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Hydrostatic pressure: A very effective approach to significantly enhance critical current density in granular iron pnictide superconductors

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Pressure is well known to significantly raise the superconducting transition temperature, T_c , in both iron pnictides and cuprate based superconductors. Little work has been done, however, on how pressure can affect the flux pinning and critical current density in the Fe-based superconductors. Here, we propose to use hydrostatic pressure to significantly enhance flux pinning and T_c in polycrystalline pnictide bulks. We have chosen $\text{Sr}_4\text{V}_2\text{O}_6\text{Fe}_2\text{As}_2$ polycrystalline samples as a case study. We demonstrate that the hydrostatic pressure up to 1.2 GPa can not only significantly increase T_c from 15 K (underdoped) to 22 K, but also significantly enhance the irreversibility field, H_{irr} , by a factor of 4 at 7 K, as well as the critical current density, J_c , by up to 30 times at both low and high fields. It was found that pressure can induce more point defects, which are mainly responsible for the J_c enhancement. Our findings provide an effective method to significantly enhance T_c , J_c , H_{irr} , and the upper critical field, H_{c2} , for other families of Fe-based superconductors in the forms of wires/tapes, films, and single crystal and polycrystalline bulks.

Iron based superconductors have revealed wonderful superconducting properties, including high values of critical temperature (T_c), critical current density (J_c), upper critical field (H_{c2}), and irreversibility field (H_{irr}). They also exhibit low anisotropy and very strong pinning, which gives rise to high J_c ($\sim 10^6$ A/cm²) in both single crystals and thin films at both low and high fields^{1–10}. The J_c and its field dependence in polycrystalline bulks and tapes/wires, however, are still lower than what is required for practical applications. Enhancement of J_c or flux pinning using various approaches has always been a main focus of research with a view to large current and high field applications. So far, three main methods have been used to increase the J_c in cuprates, MgB_2 , and iron based superconductors: 1) texturing processes to reduce the mismatch angle between adjacent grains and thus overcome the weak-link problem in layer-structured superconductors; 2) introducing point pinning centres by chemical doping and 3) high energy ion implantation or irradiation to introduce point defect pinning centres. J_c values achieved by the irradiation method have reached as high as 10^6 – 10^7 A/cm² for both low and high fields in single crystals and thin films^{11–13}. This method is not ideal, however, for J_c enhancement in polycrystalline pnictide superconductors.

As is well known, the weak-link issue is the predominant factor causing low J_c , especially at high fields in pnictide polycrystalline samples, which must be overcome. In order to improve the J_c and its field dependence in granular superconductors, the following prerequisites should be met: i) strong grain connectivity; ii) introduction of more point defects inside grains; and iii) T_c enhancement, which can increase the effective superconducting volume as well as H_{irr} and H_{c2} .

We have taken into account that the following facts relating to flux pinning mechanisms must be addressed before an effective method is introduced for polycrystalline pnictide superconductors. The coherence length is very short ($\xi \approx$ a few nm), so elimination of weakly linked grain boundaries is important to achieve high J_c ¹⁴. The nature of the pinning mechanism plays a vital role in J_c field dependence. It is noteworthy that a high pinning force can boost pinning strength and, in turn, leads to higher values of J_c . The ideal size of defects for pinning should be comparable to the coherence length¹⁵. Therefore, point defect pinning is more favourable than surface pinning, as



its pinning force is larger than for surface pinning at high field, according to the Dew-Hughes model¹⁶. Therefore, it is very desirable to induce more point defects in superconductors. Although chemical doping and high energy particle irradiation can effectively induce point defects and enhance J_c in high fields, T_c and low field J_c deteriorate greatly for various types of superconductors. Therefore, the ideal approach should be the one which can induce more point defects, with increased (or at least at no cost of) superconducting volume and T_c , as well as strongly linked grain boundaries.

