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FAST TRACK COMMUNICATION

High pressure superconductivity in iron-based layered compounds studied using designer diamonds

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Abstract
High pressure superconductivity in iron-based superconductor $FeSe_{0.5}Te_{0.5}$ has been studied up to 15 GPa and 10 K using an eight probe designer diamond anvil in a diamond anvil cell device. Four probe electrical resistance measurements show the onset of superconductivity (T_c) at 14 K at ambient pressure with T_c increasing with increasing pressure to 19 K at a pressure of 3.6 GPa. At higher pressures beyond 3.6 GPa, T_c decreases and extrapolation suggests non-superconducting behavior above 10 GPa. The loss of superconductivity coincides with the pressure induced disordering of the $Fe(SeTe)_4$ tetrahedra reported at 11 GPa in x-ray diffraction studies at ambient temperature.

(Some figures in this article are in colour only in the electronic version)

The pressure variable has always played a pivotal role in the discovery and optimization of novel superconducting materials. The high temperature superconductivity in a new class of iron-based layered compounds has received extensive attention recently because of the diversity of systems in which this phenomenon has been documented [1, 2]. Iron-based layered compounds like REOFeAs (RE = trivalent rare earth metal), AFe_2Se_2 (A = divalent alkaline earth metal), BFeSe (B = alkali metal), and simple FeSe(Te) materials have exhibited superconductivity with transition temperature (T_c) in the range of 0–55 K [1, 2]. In this broad range of iron-based layered compounds, the basic system FeSe remains of considerable interest as it is the foundation on which more complicated layered structures are built. The superconductivity in the PbO-type structure α-FeSe was discovered recently at 8 K in samples prepared with Se deficiency [3]. The occurrence of superconductivity in this simple layered FeSe compound with edge-sharing $FeSe_4$ tetrahedra has created

tremendous interest in the effects of chemical substitution and high pressure on this material system [4]. In particular, tellurium (Te) substitution has been studied [5, 6] and a maximum superconducting transition temperature of 15.2 K was measured for $FeSe_{0.5}Te_{0.5}$. Additionally, pressure induced enhancement of the superconducting transition temperature of $FeSe_{0.5}Te_{0.5}$ has been reported [6] that warrants further investigations of the high pressure structural and superconducting properties.

Powder materials of Fe (3N purity), Se (3N purity), and Te (5N purity) with appropriate stoichiometry ($FeSe_{0.5}Te_{0.5}$) were mixed in a ball mill for a duration of 1 h. The well mixed powders were cold-pressed into discs under 400 kg cm^{-2} uniaxial pressure, and then sealed in an evacuated quartz tube with pressure less than 10^{-4} Torr and heat treated at 600 °C for 20 h. The reacted bulk sample was reground into fine powders, repressed, sealed, and subsequently sintered at 650 °C for 20 h. The sample for high pressure studies was
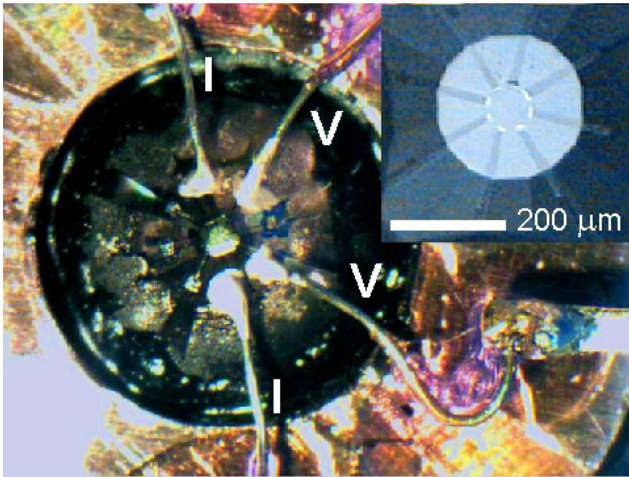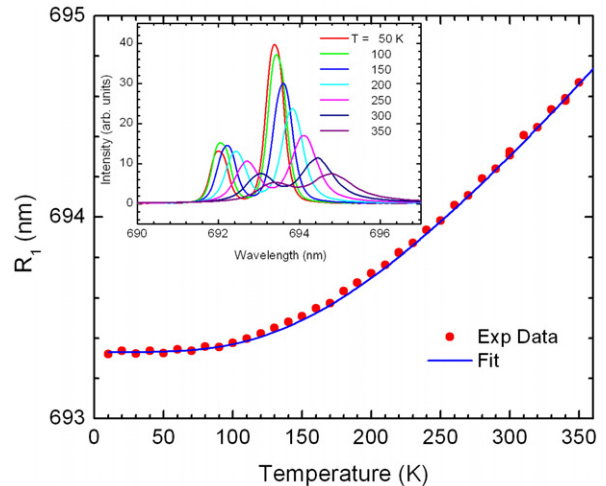
Figure 1. Eight probe designer diamond anvil used in high pressure superconductivity measurements. The current and voltage contacts in the four probe electrical measurements are indicated. The inset is the close-up of the diamond culet showing the eight shiny tungsten metal probes emerging near the center to make contact with the sample at high pressures. The metal probes are embedded in a chemical vapor deposited diamond layer elsewhere except for the contacts indicated.


