Pressure Dependence of Superconducting Properties, Pinning Mechanism, and Crystal Structure of the Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ Superconductor

Kannan Murugesan, Govindaraj Lingannan, Kento Ishigaki, Yoshiya Uwatoko, Chihiro Sekine, Yukihiro Kawamura, Hayashi JunIchi, Boby Joseph, Ponniah Vajeeston, Pankaj Kumar Maheswari, V. P. S. Awana, and Arumugam Sonachalam*

**ABSTRACT:** We have investigated the pressure (P) effect on structural (up to 10 GPa), transport [R(T): up to 10 GPa], and magnetic [(M(T): up to 1 GPa)] properties and analyzed the flux pinning mechanism of the Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ superconductor. The maximum superconducting transition temperature ($T_c$) of 22 K with the P coefficient of $dT_c/dP = +2.6$ K/GPa up to 3 GPa ($dT_c/dP = -3.6$ K/GPa, $3 < P < 9$ GPa) was evidenced from R(T) measurements. The high-pressure diffraction and density functional theory (DFT) calculations reveal structural phase transformation from tetragonal to hexagonal at 5.9 GPa, and a remarkable change in the unit cell volume is observed at ~3 GPa where the $T_c$ starts to decrease, which may be due to the reduction of charge carriers, as evidenced by a reduction in the density of states (DOS) close to the Fermi level. At higher pressures of 7.7 GPa $\leq P \leq 10.2$ GPa, a mixed phase (tetragonal + hexagonal phase) is observed, and the $T_c$ completely vanishes at 9 GPa. A significant enhancement in the critical current density ($J_c$) is observed due to the increase of pinning centers induced by external pressure. The field dependence of the critical current density under pressure shows a crossover from the $\delta$P pinning mechanism (at 0 GPa) to the $\delta T_c$ pinning mechanism (at 1.2 GPa). The field dependence of the pinning force at ambient condition and under pressure reveals the dense point pinning mechanism of Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$. Moreover, both upper critical field ($H_{c2}$) and $J_c$ are enhanced significantly by the application of an external P and change over to a high P phase (hexagonal ~5.9 GPa) faster than a Fe$_{0.99}$Ni$_{0.01}$Se$_{0.5}$Te$_{0.5}$ (7.7 GPa) superconductor.

**INTRODUCTION**

The discovery of superconductivity in iron selenide ($\alpha$-FeSe) with $T_c = 7$ K$^1$ has gained considerable attraction after cuprates$^2,3$ and iron arsenides$^4,5$ because of their simple crystal structure. The iron-based superconductors (SCs) are the most promising materials for high-magnetic-field applications due to their large $T_c$, $J_c$, and $H_{c2}$ and low anisotropy.$^6$-$^{15}$ The binary FeSe$_{1-x}$ system in which Se deficiency stabilizes the superconductivity has a similar structure to that of iron pnictide (Fe-As) SCs.$^4,5$ The Fe-Se layer plays a key role in the appearance of superconductivity in Fe-Se-based SCs$^1$ like the Fe-As layer in 1111 and 122 iron pnictide series.$^{14,15}$ Te substitution in Se sites raises the $T_c$ from 7 to 14 K in FeSe$_{0.5}$Te$_{0.5}$.$^{16,17}$ Addition of metallic elements (MEs) in Fe sites of FeSe$_{0.5}$Te$_{0.5}$ (Fe$_{1-x}$ME$_x$Se$_{0.5}$Te$_{0.5}$) is also found to enhance the superconducting properties. For example, on Cr substitution of 1%, the $T_c$ increased from 14 to 15.7 K.$^{18}$ Further, in this case, the diamagnetic shielding fraction was improved and a very high upper critical field ($H_{c2}(0) = 332$ to 1080 kOe) was observed.$^{18}$ In Ag doping of 5%, even though the $T_c$ decreased from 14 to 11 K, an excess amount of Ag induced a new pinning center with an enhancement of $J_c$ (i.e., $J_c/J_c(0)$ is increased from 1 to 1.5).$^{19}$ With 5% Sn doping, $T_c$ is increased from 9 to 15 K with a simultaneous structural phase change from the hexagonal phase to the tetragonal phase.$^{20}$ As compared with Cu substitution in Fe$_{1−x}$Te$_{0.6}$Se$_{0.4}$, Co doping increases the paramagnetic moments and magnetic moments with an increase in the doping level.$^{21}$ Further, a doping effect is reported for the composition of Fe$_{0.95}$ ME$_{0.05}$Te$_{0.8}$Se$_{0.2}$ (ME...
Fe0.97Co0.03Se0.5Te0.5, respectively, have also been reported. J_δ required for various industrial applications of SC materials. strong vortex pinning and high critical current density are δT decreases. Further, the external pressure (P) enhances the flux pinning by changing the structural parameters and shrinking unit cells.28 and enhances the c/a ratio under pressure up to ∼10 GPa.