Hydrostatic pressure has been revealed to have a positive effect on T_c in cuprate and pnictide superconductors. For instance, high pressure of 150 kbars can raise T_c of Hg-1223 significantly from 135 K to a record high 153 K¹⁷. The T_c of hole doped (NdCeSr)CuO₄ was increased from 24 to 33 K at 3 GPa by changing the apical Cu-O distance¹⁸. The enhancement of T_c for YBCO is more than 10 K at 2 GPa¹⁹. Excitingly, pressure also shows positive effects on T_c for various pnictide superconductors. Pressure can result in improvement of T_c from 28 to 43 K at 4 GPa for LaOFeAs²⁰. For Co doped NaFeAs, the maximum T_c can reach as high as 31 K from 16 K at 2.5 GPa²¹. Pressure can also enhance the T_c of La doped Ba-122 epitaxial films up to 30.3 K from 22.5 K, due to the reduction of electron scattering and increased carrier density caused by lattice shrinkage²². A huge enhancement of T_c from 13 to 27 K at 1.48 GPa was observed for FeSe, and it reached the high value of 37 K at 7 GPa^{23,24}.

Beside the above-mentioned significant pressure effects on T_c enhancement, pressure can have more advantages that are relevant to the flux pinning compared to other methods. 1) It always reduces the lattice parameters and causes the shrinkage of unit cells, giving rise to the reduction of anisotropy. 2) Grain connectivity improvement should also be expected, as pressure can compress both grains and grain boundaries. 3) The existence or formation of point defects can be more favourable under pressure, since it is well known that the formation energy of point defects decreases with increasing pressure^{25–27}. 4) Pressure can cause low-angle grain boundaries to migrate in polycrystalline bulk samples, resulting in the emergence of giant grains, sacrificing surface pinning thereafter. Hence, a higher ratio of point pinning centres to surface pinning centres is expected due to the formation energy and migration of grain boundaries under pressure. 5) The significant enhancement of T_c , as above-mentioned, means that superconducting volumes should be increased greatly below or above the T_c without pressure. Moreover, the H_{c2} , H_{irr} ,

and J_c have to be enhanced along with the T_c enhancement. These are the motivations of our present study on the pressure effects on flux pinning and J_c enhancement in polycrystalline pnictide bulks. We anticipated that hydrostatic pressure would increase the superconducting volume, H_{irr} , and H_{c2} due to T_c enhancement, increase the point defects, improve grain connectivity, and reduce the anisotropy in pnictide polycrystalline bulk samples.

There is some evidence for J_c enhancement under pressure in YBa₂Cu₃O_{7-x} (YBCO) single crystal, which emphasizes the pressure effects on transport J_c for different angle grain boundaries. A recent report also shows enhanced J_c in a pnictide single crystal which is free of grain boundaries^{28–30}. As mentioned earlier, polycrystalline superconducting materials are commonly used in practical applications, as they are easy to fabricate at low cost as compared to single crystals/thin films. Their superconducting performance is hindered by grain boundaries, however, due to granularity. Therefore, it is more important to use an efficient approach to enhance the J_c in polycrystalline bulk samples. In this study, we chose a polycrystalline Sr₄V₂O₆Fe₂As₂ sample to demonstrate the significant effects of the hydrostatic pressure on flux pinning and the significant enhancement of J_c and T_c in this granular sample. It has been reported that the T_c for this compound can range from 15–30 K, depending on fabrication process and carrier concentration³¹. Generally, T_c under pressure remains nearly constant (or little increase) for optimal doped superconductors and decreases linearly in the overdoped range. Under doped superconductors under pressure have dome-like plots for T_c vs. pressure, so we chose a Sr₄V₂O₆Fe₂As₂ sample with the low T_c of 15 K for the proposed pressure effect investigation to ensure a clear pressure effect on T_c ³². Our results show that pressure can enhance the J_c by more than 30 times at 6 K and high fields in polycrystalline Sr₄V₂O₆Fe₂As₂, along with T_c enhancement from 15 to 22 K at 1.2 GPa and H_{irr} enhancement by a factor of 4. Our analysis shows that pressure induced point defects inside the grains are mainly responsible for the flux pinning enhancement.

