The pair density wave (PDW) is an extraordinary superconducting state in which Cooper pairs carry non-zero momentum\(^1,2\). Evidence for the existence of intrinsic PDW order in high-temperature (high-\(T_c\)) cuprate superconductors\(^3,4\) and kagome superconductors\(^5\) has emerged recently. However, the PDW order in iron-based high-\(T_c\) superconductors has not been observed experimentally. Here, using in situ scanning tunnelling microscopy and spectroscopy, we report the discovery of the PDW state in monolayer iron-based high-\(T_c\) Fe(Te,Se) films grown on SrTiO\(_3\) (001) substrates. The PDW state with a period of \(\lambda = 3.6 \ell_{\text{c}}\) (\(\ell_{\text{c}}\) is the distance between neighbouring Fe atoms) is observed at the domain walls by the spatial electronic modulations of the local density of states, the superconducting gap and the \(\pi\)-phase shift boundaries of the PDW around the vortices of the intertwined charge density wave order. The discovery of the PDW state in the monolayer Fe(Te,Se) film provides a low-dimensional platform to study the interplay between the correlated electronic states and unconventional Cooper pairing in high-\(T_c\) superconductors.

### Domain walls in the one-unit-cell Fe(Te,Se) film

The high-quality 1-UC Fe(Te,Se) films were grown on STO substrates by molecular-beam epitaxy (MBE) in an ultrahigh-vacuum system (Methods). Fig. 1a shows a typical atomically resolved STM topography of the 1-UC Fe(Te,Se) film. The nominal stoichiometry of 1-UC Fe\(_x\)Te\(_{1-x}\)Se\(_x\) (\(x = 0.5\)) is estimated from the thickness of the second-layer film\(^27,30,31\) (Extended Data Fig. 1). A typical tunnelling spectrum is displayed in Fig. 1b. In this work, all STS measurements were carried out at 4.3 K. The U-shaped feature indicates the fully gapped superconductivity and two pairs of coherence peaks are marked by dashed lines around \(\Delta_1 \approx 11\) meV and \(\Delta_2 \approx 18\) meV, consistent with previous reports\(^3,10,32\). The spatially uniform superconductivity of the crystalline film is confirmed by the tunnelling spectra taken along an 11.5 nm line-cut (Extended Data Fig. 1e).

A bright line in the topography of the 1-UC Fe(Te,Se) film indicates the domain wall that emerges naturally during the growing process\(^3,37\) (Fig. 1c). The orientation of the domain wall is about 45° from the direction of the Te/Se lattice, that is, along the orientation of the Fe–Te lattice. Tunnelling spectra taken on and off the domain wall show that the superconducting gaps decrease on the domain wall (Fig. 1d and Supplementary Fig. 1). In some regions of the domain wall, non-zero local density of states (LDOS) can be detected around zero bias, further confirming the suppression of superconductivity (green curve in Fig. 1d). Comparing with previous STM studies at the domain walls in bulk Fe(Te,Se)\(^3\), we do not observe clear evidence for propagating Majorana states, possibly because of the absence of 2D topological

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surface states in 1-UC Fe(TE,Se)/STO and its different band structure compared with bulk Fe(TE,Se). Figure 1e is the zoom-in-view of the region boxed in Fig. 1c, which characterizes the atomic structure of the domain wall. The grid imprinted in Fig. 1e simulates the Te/Se lattice of the domain on the right-hand side. Across the domain wall, a nearly 1/2 d_{Te,Se} shift (d_{Te,Se} is the lattice constant of the top Te/Se surface along the [110] direction) between the grid and the Te/Se lattice of the left domain can be observed (Methods). The lattice shift reveals the local compression of Te/Se lattice by about 1/2 d_{Te,Se} across the domain wall (Fig. 1f).

**LDOS modulation at the domain wall**

The electronic properties of the domain wall are examined by tunnelling conductance mapping. Figure 2a,b shows the drift-corrected and atomically resolved topography (Methods) of the domain wall D1 (zoom-in view of Fig. 1c) and the magnitude of its Fourier transform. The zero-bias conductance (ZBC) dI/dV(r, V = 0 mV) = g(r, V = 0 mV) around the domain wall is mapped in Fig. 2c, where r is the real space position and V is the bias voltage. Notably, a spatial modulation of the ZBC g(r, V = 0 mV) is observed at D1, which is absent in the topography (Fig. 2a,b). The wavevector of the LDOS modulation is independent of energy, indicating an origin of electronic order rather than quasiparticle interference (QPI) (Methods). This unidirectional LDOS modulation is absent in areas without the domain wall (Supplementary Fig. 4).

In addition to atomic Bragg peaks, the Fourier transform of the ZBC map (Fig. 2d) shows two Fourier peaks at $Q = 0.25 − 0.3 \text{Q}_{\text{Te}}$ and $Q = 2\pi/a_{\text{Fe}}$, corresponding to a unidirectional spatial modulation with a period of $\lambda = 3.3 − 4 \text{a}_{\text{Fe}}$ along the domain wall. The broadening of the Fourier peaks at Q and Q_{average} (Fig. 2d) results from spatial confinement of the modulation at the domain wall.

To further characterize the spatial variation of the electronic modulation at wavevector Q, a 2D lock-in technique is applied (Methods). The complex amplitude of r-space modulations at Q can be estimated by $A_Q(r) = \int \text{dR} A(R) e^{i \mathbf{R} \cdot \mathbf{r}} e^{−i \mathbf{Q} \cdot \mathbf{r}}$, where A(R) is an arbitrary r-space image and $\sigma$ is the averaging length scale. The amplitude $|A_Q(r)|$ and phase $\phi_Q(r)$ of the modulation at Q are given by $|A_Q(r)| = A_Q(0) e^{i \phi_Q(r)}$. The coherence peak height modulation $\phi_Q(r)$ at Q is determined by maximizing the distribution of amplitude $|A_Q(r)|$ and phase $\phi_Q(r)$ of observed modulations at Q = 2.7Q_{Te}. The value of Q is determined by maximizing the spatial uniformity of the phases at the domain wall (Methods). As shown in Fig. 2e,f, the modulation amplitude at 0.27Q_{Te} is confined to the domain wall with uniform phases, consistent with the spatial distribution of the periodic ZBC modulation in Fig. 2c.