Figure 2. The measured temperature dependence of the wavelength of the ruby R_1 emission line used in the calibration of pressure at low temperatures. The inset shows the ruby emissions at various temperatures. The solid curve is a fit described in the text.

cut from the sintered pellet and loaded in a gas membrane diamond anvil cell for both structural and superconducting property investigations. The $FeSe_{0.5}Te_{0.5}$ compound belongs to the PbO-type structure (space group $P4/nmm$), tetragonal with 4 atoms/cell. Fe atoms occupy the 2a positions (0, 0, 0) and (1/2, 1/2, 0) and Se and Te atoms at random occupy 2c positions (0, 1/2, z), and (1/2, 0, −z). The parameter z has been determined to be 0.2715 based on earlier work on $Fe_{1+y}Se_xTe_{1-x}$ compounds [7]. The measured lattice parameters for the tetragonal phase of the $FeSe_{0.5}Te_{0.5}$ sample at ambient conditions are $a = 3.7924$ Å and $c = 6.0112$ Å with volume $(V_0) = 21.614$ Å^3/atom. It is to be noted that our materials' synthesis method resulted in a phase pure sample with a well crystallized PbO-type tetragonal phase, and no detectable contamination of any impurity phases were observed in x-ray diffraction studies in the diamond anvil cell.

Figure 1 shows an eight probe designer diamond anvil [8, 9] employed in high pressure superconductivity measurements on the layered iron-based compounds. The eight tungsten microprobes are encapsulated in a homoepitaxial diamond film and are exposed only near the tip of the diamond to make contact with the $FeSe_{0.5}Te_{0.5}$ sample at high pressure (figure 1 inset). Two electrical leads are used to send constant current through the sample and the two additional leads are used to monitor the voltage across the sample (figure 1). The pressure was monitored by the ruby fluorescence technique and care was taken to carefully calibrate the ruby R_1 emission to a low temperature of 10 K, as shown in figure 2. The following equation describes the reference value for R_1 that is utilized in the pressure measurement [10].

$$R_1(T) = R_1(0) + \alpha \left(\frac{T}{\theta}\right)^4 \int_0^{\theta/T} \frac{x^3 \, dx}{e^x - 1},$$
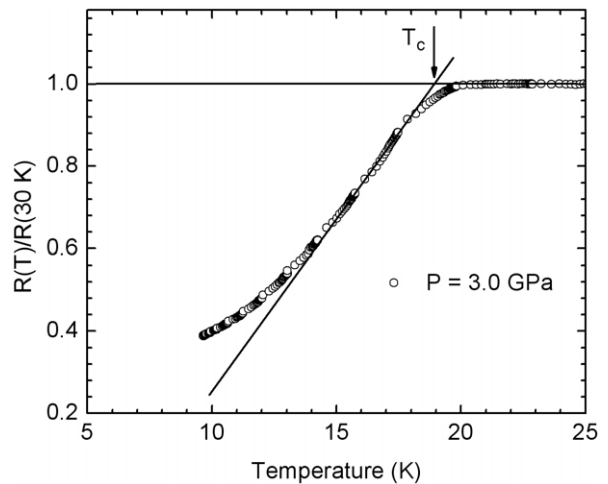
Figure 3. The normalized four probe electrical resistance of $FeSe_{0.5}Te_{0.5}$ as a function of temperature at a pressure of 3 GPa. The measured superconducting transition temperature T_c is 19.2 K.