Results and Discussion

Figure 1 shows the temperature dependence of the zero-field-cooled (ZFC) and field-cooled (FC) moments for Sr₄V₂O₆Fe₂As₂ at different pressures. Pressure causes little change to the field-cooled branch, indicating that strong pinning is retained under pressure. The T_c without pressure is about 15 K, very similar to that of underdoped samples reported for Sr₄V₂O₆Fe₂As₂ bulks³¹. Pressure enhances T_c

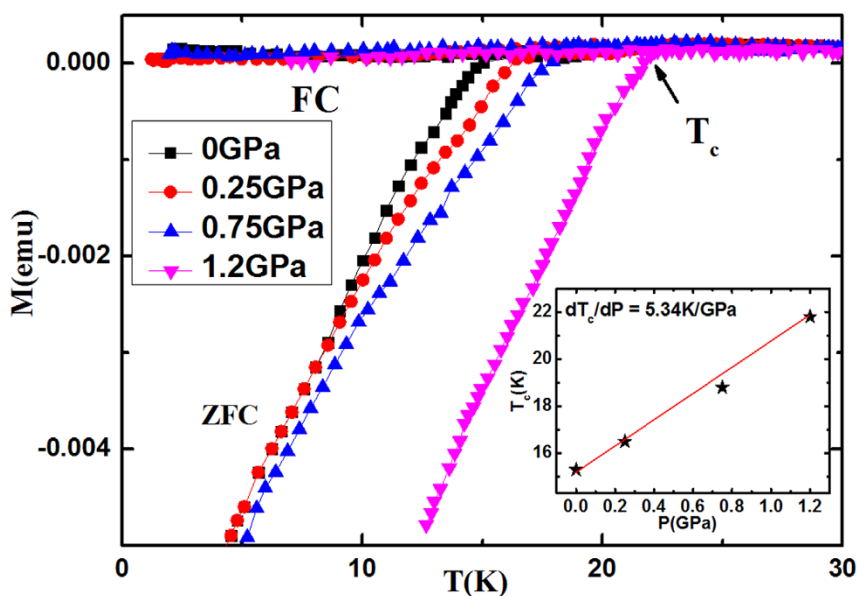


Figure 1 | Temperature dependence of ZFC and FC moments at different pressures for Sr₄V₂O₆Fe₂As₂. The inset shows the pressure dependence of T_c .

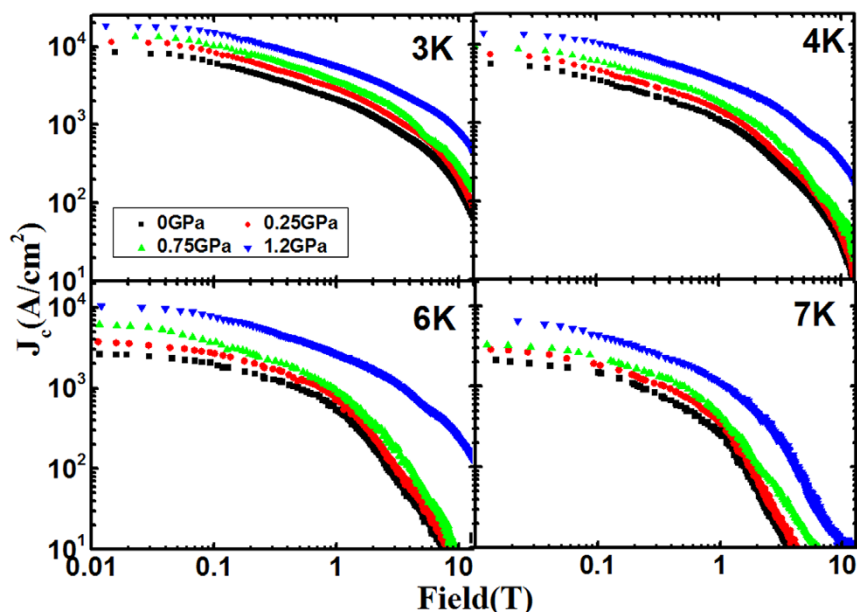


Figure 2 | Field dependence of J_c under different pressures at 3, 4, 6, and 7 K.