In addition to the doping of elements (chemical pressure), external pressure (P) is an effective way to enhance/decrease the SC properties by changing the structural parameters and pinning centers. Further, the external P reduces anisotropy by shrinking unit cells28 and enhances the J_c by inducing more pinning centers.29,30 As far as the chemical pressure is concerned, for β-FeSe, T_c increases (8.5 to 36.7 K under an applied P of 8.9 GPa) and then decreases at higher P followed by a decreasing trend, which is associated with the remarkable decrease in volume of the unit cell. It is due to the collapse of the space between the Fe2Se2 planes, and a phase transition occurs from the tetragonal to hexagonal phase.31 Pressure 10 GPa. Further, on compression above 11.9 GPa, a distorted monoclinic structure is observed, and it led to the suppression of superconductivity. Further, for FeSe_1−xTe_x at x = 0.5, T_c (∼14 K) is enhanced (27−37 K) by applying an external pressure.31,33−35 For the Co-doped sample, a significant enhancement of T_c (from 11.5 to 17 K) and J_c has been reported up to 1.2 GPa.36 Further, the P effect on the flux pinning mechanism has also been reported for Fe0.99Co0.01Se0.5Te0.5.36 Generally, from the application point of view, SCs with high T_c, high J_c and high H_c, are preferred; however, it is still a huge obstacle to enhancing the J_c along with vortex pinning and other superconducting properties. Evidently, the J_c, H_c, and flux pinning were enhanced by both chemical and physical pressures on this iron-selenide system.25−27 The ionic radius of Mn is higher than that of Ni and also the Fe0.99Ni0.01Fe0.5Te0.5 sample to have a negative chemical pressure and the corresponding change in T_c. Further, it exhibits dT_c/dP = +5 K/GPa up to 3 GPa followed by a decrease of T_c and suppression at 10 GPa (quasi-hydrostatic pressure). Hence, we investigated the Fe0.99Mn0.01Se0.5Te0.5 sample under a hydrostatic pressure of up to 12 GPa and compared the results with the Ni-doped one to understand the P dependence of T_c on both samples. In this context, the P effect on the pinning mechanism has been reported only for FeSe0.5Te0.5 and Fe0.97Co0.03Se0.5Te0.5 but not for Mn-doped FeSe0.5Te0.5. Hence, we investigated the SC properties and flux pinning properties of the Fe0.99Mn0.01Se0.5Te0.5 superconductor under an external P. Further, we also investigated the P and field dependence of the critical current density.

RESULTS AND DISCUSSION

Structural Properties of Fe0.99Mn0.01Se0.5Te0.5 under Pressure 10 GPa. The in situ powder X-ray diffraction data were collected at ambient pressure and various P (0−10 GPa) at RT in the ELETTRA synchrotron radiation Xpress beamline for the Fe0.99Mn0.01Se0.5Te0.5 sample, as shown in Figure 1a. Exploration of the diffraction profile shows a tetragonal structure (space group: P4/nmm) in the Fe0.99Mn0.01Se0.5Te0.5
superconductor at ambient $P$ (see Figure 1b). The observed powder diffraction profiles under various $P$ reveal that no structural change occurs and continue to exhibit tetragonal structures in the $P$ range of 0.1–7.7 GPa. Further, a monotonic decrease of the lattice parameters until 3.28 GPa followed by a moderate decrease until 10 GPa (Figure 1d) and found a sudden change in the $c/a$ ratio at 3.28 GPa (tetragonal cell parameters from 1.58456 to 1.54381 (2.4%)) (Figure 1d) and a 10% reduction in the volume at 3.28 GPa (Figure 2) as observed for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$. These results reveal a possible collapse of the van der Walls-bonded region that separates Fe$_x$Se$_y$ planes, similar to structural studies reported for the FeSe compound.37 Between 7.7 and 10.2 GPa, a more complicated structure is observed. At 7.7 GPa, the diffraction profile shows a partial transformation to a hexagonal NiAs-type structure—the same transformation reported for FeSe$_{31,38}$ and also for Fe$_{0.99}$Ni$_{0.01}$Se$_{0.5}$Te$_{0.5}$ at different $P$. A characteristic first-order transition of mixed-phase (tetragonal + hexagonal) coexistence is seen in the wide $P$ region: 5.9 GPa $\leq$ $P$ $\leq$ 10 GPa. It is noted that for FeSe the mixed phase started at 8.5 GPa and a complete transformation to the hexagonal phase was observed at 12 GPa.39 However, the $P$ dependence of lattice parameters was strongly anisotropic ($a$ and $c$ contract by 3.3 and $\sim$8%, respectively), revealing a significant shrinkage in the interlayer spacing compared to the intralayer dimensions. As $P$ increases, the $c/a$ ratio smoothly decreases, as shown in Figure 1d. Further, in the $P$ region 7.7–10 GPa shows a mixed phase. For the case of Fe$_{0.99}$Ni$_{0.01}$Se$_{0.5}$Te$_{0.5}$, the mixed phase was identified at 7.5 GPa, and at 15 GPa, the tetragonal structure was completely transformed to a hexagonal structure.39 A close inspection shows that all of the peaks are shifted to a higher angle (Figure 1c), which reveals the reduction in the unit cell volume. Figure 2 shows the pressure dependence of the unit cell volume of Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$. There is a discontinuous change in the unit cell volume between 3 and 4 GPa. The equation of state (EOS) fit to the data is undertaken with a second-order Birch–Murnaghan equation40 as follows

$$P(V) = \frac{3}{2} B_0 \left( \left( \frac{V_0}{V} \right)^{7/3} - \left( \frac{V_0}{V} \right)^{5/3} \right)$$