**Coherence peak height modulation**

The dI/dV maps measured at various bias voltages show that the electronic ordering-induced LDOS modulations mainly exist in the energies within the superconducting gap (Methods). Previous studies of PDWs in high-T_c cuprates and kagome superconductors have reported a strong correlation between the coherence peak height and the superconducting order parameter. We thus measure the coherence peak height along the domain wall labelled as D2 with its corresponding magnitude of Fourier transform (Fig. 3c).

Further analysis using the 2D lock-in technique shows the modulation wavevector Q = 2.7Q_{Te} (Fig. 3d,e). Figure 3f illustrates the tunnelling spectra measured along a line-cut parallel to the Fe–Fe bond direction (light grey arrow in Fig. 3a), which is the direction of Q and roughly along the domain wall. The coherence peak height modulation with a period of $\lambda = 3.6 \text{a}_{\text{Fe}}$ can be clearly distinguished (Fig. 3g), consistent with the modulation of the LDOS (Fig. 3b,c). The coherence peak height modulation is also detected in another domain wall labelled as D7 (Supplementary Fig. 5). Furthermore, the dI/dV map (Supplementary Fig. 6a) and its Fourier transform (Supplementary Fig. 6b), measured near the superconducting gap energy at the domain wall, show clear periodic modulations. The coherence peak height modulations with nearly identical wavevector Q as the LDOS suggest spatial modulations of the superconducting order parameter and the existence of PDW order.

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**Fig. 1** Domain wall along the Fe–Fe bond direction in 1-UC Fe(TE,Se)/STO. **a** An STM topography of the 1-UC Fe(TE,Se)/STO (10 × 10 nm², $V_s = 0.1$ V, $I_s = 0.5$ nA). **b** A typical U-shaped tunnelling spectrum measured on the 1-UC Fe(TE,Se) film at 4.3 K, which shows fully gapped superconductivity; two pairs of coherence peaks are marked by dashed lines ($\Delta_T = 11$ mV, $\Delta_c = 18$ mV). **c** An STM topographic image of the domain wall structure (bright line) in 1-UC Fe(TE,Se)/STO (16 × 16 nm², $V_s = 0.1$ V, $I_s = 0.5$ nA). **d** Typical tunnelling spectra taken on and off the domain wall (labelled as D1) at 4.3 K. The red, purple and green curves correspond to the spectra taken at the red, purple and green points, respectively. The superconducting gap is smaller on the domain wall, indicating the suppression of superconductivity. The inset shows the zoom-in tunnelling spectra at low bias. **e** A zoom-in of the light-grey dashed box region in **c**. The zoom-in topography is rotated to show the lattice shift across the domain wall. The spots are illustrative lattice points corresponding to the right domain. The mismatches between the spots and the lattice of the left domain show a shift around 1/2 d_{Te,Se} (d_{Te,Se} is the Te/Se lattice distance along the [110], that is, the Fe–Fe direction on the top surface) across the domain wall. Two brighter Te/Se atoms in the orange dashed rectangle marked by white arrows are guides to the eye for the nearly 1/2 d_{Te,Se} shift across the domain wall. **f** The schematic of the domain wall in 1-UC Fe(TE,Se)/STO, illustrating the compression across the domain wall.
The spatial modulation of the LDOS at the domain wall labelled as D1. a. Zoom-in image (7.7 × 7.7 nm²) of Fig. 1c, clearly showing the topography of the domain wall T(ρ). b. Magnitude of the Fourier transform |F(χ)| of a. Orange crosses are at χ = (±Q_g, 0), (0, ±Q_g). Bragg peaks of the Te/Se lattice are circled in red. c. ZBC map g(ρ, V = 0 mV) taken at the same area in at 4.3 K, which shows an emergent electronic modulation along the domain wall. d. Magnitude of the Fourier transform |g(χ, V = 0 mV)| of the ZBC map in c. The modulation wavevector Q = 0.25–0.3Q_g circled in blue shows a spatial modulation with a period of around 3.7a_F along the direction of the domain wall. e. Spatial distribution of the modulation g_4(χ, V = 0 mV) (calculated by the 2D lock-in method, showing that modulations at Q_g g(ρ, V = 0 mV) occur only within the domain wall region. f. Spatial distribution of the modulation g_4(χ) phase φ_g(χ, V = 0 mV). The wavevector Q = 0.27Q_g used in e and f is determined by analysing the uniformity of the phase at the domain wall. The averaging length scales in e and f are denoted by dashed circles. Black dashed lines in a, e, f mark the edges of the domain wall.

Superconducting gap energy modulation
Apart from the coherence peak height modulation, the direct observation of superconducting gap energy modulation Δ(ρ) has also been recognized as compelling evidence of the multi-Q PDW, in contrast to the helical Fulde–Ferrell (FF) state in which only the phase of the order parameter modulates spatially. However, the modulation of Δ(ρ) can be challenging to detect in the presence of strong tunnelling spectra variations induced by disorder that can obscure the small amplitude of PDW modulations, compared with the background signal. To investigate the possible gap modulations and further confirm the PDW order, we first obtained the ZBC g(ρ, V = 0 mV) map (Fig. 4b) and its
Fourier transform (Fig. 4c) in the region around the domain wall labelled as D3 (Fig. 4a). A clear periodic spatial modulation along the domain wall was revealed, consistent with the modulations observed at other domain walls (Figs. 2c and 3b, Extended Data Fig. 2d and Supplementary Fig. 3e). By extracting the superconducting gap energy from the tunnelling conductance spectrum at every pixel in Fig. 4b (Methods), the spatial distribution of $\Delta_{g}(r)$ (gap map) measured in the same area as in a. $\Delta_{g}$ is the smaller superconducting gap. The inset in Fig. 4f plots the magnitude of the Fourier transform of $d(r)$ after filtering out Bragg peaks and the small-$q$ noise in f, showing clear gap modulation. Spatial variation of the amplitude $|A^{g}_{Q}|(r)$ of the gap energy modulation at $Q = 0.28Q_{Fe}$. The large amplitude at the domain wall justifies the gap modulation. Spatial variation of the phase $\phi^{g}_{Q}(r)$ of gap modulations in d at $Q = 0.28Q_{Fe}$. The averaging length scales in b and d are denoted by dashed circles. Black dashed lines in a, b, d, g–i mark the domain wall boundaries.
modulation with $2Q$ periodicity. By analysing the spatial variation of the amplitude and phase of this secondary $2Q$ CDW, the influence of the topmost Te/Se atomic lattice can be excluded (Methods and Extended Data Figs. 7 and 8). Second, a $\pi$-phase shift in the PDW order around a half-dislocation (black circle in Fig. 5a) is predicted to nucleate at a topological defect with a $2\pi$ phase-winding (that is, a vortex) in the induced $2Q$ CDW order, as indicated by the black circle in Fig. 5c. For a pure PDW order depicted in Fig. 5a, the $\pi$-phase shift boundary can be simulated explicitly. Figure 5b,d displays the phases of the PDW and the CDW order parameters, showing the $\pi$-phase shift boundary in the PDW phase (Fig. 5b) and the corresponding $2\pi$-phase winding around the vortex of the induced $2Q$ CDW (Fig. 5d). The phase of the PDW is usually locked to that of the uniform superconducting background, making it energetically costly to nucleate a PDW $\pi$-phase shift. In 1-UC Fe(Se, Te)/STO, the structural $\pi$-phase-shifted domains may induce a $\pi$-phase shift in the superconducting order parameter across the domain wall as reported in bulk Fe(Se, Te)\(^\text{15}\). This makes the PDW $\pi$-phase shift possible in the domain wall region where the uniform superconductivity is weakened (Methods). Consequently, detecting the $\pi$-phase shift boundary in the PDW phase bound to the $2Q$ CDW vortex can provide further evidence for the primary PDW order.

