Enhanced critical current density in the pressure-induced magnetic state of the high-temperature superconductor FeSe

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We investigate the relation of the critical current density \(J_c\) and the remarkably increased superconducting transition temperature \(T_c\) for the FeSe single crystals under pressures up to 2.43 GPa, where the \(T_c\) is increased by \(-8\) K/GPa. The critical current density corresponding to the free flux flow is monotonically enhanced by pressure which is due to the increase in \(T_c\), whereas the depinning critical current density at which the vortex starts to move is more influenced by the pressure-induced magnetic state compared to the increase of \(T_c\). Unlike other high-\(T_c\) superconductors, FeSe is not magnetic, but superconducting at ambient pressure. Above a critical pressure where magnetic state is induced and coexists with superconductivity, the depinning \(J_c\) abruptly increases even though the increase of the zero-resistivity \(T_c\) is negligible, directly indicating that the flux pinning property compared to the \(T_c\) enhancement is a more crucial factor for an achievement of a large \(J_c\). In addition, the sharp increase in \(J_c\) in the coexisting superconducting phase of FeSe demonstrates that vortices can be effectively trapped by the competing antiferromagnetic order, even though its antagonistic nature against superconductivity is well documented. These results provide new guidance toward technological applications of high-temperature superconductors.

The technological application of superconductors hinges on how to preserve a zero-resistance state at high temperature while maintaining large electrical currents. The discovery of copper-based high-temperature superconductors (HTSs) brought great excitement not only because of its unconventional superconducting nature, but also because of its high superconducting transition temperature \(T_c\), which was expected to open the door for revolutionary applications at temperatures higher than liquid nitrogen temperature (=77 K) (refs 1–3). A key issue for practical applications of superconductors is the necessity to increase the value of the depinning critical current density \(J_c\), at which magnetic flux lines (or vortices) start to flow and energy dissipation occurs. For decades, several approaches effectively enhanced the \(J_c\) of HTSs by introducing and/or manipulating the extrinsic defects that suppress superconductivity4,5. Because the

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flux lines have a normal state within the core, they tend to be pinned at defects where superconductivity is suppressed, i.e., extrinsic pinning effects.

Another possible approach to improve the $I_c$ is associated with an intrinsic property of materials, e.g., a coexisting order with superconductivity as an intrinsic pinning source. Recently, it has been proposed that magnetism may be conducive to holding the vortex, which leads to the enhancement of the $I_c$ (refs 6–11). Several high-$T_c$ superconductors, such as La$_{2−x}$Sr$_x$CuO$_4$ and Ba(Fe$_{1−y}$Co$_y$)$_2$As$_2$, are candidate materials for the intrinsic pinning because superconductivity occurs in the vicinity of an antiferromagnetically ordered state$^{6–8}$. Superconductivity in those materials, however, requires a chemical substitution that inherently induces defects or site disorder, intertwining the effects of impurities and intrinsic pinning on $I_c$. In addition, it is still controversial if the magnetic order arises from macroscopically phase separated domains or from an intrinsic coexisting phase on a microscopic level. Therefore, in order to clarify the role of the intrinsic pinning on $I_c$, it is crucial to perform a systematic study on a high-$T_c$ compound that is superconducting in stoichiometric form and tunable between superconducting and magnetic ground states by non-thermal control parameters.

The binary high-$T_c$ superconductor FeSe is a promising candidate to probe the effects of the intrinsic pinning and the $T_c$ on the $I_c$, because superconductivity which appears at $\sim$10 K without introducing a hole or electron in the parent compound is greatly tunable up to 37 K by application of pressure$^{10,11}$. In addition, an emergence of magnetic state at pressure $\sim$0.8 GPa makes it a more interesting material in its basic properties and application issues$^{10,11}$. A $T_c$ above 100 K in FeSe monolayer shows its promising potential for the possibility of application$^{12}$. In the following, we report the evolution of the critical current density ($J_c$) of FeSe single crystals as a function of pressure in connection with the increase of $T_c$.