linearly from 15.3 K for $P = 0$ GPa to 22 K for $P = 1.2$ GPa, with the pressure coefficient, $dT_c/dP = 5.34$ K/GPa.

The M - H curves measured under different pressures indicate that the moment increases with increasing pressure. The field dependence of J_c at different temperatures obtained from the M - H curves by using Bean's model under different pressures is shown in a double-logarithmic plot [i.e. Figure 2]. The remarkable effect of pressure towards the enhancement of J_c can be clearly seen. For $P = 1.2$ GPa, the J_c is significantly enhanced by more than one order of magnitude at high fields at 4 and 6 K, respectively, as shown in Figure 3.

The J_c at 6 K as a function of pressure at different fields is plotted in Figure 4. The solid lines in Figure 4 show linear fits to the data, which give the slopes (i.e. $d(\ln J_c)/dP$) of 1.09, 1.69, and 2.30 GPa^{-1} at 0, 2, and 4 T, respectively, indicating that the effects of pressure towards the enhancement of the J_c are more significant at high fields.

We also found that the H_{irr} of $\text{Sr}_4\text{V}_2\text{O}_6\text{Fe}_2\text{As}_2$ is greatly increased by pressure. The H_{irr} is defined as a field where J_c reaches as low as 10 A/cm^2 in J_c vs field curves for different pressures and temperatures. As shown in Figure 5, the H_{irr} increases gradually with pressure and rises to 13 T from 3.5 T at 7 K. The J_c vs. reduced temperature $(1-T/T_c)$ at zero field and different pressures is plotted in Figure 6, which shows a rough scaling behaviour as $J_c \propto (1-T/T_c)^\beta$ at different pressures. The slope of the fitting line, β , depends on the magnetic field. The exponent β (i.e. slope of the fitting line) is found to be 2.54, 2.73, 2.96, and 3.13 at 0, 0.25, 0.75, and 1.2 GPa, respectively. According to Ginzburg-Landau theory, the exponent " β " is used to identify different vortex pinning mechanisms at specific magnetic fields. It was found that $\beta = 1$ for non-interacting vortices, while $\beta \geq 1.5$ indicates the core pinning mechanism³³. The different values of β (i.e. 1.7, 2, and 2.5) were also reported for YBCO films which show