Applying the 2D lock-in technique to the experimental data, the phase-resolved image of the induced $2Q$ CDW $\rho^{\text{2D}}_{a}(r)$ (Methods) is shown in Fig. 5e, where the topological vortices are marked by the black dots. Over the same region, the obtained PDW phase map $\phi^{\text{PDW}}_{a}(r)$ is shown in Fig. 5f and imprinted with the locations of the CDW vortex (black dots in Fig. 5e). The $\pi$-phase shifts in the PDW phase $\phi^{\text{PDW}}_{a}(r)$, marked by the arrowed cuts in Fig. 5f and inset, are indeed observed near the vortices of the $2Q$ CDW. More analyses of the $\pi$-phase shift boundaries are provided in Methods. The observation of the $\pi$-phase shift boundaries bound to vortices of the induced CDW provides further evidence for the observed primary multi-$Q$ PDW in the monolayer Fe-based superconductor.

**Discussion**

The interplay among CDW, PDW and uniform superconductivity plays a key role in the nature of the PDW phenomenon. The coexistence of a primary CDW order with uniform superconductivity can induce a secondary PDW order. This has been observed in NbSe\(_2\) (ref. 17), where the wavevector $Q$ of the PDW is identical to that of the CDW. By contrast, in the current 1-UC Fe(Se, Te) films, a primary CDW state is absent (Methods). The observed electronic modulations are thus expected to originate from a primary PDW nucleated at the domain walls. Moreover, the spatial gap modulations at the domain wall (Fig. 4) suggest a unidirectional multi-$Q$ PDW state described by $\Delta_{\text{PDW}}(r) = \Delta_{Q}e^{i2\pi r} + \Delta_{Q}e^{-i2\pi r} \cos(Q \cdot r)$. The possible scenarios for the PDW order are discussed in Methods. Describing the domain wall as an embedded quantum structure\(^{40}\), we find that a new equal-spin pairing PDW state can emerge at the domain wall, which is consistent with our observations and supported by effective model calculations in Supplementary Information.

In summary, we have detected an incommensurate PDW state with a period of $A = 3.6d_0$, located at the domain walls in the 1-UC Fe(Se, Te)/STO. Our findings demonstrate that quantum structures such as naturally appearing domain walls embedded in 2D FeSCs provide a new material platform to study the PDW state and its interplay with the topological electronic states and unconventional high-$T_c$ superconductivity.

**Online content**

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Methods

Sample preparation and measurement
Our experiments were carried out in an ultrahigh-vacuum MBE–STM system (Scienta Omicron), The Nb-doped SrTiO3(001) (wt 0.7%) substrates were thermally boiled in deionized water at 90 °C for 50 min and chemically etched in 12% hydrochloric acid for 45 min. Then the substrates were transported into the MEBE chamber with a base pressure of about 2 × 10⁻⁹ mbar and underwent Se-flux treatment at 950 °C to expose an atomically flat TiO₂-terminated surface. High-quality monolayer FeTe₀.₇Se films were grown on pretreated Nb-doped SrTiO₃ substrates by codepositing high-purity Fe(99.99%), Te(99.999%) and Se(99.999%) while the substrates were kept at 340 °C. Then the films were annealed at 380 °C for 3 h and transported to the in situ STM chamber after annealing. The thickness of the 1-UC FeT e₁−xSe films is around 0.59 nm (Extended Data Fig. 1), corresponding to the composition x = 0.5 (ref. 2). All the results shown were acquired at 4.3 K with a polycrystalline PtIr tip by the standard lock-in technique. The modulation voltage was Vₘ₀d = 1 mV at 1.7595 kHz. The set-up of all results was Vₛ = 0.04 V and Iₛ = 2.5 nA unless stated otherwise.

Characterization of the domain wall
We characterized the crystal structure of the domain wall by analysing the top Te/Se lattice on both sides of the domain wall. The lattice in each domain can be described by \( \mathbf{T}(r) = \cos(q_x \cdot r + \theta_{q_x}) + \cos(q_y \cdot r + \theta_{q_y}) \), where \( q = 1, 2 \) labels two domains, \( \theta_{q_x} \) is the reciprocal lattice vector of the top Te/Se lattice along the \( x \) direction and \( \theta_{q_y} \) is the phase of the lattice. In the area far from the domain wall, the reciprocal lattice vectors of top Te/Se lattice are the same, that is, \( \theta_{q_x} = \theta_{q_y} \). As shown in Extended Data Fig. 2, regions as large as possible inside each domain but far from the domain wall are selected, in which \( \theta_{q_x} = \theta_{q_y} \). The periodical perfect lattices calculated using the \( \theta_{q_x} \) and \( \theta_{q_y} \) are shown by white dots in Extended Data Fig. 2. The phase difference \( \Delta \theta_{q} = \theta_{q_x} - \theta_{q_y} \) between the two domains represents a displacement along the \( x \) direction, which turns out to be a compression along the Fe–Fe direction across the domain wall. Besides, we also calculated the phase maps using the 2D lock-in technique around the Bragg peaks\(^4\), as shown in Supplementary Fig. 2. The phase shifts obtained by these two methods are consistent.