The current-voltage ($I$–$V$) characteristic curves as well as temperature dependences of the electrical resistivity show a sharp contrast across the critical pressure ($P_c = 0.8$ GPa) above which $\mu$SR measurements reported a pressure-induced AFM state that coexists with superconductivity$^{14,15}$. There are a few interesting behaviours. First, the superconducting (SC) transition is sharp at low pressures, but becomes broader in the coexisting SC state for $P > P_c$. Secondly, temperature dependence of the critical current density follows the prediction by the $\delta T_c$-pinning at low pressures ($P < P_c$), while the $\delta \rho$-pinning becomes more effective at higher pressures. Thirdly, amplitude of $I_c$ is strongly enhanced in the coexisting state. The fact that physical pressure does not induce extra disorder suggests that the enhancement in $I_c$ as well as the change in the pinning mechanism in the coexisting phase arises from the antiferromagnetically ordered state.

**Results**

Figure 1(a,b) representatively shows the in-plane electrical resistivity ($\rho_{ab}$) of FeSe as a function of temperature for several pressures. For clarity, $\rho_{ab}(T)$ for different pressures was rigidly shifted upwards. At ambient pressure, a change in the slope of $\rho_{ab}$ occurs at 75 K due to the tetragonal to orthorhombic structural phase transition. Unlike other iron-based superconductors, this structural transition is not accompanied by a magnetic phase transition. The structural transition temperature ($T_s$), which is assigned as a dip in $d\rho_{ab}/dT$, progressively decreases with increasing pressure at a rate of $\sim$36.7 K/GPa and is not observable for pressures above 1.3 GPa where $T_s$ becomes equal to the superconducting transition temperature $T_c$, as shown in Fig. 1(d). With further increasing pressure, an additional feature appears in the normal state as a dip or a slope change in $d\rho_{ab}/dT$, as shown in Fig. 1(e). In contrast to $T_s$, this new characteristic temperature linearly increases with pressure and is nicely overlaid with the $T_c$ determined from $\mu$SR results$^{14}$, showing that the resistivity anomaly arises from the paramagnetic to antiferromagnetic phase transition, as described in Fig. 1(f).

Figure 1(c) presents that the temperature for the onset of the superconducting transition ($T_{c\text{on}}$) gradually increases with increasing pressure at a rate of 8 K/GPa. Also, the transition width $\Delta T_c$, which was defined as the difference between the 90 and 10% resistivity values of the normal state at $T_{c\text{on}}$, decreases with increasing pressure at low pressures because of the enhanced superconductivity under pressure. At pressures $P > 0.8$ GPa, where superconductivity coexists with a magnetically ordered state on a microscopic scale$^{14,13}$, $\Delta T_c$ becomes broader even though $T_{c\text{on}}$ increases with increasing pressure. The dichotomy between $T_{c\text{on}}$ and $\Delta T_c$ in the coexisting phase suggests that the pressure-induced antiferromagnetic phase acts as an additional source for breaking Cooper pairs.

Correlation between the anomalous broadening in the $\Delta T_c$ and the magnetic phase is further supported by a qualitative difference in the current-voltage ($I$–$V$) curves of FeSe across the critical pressure $P_c$. As shown in Fig. 2(a–d), the voltage curve sharply decreases with decreasing current at 0.41 GPa, i.e., the pressure where superconductivity itself only exists. In the coexisting phase ($P > P_c$), on the other hand, the voltage curve develops a knee with decreasing current. Figure 2(d) summarizes pressure evolution of the transition broadening in the $I$–$V$ curve at 7 K. These anomalous broadenings in the $I$–$V$ curves are also considered due to the pressure-induced antiferromagnetic state.

**Discussion**

Two characteristic critical currents, $I_c$ and $I_{\delta \rho}$ from the $I$–$V$ curves, are marked by the two arrows in Fig. 2(d). The depinning critical current ($I_c$) was obtained from the 1 $\mu$V criterion where the vortices start to move and the free-flux-flow (FFF) current ($I_{\delta \rho}$) was obtained from the point where vortices are no longer affected by the pinning sites and therefore move freely$^{10,17}$. Figure 3(a,b) describes the
temperature dependence of the critical current densities $J_\parallel$ and $J_\perp$ estimated from $I_{\parallel}$ and $I_{\perp}$, respectively. Both $J_\parallel$ and $J_\perp$ were significantly improved with increasing pressure. The FFF current density $J_\parallel(T_c,0)$, which is concerned with thermally activated flux flow with increasing $T_c$, is best explained by the empirical relation $J_\parallel(T_c,0) \sim [1 - (T_c/T_{c,0})^n]^2$, with $n = 2.6 \pm 0.2$ indicated by solid lines in Fig. 3(a). The curves all collapse onto a single curve, as shown in Fig. 3(c), which cannot be explained by the depairing current density $J_d(\rho)$, nor by the Joule heating $\Delta T \propto I^2$ which is caused by the contact resistance (dotted line). Rather, they collapse onto the curve expected from the $\delta T_c$-pinning mechanism (solid line), $J_\parallel(t) \sim (1 - t^2)^3/(1 + t^2)^{1/2}$, suggesting that the temperature dependence of the FFF current density is primarily determined by spatial variations in $T_c$ (refs 20,21).

