A strongly inhomogeneous superfluid in an iron–based superconductor

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Although the possibility of spatial variations in the superfluid of unconventional, strongly correlated superconductors has been suggested1–7, it is not known whether such inhomogeneities—if they exist—are driven by disorder, strong scattering or other factors. Here we use atomic-resolution Josephson scanning tunnelling microscopy to reveal a strongly inhomogeneous superfluid in the iron-based superconductor FeTe0.55Se0.45. By simultaneously measuring the topographic and electronic properties of the superconductor, we find that this inhomogeneity in the superfluid is not caused by structural disorder or strong inter-pocket scattering and is not correlated with variations in the energy required to break electron pairs. Instead, we see a clear spatial correlation between the superfluid density and the quasiparticle strength (the height of the coherence peak) on a local scale. This result places iron-based superconductors on equal footing with copper oxide superconductors, where a similar relation has been observed on the macroscopic scale. Our results establish the existence of strongly inhomogeneous superfluids in unconventional superconductors, excluding chemical disorder and inter-band scattering as the causes of the inhomogeneity, and shed light on the relation between quasiparticle character and superfluid density. When repeated at different temperatures, our technique could further help to elucidate what local and global mechanisms limit the critical temperature in unconventional superconductors.

Superconductivity emerges when electrons pair up to form Cooper pairs and then establish phase coherence to condense into a macroscopic quantum state, the superfluid. Cooper pairing is governed by the binding energy of the pairs, \( \Delta_{CP} \), and the phase coherence (or stiffness) governs the superfluid density \( n_{SF} \) (see Extended Data Table 1 for symbol definitions). For conventional superconductors, like aluminium or lead, the superfluid density is spatially homogeneous because

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**Fig. 1 | Principles of Josephson STM.**  
(a) Schematic of a Josephson junction consisting of the tip and the sample (see Extended Data Table 1 for variable definitions).  
(b) Schematic energy diagram of quasiparticle tunnelling between the tip and the sample. Solid black lines indicate the density of states (horizontal axis) as a function of energy (vertical axis); filled (empty) states are denoted with blue (red); dashed lines indicate the Fermi level, \( E_F \). When the voltage bias \( V_B \) is larger than \( (\Delta_{CP,t}+\Delta_{CP,s})/e \), quasiparticles can tunnel (\( e \) is the electron charge).  
(c) Current–voltage characteristic curve (blue) for quasiparticle tunnelling. The dashed lines indicate zero values. The arrow indicates \( 2(\Delta_{CP,t}+\Delta_{CP,s})/e \).  
(d) Equivalent circuit diagram of the Josephson junction; the complex impedance \( Z(\omega) \) represents the electromagnetic environment.  
(e) Schematic of inelastic Cooper pair tunnelling in a Josephson junction. A Cooper pair interacts with the environment by emitting energy of \( h \nu = 2eV_B \) (wavy arrow; \( h \), Planck constant; \( \nu = \omega/2\pi \), frequency) and subsequently tunnels across the junction.  
(f) Simulated current–voltage curves for Cooper pair tunnelling, obtained using the IZ and \( P(E) \) models. Both curves exhibit a maximum current \( I_{max} \) at a finite bias proportional to \( I_C^2 \) (\( I_c, \) critical supercurrent).

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the lattice constant is much smaller than the size of the Cooper pair (usually hundreds of nanometres) and because the large superfluid density guarantees a high phase stiffness. In unconventional, strongly correlated superconductors the situation is very different from that in conventional superconductors for the following reasons: (i) the size of the Cooper pairs, which is roughly given by the coherence length, is generally smaller; (ii) the superfluid density is smaller; (iii) higher disorder exists, owing to dopant atoms or intrinsic tendencies for phase separation or charge order; and (iv) the sign of the superconducting gap changes. Despite much progress, we lack a theoretical understanding of strongly correlated superconductors. It has been proposed that, in principle, spatial variations can exist in the superfluid density. Very similar ideas have been discussed thoroughly in the context of superconductor–insulator transitions or Bose–Einstein condensation of electronic liquids. However, little is known about the local physics in such systems because of the technical challenges associated with visualizing the superfluid density on the atomic scale, especially when simultaneously probing the density of states to investigate the origin of the inhomogeneity.