Correction on distortions in topography \( T(r) \) and differential conductance map \( g(r, V) \)
The spectroscopic imaging STM experiments are time-consuming, thus the piezo creep (and many other effects) usually causes remarkable distortions on the dataset. In the well-known Lawler–Fujita algorithm\(^4\), the total displacement field \( u(r) \) is considered to be roughly a constant over a coarsening length scale \( 1/\Lambda \), where \( \Lambda \) is the characteristic broadening length of Bragg peak. However, in some of our datasets, \( u(r) \) varies quickly with space, leading to a large \( \Lambda \) that doesn’t satisfy the condition \( \Lambda << \Theta_q \). The distortion information is broadened to a comparatively large range in \( q \) space and it is hard to define unambiguous Bragg peaks. Besides, the large distortions usually result in the severely wrapped phase in the Lawler–Fujita algorithm. Thus, it is not suitable to directly use the Lawler–Fujita method to correct these datasets. Considering that the distortions caused by piezo creep are accumulated with time, we subtracted a creep-caused displacement assuming that the creep exponentially decays with time. After this pretreatment, we applied the Lawler–Fujita algorithm to eliminate the residual picometre-scale distortions. In this way, we calculated \( u(r) \) for topography and corrected \( T(r) \) and \( g(r, V) \) by subtracting \( u(r) \), as shown in Supplementary Fig. 3.

Characterization of the LDOS modulation induced by electronic ordering
The electronic ordering origin of the LDOS modulations can be confirmed by the differential conductance map \( g(r, V) \) with sweeping bias. The possibility of the LDOS modulations in \( g(r, V) \) originating from QPI can be excluded. The wavevectors of the QPI pattern connect the constant energy contours of quasiparticles with energy \( E = e \times V \). The constant energy contours vary with energy \( E \), thus the wavevectors of QPI are dispersive with bias \( V \). However, as shown in Extended Data Fig. 3b, the LDOS modulations at the domain wall are non-dispersive and exist mainly within the superconducting gap, which is the decisive evidence of electronic ordering rather than QPI origin.

The quantum confinement effect can also induce LDOS modulations by the scattering and interference of the confined quasiparticle. The finite width of the domain wall (about 2–3 nm) may lead to LDOS modulations with the wavevector perpendicular to the domain wall. Nevertheless, the observed LDOS modulations are approximately along the domain wall (specifically, along the Fe–Fe direction of the film), which cannot be induced by the perpendicular restriction. In addition, if the LDOS modulations along the domain wall are from the finite length of the domain wall (about 10 nm), then the wavevector of such LDOS modulations is expected to disperse with energy, which disagrees with the non-dispersive LDOS modulations observed at the domain wall (Extended Data Fig. 3b). Furthermore, the quantum confinement-induced LDOS modulations appear in a large energy range, whereas the observed modulations at the domain wall mainly appear within the superconducting gap energy (Extended Data Fig. 3 and Supplementary Fig. 8). Therefore, the quantum confinement origin can be excluded.

Recently, the smectic phase has been detected in uniaxial strained LiFeAs\(^1\) and the correlation between the charge-ordered stripes and local anisotropic strain has been studied in FeSe multilayers grown on STO\(^1\). This raises the question of whether the PDW state in the 1-UC Fe(T e,Se) films is induced by a primary CDW state coexisting with superconductivity. The primary CDW state usually appears as spatial modulations in STM topographic images. For example, the CDW-like stripe order has been observed in the topography of roughly 30-UC FeSe films grown on STO\(^1\). However, no spatial modulation can be detected in the topography of the 1-UC Fe(T e,Se) films. Moreover, the stripe order in FeSe multilayers can be detected in the \( dV/dI \) mapping at a large bias (≥100 mV)\(^1\), which is another characteristic of the primary CDW state. As shown in Extended Data Fig. 3, the PDW state observed in the 1-UC Fe(T e,Se) films mainly emerges at the energy within the superconducting gap (around 10 meV), which further excludes the possibility of the primary CDW-induced PDW state in 1-UC Fe(T e,Se) films.

Two-dimensional lock-in technique
Consider a real-space image containing a series of modulation wavevectors: \( \mathbf{A}(r) = \sum_\mathbf{q} a_\mathbf{q}(r)e^{i q \cdot r} \), where \( a_\mathbf{q}(r) \) is the complex amplitude at wavevector \( \mathbf{q} \) and position \( r \) is the Euler's number and \( i \) is the imaginary unit. Then the Fourier transform of \( \mathbf{A}(r) \) \( (\mathcal{F}(\mathbf{A}(r))) \) can be written as:

\[
\mathcal{A}(\mathbf{q}) = \mathcal{F}[\mathbf{A}(r)] = \int \mathbf{q} d\mathbf{q} (a_\mathbf{q}(r)) \delta(\mathbf{Q} - \mathbf{q} + \mathbf{q}' = \sum_\mathbf{q} a_\mathbf{q}(\mathbf{q} - \mathbf{Q})
\]

where \( a_\mathbf{q}(\mathbf{q} - \mathbf{Q}) \) is the Fourier transform of the complex amplitude centred at \( \mathbf{Q} \). Defining \( \Lambda_q \) as a characteristic broadening length of \( a_\mathbf{q}(\mathbf{q}) \), \( a_\mathbf{q}(\mathbf{q}) \) is almost zero for \( |\mathbf{q}| \geq \Lambda_q \) and \( a_\mathbf{q}(r) \) is roughly a constant over a coarsening length scale \( 1/\Lambda_q \). \( \mathbf{Q} \) represents a typical separation between different wavevectors. In this case, the Fourier transforms of the complex amplitudes at different wavevectors are clearly separated. \( a_\mathbf{q}(\mathbf{q}) \) can be approximately extracted by shifting it back to the centre and multiplying a Gaussian window with a cutoff length \( \Lambda_q \). The average length scale in \( r \) space. Then approximate complex amplitude in real space \( a_\mathbf{q}(r) \) can be obtained by inverse Fourier transform \( (\mathcal{F}^{-1}) \). In practice, the 2D lock-in technique\(^1\) is realized as below:

\[
A_\mathbf{Q}(\mathbf{r}) = \mathcal{F}^{-1}[A_\mathbf{Q}(\mathbf{q})] = \mathcal{F}^{-1} \left\{ \mathcal{F}[\mathbf{A}(r)e^{i\mathbf{q} \cdot \mathbf{r}}] \cdot \frac{1}{\sqrt{2\pi} \sigma_\mathbf{q}} e^{-\frac{q^2}{2\sigma_\mathbf{q}^2}} \right\}
\]
which is equivalent to the processes discussed above. Using the 2D lock-in technique, the PDW wavevector is carefully analysed, as shown in the main text.

**Determination of modulation wavevector Q**

For a real-space image $A(r)$ in a limited field of view (FOV) $L_x \times L_y$, the discrete Fourier transform $A(q)$ has a pixel size of $\frac{2\pi}{L_x}$ \times $\frac{2\pi}{L_y}$. Limited by the domain wall size, the FOVs in our experiments are typically smaller than 16 nm \times 16 nm, which severely limits the resolution in $q$ space. As the PDW state is localized at the domain wall, the inhomogeneous distribution of the modulation amplitude also makes the peaks in $q$ space broaden into a small region around the wavevector. Thus, it is hard to directly obtain the wavevector $Q$ using Fourier transformation. To determine the wavevector $Q$, we used the 2D lock in technique to solve the phase map under a tentative $Q$:

$$A_Q (r) = F^{-1} [A_Q (q)] = F^{-1} [F[A(r)e^{iq \cdot r}] \cdot \frac{1}{2\pi} e^{-i q \cdot \frac{r^2}{2 \sigma^2}}]$$

$$\phi_Q^i (r) = \tan^{-1} \left( \frac{\text{Im}(A_Q (r))}{\text{Re}(A_Q (r))} \right)$$

where $\sigma$ is the $q$-space cutoff length. $\phi$ was carefully chosen to exclude the influence of other wavevectors, which contain the low-frequency information of the slowly varied modulation amplitude and phase. For $Q = Q$, $\phi_Q^i (r) = (Q \cdot Q) \cdot r + \phi$ which roughly approaches a constant only when $Q = Q$. As shown in Extended Data Fig. 4 and Supplementary Figs. 9 and 10, by counting the density distribution of $\phi_Q^i (r)$ at domain walls D1 and D3 for a series of tentative $Q$, we determined the wavevector $Q$ that corresponds to the minimum of the ratio at half maximum of the peak in the $\phi_Q^i (r)$ distribution histogram ($r_{int}$ represents the area of the domain wall).

We further determined the $Q$ value of the LDOS modulations in Fig. 2 by using different cutoff lengths in the phase calculations. The determined $Q$ value is stable when we use different cutoff lengths (shown in Supplementary Fig. 11), which confirms that the determination method is reliable.

**Measurement of gap modulation**

We measured tunnelling spectra at each position $r$, and estimated the smaller gap $\Delta_1 (r)$ as follows:

1. Take the second derivative of the differential conductance spectra.
2. Find the bias $V_0$ that has the minimal value of $\frac{d^2}{dV^2}$$r (r, V)$.
3. Use three data points of $\frac{d^2}{dV^2}$$r (r, V)$ in the neighbourhood of $V_0$, namely $[V_+, V_0, V_-]$, to fit a quadratic function and take the apex $eV_0$ as the gap value $\Delta_1$.
4. Select the smaller gap values obtained at each position as $\Delta_1$. A cutoff gap energy $\Delta_1 = 10.5$ meV is used as the upper limit of $\Delta_1$. The cutoff $\Delta_1$ is determined by the statistical histogram of gaps (dashed line in Supplementary Fig. 12).

Then the gap modulation was analysed by calculating the Fourier transform $\Delta_1 (Q)$ and 2D lock-in signals $|A_Q^i (r)|$ and $\phi_Q^i (r)$.

**Existence of the secondary charge density wave state**

Consider the case of coexisting zero-momentum and multi-Q PDW order parameters, namely $\Delta (r) = \Delta_0 + \Delta_Q e^{i q \cdot r} + \Delta_Q e^{-i q \cdot r}$. The secondary CDW order is induced by the product of the PDW order parameters:

$$\rho_{\text{CDW}}^{\pm \Delta Q} = \rho_{\text{PDW}}^{\Delta Q}$$

where $\Delta Q$ denotes the complex conjugate. The secondary 2Q CDW signal is expected to be weaker and less coherent than the 1Q CDW $\rho_{\text{PDW}}^{Q} - \rho_{\text{PDW}}^{Q*} = \Delta_1^2$. To enhance the signal intensity of the 2Q CDW, we obtain the $r$-map using $\rho (r, V) = \Delta (r, V) = \frac{1}{2} \int N (r, f) dE$ that is, the integrated density of states within $eV$, and analyse it in Extended Data Fig. 7a. The integration suppresses the QPI signal, which is phase incoherent at different bias voltages, and enhances the signal of the phase-coherent charge order-induced LDOS modulations. Fourier transform of the $\rho (r, V)$ map exhibits maxima around $\pm 2Q$ (Extended Data Fig. 7b), indicating the 2Q CDW modulations. The Fourier maxima are a little broadened, which may be attributed to the small FOV limited by the size of domain wall. To further confirm the existence of 2Q CDW, 2D lock in analysis is applied. As shown in Extended Data Fig. 7c, d, the amplitude of the 2Q modulation is confined to the domain wall region and the phase of the modulation is nearly uniform in the domain wall region, supporting the existence of the 2Q CDW order.