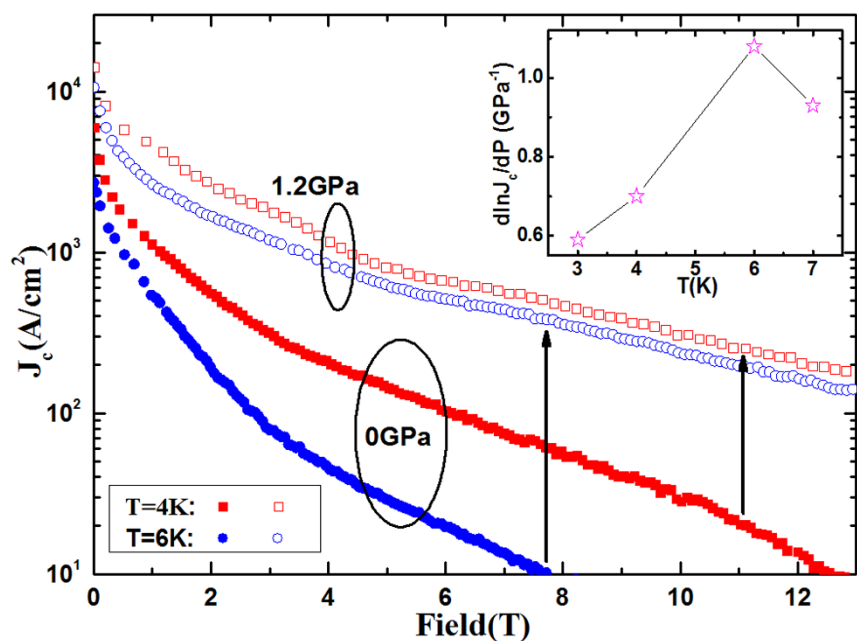


Figure 3 | Comparison of J_c at 0 and 1.2 GPa at 4 and 6 K. The inset shows $d(\ln J_c)/dP$ versus temperature, indicating enhancement of J_c at a rate of 1.08 GPa^{-1} at zero field.

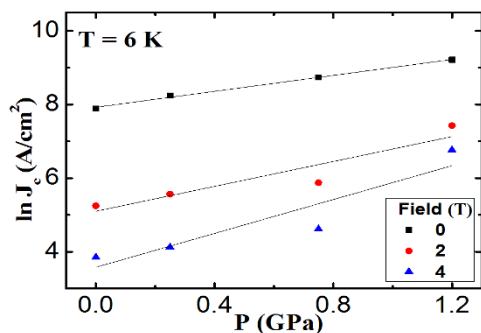


Figure 4 | Pressure dependence of J_c (logarithmic scale) at 0, 2, and 4 T at the temperature of 6 K.

the functioning of different core pinning mechanisms^{34,35}. In addition, the exponent β values that we obtained are higher at higher pressures in our sample, indicating stronger improvement of J_c with temperature at high pressures.

For polycrystalline samples, high pressure can modify the grain boundaries through reducing the tunnelling barrier width and changing the tunnelling barrier height. The Wentzel-Kramers-Brillouin (WKB) approximation applied to a potential barrier gives the following simple expressions³⁶:

$$J_c = J_{c0} \exp(-2kW) \quad (1)$$

Where W is the barrier width, $k = (2 \text{ mE})^{1/2}/\hbar$ is the decay constant, which depends on the barrier height L , \hbar is the Planck constant, and J_{c0} is the critical current density for samples with no grain boundaries. The relative pressure dependence of J_c can be obtained from Eq. (1) as:

$$\begin{aligned} \frac{d \ln J_c}{dP} &= \frac{d \ln J_{c0}}{dP} - \left[\left(\frac{d \ln W}{dP} \right) \ln \left(\frac{J_{c0}}{J_c} \right) \right] - \frac{1}{2} \left[\left(\frac{d \ln L}{dP} \right) \ln \left(\frac{J_{c0}}{J_c} \right) \right] \\ &= \frac{d \ln J_{c0}}{dP} + \kappa_{GB} \ln \left(\frac{J_{c0}}{J_c} \right) + \frac{1}{2} \kappa_L \ln \left(\frac{J_{c0}}{J_c} \right) \end{aligned} \quad (2)$$

Where the compressibility in the width and height of the grain boundary are defined by $\kappa_{GB} = -d \ln W/dP$ and $\kappa_L = -d \ln L/dP$, respectively. To estimate their contributions to the second and the third terms of Eq. (2) for J_c enhancement, we assume to a first approximation that κ_{GB} and κ_L are roughly comparable to the average linear compressibility values $\kappa_a = -d \ln a/dP$ ($\kappa_{GB} \approx \kappa_a$) and $\kappa_c = -d \ln c/dP$ ($\kappa_L \approx \kappa_c$) of $\text{Sr}_4\text{V}_2\text{O}_6\text{Fe}_2\text{As}_2$ in the FeAs plane, where a and c are the in-plane and out-of-plane lattice parameters, respectively. We