where $R_1(T)$ is the wavenumber of the R_1 band at temperature T and ambient pressure; $R_1(0)$ is the wavenumber of the R_1 band at 0 K and ambient pressure; θ and α are the effective Debye temperature and an electron–phonon coupling parameter. We used the following fit parameters based on our measurements: $\alpha = -440$ cm^{-1}; $\theta = 760$ K; $R_1(0) = 14\,423.2$ cm^{-1}. The high pressure x-ray diffraction experiments were carried out at the beam-line 16-BM-D, HPCAT, Advanced Photon Source, Argonne National Laboratory. An angle dispersive technique with an image plate area detector was employed using an x-ray wavelength $\lambda = 0.3875$ Å and sample detector distance of 555.5 mm. An internal copper pressure marker with a known equation of state [11] was utilized in our high pressure x-ray diffraction experiments.

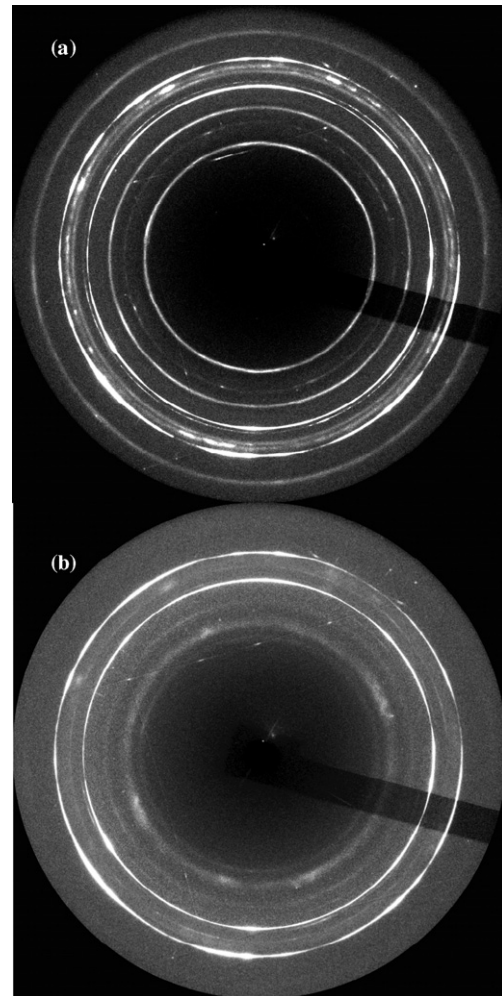Figure 3 shows the measured four probe resistance of the $FeSe_{0.5}Te_{0.5}$ sample as a function of temperature at a
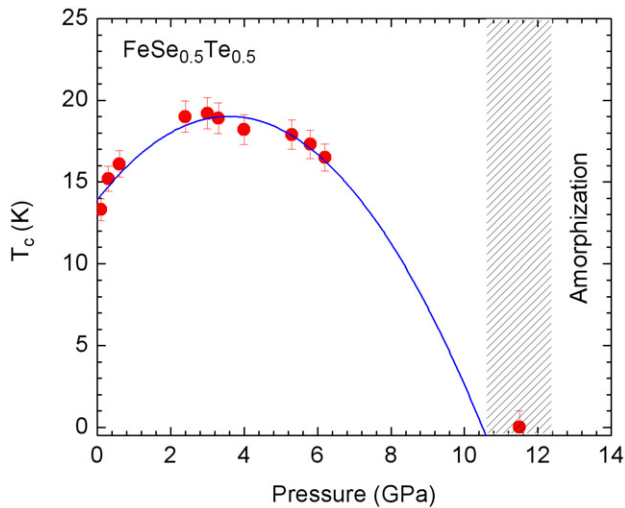
Figure 4. The measured superconducting transition temperature for FeSe$_{0.5}$Te$_{0.5}$ as a function of pressure to 14 GPa. The solid curve is a quadratic fit to the data and is described in the text. The amorphization pressure range of 11.5 ± 1 GPa at room temperature is also indicated.