In 1-UC Fe(Fe,Se) films, the wavevector of the PDW state is about $0.28Q_{\text{ns}}$, and the wavevector of the secondary 2Q CDW should be close to $0.56Q_{\text{ns}}$. However, the topmost Te/Se atomic lattice shows a reciprocal lattice vector of $0.5Q_{\text{ns}}$ when projected to the Fe–Fe direction, which is close to 2Q and may mix with the 2Q CDW. To distinguish the 2Q CDW signal from the Te/Se atomic lattice projection, we calculate the spatial variation of the phase $\phi_{\text{PDW}}^Q (r)$ (Extended Data Fig. 8a) and the amplitude $|A_{\text{PDW}}^Q (r)|$ of the STM topography $T (r)$ (Extended Data Fig. 8c) at precisely $0.5Q_{\text{ns}}$. The set point of the STM topography is at 40 mV, where the contribution of the 2Q CDW modulation is negligible.

As shown in the phase map $\phi_{\text{PDW}}^Q (r)$ (Extended Data Fig. 8a), the phase of $0.5Q_{\text{ns}}$ modulation in the topography (that is, topmost Te/Se atomic lattice) is not distributed uniformly at the domain wall, which is different from the uniform phase of 2Q CDW $\phi_{\text{PDW}}^{2Q} (r)$ (Extended Data Fig. 8b). Furthermore, the amplitude of $0.5Q_{\text{ns}}$ modulation $|A_{\text{PDW}}^{Q/2} (r)|$ is also randomly distributed in the whole FOV (Extended Data Fig. 8c), whereas the amplitude of 2Q CDW $|A_{\text{PDW}}^{2Q} (r)|$ (Extended Data Fig. 8d) is restricted to the domain wall area and consistent with the uniform 2Q CDW phase $\phi_{\text{PDW}}^{2Q} (r)$. The topological defects with 2Q phase winding are further marked in $\phi_{\text{PDW}}^{Q/2} (r)$ and $\phi_{\text{PDW}}^{2Q} (r)$ (black dots in Extended Data Fig. 8a,b). The topological defects in $\phi_{\text{PDW}}^{Q/2} (r)$ are randomly distributed in the whole FOV, whereas the topological defects in $\phi_{\text{PDW}}^{2Q} (r)$ are mainly around the edge of the domain wall. These results indicate that the 2Q CDW is independent of the atomic lattice.

**n-phase shift boundaries in the pair density wave phase**

Consider that the PDW order $\Delta (Q, Q) = \Delta_{\text{PDW}} (e^{i \theta} e^{-i \phi})$ coexists with the uniform superconducting order $\Delta_0 e^{i \theta}$, where $\theta$ and $\phi$ are U(1) phases. The phases of the $Q$ and $-Q$ PDW order Parameters $\Delta_0$ can be expressed as $(Q \cdot r + \phi) Q (r)$ and $(-Q \cdot r + \phi) Q (r)$. The magnitude of the order parameter can be expressed as:

$$|\Delta| = |\Delta_0 e^{i \theta} + \Delta_{\text{PDW}} (e^{i \theta} e^{-i \phi})|$$

$$= |\Delta| + |2 \Delta_{\text{PDW}} | \cos \theta \frac{\phi_0}{2} \frac{\phi_0 - \phi}{2} \cos \left( Q \cdot r - \frac{\phi_0 + \phi}{2} \right)$$

One half of the relative phase between $Q$ and $-Q$ PDW order parameters $\phi_{\text{PDW}} (r) = Q \cdot r + \frac{\phi_0 (r) - \phi (r)}{2}$ is extracted from the second term, which spatially modulates as $\cos (\phi_{\text{PDW}} (r))$. During the 2D lock-in calculation process, a relative phase $Q \cdot (r - r_0)$ is subtracted and the phase value in the obtained phase map is $\phi (r) = \frac{2 \Delta_{\text{PDW}} (e^{i \theta} e^{-i \phi})}{2} + Q \cdot r_0$, which depends on the selected origin ($r_0$). Note that the phase shift between two points $\phi (r_1) - \phi (r_2)$ or along a closed path $\phi dP$ is independent of $r_0$. Thus, the phase shift, which contains the information of topological features, is meaningful and discussed here ($Q \cdot r_0$ can be ignored).
We further introduce $y = (\phi_d + \phi_c)/2$ that appears in the supercurrent formula, and $Q \cdot d = \phi(r) - (\phi_d - \phi_c)/2$, which is caused by the displacement ($d$). For the pure PDW case ($Q_d = 0$), the $\pi$-phase winding of $Q \cdot d$ is observable by STM and corresponds to $2\pi$-phase winding in the 2Q CDW order. The single-valuedness of the order parameters implies that $\phi_d - \phi_c = y \cdot Q \cdot d$ must wind by integer multiples of $2\pi$, which leads to the $\pi$-phase shift of $y$ and leads to what was often referred to as a half-vortex.

In Fig. 5f, the boundaries across which the phase changes by $\pi$ can be observed in the PDW phase ($Q \cdot d$). The topological defects with $2\pi$-phase winding (vortices) in 2Q CDW phase (Fig. 5e) are also detected. In the Fourier-filtered gap (Supplementary Fig. 14) and $\rho(r, V)$ map (Supplementary Fig. 15), the real-space evolutions of the gap and 2Q $\rho(r, V)$ modulations are consistent with the simulated PDW $\pi$ phase boundary (Fig. 5a,b) and Supplementary Fig. 14d–g. However, despite non-zero centre-of-mass momentum $Q$, $\pi$-phase shift can arise from pairing with finite centre-of-mass momentum $Q$, and $\pi$-phase shift cannot observed in the PDW phase ($Q = 0$). The appearance of the PDW order is mainly confined to the domain walls (Extended Data Fig. 9a), which offers some insights for its origin because each spin pairing channel contains only one momentum, either $Q$ or $-Q$, and cannot produce the observed gap modulation. It is crucial to realize that the observed domain wall in Fig. 5 breaks the mirror symmetry about the one-dimensional axis because of the domain wall structure in Fe(Te,Se), the imperfect wall shape and disorder, and the two nearly $\pi$-phase-shifted bulk superconducting order on either side of the domain wall as reported in bulk Fe(Te,Se).