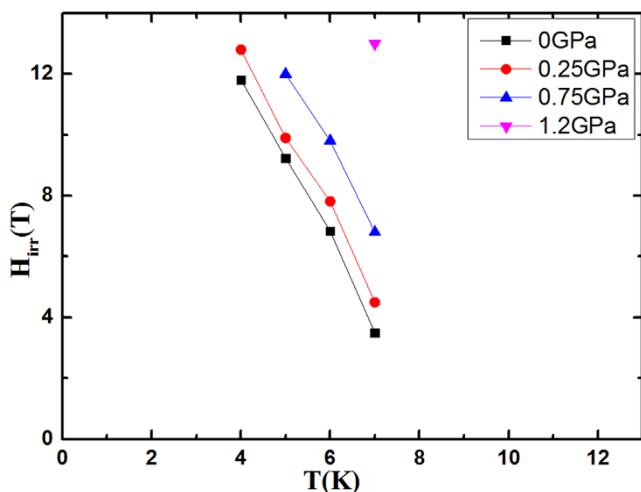


Figure 5 | H_{irr} vs. T for different pressures.

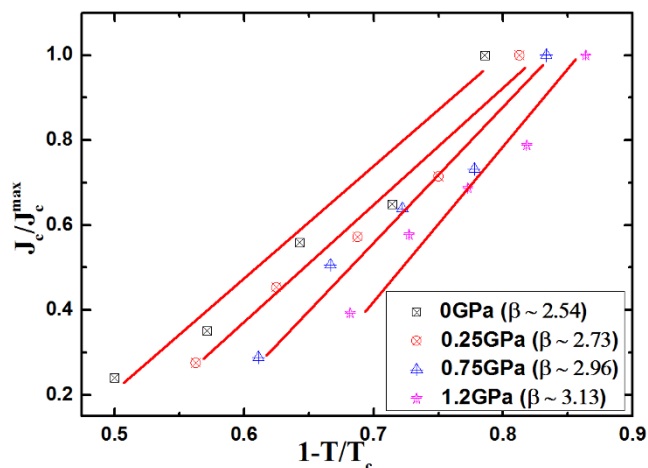


Figure 6 | Logarithmic plot of J_c as a function of reduced temperature at different pressures and fields.

assume to a first approximation that $\kappa_a = -d \ln a/dP = -0.029 \text{ GPa}^{-1}$, $\kappa_c = -d \ln c/dP = -0.065 \text{ GPa}^{-1}$ ²⁴, and the in-plane $J_{c0} \cong 10^5 \text{ A/cm}^2$ for a sample with no grain boundaries (single crystal)³⁷. Setting $J_c \cong 2 \times 10^3 \text{ A/cm}^2$ at the temperature of 6 K and ambient pressure, we find that $(-d \ln W/dP) \ln(J_{c0}/J_c) \approx 0.11$ and $-0.5(d \ln L/dP) \ln(J_{c0}/J_c) \approx 0.13$, with both values adding up to 75% less than the above experimental value $d \ln J_c/dP = 1.08 \text{ GPa}^{-1}$. This result suggests that the origin of the significant increase in $J_c(T)$ under pressure does not arise from the compression of the grain boundaries. Therefore, Eq. (2) suggests that the main reason for the rapid increase of J_c with pressure is through point defects induced under pressure, i.e., $d \ln J_{c0}/dP$ is responsible for approximately 75% of the total increase in the J_c with pressure.

In order to further understand the J_c enhancement under pressure, the pinning force $F_p = B \times J_c$ is calculated, and the scaling behaviour for the normalized pinning force $f_p = F_p/F_{p,max}$, is analysed for $h = H/H_{irr}$. The results are shown in Figure 7 at 4 and 6 K under 0, 0.75, and 1.2 GPa. For the scaling, we can use the Dew-Hughes formula, i.e. $f_p(h) = Ah^p(1-h)^q$, where p and q are parameters describing the pinning mechanism¹⁶. In this model, $p = 1/2$ and $q = 2$ describes surface pinning while $p = 1$ and $q = 2$ describes point pinning, as was predicted by Kramer³⁸. At ambient pressure in the temperature range of 3–7 K, the best fits of the curves are obtained with $p = 0.51 \pm 0.03$, $q = 1.86 \pm 0.03$, which suggests that surface pinning is the dominant pinning mechanism in our sample. At 1.2 GPa, the best obtained values for p and q were 0.9 ± 0.1 and 2 ± 0.1 , respectively, within the studied temperature range. This means that the dominant pinning mechanism is normal core point pinning for high pressures. Therefore, our results show that the pressure has induced a clear transformation from surface to point pinning.