(1)

where $B_0$ is the bulk modulus and $V_0$ is the unit cell volume at ambient pressure at atm conditions. The fittings yield $B_0 = 33$ GPa below 3.28 GPa and $B_0 = 80.7$ GPa above 3.28 GPa. The $B_0$ values we obtain are comparatively higher than those of FeSe$_{32,37}$ and FeSe$_{0.5}$Te$_{0.5}$ (30.9 and 36.6 GPa, respectively) in the tetragonal phase. In the pressure-released state (0.02 GPa), although the intensities are not restored, all Bragg peaks are almost reverted to their ambient $P$ condition, thus showing the elasticity of Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$.

DFT Calculations. The structural parameters of FeSe in the hexagonal and tetragonal (P4/mnm) phases were taken from the ICSD reference database$^{41}$ for the initial structure generation. The occupancy of Se and Te in both systems is 50:50. For the partial occupancy simulation, the ab initio random searching structure (AIRSS)$^{42}$ approach was employed to create possible suitable model structures for the chosen substitution in the Fe$_{64}$Se$_{32}$Te$_{32}$ matrix, along with VASP calculations. As a result, in the acquired AIRSS model, Mn replaces one Fe, and the structure is entirely relaxed. In this case, the theoretical composition (Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$) is nearly identical to the experimental composition (Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$). The resulting structures are depicted in Figure 3. To get a better understanding of the relative stability, total energy versus unit cell volume (see Figure 4a) was calculated for both phases. Figure 4b shows the measured cell volume of 86.2 Å$^3$ f.u.$^{-1}$,
and the tetragonal polymorph has the lowest energy structure, which is compatible with the experimental findings. Pressure-induced structural phase transformation from $P4/nmm$ to $P6_3/mmc$ was observed, and this entails reconstructive rearrangements of the cation and anion sublattices. The measured energy-volume data yields bulk module values of 36.8 and 95.3 GPa for $P4/nmm$ and $P6_3/mmc$, which agree well with experimental values of 33 and 80.7 GPa, respectively. Figure 5 shows the total density of states (DOS) for both $P4/nmm$ and $P6_3/mmc$ phases at equilibrium volumes and phase transition points.

Figure 5. Estimated DOS as a function of energy at $P = 0$ GPa (for the tetragonal phase $P4/nmm$) at higher $P$ (where the transition occurs).

and $P6_3/mmc$ phases at equilibrium volumes and phase transition points. $P4/nmm$ has more electrons at Fermi energy ($E_F$) than $P6_3/mmc$. As $P$ increases, the electron density increases at $E_F$ in the tetragonal phase ($P4/nmm$) (see Figure 6). Despite the fact that $P6_3/mmc$ is not an energetically desirable structure in the pressure range 0−4 GPa, it exhibits a similar tendency at $E_F$. This observation agrees with the experimental results that $T_c$ decreases with increasing pressure due to a structural transition, together with a decrease in charge carriers in the density of states at the Fermi surface.

Heat Capacity Studies. Figure 7a shows the temperature-dependent heat capacity ($C_p$) measurement for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ (inset) from 2 to 300 K without a magnetic field. There is a small heat capacity jump observed at $T_c = 14$ K showing the absence of bulk superconductivity. The $T$ dependence of $C_p$ can be explained by the Debye equation

$$C_p = \frac{\gamma}{T} + \frac{\beta}{T^3} + \frac{C}{T^5},$$

where $\gamma$, $\beta$, and $C$ are the electronic, lattice heat capacity, and anharmonic impurity contribution, respectively. Further, the equation well fits in the low-temperature region (Figure 7b) and gives fitting parameters of $\gamma = 25$ mJ mol$^{-1}$ K$^{-2}$ and $\beta = 0.45$ mJ mol$^{-1}$ K$^{-4}$; the estimated $\gamma$ value is in agreement with the theoretical value. The Debye temperature, $\Theta_D = (1944/\beta)^{1/3}$, estimated from the fitting parameter $\beta$ is 162 K. ($C - \gamma T)/T^3$ vs $T$ shows a broad maximum at $T_{max} \approx 13$ K. This characteristic temperature is described as the Einstein temperature, $\Theta_E \approx 4.92 T_{max}$ and it is estimated to be $\Theta_E \approx 64$ K. We analyzed the results using the MM theory and showed that Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ samples fall under the phonon-mediated SC mechanism and $T_c$ can be described as