The broken mirror symmetry introduces an extra Dresselhaus SOC, such that each of the Rashba split bands now contains both spin projections. As shown in Extended Data Fig. 9c, the equal-spin pairing interaction leads to order parameters $(c_k, c_{-k}^\dagger)$ and $(c_{k', c_{-k'}^\dagger})$ $(o$ is the spin index), which ensure that Cooper pairs in each spin sector carry both momentum $Q$ and $-Q$, giving rise to a new equal-spin pairing multi-Q PDW order with superconducting gap modulations. Model calculations (see Supplementary Information for detailed discussions) of the tunnelling density of states and the spatial modulation of the superconducting gap are shown in Extended Data Fig. 9d–g for two examples of PDW momentum $Q = 2n/12$, and $Q = 2n/4$ close to the experimentally observed incommensurate wavevector.

We cannot rule out other possibilities based on the current experimental data. For example, similar to the magnetic field-induced PDW state in vortex halos of cuprates, the PDW state may also exist throughout the 1-UC Fe(Te,Se) film; it may difficult to detect because of the predominant bulk superconductivity, but becomes visible at the domain walls because of the weakened bulk superconductivity. The microscopic description of the origin and properties of the domain-wall PDW requires further theoretical and experimental investigations.

**Possible scenarios for the pair density wave state at the domain wall**

The appearance of the PDW order is mainly confined to the domain walls (Extended Data Fig. 9a), which offers some insights for its origin in 1-UC Fe(Te,Se) films. The local lattice distortion around the domain wall breaks spatial symmetries and results in strong spin–orbit coupling (SOC). To illustrate how this may generate a quasi-one-dimensional PDW, consider a simple band of domain wall states, which is split by Rashba SOC in momentum along the domain wall direction ($k_z$). If time-reversal symmetry is broken, for example by an induced ferromagnetic order around the domain wall, a spin-singlet FF helical state $\Delta_{iy}(r) = \Delta_{iy}(r)^{\uparrow\downarrow}$ can arise from pairing with finite centre-of-mass momentum $Q_y$ on the same Zeeman-shifted Fermi surface, where $Q_y = \frac{2\pi}{12}E_F$ is the Zeeman energy and $E_F$ is the Fermi velocity. However, our findings do not support this scenario. The observed PDW wavevector is rather large, $Q = 0.28Q_{iy}$. If we choose $hV_F = 0.54eV \cdot Å$ (ref. 34), a large Zeeman energy ($E_F = 340$ meV) is needed. However, we find no evidence suggesting such a notable spontaneous magnetism around the domain wall.

We therefore propose a different scenario in which a time-reversal invariant multi-Q PDW order is created at the domain wall and accounts for the observed gap modulations. Analogous to the atomic line defect of missing Te/Se atoms hosting zero-energy bound states at both ends, the domain wall can be considered as an embedded quantum structure in the unconventional superconductor. The pairing interaction can be generally described by equal-spin and opposite-spin pairing across the Fermi points of the Rashba split bands shown in Extended Data Fig. 9b. Owing to the microscopic coherent quantum tunnelling processes and SOC, triplet equal-spin pairing can be greatly enhanced for the domain wall states because each spin pairing channel contains only one momentum, either $Q$ or $-Q$, and cannot produce the observed gap modulation. It is crucial to realize that the observed domain wall in Fig. 5 breaks the mirror symmetry about the one-dimensional axis because of the domain wall structure in Fe(Te,Se), the imperfect wall shape and disorder, and the two nearly $\pi$-phase-shifted bulk superconducting order on either side of the domain wall as reported in bulk Fe(Te,Se).
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Additional information

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Extended Data Fig. 1 | More information about the 1-UC Fe(Te,Se)/STO.  

a, An STM topography of 1-2 UC Fe(Te,Se)/STO at large-scale (200×200 nm², \( V_s = 1 \) V, \( I_s = 0.5 \) nA) with terraces of the STO substrate (lighter color means the higher height). The cyan area at the edge of the lower STO terrace shows the 2nd-UC Fe(Te,Se) film.  

b, Line profile taken along the orange curve in a. The thickness of the 1-UC FeTe\(_{1-x}\)Se\(_x\) film is approximately 0.59 nm, corresponding to the composition \( x \approx 0.5 \).  

c, An STM image of 1-UC Fe(Te,Se)/STO (10×10 nm², \( V_s = 0.1 \) V, \( I_s = 0.5 \) nA).  

d, Averaged tunneling spectrum (averaged over ~100 spectra) taken along the light grey arrow in c, which shows two pairs of coherence peaks at \( \Delta_1 \approx 11 \) meV and \( \Delta_2 \approx 19 \) meV.  

e, Waterfall plot of \( \text{d}I/\text{d}V \) curves taken along the light grey arrow in c.
Extended Data Fig. 2 | Characterization of domain walls. 

(a, b) STM images (16 × 16 nm², $V_s = 0.1\, V$, $I_s = 0.5\, nA$) of two domain walls labeled as D1 (a) and D4 (b). 

(c, d) $dI/dV$ maps of two domain walls. The lattice vectors $q_{i,x,y}$ of two regions marked by white spots in a and b satisfy $q_{i,x,y} = q_{j,x,y}$. $dI/dV$ maps shown in c and d are measured at the red dashed box in a and b, respectively.
Extended Data Fig. 3 | Non-dispersive PDW and absence of PDW in high energy \( \frac{dI}{dV} \) maps. a, The STM topography of the domain wall D3 shown in Fig. 4. b, The bias dependence of the wavevector of the LDOS modulation at the domain wall. c, d, \( \frac{dI}{dV} \) map (c) taken at 80 mV over the same FOV of a and corresponding magnitude of the Fourier transform (d). There is no spatial modulation in the \( \frac{dI}{dV} \) map (c) and no FFT peak at around \( Q \approx 0.28Q_\text{Fe} \) at the energy much higher than the superconducting gap (d). e, f, Spatial variation of the amplitude (e) and phase (f) of the LDOS modulation at bias voltage from 0 to 14 mV. The averaging length scales in e and f are denoted by dashed circles. The LDOS modulations mainly exist in the energies within the superconducting gap, which is also observed at domain wall D2 (Supplementary Fig. 13). Dashed lines in a, c, e and f mark the edges of the domain wall.
Extended Data Fig. 4  Determination of modulation wavevector \( Q \) at
domain wall D1. a, The STM topography of the domain wall D1. b, Zero-bias
conductance (ZBC) map \( g(r, V = 0 \text{ mV}) \) taken at the same area in a. c, The
magnitude of the Fourier transform \( g(q, V = 0 \text{ mV}) \) of the ZBC map in b.
d–f, Spatial distribution of the modulation \( \phi_Q(r) \) phase \( \phi_Q(r) \). The averaging
length scales in d–f are denoted by dashed circles. The domain wall area is marked
by black dashed lines in a, b and d–f. g–i, Statistical histogram of \( \phi_Q(r_{\text{DW}}) \)
\( (r_{\text{DW}} \text{ represents the area of the domain wall}) \) at the domain wall D1 for a series of
tentative \( Q \). Only the points inside the domain wall area in d–f are counted. Black
dashed lines in g–i are located at the half maximum of the peak. The determined
wavevector \( Q \) corresponds to the minimum of the full width at half maximum
(FWHM) of the peak in the distribution histogram \( \phi_Q(r_{\text{DW}}) \), as shown in h.
Extended Data Fig. 5 | Superconducting gap energy modulations at the domain wall labeled as D8 in another 1-UC Fe(Te,Se)/STO sample (s2).