The pair-breaking gap (the energy required to break a Cooper pair) and the superfluid density should be accessible through two distinct spectroscopic signatures in a tunnelling junction between superconductors (Fig. 1a). The first one is visible in the single-particle channel, where Bogoliubov quasiparticles with energies larger than the pair-breaking gaps transport the charge, as shown in Fig. 1b. In the case of the scanning tunnelling microscopy (STM) configuration relevant to this Letter, one of the superconductors is the tip (with gap \( \Delta_{CP,t} \)) and the other is the sample (with gap \( \Delta_{CP,s} \)), leading to a total energy gap of \( 2(\Delta_{CP,t} + \Delta_{CP,s}) \) (Fig. 1c). The second spectroscopic feature is observed at bias energies close to the Fermi energy, \( E_F \), where one can access the Cooper pair channel that yields information about the superfluid density. Voltage-biased Josephson tunnelling in our STM configuration differs from the case of planar junctions in that: (1) the capacitive energy \( E_C \) is much bigger than the Josephson energy, \( E_J \), turning the environmental impedance into a relevant quantity, and (2) the thermal energy is relatively high. Figure 1d shows the equivalent circuit for a generic junction in an STM environment.

We calculate the current–voltage characteristics of Josephson tunnelling using two different theoretical frameworks: IZ and \( P(E) \). The former (named after its developers, Ivanchenko and Zilberberg) models the environment as Ohmic and assumes that the thermal energy exceeds the Josephson energy\(^{11} \). The latter (named after the probability function, which is central to this theory) is a quantum mechanical treatment of Cooper pair tunnelling in ultrasmall junctions\(^{15} \). For our configuration, the qualitative predictions obtained from both theoretical descriptions are similar: a Josephson current flows at small bias and exhibits a maximum within a few microvolts of the Fermi energy (Fig. 1e), which is reflected in a conductance spectrum that shows a peak at zero applied bias. The maximum Josephson current (arrow in Fig. 1f) is proportional to the square of the critical current \( I_C \) of the junction. In single-band, s-wave superconductors, the superfluid density is then proportional to \( (I_C R_N)^2 \), where \( R_N \) is the normal-state resistance, and is interpreted as the density of condensed Cooper pairs\(^{12} \). In multi-band or unconventional superconductors, the superfluid density defined this way represents the superposition of different contributions from different bands, with weights depending on the relative phases, \( I_C R_N \propto \sum n_i (\Delta_{CP,s})^{1/2} \cos \chi_i \), where \( n_i \) are individual superfluid densities of the different bands and \( \chi_i \) their relative phases (see also Methods). When tunnelling locally, one has to convert from a band basis to an orbital basis and consider the overlap of each kind of orbital with the different bands, as well as the individual tunnelling matrix elements for the different orbitals. One can still extract spatial variations in the superfluid using the definition above, if the ratios between the tunnelling matrix elements are spatially constant or when the superconducting phase is not strongly related to the orbitals. Importantly, the superfluid density thus defined cannot be simply interpreted as the total density of Cooper pairs for unconventional or multi-band superconductors, including the one investigated here. Notably, the multiplication with \( R_N \) in \( (I_C R_N)^2 \) further allows us to disentangle the measured superfluid density from variations in the coupling between the tip and the superfluid, which might vary spatially\(^{15,14} \). Spatially imaging a superfluid using Josephson STM techniques\(^{15} \) has thus far been achieved in two instances. First, a pair density wave was discovered in a copper oxide sample\(^{13} \), by exfoliating pieces of the sample onto the STM tip and imaging it with a resolution of about 1 nm. Second, the superfluid of a Pb(111) surface was resolved with atomic resolution, by using the sample material to coat the STM tip\(^{14} \).

In this study, we investigate the unconventional iron-based superconductor FeTe\(_{0.55} \)Se\(_{0.45} \). Iron-based superconductors are moderately strongly correlated, with Hund’s rule and orbital selectivity playing important roles\(^{16} \). We chose FeTe\(_{0.55} \)Se\(_{0.45} \) because it encompasses the key properties of unconventional superconductivity. Furthermore, its nodless gap structure\(^{17,18} \) and the possibility to scan at low junction resistances facilitate Josephson experiments. FeTe\(_{0.55} \)Se\(_{0.45} \) is considered not to be in the ‘dirty’ Bardeen–Cooper–Schrieffer (BCS) limit and has a low average superfluid density similar to that of copper oxide high-temperature superconductors\(^{19,20} \). We cleave the single crystals at 30 K and insert the samples into our cryogenic STM system with rigorous electronic filtering (see Extended Data Fig. 3). All measurements are performed at an effective electron temperature of 2.2 K. The topograph (Fig. 2a) shows atomic resolution and contrast differences that stem from the tellurium or selenium inhomogeneities; we
Further verify that the interstitial iron concentration is negligible. We use a mechanically sharpened platinum–iridium wire with its apex coated with lead, which is an $s$-wave superconductor with a relatively large gap\(^{14}\) of about 1.3 meV. We characterize its properties on an atomically flat Pb(111) surface (see Methods).