$$T_c = \frac{\Theta_D}{1.45} \exp \left[ -\frac{1.04(1 + \lambda)}{\lambda - \mu^* (1 + 0.62\lambda)} \right]$$

where $\Theta_D$, $\lambda$, and $\mu^*$ are the Debye temperature, the electron−phonon coupling constant, and the Coulomb pseudo-potential, respectively, and $\lambda$ is estimated to be 1.12. Hence, the superconductivity for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ could be understood based on the strong-coupling BCS theory as in the cases of FeSe$_{0.88}$ and FeSe$_{0.6}$Te$_{0.4}$ where the estimated $\lambda$ values were greater than 1.
the Fe0.99Mn0.01Se0.5Te0.5 sample was measured using different types of pressure cells with various fixed pressures up to 10 GPa using a hybrid clamp-type piston cylinder pressure cell (3 GPa), DAC (5 GPa), and a cubic anvil press (10 GPa), shown, respectively, in (a), (b), and (c) of Figure 8. At ambient pressure, R(T) results show a little increase of resistance as T decreases from 300 to 200 K, which indicates the semimetallic behavior of the sample. However, the resistance decreases from 200 to 14.1 K, which indicates the metallic behavior; further decrease of the temperature resistance suddenly drops to zero at $T_c = 14.1$ K, and it is consistent with the measured magnetization data and reported value. 46 The behavior of normal state resistance is similar to that of FeSe0.5Te0.5.47,48 However, it is to be noted that the resistance curve gets flattened at the same time as doping of Co and Ni at the Fe site in FeSe0.5Te0.5 and $T_c$ also gets decreased for Co ($T_c = 7$ K) and Ni ($T_c = 5.2$ K).47 The significantly large overlap between electron and hole bands at the Fermi surface modifying the density of the charge carrier reflects the curved nature above $T_c$.48 Further, on increasing P (>3 GPa), the resistance curves were flattened and the $T_c$ increased monotonically from 14.1 to 22 K. The P dependence of $T_c$ ($P$–$T_c$ phase diagram) obtained from the R(T) measurement under various Ps was found to be domelike. Figure 8a–c clearly reveals that $T_c$ was enhanced up to a P of ~3 GPa, at which it reached the highest $T_c$ of 22 K and a positive temperature coefficient of $dT_c/dP = +2.6$ K/GPa (P range 0 ≤ P ≤ 3 GPa). At higher Ps, $T_c$ begins to turn down and the pressure coefficient becomes negative ($dT_c/dP = −3.6$ K/GPa (3 ≤ P ≤ 9 GPa)). Further, application of high P to 10 GPa leads to favoring the suppression of superconductivity. The phase diagram of $T_c$–P for Fe0.99Mn0.01Se0.5Te0.5 shown in Figure 9 reveals a change in the sign of the pressure coefficient from positive to negative that occurs at 3 GPa. A similar domelike behavior has been reported in FeSe and Fe1.03Se0.43Te0.57 with maximum $T_c$ values of 36 and 26 K,32,34,38 respectively.

Understanding the dome-shaped $T_c$–P phase diagram is challenging to have a deeper look into the mechanism of superconductivity in these materials. Effect of P on the FeSe system by varying the unit cell parameters interlayer/intralayer and interatomic distance40 leads to modification of the electronic structure and strengthens the interaction among the electrons in the unit cells and the SC state affected by these parameters.

There are other possible reasons for the behavior of $T_c$. (i) At modest P until 4.5 GPa, an increase in N ($E_F$) enhances the superconductivity and reaches a high $T_c$ (22 K at 3 GPa in R(T)); however, at high P (>5 GPa), the lattice stiffening effect becomes detrimental to $T_c$ or the P response of volume markedly changes at 3–4 GPa, revealing the structural distortion, which leads to a decrease in $T_c$. (ii) The Fe0.99Mn0.01Se0.5Te0.5 exhibits a partial structural phase transition (tetragonal to hexagonal phase), which reveals either an enhancement of the lattice stiffening or a transformation to a high-pressure phase, and charge carriers are reduced in the...
DOS in the Fermi surface, hence resulting in a decreased $T_c$. For the case of the Fe$_{0.99}$Ni$_{0.01}$Se$_0.5$Te$_0.5$ sample, the decrease of $T_c$ due to the structural transition leads to the reduction of charge carriers in DOS on the Fermi surface. For FeSe and FeSe$_0.5$Te$_0.5$, the observed $T_c$ increase seems to be caused by the pressure-induced changes in the tetragonal phase and the imperfect Fermi surface nesting of corrugated cylinders, which enhanced with increasing $P$, leading to higher values of $T_c$. The hybridization of Fe 3d electrons with chalcogenide 3p/4p states concurrently increases the DOS at the Fermi level [from 1.49 and 1.73 eV$^{-1}$ (@ 0 GPa) to 1.54 eV$^{-1}$ (@ ∼9 GPa) and 1.77 eV$^{-1}$ (@ 2 GPa) for FeSe and FeSe$_0.5$Te$_0.5$, respectively] under $P$, thus improving the SC properties.