a, The STM topography of the domain wall. (4.8×4.8 nm², Vₛ = 0.04 V, Iₛ = 2.5 nA).

b, Spatial distribution of Δ₁(r) measured in the same area in a. White dashed lines in b are guides to the eye.

c, The magnitude of the Fourier transform of b. Orange crosses are at q = (±Q₁₁, 0), (0, ±Q₁₁). The modulation wavevector Q ≈ ±0.28Q₁₁ is marked by dashed blue circles. Black dashed lines in a and b mark the edges of the domain wall.
Extended Data Fig. 6 | Spatial modulations of the LDOS, superconducting gap energy and coherence peak height at one domain wall labeled as D9 in another 1-UC Fe(Te,Se) film (s2). a, The STM topography of the domain wall. (4.3×5.4 nm², V = 0.04 V, I = 2.5 nA). b, dI/dV map g(r, V = 3 mV) taken at the same area in a. c, The magnitude of the Fourier transform of b. d, Spatial distribution of Δ₁(r) measured in the same area in a. e, The magnitude of the Fourier transform of d. f, Spatial distribution of superconducting coherence peak height measured in the same area in a. g, The magnitude of the Fourier transform of f. h, Line profiles along the (0,0) to (1,0) Q_Fe direction in c, e, and g. Peaks at Q = 0.28Q_Fe appear for all curves. The profiles are normalized and shifted vertically for comparison. Orange crosses in c, e, and g are at q = (±Q₁Fe,0), (0,±Q₁Fe). Black dashed lines in a, b, d and f mark the edges of the domain wall. Orange dashed lines in b, d and f are guides to the eye.
Extended Data Fig. 7 \(\rho(r, V)\) map (a), corresponding Fourier transform \(\rho(q, V)\) (b), spatial variation of the amplitude \(A^{2Q}(r)\) (c) and the phase \(\phi^{2Q}(r)\) (d) of the 2Q \(\rho(r)\) modulation at 2 mV at the domain wall shown in Fig. 4 (D3). \(\rho(q, V)\) exhibits maxima at around \(\pm 2Q\) (marked by dashed blue circles). The amplitude of the modulation is restricted to the domain wall region and the phase of the 2Q modulation is nearly uniform in the domain wall region, indicating the existence of the 2Q CDW. The averaging length scales in c and d are denoted by dashed circles. The edge of the domain wall is marked by the dashed lines in a, c and d.
Extended Data Fig. 8 | Comparison between the atomic lattice and 2Q CDW state at the domain wall shown in Fig. 4 (D3). a, b, Spatial variation of the phase $\psi^{T(r)}(r)$ of the STM topology $T(r)$ modulation at $0.5\mathbf{Q}_\perp$ (a) and the phase $\psi^{\rho(2mV)}(r)$ of the $\rho(r, V = 2\, mV)$ (charge density) modulation at $2\mathbf{Q}$ (b). Topological defects in a and b are marked by black dots. c, d, Spatial variation of the amplitude $|A^{T(r)}_{Q_\perp}(r)|$ of the STM topology $T(r)$ modulation at $0.5\mathbf{Q}_\perp$ (c) and the amplitude $|A^{\rho(2mV)}_{Q}(r)|$ of the $\rho(r, V = 2\, mV)$ modulation at $2\mathbf{Q}$ (d). The spatial variations of the phase and amplitude show many differences between the atomic lattice and 2Q CDW, indicating that the 2Q CDW is independent of the atomic lattice. The averaging length scales are denoted by dashed circles. The edge of the domain wall is marked by the dashed lines.
Extended Data Fig. 9 | See next page for caption.
Extended Data Fig. 9 | Possible theoretical scenarios for the PDW state at the domain wall.

a, Schematic of the PDW state at the domain wall. \( \Delta_{SC}^{1,2} \) and \( \Delta_{SC}^{DW} \) represent the zero-momentum superconductivity in the domains (1 and 2) and at the domain wall, respectively. 

b, Illustration of the finite center of mass momentum, equal-spin pairing in the presence of only Rashba SOC. \( \langle \langle \sigma_{k,k+Q}, \uparrow \rangle \rangle \) and \( \langle \langle \sigma_{k,k-Q}, \downarrow \rangle \rangle \) are equal-spin pairing order parameters where one spin sector carries momentum +Q and the other spin sector −Q.

c, Illustration of the finite center of mass momentum equal-spin pairing in the presence of both Rashba and Dresselhaus SOC. In this case, the triplet equal-spin pairing order parameters \( \langle \langle \sigma_{k,k+Q}, \uparrow \rangle \rangle \) and \( \langle \langle \sigma_{k,k-Q}, \downarrow \rangle \rangle \) can have both spin components \( \sigma \), i.e., each equal-spin pairing sector carries both nonzero momentum +Q and −Q, giving rise to the equal-spin pairing PDW state. 

d, f, Calculated tunneling density of states \( \rho_i(\omega) \) with \( i \) the site index, for two examples with \( Q = 2\pi/12 \) (d) and \( Q = 2\pi/4 \) (f), where the insets show the zoom-in feature around the coherent peaks. 

e, g, The spatial modulations of the superconducting gap extracted from the corresponding coherence peak positions for the \( Q = 2\pi/12 \) (e) and \( Q = 2\pi/4 \) (g).