Moreover, it is noteworthy that pressure can induce a reduction in anisotropy. The anisotropy is defined as $r = \zeta_{ab}/\zeta_c$ where ζ_{ab} is a coherence length along ab plane and ζ_c along c plane. At high temperatures, the pressure dependence of T_c , unit cell volume (V), and anisotropy (r) are interconnected through the following relation³⁹,

$$\frac{-\Delta T_c}{T_c} = \frac{\Delta V(T_c)}{V(T_c)} + F(\gamma) \quad (3)$$

Where $F(\gamma) = [\gamma(P) - \gamma(0)]/\gamma(0)$. Although, no report for bulk modulus of $\text{Sr}_4\text{V}_2\text{O}_6\text{Fe}_2\text{As}_2$ is yet available, we have tentatively used the bulk modulus ($K = 62 \text{ GPa}$) of a similar superconductor i.e., SrFe_2As_2 , to estimate $\Delta V(T_c)/V(T_c)$, which is found to be ~ -0.016 at $\Delta P = 1 \text{ GPa}$, as it can be related to the bulk modulus as $\Delta V/V = -\Delta P/K$ ⁴⁰. Our experimental results yield a value of 0.486 for $\Delta T_c/T_c = [T_c(P) - T_c(0)]/T_c(0)$. By using these results, we can obtain