Figure 3(b) shows the pressure evolution of the depinning critical current density $I_c$, usually called the critical current density, as a function of temperature. At 1.8 K, the lowest temperature measured, $I_c$ increases in commensurate with $T_{c,0}$ with increasing pressure, while $I_c$ in the coexisting phase is strongly enhanced from 1.89 kA/cm$^2$ at 0.41 GPa (red circles) to 3.24 kA/cm$^2$ at 1.22 GPa (blue triangles). Here, we used the zero-resistivity SC transition temperature $T_{c,0}$ with applied current density $I_c \sim 1$ A/cm$^2$. Resistance is not zero any more above the $I_c$ where vortices start to move, which is significantly influenced on the pinning properties of samples, such as pinning strength, density of pinning sites, and so on. Therefore, the $I_c$ comparison by the $T_{c,0}$ is reasonable than the comparison by the $T_{c,0}$. Considering that the increase in $T_{c,0}$ is negligible at 1.2 GPa, the anomalous jump in $I_c$ as shown in Fig. S1 in SI, deviates from the trend in $I_c$ as a function of $T_{c,0}$, underlining that an additional source of pinning is indeed required to explain this anomaly. The possibility of the enhancement in $I_c$ due to improved grain boundary connectivity has been reported in some high-$T_c$ cuprate superconductors or in the iron-based polycrystalline superconductor Sr$_2$V$_2$O$_7$Fe$_2$As$_2$ (ref. 24). Because the studied FeSe samples are single crystalline specimens, however, the lack of a weak-link behaviour in the field dependence of $I_c$ rules out the possibility of grain boundary as the additional pinning source (see Fig. S2 in SI). Rather, the simultaneous enhancement in $I_c$ and appearance of antiferromagnetism indicate that the...
Pressure-induced magnetic state leads to an inhomogeneous SC phase and is conducive to the trapping of magnetic flux lines. With further increase in pressure, both $J_c$ and $T_{c,0}$ increase. The additional flux pinning caused by the antiferromagnetic (AFM) order in the FeSe is reflected in the different temperature dependence of $J_c$ across the critical pressure $P_c$. As shown in Fig. 3(d), the normalized self-field critical current density $J_c(t_0)$ as a function of the reduced temperature ($t_0 = T/T_{c,0}$) is well described by the $\delta T_c$-pinning mechanism (solid line) for $P < P_c$, where the $T_c$ fluctuates due to defects, such as Se deficiencies and point defects, which are the main sources for trapping the vortices. For $P \geq P_c$, however, $J_c(t_0)$ shows a completely different behaviour: the curvature of $J_c$ near $T_{c,0}$ is positive, while it is negative at lower pressures. Also with increasing pressure, $J_c$ deviates further away from the $\delta T_c$-pinning and at 2.43 GPa becomes close to the curve predicted by $\delta l$-pinning (dashed line), $J_c(t) \propto (1-t^2)^{3/2}(1+t)^{-1/2}$, suggesting that spatial fluctuations in the mean free path ($l$) of the charge carrier becomes important for flux pinning at high pressures. As shown in Fig. S3 in SI, the pressure-induced crossover in $J_c(T)$ is almost independent of the magnetic field, indicating that the vortex pinning within the AFM phase is robust against variations in the magnetic field strength.

A similar crossover from $\delta T_c$-pinning to $\delta l$-pinning has been reported in MgB$_2$ when additional pinning sources, such as grain boundaries or inclusions of nanoparticles by chemical doping, were introduced or hydrostatic pressure was applied. In the present study, a broadening of superconducting transition with the pressure-induced AFM state is important for the crossover. A possibility of enhanced mean free path ($l \propto 1/\xi$) fluctuations due to the competition between superconducting and AFM order parameters and change in the superconducting coherence length ($\xi$) with pressure may be closely related to the crossover because the disorder parameter that characterizes the collective vortex pinning properties is proportional to $\xi$ and to $1/\xi^2$ for $\delta T_c$- and $\delta l$-pinning, respectively. As shown in Fig. S4 in SI, the values of the upper critical field $H_{c2}(0)$ increase with applied pressure, indicating that the change in $\xi$ may be of some relevance to the crossover.

