Correlation between Fermi surface reconstruction and superconductivity in pressurized FeTe$_{0.55}$Se$_{0.45}$

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Here we report the first results of the high-pressure Hall coefficient ($R_H$) measurements, combined with the high-pressure resistance measurements, at different temperatures on the putative topological superconductor FeTe$_{0.55}$Se$_{0.45}$. We find the intimate correlation of sign change of $R_H$, a fingerprint to manifest the reconstruction of Fermi surface, with structural phase transition and superconductivity. Below the critical pressure ($P_C$) of 2.7 GPa, our data reveal that the hole - electron carriers are thermally balanced ($R_H=0$) at a critical temperature ($T^*$), where $R_H$ changes its sign from positive to negative, and concurrently a tetragonal-orthorhombic phase transition takes place. Within the pressure range from 1bar to $P_C$, $T^*$ is continuously suppressed by pressure, while $T_C$ increases monotonically. At about $P_C$, $T^*$ is indistinguishable and $T_C$ reaches a maximum value. Moreover, a pressure-induced sign change of $R_H$ is found at ~$P_C$ where the orthorhombic-monoclinic phase transition occurs. With further compression, $T_C$ decreases and disappears at ~ 12 GPa. The correlation among the electron-hole balance, crystal structure and superconductivity found in the pressurized FeTe$_{0.55}$Se$_{0.45}$ implies that its nontrivial superconductivity is closely associated with its exotic normal state resulted from the interplay between the reconstruction of the Fermi surface and the change of the structural lattice.
The discovery of Fe-based superconductors provides a new platform not only for understanding the microscopic mechanism of high-temperature superconductivity beyond the copper oxide superconductors [1,2], but also for finding new phenomena from correlated electron systems. Among Fe-based superconductors, iron selenide (FeSe) is distinct; it has the simplest crystal structure [3] and shows sensitive effect of pressure on the superconducting transition temperature ($T_C$) [4,5]. The isovalent substitution Se with Te in FeSe superconductors can increase $T_C$ from 8 K to about 15 K [6-8], and more attractively, an unusual interplay between the resonance and the incommensurate magnetism has been found only in the crystals with an average composition near $\text{FeTe}_{0.5}\text{Se}_{0.5}$ [7,9,10]. Intriguingly, recent high-resolution angle resolved photoelectron spectroscopy and scanning tunneling spectroscopy experiments find the evidences for Dirac-cone type spin-helical surface states [11] and Majorana bound states in $\text{FeTe}_{0.55}\text{Se}_{0.45}$ superconductor [12], which is a signature of topological superconductivity. These new findings have renewed research interests of this material. One particularly interesting direction is to explore the variation of its electronic state with lattice structure. Results from such work are expected to reveal insights into the nature of the topological superconductivity of this material.

In general, the unconventional superconductivity of a given material is dictated by multiple degrees of freedom of charge, spin, orbital and lattice. These degrees of freedom as well as the interactions among them can be manipulated by control parameters such as pressure, magnetic field and chemical doping [13-18]. Pressure tuning is a clean way to provide significant information on co-evolution among
superconductivity, electronic state and crystal structure without changing the chemistry, and to result in a deeper understanding on the underlying physics of the exotic state emerging from ambient-pressure materials. In this study, we performed in-situ high pressure transport measurements on the high quality single crystals of FeTe\(_{0.55}\)Se\(_{0.45}\), with the attempt to find such kind of co-evolution information.

The single crystals with nominal composition of FeTe\(_{0.55}\)Se\(_{0.45}\) were synthesized using a flux method [19]. High pressure was generated by a diamond anvil cell made of BeCu alloy with two opposing anvils. A four-probe method was applied for our resistance measurements. Diamond anvils with 300 µm and 400 µm culets (flat area of the diamond anvil) were used for several independent measurements. In the experiments, we employed platinum foil as electrodes, rhenium plate as gasket, cubic boron nitride as insulating material and NaCl as pressure medium. High-pressure Hall coefficient was measured through Van der Pauw method under magnetic field generated from a superconducting coil. The longitudinal component in measured Hall resistivity was eliminated through symmetrizing of Hall resistivity data measured under positive and negative fields. Pressure in all measurements is determined by the ruby fluorescence method [20].