Magnetic Properties under Pressure up to ∼1.2 GPa. Figure 10a shows the temperature dependence of magnetization ($M(T)$) at various pressures for the sample Fe$_{0.99}$Mn$_{0.01}$Se$_0.5$Te$_0.5$. At $P = 0$, bulk superconductivity is observed with the $T_c$ onset of 13.5 K (see the inset of Figure 10a). Figure 10b shows two kinds of $T_c$ (onset, offset) with respect to the applied $P$ up to 1.2 GPa. The offset of $T_c$ is almost consistent with the offset $T_c$ of ∼14.1 K in $R(T)$ measurements. The $T_c$ increased monotonically from 13.9 to 19.1 K having a positive $P$ coefficient of $dT_c/dP = +4.6$ K/GPa. The initial positive $P$ coefficient is ∼4.5 K/GPa ($T_c = 11$ K @ 0 GPa), and the highest $T_c$ of 17 K was achieved at 1.2 GPa for Fe$_{0.97}$Co$_{0.03}$Fe$_0.5$Te$_0.5$. A similar nature has been reported for FeSe$_0.5$Te$_0.5$ and FeSe$_0.88$. These same behaviors of the positive $P$ coefficient were observed in Fe-based high-temperature superconductors.

Figure 11a–c shows the isothermal magnetization curves ($M[H]$) at different temperatures of the Fe$_{0.99}$Mn$_{0.01}$Se$_0.5$Te$_0.5$.
sample under 0, 0.5, and 1.2 GPa, respectively. A clear diamagnetic signal can be seen, suggesting a typical type II superconducting nature for all of the temperatures below $T_c$. Figure 11d shows the $M[H]$ curves at 2 K under 0, 0.5, and 1.2 GPa, and it reveals the enhancement of SC. From the $M[H]$ plot, we measured a lower critical field of $H_{c1} = 450$ Oe at 2 K for ambient pressure, and it was found to increase with pressure. For evaluating $H_{c2}(0)$ at zero temperature, we measured $H_{c1}$ for $P$ of 0, 0.5, and 1.2 GPa using the relation $H_{c1}(T) = H_{c1}(0)[1 - (T/T_c)^2]$ (not shown here). Extrapolated $H_{c1}(0)$ values are 518, 563, and 617 Oe, respectively, for 0, 0.5, and 1.2 GPa. Further, the large opening of the $M[H]$ plot until 5 Tesla reveals the high $H_{c2}$ for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ at 2 K for all pressures. The values of $H_{c2}$ were obtained from the field-dependent $R(T)$ for all of the applied pressures as shown in the inset of Figure 12, and it reveals the positive effect of $P$ on the $H_{c2}$ value similar to the Co-doped sample.52 $H_{c2}(0)$ is obtained through the Werthamer–Helfand–Hohenberg (WHH) equation $H_{c2}(0) = -0.693 T_c(\partial H^2/\partial T)_{T=T_c}$. In Figure 12, the red line indicates the extrapolation of the Ginzburg–Landau formula $H_{c2}(T) = H_{c2}(0)(1 - T^2/1 + T^2)$, where $t = T/T_c$ and the blue line corresponds to the WHH equation. Obviously, the calculated $H_{c2}(0)$ values (given in Table 1) exceed the Pauli paramagnetic limit of 1.84$T_c$, suggesting that Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ shows a heavy pinning or exotic nature. The $G–L$ coherence length is calculated from the relation $\xi(0) = [\pi \rho_0 H_{c2}(0)]^{1/2}$, where $\rho_0$ is defined as a flux quantum ($\rho_0 = 2.0678 \times 10^{-15}$ T-m$^2$). The calculated $\xi(0)$ at zero temperature at ambient $P$ is comparable to the value reported for Cr$^{18}$ and Co-doped samples,52 and the obtained values of $\xi(0)$ for all pressures are given in Table 1.

The thermodynamic critical field $H_{c1}(0)$ is calculated by the formula $H_{c1}(0) = (H_{c1}^* H_{c2})^{1/2}$, where $H_{c1}$ and $H_{c2}$ are the lower and upper critical fields at zero temperature. Further, the thermodynamic field and the upper critical field are connected by the relation $H_{c2} = 2^{1/2} k T_c$, where $k$ is the kappa factor, also called the $G–L$ parameter according to the Ginzburg–Landau theory, and the calculated $k$ values are 29.23, 31.11, and 31.7 for 0, 0.5, and 1.2 GPa, respectively. Such a large value ($>1/\sqrt{2}$) indicates that the type II SC is exhibited in Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$. Further, for Co-doped52 (33.6) and Fe-chalcogenide SCs, the $k$ value lies in the same line. The London penetration depth $\lambda(0)$ is obtained through the equation $\lambda(0) = \kappa \xi(0)$, and it is found to be 563 Å for the ambient condition, comparable to 616 Å for the Co-doped52 one, and $\lambda(0)$ values for all of the applied $P$ are given in Table 1.