Figure 5. Image plate x-ray diffraction data collected at a synchrotron source using an x-ray wavelength $\lambda = 0.3875$ Å and sample to detector distance of 555.5 mm. (a) Powder diffraction rings from the tetragonal phase of FeSe$_{0.5}$Te$_{0.5}$ along with a copper pressure marker at a pressure of 9.25 GPa. (b) Powder diffraction patterns showing broad diffuse bands from the amorphous phase at 15.2 GPa, the only two remaining sharp peaks are from the copper pressure marker. All x-ray diffraction spectra have been recorded at ambient temperature.

fixed pressure of 3 GPa. The sample pressure was monitored continuously by an *in situ* ruby fluorescence system during cooling and warming cycles to detect any changes. The pressure values reported are averages over the values recorded during the superconducting transition. The measured four probe resistance shows a plateau at low temperatures before decreasing at the onset of superconductivity. The onset temperature T_c is determined by the intersection of the two linear fits to the data above and below T_c, as shown in figure 3 at a pressure of 3 GPa. The measured T_c at 3 GPa is 19.2 K. The variation of T_c measured in this way is plotted as a function of pressure in figure 4. Our experimental data point at 11.5 GPa does not show any evidence of superconductivity. The measured T_c variation can be fitted by the following quadratic equation over the entire pressure range.

$$T_c \text{(in kelvin)} = -0.40P^2 + 2.86P + 13.97,$$

$$P \text{ is pressure in GPa.}$$

Our measured value of dT_c/dP at ambient pressure ($P \approx 0$) is 2.86 GPa K^{-1}. This is lower than the value of dT_c/dP of 4.87 GPa^{-1} reported earlier in measurements confined to the lower pressure region between 1 and 2 GPa [6]. The maximum T_c from the fit is at 3.6 GPa and has a value of 19.1 K. The measured extrapolation of the parabolic fit to the T_c data predicts that material will be non-superconducting above a pressure of 10 GPa. This prediction coincides with the observations in our x-ray diffraction studies that the FeSe$_{0.5}$Te$_{0.5}$ sample becomes amorphous under high pressures above 11.5 ± 1 GPa at ambient temperature. We show this amorphization pressure range in figure 4. It should be added that amorphization pressure indicated in figure 4 is an approximation and does not include the temperature dependence of this phase boundary. In figure 5, we show image plate x-ray diffraction data in the tetragonal phase at 9.25 GPa and in the amorphous phase at 15.2 GPa at ambient

temperature. The tetragonal phase is identified by the lattice parameters of $a = 3.626$ Å and $c = 5.602$ Å with volume $(V_0) = 18.409$ Å^3/atom at 9.2 GPa. The amorphous phase shows only a single broad band in the 2θ diffraction angle range of 15° accessible in our diamond anvil cell experiments with interplanar spacing for the amorphous phase $d = 2.655$ Å at 15.2 GPa.

The superconducting transition temperatures increase with increasing pressure in the PbO tetragonal phase and the observed decrease in T_c at higher pressure is due to the onset of the disordering of Fe(SeTe)$_4$ tetrahedra layers. Recently, we have carried out additional studies on a FeSe$_{0.8}$Te$_{0.2}$ sample and the maximum in superconducting transition is shifted to higher pressures as the amorphization pressure increases for this composition. Furthermore, the amorphous phase was found to be semiconducting with the electrical resistance

increasing with decreasing temperature. The iron-based layer superconductors are represented by a quasi-two-dimensional system from the point of view of their electronic properties. In addition, structural distortions giving rise to antiferromagnetic ordering occur at low temperatures of below 100 K in $FeSe_{0.5}Te_{0.5}$ at ambient pressure [6]. The application of external pressure can bring the tetrahedral layers together and also affect the onset of low temperature structural distortions. The Cooper pairing of electrons that is so essential for the superconductivity and the pairing depends on the electronic structure and structural distortions. Therefore, further x-ray diffraction studies are needed to establish crystal structures at low temperature and high pressure.

In this letter, we have presented high pressure superconductivity data on the iron-based layered superconductor $FeSe_{0.5}Te_{0.5}$ to pressures up to 15 GPa and temperatures to 10 K. The superconducting transition temperature increases with increasing pressure to 3.6 GPa and then shows a decrease beyond this pressure. The extrapolation of superconducting transition temperature data suggests that a non-superconducting state is favored above 10 GPa. This observation coincides with the x-ray diffraction studies, indicating that the $FeSe_{0.5}Te_{0.5}$ sample undergoes a phase transformation from a tetragonal to amorphous phase above 11 GPa. It is likely that this perturbation of superconducting states due to a disordering of $Fe(SeTe)_4$ tetrahedra is a fairly common phenomenon in other iron-based superconductors under high pressure. Further high pressure structural studies at low temperatures are needed to firmly correlate the crystalline–amorphous phase boundary with the vanishing superconductivity in these materials.

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