Figure 1 displays the temperature dependence of electrical resistance at different pressures. We find that the superconducting transition temperature \((T_c)\) of sample 1 increases upon elevating pressure and then decreases upon further compression (Fig.1a and 1b), in good agreement with the results reported previously [21-26]. Similar results were obtained in the measurements on sample 2 (Fig.1c and 1d), i.e. \(T_c\) first shows an
increase in the low pressure range, reach a maximum value and then decrease with further pressurizing. At about 12 GPa, the superconductivity is completely suppressed (Fig.1d). We repeated the measurements with new samples in five independent experiments and obtained reproducible results.

To know the connection between the superconductivity and the electronic state in FeTe\(_{0.55}\)Se\(_{0.45}\), we performed high-pressure measurements on Hall resistance \(R_{xy}\) by sweeping the magnetic field \(B\), applied perpendicular to the \(ab\) plane, from 0 T to 2 T on a single-crystal sample at various temperatures, as shown in Fig. 2a-e. \(R_{xy}(B)\) is negative below 33 K at 0.5 GPa, 28 K at 1.8 GPa, 23 K at 2.4 GPa, respectively. However, at the pressures above 3.4 GPa, the plots of the \(R_{xy}(B)\) are positive within the temperature range investigated. These results indicate that an electron-hole carrier balance \((R_{xy}(B)=0)\) at the critical temperatures \((T^*)\) occurs only below 3.4 GPa. If the temperature is fixed at \(\sim 23\) K, the critical pressure for \(R_{xy}(B)=0\) can be estimated to be \(\sim 2.7\) GPa (Fig.2f and Fig. S1).

To visualize the correlation between \(T_C\) and electronic state in FeTe\(_{0.55}\)Se\(_{0.45}\), we summarize our experimental results in Fig. 3, which demonstrates \(R_H\), \(T_C\) and structure information of the FeTe\(_{0.55}\)Se\(_{0.45}\) at different pressures. It is seen that \(T_C\) is significantly enhanced upon increasing pressure in the pressure range of \(0 < P < P_C\) (\(\sim 2.7\) GPa), as shown in the lower panel of Fig.3, while the \(R_H\) derived from the Hall resistance \(R_{xy}\) becomes less negative (see upper panel of Fig.3 and Fig. S1), reflecting that the contribution of hole carriers to the \(T_C\) enhancement is increased.

The connection between the electron state and the lattice structure is one of the key...
issues for understanding the emergence of the exotic phenomena in correlated electron materials [27, 28]. Interestingly, we noted that the high resolution X-ray diffraction measurements find a temperature-induced structural transition of the tetragonal-orthorhombic (T-O) phase at ~ 40 K in FeTe$_{0.43}$Se$_{0.57}$ superconductor [24]. Also, a pressure-induced transition from O phase to monoclinic (M) phase was observed in the same sample at ~2.5 GPa below 40 K [24]. Because the composition of the superconductor used for the high pressure XRD measurements is nearly the same as that of our sample, and, in particular, its ambient-pressure transition temperature (~ 40 K) of the T-O phase and the pressure-induced O-M phase transition at low temperature (at ~2.5 GPa) fall on the line of our $T^*(P)$ (upper panel of Fig.3), we propose that our samples should share the same structure phase transitions to that of the FeTe$_{0.43}$Se$_{0.57}$ superconductor upon cooling at ambient pressure or at the $P_C$ (~ 2.5 GPa) in the low temperature range. We find that $T^*$ decreases with increasing pressure below $P_C$ (blue region of the upper panel) until undetectable at ~ $P_C$ where the O-M phase transition takes place [24]. This implies that below $P_C$ the transport property of the normal state becomes more $p$ type. Meanwhile, $T_C$ of the orthorhombic superconducting phase is around a maximum. On further compression above $P_C$, $T_C$ decreases, while $R_{H}(P)$ undergoes a sign change from negative to positive, as signified by the change of the color from blue to red (see upper panel of Fig.3 and Fig. S1).