Figure 4(a) shows a contour plot of the free-flux-flow current density ($J_f$) for FeSe as a function of temperature and pressure at zero field, where the colours represent different values of $J_f$. Also plotted are the structural and the magnetic phase boundaries that are obtained from the electrical resistivity measurements; these boundaries are consistent with those reported in previous works. The contour of $J_f$ monotonically increases with an increase in $T_c$ by pressure, while $J_c$ deviates from the monotonic pressure evolution of $J_f$. Instead, the contour of $J_f$ reflects the appearance of the pressure-induced AFM
phase, as shown in Fig. 4(b). The \( J_c \) as well as the \( T_{c,0} \) gradually increases with increasing pressure, however near the critical pressure where AFM phase is induced, \( J_c \) shows a high increase compared to \( T_{c,0} \), as mentioned in Fig. 3(b). We note that \( J_c \) shows a dome shape centred around 2.1 GPa, the projected critical pressure where the tetragonal to orthorhombic structural phase transition temperature is extrapolated to \( T = 0 \) K inside the dome of superconductivity. A Possibility of flux pinning by structure transition had been reported in the superconducting A15 compounds such as \( V_3Si \) (refs 28, 29), and further work is in progress to better understand the role of structural fluctuations in producing the peak in \( J_c \).

**Conclusions**

In conclusion, we studied the correlation between superconducting transition temperature and critical current density for the high-\( T_c \) superconductor FeSe. Both \( T_{c,0} \) and \( J_c \) increase with pressure, which is insensitive to the presence of AFM states, on the other hand, the superconducting transition width becomes considerably broader with the emergence of the AFM phase, and \( J_c \) is prominently enhanced in the coexisting phase. This behaviour reflects that the AFM phase not only provides an additional source of vortex pinning, but also makes the system susceptible to the inhomogeneous SC phase. Even though these observations are only specific to FeSe, they are expected to guide theoretical as well as experimental efforts to better understand the vortex pinning in the high-\( T_c \) superconductors where competing orders coexist on a microscopic scale. Further, when combined with well-known extrinsic pinning techniques, intrinsic magnetic pinning will provide a blueprint for greatly enhancing the critical current density, thereby bringing one step closer to the technological applications of high-temperature superconductors.
Methods

The c-axis-oriented high-quality FeSe$_{1-\delta}$ ($\delta = 0.04 \pm 0.02$) single crystals with a tetragonal structure (space group $P4/nmm$) were synthesised in evacuated quartz tubes in permanent gradient of temperature by using an AlCl$_3$/KCl flux. The synthesis technique used to fabricate the FeSe single crystals and their high-quality are described in detail elsewhere. The current-voltage ($I-V$) characteristics of FeSe were measured under hydrostatic pressures of 0.00, 0.41, 1.22, 1.72, 2.00, and 2.43 GPa. The physical pressure was applied by using a hybrid clamp-type pressure cell with Daphne 7373 as the pressure-transmitting medium, and the value of the pressure at low temperatures was determined by monitoring the shift in the $T_c$ of high-purity lead (Pb) as a manometer. The $I-V$ characteristic measurements under pressure were performed in the physical property measurement system (PPMS 9T, Quantum Design), where the electrical current was generated by using an Advantest R6142 unit and the voltage was measured by using an HP34420A nanovoltmeter. The depinning critical current ($I_c$) was obtained from the $1\mu V$ criterion instead of $1\mu V/cm$ in the $I-V$ curves due to a small size of FeSe single crystals. A few layers of FeSe in the FeSe single crystals were easily exfoliated by using adhesive tape, which is similar to the exfoliation technique that is used for graphene. The size of the measured crystals is typically $590 \times 210 \times 5 \mu m^3$. Quasi-hydrostatic pressure was achieved by using a clamp-type piston-cylinder pressure cell with Daphne oil 7373 as the pressure-transmitting medium. The magnetic fields were applied parallel ($H//ab$) to the $ab$-plane of the samples.

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Author Contributions

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Supplementary information

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