The critical current density, $J_c$ (A/cm$^2$), is calculated from Bean’s critical state model $J_c = 20 \Delta M/[a(1 - (a/3b))]$,
where $\Delta M$, $a$, and $b$ are the difference in the magnetization, the width, and the thickness of the used sample, respectively.\textsuperscript{53,54}

Field-dependent $J_c(H)$ plots for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ at various temperatures for 0, 0.5, and 1.2 GPa are shown in Figure 13a–c, and d shows the $J_c(t)/J_c(0)$ versus $T/T_c$ under various pressures for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$, in good agreement with $\delta l$ and $\delta T_c$ pinning.

Figure 13. Field-dependent critical current density, $J_c(H)$, for various temperatures 2, 2.5, 3, 4, 5, 6, 8, 10, 11, 12, and 13 K at (a) ambient, (b) 0.5 GPa, and (c) 1.2 GPa pressures of Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$. (d) Field-dependent critical current density, $J_c(H)$, at 2 K under ambient, 0.5, and 1.2 GPa pressures.

Field-dependent $J_c(H)$ plots for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ at various temperatures for 0, 0.5, and 1.2 GPa are shown in Figure 13a–c, and d shows the $J_c(t)/J_c(0)$ versus $T/T_c$ under various pressures for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$, in good agreement with $\delta l$ and $\delta T_c$ pinning.

Figure 14. (a) Double logarithmic plot of $J_c$ versus $(1 - T/T_c)$ at 0 T and (b) for 5 T under various pressures of 0, 0.5, and 1.2 GPa for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$. (c) $J_c(t)/J_c(0)$ versus $T/T_c$ under various pressures for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$, in good agreement with $\delta l$ and $\delta T_c$ pinning.

The observed critical current densities $J_c(0)$ are $\approx 5000$ and $15,000$ A/cm$^2$ at 6 K for FeSe$_{0.5}$Te$_{0.5}$ and Fe$_{0.99}$Co$_{0.01}$Se$_{0.5}$Te$_{0.5}$, respectively.\textsuperscript{27} The irreversible fields ($H_{irr}$) of 0, 0.5, and 1.2 GPa; the high-field $J_c$ is also enhanced from $4 \times 10^4$ to $17 \times 10^4$ A/cm$^2$. The significant enhancement of $J_c$ at high field and high temperature by $P$ leads to the improvement of in-field performance. The highest $J_c$ at 1.2 GPa for 2 K is higher than that of Fe$_{0.9}$Co$_{0.01}$Se$_{0.5}$Te$_{0.5}$ at both low fields and high fields.\textsuperscript{27}
GPa are determined from the $M-H$ loop using the criteria of the zero critical current density, $J_c(0)$, because of depinning of the magnetic flux lines, and it was found that $H_{irr}$ increases with $P$.

**Pinning Mechanism.** From Figure 11d, when compared to the loop at $P = 0$ GPa, $M-H$ loops under $P$ are significantly widened at both low and high fields, suggesting that the widening can be attributed to a huge enhancement of flux pinning under $P$. To differentiate vortex pinning mechanisms in Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$, the Ginzburg–Landau theory is employed. Based on the theory, $J_c$ obeys the power law $J_c \propto (1 - T/T_c)^\delta$, where $\delta > 1.5$ for strong vortex core pinning and $\delta = 1$ for individual, noninteracting vortices. Figure 14a,b shows the double logarithmic $J_c$ versus $(1 - T/T_c)$ plots for various $P$, up to 1.2 GPa. Figure 15 shows the normalized pinning force $F_p$ $(F_p/F_{p_{max}})$ versus normalized field $H/H_{irr}$ for various fixed $P$ values, and we used the Dew–Hughes model for scaling, i.e., $F_p \approx h^q (1 - h)^p$, where $p = 1/2$ and $q = 2$; moreover, $p = 1$ and $q = 2$ describe surface pinning and point pinning, respectively. The experimental results show that the point pinning mechanism exhibits in Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ both at ambient and at $P$. The best fit of the curves is obtained with an $f(h)$ dependence given by $h^{1.35} (1 - h)^{2.7}$ for $P = 1.2$ GPa. The obtained $p = 1.3$ and $q = 2.7$ are similar to the observed values of $p = 1.35$ and $q = 2.7$, and 3.06 and 3.0 in Fe$_{1.04}$Te$_{0.8}$Se$_{0.2}$ and Fe$_{0.99}$Co$_{0.01}$Se$_{0.5}$Te$_{0.5}$ respectively. In the Dew–Hughes model, it should be noted that $h_{max} = (p/(p + q)) = 0.20$ and 0.33 correspond to surface pinning and point core pinning, respectively, for a perfect center of core interaction. In the case of Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ under pressure, $h_{max}$ is 0.32, and it corresponds to small-size normal point pinning.

Further, the matching field, where the defect spacing matches the vortex lattice spacing, gradually increases with increasing pressure as in the case of the Fe$_{0.99}$Co$_{0.01}$Se$_{0.5}$Te$_{0.5}$ sample in which the field-dependent intervortex distance decreases, consequently showing that the defect density increases with increasing pressure. Therefore, more point defects are expected under pressure in Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$.