The sign change of $R_H$ in materials is usually associated with a reconstruction of the electronic structure on the Fermi surface (FS) [29-32], so that it can be taken as a fingerprint to manifest the FS reconstruction. Our results demonstrate a close
correlation between the FS reconstruction and the T-O or the O-M phase transition. It is interesting to note that the ambient-pressure neutron scattering measurements on superconducting Fe$_{1.08}$Te$_{0.64}$Se$_{0.33}$ [33] and FeTe$_{0.5}$Se$_{0.5}$ [9], whose compositions are similar to that of our sample, show that there are no long-range magnetic order exist in the samples, but the short-range magnetic correlations with the incommensurate excitation in the superconducting phases. Moreover, angle-resolved photoemission spectroscopy (ARPES) studies found that the normal state of the FeTe$_{0.58}$Se$_{0.42}$ superconductor presents a strongly correlated metallic feature, which hosts the effective carrier mass up to 16$m_e$ [34]. Based on our results and analysis, we propose that the nontrivial superconductivity of this class of materials [11,12] may be associated with the interplay between FS reconstruction and the lattice change, which generates the unusual normal state with an incommensurate magnetism.

In addition, at the pressure above $P_C$, the sample loses its nontrivial superconductivity due to an O-M phase transition [35-38]. Considering no significant change in $R_H(P)$ in the M phase (see upper panel of Fig.3 and Supplementary Material), we suggest that the pressure-induced instability, i.e. the extent of its lattice distortion, of M phase is responsible for the $T_C$ decrease.

In conclusion, an intimate correlation among the sign change of $R_H$ (a fingerprint for the reconstruction of the Fermi surface), structural phase transition and $T_C$ in the putative topological superconductor FeTe$_{0.58}$Se$_{0.45}$ has been revealed by our high pressure studies for the first time. We find that a noticeable sign change in $R_H$ influences its superconducting transition temperature remarkably. The nontrivially topological
superconductivity can be stabilized up to 2.7 GPa \( (P_C) \), but it no long exists above \( P_C \) due to a crystal structural phase transition. Our results suggest that the nontrivial superconductivity in this material may be associated with its unusual normal state featured by the dramatic interplay between the electronic state and the lattice change. We hope that the correlation among the sign change of \( R_H \), structural phase transition and \( T_C \) found in this study will shed new light on understanding the entangling state among superconductivity, electronic and lattice structure, and such an entangling state should be responsible for the presence of the nontrivially topological nature of this topological superconductor.

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Figure 1. The superconducting behavior of FeTe$_{0.55}$Se$_{0.45}$ at high pressures. (a) Temperature dependence of the resistance in the pressure range of 0.5 GPa–9.6 GPa for the sample 1. (b) Enlarged views of the resistance in the lower temperature for the sample 1. (c) Resistance as a function of temperature for pressures ranging from 0.8 GPa to 12 GPa for the sample 2. (d) Resistance versus temperature near the superconducting transition of the sample 2.
Figure 2 Hall resistance ($R_{xy}$) as a function of magnetic field ($B$) for the FeTe$_{0.55}$Se$_{0.45}$ single crystals. Plots of $R_{xy}$ versus $B$ at different temperatures in the pressure range of (a) 0.5 GPa, (b) 1.8 GPa, (c) 2.4 G Pa, (d) 3.4 GPa and (e) 4.1 GPa. (f) $R_{xy}$ versus $B$ at 23 K for pressures ranging from 0.5 GPa to 3.4 GPa. The dashed line indicates $R_{xy}(B)=0$ where the pressure is estimated to be ~ 2.7 GPa.
Figure 3 Hall coefficient ($R_H$), structure and superconducting transition temperature ($T_C$) information of the FeTe$_{0.55}$Se$_{0.45}$ at different pressures. Upper panel presents the mapping information of temperature and pressure dependent $R_H$, shown in color scale. Here $T^*$ represents the temperature of the electron-hole carrier balance. T, O and M stand for the tetragonal, orthorhombic and monoclinic phases, respectively. Lower panel displays $T_C$ as a function of pressure. SC$_{NT-e}$ and SC$_{T-h}$ represent the nontrivial superconducting phase with the dominance of electron-carriers and the trivial
superconducting phase with the dominance of hole-carriers, respectively. $T_c(R1)$, $T_c$
(R2), $T_c(R3)$, $T_c(R4)$ and $T_c(R5)$ stand for the $T_c$ obtained by the resistance
measurements for the sample 1, sample 2, sample 3, sample 4 and sample 5. $T_c(ac)$ and
$T_c(RT)$ represent the $T_c$ obtained by the $ac$ susceptibility and resistance measurements.