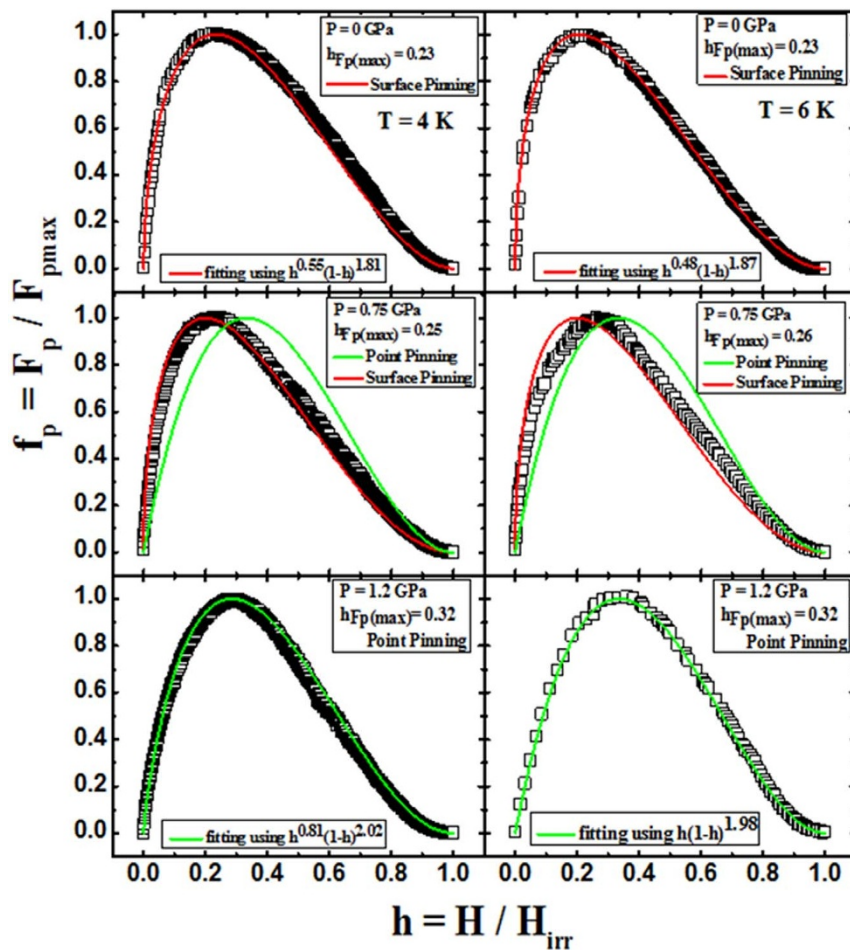


Figure 7 | Plots of f_p vs. H/H_{irr} at different pressures (0, 0.75, and 1.2 GPa) for 4 (left) and 6 K (right) temperature curves. The experimental data is fitted through the Dew-Hughes model, and the parameters are shown.

$\gamma(P) \approx 0.53\gamma(0)$. Thus, we can conclude that the anisotropy has been reduced by almost half at high temperature by pressure. The decrease in the unit cell parameters suppresses its volume, leading to an increase in the Fermi vector $k_F = (3\pi^2N/V)^{1/3}$, where N is the total number of electrons in the system. The increase in the Fermi vector promotes enhancement of the coherence length along the c -axis ($\xi_c = \hbar^2 K_F / \pi m \Delta$ where Δ is the uniform energy gap which, in turn, leads to the suppression of anisotropy.

In summary, hydrostatic pressure is a very effective means to significantly enhance T_c , J_c , H_{irr} , and flux pinning in the granular pnictide superconductor $Sr_4V_2O_6Fe_2As_2$. We demonstrate that the hydrostatic pressure can significantly increase T_c from 15 to 22 K, as well as increasing J_c by up to 30 times at both low and high field and increasing H_{irr} by a factor of 4 at $P=1.2$ GPa. Pressure introduces more point defects inside grains, so that it is mainly responsible for J_c enhancement. In addition, we found that the transformation from surface pinning to point pinning induced by pressure was accompanied by a reduction of anisotropy at high temperatures. Our findings provide an effective method to significantly enhance T_c , J_c , H_{irr} , and H_{c2} for other families of Fe-based superconductors in the forms of wires/tapes, films, and single and polycrystalline bulks.

Methods

For the polycrystalline $Sr_4V_2O_6Fe_2As_2$ sample synthesis, the Fe (Alfa Aesar, 99.2%) and As (Alfa Aesar, 99%) chips were sealed in evacuated quartz tube and heat treated for 12 hours at 700°C. Later, the stoichiometric amounts of V_2O_5 (Aldrich, 99.6%) + $\frac{1}{2} \times SrO_2$ (Aldrich) + $7/2 \times Sr$ (Alfa Aesar, 99%) + $2 \times FeAs$ were weighed, mixed, grounded thoroughly and palletized in rectangular form in a glove box in a high purity Ar atmosphere. The pellet was further wrapped in tantalum foil and then sealed in an evacuated (10–5 torr) quartz tube and put for heat treatments at 750 and

1150°C in a single step for 12 and 36 hours respectively. Finally, the quartz ampoule was allowed to cool naturally to room temperature.

The temperature dependence of the magnetic moments and the M-H loops at different temperatures and pressures were performed on Quantum Design Physical Property Measurement System (QD PPMS 14T) by using Vibrating Sample Magnetometer (VSM). We have used HMD High Pressure cell and Daphne 7373 oil as a pressure transmitting medium to apply hydrostatic pressure on a sample. The critical current density was calculated by using the Bean approximation.

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Author contributions

X.L.W. conceived the pressure effects and designed the experiments. B.S. performed high pressure measurements and collected the data. C.S. and O.N.S. provided samples for this work. X.L.W., B.S., S.X.D., and R.G. contributed to the discussions and analysis of the data. X.L.W., B.S. and R.G. co-wrote the paper.

Additional information

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