The maximum $T_c$ of Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ is 22 K, while that of the Fe$_{0.99}$Ni$_{0.01}$Se$_{0.5}$Te$_{0.5}$ sample is 30.5 K up to 3 GPa, and the behavior of $T_c(P)$ data to low temperature also reveals a different behavior. The Mn-doped sample exhibits a low-temperature metallic behavior, $T < 140 K$ ($T > 140$ reveals a semimetal nature), whereas the Ni-doped sample exhibits a low-temperature upturn $T < 100 K$, and at $T > 100 K$, it is metallic. However, the Mn-doped sample is more preferred for practical applications than the Ni-doped sample. Further, structural studies for both systems show a tetragonal structure at ambient pressure and the mixed phase (tetragonal + semimetal nature) is observed in the $P$ region at $\sim 7.5$–14 and $\sim 7.7$–10 GPa for Ni- and Mn-doped samples, respectively. The DFT calculations show a higher electron density in $P4/nmm$ (ambient phase) than in $P6_3/mmc$ (high $P$ phase), where the phase transition initiation for both samples is different (for Mn-doped sample: $5.9$ GPa; for Ni-doped sample: $9.8$ GPa). These high-$P$ studies reveal that the Mn-doped sample reaches the high-pressure phase faster than the Ni-doped sample.

### CONCLUSIONS

In conclusion, for Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$, $T_c$ attains a broad maximum of 22 K at $\sim 3$ GPa and then decreases. $T_c$ is found to vanish at 9 GPa, where the pressure induces a structural phase transition from the tetragonal phase ($P4/nmm$) to the hexagonal phase ($P6_3/mmc$). This structural phase transition is expected to reduce the charge carrier in the density of states at the Fermi surface. The upper critical field is found to exceed the universal Pauli's limiting field, thus suggesting that an unconventional superconductivity mechanism is exhibited by this compound. These results prove that a considerable increase of the $J_c$ value with the application of pressure reveals the enhancement of magnetic flux pinning. Interestingly, $\delta T_c$ pinning is shown as a dominant pinning mechanism at 0 GPa and $\delta T_c$ pinning at 1.2 GPa in Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$. The normalized pinning force shows dense point pinning in this compound both at ambient pressure and under $P$. 

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**Figure 15.** Normalized pinning force $F_p$ versus $H$ at various pressures 0, 0.5, and 1.2 GPa, in good agreement with the dense point pinning mechanism for the high pressure at 2 K.
Experimental Methods. Single crystals of Fe$_{0.99}$Mn$_{0.01}$Se$_{0.5}$Te$_{0.5}$ were grown by a self-flux melt growth method. High-purity (Alfa Aesar, 99.99%) Fe, Se, Te, and Mn powders were weighed, mixed in stoichiometric amounts, and ground thoroughly in an argon-filled glovebox. The procedure is the same as reported by Maheswari et al. The in situ powder X-ray diffraction method was performed under various P values up to 10 GPa at the Xpress beamline of the Elettra synchrotron radiation facility (Trieste, Italy). The wave-length and diameter of the synchrotron radiation used were \(\sim 0.4957 \text{ Å} \) and \(\sim 50 \mu\text{m}\), respectively, and the maximum P of 10 GPa was generated/applied using a gear-driven Boehler-Almax diamond anvil cell (DAC). The DAC is composed of a 500 \(\mu\text{m}\) paired culet. The sample chamber of the DAC was prepared by preindenting the gasket from 200 to \(\sim 110 \mu\text{m}\) thickness, and the center of the indented region was drilled by electrical discharge to a diameter of \(\sim 200 \mu\text{m}\). Fine-ground powders were loaded inside the sample chamber to collect diffraction data to orientation effects. Silicone oil was used as a pressure-transmitting medium (PTM). Silicone oil is one of the common PTMs used in DAC for structural investigations due to its ease in managing the loading. Although hydrostatic conditions are not optimal when using silicone oil, it is found that even subtle structural anomalies can be observed with this PTM. The pressure was calibrated using the shift in the ruby fluorescence techniques. A MAR-345 imaging plate detector was used for collecting the diffractograms. The data were collected from the imaging plate with an angular range between 4 and 30\(^\circ\), and fit2D software was employed to convert data into 2\(\theta\)-intensity plots. General Structure Analysis System (GSAS) software with the Rietveld method was used to refine the structural analysis.

