 1.3 \times 10^2 \text{ m}^{-3} \) at ambient pressure [24]. We have observed a similar concomitant enhancement of \( n_e \) and \( T_c \) in the SC-II phase of (Li\(_{1-x}\)Fe\(_x\))OHFeSe [23], thus implying a common mechanism controlling the \( T_c \) of SC-II phase in these HED FeSe-derived materials. As illustrated in Fig. 4(c), \( T_c(\rho_{xy}) \) of the SC-II phases for both compounds indeed scales linearly with the inverse Hall coefficient \( R_H/R_{H0}(P_e) \) or the electron charge-density \( n_e \), similar to the well-known Uemura’s law [29]. In addition, these two curves are nearly parallel with each other, further elaborating a common origin for the SC-II phase. It should be noted that despite the dramatic enhancement of carrier density the relatively small resistivity change above 2 GPa should be caused by the decrease of mobility under pressure.

As mentioned above, the electron doping plays an essential role to raise the \( T_c \) of bulk FeSe, giving rise to a variety of HED FeSe-derived bulk materials with an optimal \( T_c \) reaching \( \sim 46 \text{ K} \) at ambient pressure. The observed antiferromagnetic insulating behavior in the overdoped regime suggests the presence of some threshold for band filling to approach the Mott transition [18,30]. This sets an upper limit of \( T_c \) for these HED FeSe bulk materials at ambient pressure if the band-structure or Fermi-surface topology remains intact under pressure. Then, high pressure usually broadens the bandwidth, reduces the effective density of states at the Fermi level, and leads to a gradual reduction of \( T_c \) as seen in the SC-I phase.

Since the structural transition has been excluded around \( P_c \) in (Li\(_{1-x}\)Fe\(_x\))OHFeSe [23] and is unlikely to occur at such a low pressure of 2 GPa in Li\(_{10.36}\)(NH\(_3\))\(_3\)Fe\(_2\)Se\(_2\) [31], the sudden reversal of \( T_c(P) \) above \( P_c \) and the emergence of SC-II should be ascribed to an electronic origin, presumably associated with a Fermi-surface reconstruction. The observed higher carrier concentration in SC-II than that of SC-I, e.g., \( 1.5 \times 10^2 \text{ m}^{-3} \) at 6 GPa versus \( 1.3 \times 10^2 \text{ m}^{-3} \) at ambient pressure [24], indicates that the band structure of SC-II would allow for more band filling before reaching the Mott transition. It is likely that the Fermi-surface volume is enlarged above \( P_c \). According to a recent ARPES study on FeSe films by Phan et al. [32], a compression strain realized in FeSe/CaF\(_2\) will enlarge significantly both hole and electron Fermi surfaces in comparison with the strain-free FeSe. Similarly, in these HED FeSe materials, there may exist a critical \( P_c \) above which the compression on the FeSe plane can result in a sudden Fermi-surface reconstruction or Lifshitz transition leading to a larger Fermi-surface volume. The Se-Fe-Se angles and the anion height that can be tuned by pressure should be the key factors governing such a transition [33]. In addition, the observed concomitant enhancement of \( T_c \) and \( n_e \) in SC-II suggests that the Fermi-surface topology in the SC-II phase allows for above \( P_c \) a gradual recovery of the density of states that has been reduced in the SC-I phase. It is interesting to note that the two-dome-shaped superconducting phases have been observed in the overdoped LaFeAsO\(_1-x\)F\(_x\), \( 0 \leq x \leq 0.53 \) [34] and LaFeAsO\(_1-x\)F\(_x\), \( 0 \leq x \leq 0.75 \) [35] at ambient pressure. The two superconducting phases were found to be adjacent with two distinct antiferromagnetically ordered states \( 0 < x < 0.53 \) [34] and LaFeAsO\(_1-x\)F\(_x\), \( 0 < x < 0.75 \) [35] at ambient pressure. The two superconducting phases have proposed that the observed two superconducting phases under pressure may have different pairing symmetries associated with a renormalization of Fermi-surface topology [37]. Although much endeavor is needed to figure out the underlying physics.
mechanisms, our present study together with those previous high-pressure studies demonstrate that these HED FeSe-derived materials would universally emerge a pressure-induced SC-II phase with the maximum $T_c$ about 10 K higher than that of the SC-I phase [20,22,23]. This offers an alternative route to further raise the $T_c$ of these HED FeSe-derived materials.

To summarize, we have performed the magnetotransport measurements on a $\text{Li}_0.36(\text{NH}_3)_2\text{Fe}_2\text{Se}_2$ single crystal under hydrostatic pressure up to 12 GPa and constructed the $T$-$P$ phase diagram featured by the emergence of an SC-II phase above $P_c \approx 2$ GPa. We have achieved the highest $T_{c\text{expt}} \approx 55$ K above 10 GPa among the FeSe-based bulk materials. In addition, we obtained a nearly parallel scaling behavior between $T_{c\text{expt}}$ and the inverse Hall coefficient for the SC-II phase of both $\text{Li}_0.36(\text{NH}_3)_2\text{Fe}_2\text{Se}_2$ and (Li,Fe)OHFeSe. Our present study thus demonstrates a way for high pressure to further raise $T_c$ of these HED FeSe-based materials by increasing the effective charge-carrier concentration via a possible Fermi-surface reconstruction at $P_c$.

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[31] We have attempted to measure the high-pressure synchrotron x-ray powder diffraction on Li$_{0.36}$(NH$_3$)$_y$Fe$_2$Se$_2$, but our effort was hampered by the fact that the intercalated ammonia easily escapes during the sample preparation.