Temperature dependence of electrical resistance (\(R(T)\)) was carried out for both ambient pressure and various hydrostatic pressures (up to \(\sim 3\) GPa) down to 4 K temperature using the standard four-probe method using the clamp-type piston cylinder hybrid hydrostatic pressure cell and the physical property measurement system (PPMS Quantum Design). Daphene oil (#7373) was used as an efficient pressure-transmitting medium (PTM) to maintain the hydrostaticity up to \(\sim 3\) GPa. Bismuth (Bi) structural transition (I-II, II-III) under \(P\) at room temperature was used as a calibrant to estimate the actual pressure exhibits in the P cell. The onset of the superconducting transition temperature was determined using the criteria of inflection point of resistance marked where the two slopes intersect in \(R(T)\) measurements. The temperature dependence of heat capacity measurement from 300 to 4 K under ambient \(P\) was carried out using the physical property measurement system (Quantum), and \(R(T)\) measurements under \(P\) (up to 5 GPa) were also carried out using 1000 \(\mu\text{m}\) culet diamond pairs and DAC at the Muroran Institute of Technology, Muroran Japan. A stainless steel gasket with thickness 300 \(\mu\text{m}\) was preindentated to 100 \(\mu\text{m}\), a miniature drill was used to make \(\phi = 200 \mu\text{m}\) at the center of the indentation, and fine powders of high-purity NaCl were used as PTMs. A small thin crystalline sample (\(\sim 80 \times 20 \times 40 \mu\text{m}^3\)) was placed on the NaCl medium, and the four contact leads were made by a 10 \(\mu\text{m}\) gold foil. The ruby fluorescence approach was used for monitoring the pressure in situ; two 5 \(\mu\text{m}\) ruby chips were placed along with the sample in the pressure cell. Pressure was measured at room temperature using the Ruby R1 line shift. \(R(T)\) measurements up to \(\sim 12\) GPa were done in the cubic anvil press at ISSP, Japan. As a PTM, Daphene oil (#7474) was used to have a good hydrostatic \(P\), and \(P\) calibration was carried out using bismuth (Bi) structural (I-II, II-III) transitions up to 5 GPa at room temperature. Four electrical contact leads were made on the 0.7 \(\times 0.4 \times 0.3 \text{ mm}^3\) sample by a gold wire of \(\phi = 20 \mu\text{m}\) with high-quality silver paste.

Temperature-dependent DC magnetization (\(M(T)\)) was measured in ZFC and FC modes with a magnetic field of 10 Oe parallel to the c-axis of the single crystal in the temperature range from 20 to 2 K at ambient pressure and various hydrostatic \(P\) in a magnetic property measurement system (MPMS Quantum Design) and a hydrostatic piston cylinder pressure cell at ISSP Japan. A mixture of Fluorinert FC #77 and FC #70 with an equal ratio was used as a PTM. The in situ \(P\) in the piston cylinder P cell was estimated from the \(P\) dependence of \(T_c\) of pure Sn. The critical current density, \(J_c\), was determined from the field-dependent magnetization (\(M-H\)) data by the Bean model, \(J_c = 20 \Delta M/[a(1 - (a/3b))]\), where \(\Delta M\), \(a\), and \(b\) are the height difference in the \(M-H\) hysteresis loop, the width, and the length of the sample perpendicular to the applied field, respectively.

Theoretical Method. The projected-augmented plane-wave (PAW) method, as implemented in the Vienna ab initio simulation (VASP) package, was employed to perform DFT calculations. For crystal structure relaxation, the PBE (Perdew, Burke, and Ernzerhof) functional was used, and for further simulations, the exchange-correlation functional with Hubbard parameter correction (GGA + U) was used. The Fe-d and Mn-d states were given as \(U\) values of 5.3 and 6.1 eV (with \(J = 1\) eV), respectively. The conjugate-gradient algorithm with a force convergence threshold of \(10^{-5}\) eV Å\(^{-1}\) was used to estimate ground-state geometries by optimizing stresses and Hellman–Feynman forces. Throughout all relaxations, Brillouin zone integration was carried out with a Gaussian broadening of 0.1 eV, and the various sets of calculations revealed that 1152 k-points in the Brillouin zone for the structure with a 600 eV plane-wave cutoff were adequate to guarantee maximum accuracy in the computed values. For structural optimization, the k-point mesh was generated using the Monkhorst–Pack method with a grid size of \(12 \times 12 \times 8\). To measure the total energy as a function of volume, a comparable density of k-points and energy cutoff was used for all of the structures considered in these calculations. Iteration was carried out for the relaxation of atomic positions until the variation of total energy less than 1 meV/cell was achieved between successive steps.

The volume-dependent total energy was calculated and fitted with the universal equation of state (EOS). The Gibbs free energy vs pressure curves are often used to determine the transition pressures. The Gibbs free energy is obtained by fitting the total energy versus volume curve to the universal EOS equation. The \(P\) dependence of volume can be calculated using the formula \(\Delta V = V_0/[(1 + (B_0/B_0)P)^{1/3}]\), where \(V_0\), \(B_0\), and \(B_0\) denote the equilibrium volume, the bulk modulus, and its derivative with respect to \(P\), respectively. The scan over the \(P\) values yields the change in enthalpy between two sets of data.

AUTHOR INFORMATION

Corresponding Author

Arunagam Sonachalam – Centre for High Pressure Research, School of Physics, Bharathidasan University, Tiruchirappalli 620024 Tamil Nadu, India; orcid.org/0000-0002-2589-...

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