These preparations enable us to acquire Josephson tunnelling spectra and maps on FeTe\(_{0.55}\)Se\(_{0.45}\). Figure 2 shows current and differential conductance spectra acquired at the location marked by a cross in Fig. 2a. The data agree well with expectations from the IZ and $P(E)$ models, and reproduce small oscillation features seen previously in elemental superconductors and explained by a tip-induced antenna mode\(^{14,21}\). Decreasing the junction resistance shows an increase in the critical current expected for a Josephson tunnelling junction (Extended Data Fig. 4). The rate of the increase is lower than that expected for simple $s$-wave junctions but more consistent with theoretical predictions for an $s_\pm$ pairing symmetry in the sample, where states with both positive and negative gaps tunnel\(^{22}\). We further note a small kink in the Josephson current at 25 µeV of yet unknown origin.

In Fig. 3a, b we show an atomic-resolution map of the superfluid density as defined above, extracted from about 16,000 individual spectra, as well as the topographic image, with the two images aligned to each other on the atomic scale at each point. The most striking finding of our experiment is the strong inhomogeneity of the superfluid over length scales of the order of the coherence length, that is, a few nanometres. To illustrate this, we show in Fig. 3c a series of individual raw spectra normalized by the normal-state resistance in the superfluid. The most obvious possible causes are structural disorder and strong quasiparticle scattering. The structural disorder stems from the effective FeSe and FeTe alloying, which is clearly visible in the topographic images (Figs. 2a, 3a). Surprisingly, the variations in the superfluid are not correlated to these structural features, with the exception of a few impurity atoms that lead to a strong suppression of the Josephson current (see Extended Data Fig. 10). The strength of the quasiparticle scattering is visible in the quasiparticle interference (QPI) pattern and is dominated by inter-pocket scattering in FeTe\(_{0.55}\)Se\(_{0.45}\) (ref. \(^{17}\)). In Fig. 3d we identify areas of strong scattering with red contours, which are obtained by Fourier-filtering the QPI data, to distinguish between strong- and weak-scattering regions (see Methods). Again, there is no correlation between these regions and the superfluid density. We cannot exclude that the superfluid density is influenced by potential scatterers that are not visible in our measurement, remnant short-range magnetic order, or possible phase separations at higher energies. Given the putative $s_\pm$ pairing symmetry of the sample, as mentioned above, one could also consider a scenario involving spatially varying tunnelling matrix elements between the tip and orbitals that are coupled to gaps with opposite signs, leading to a spatially varying suppression of the Josephson current\(^{22}\). However, in FeTe\(_{0.55}\)Se\(_{0.45}\) the gap sign is not strongly related to the orbital character\(^{18,23}\), and we do not observe the imprint that a relative change in the tunnelling matrix elements of the different orbitals would leave on the local density of states and the topography. More generally, the fact that prominent effects such as the chemical disorder and the inter-pocket QPI do not influence the superfluid indicates that the inhomogeneity in the superfluid density is intrinsic.

We now return to the relation between the pair-breaking gap and the superfluid density. We extract the pair-breaking gap energy, as well as the height of the coherence peaks (which will prove to be important later) by fitting the coherence peaks of each spectrum to find the energy

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**Fig. 3** Visualizing the superfluid in FeTe\(_{0.55}\)Se\(_{0.45}\). a, $25 \times 25$ nm\(^2\) topographic image of FeTe\(_{0.55}\)Se\(_{0.45}\) ($V_{\text{set}} = -6$ mV, $I_{\text{set}} = 0.12$ nA). b, Spatially resolved map of ($I/R_0$)$^2$, representing the superfluid density as discussed in the text ($V_{\text{set}} = -6$ mV, $I_{\text{set}} = 5$ nA, $V_{\text{mod}} = 30$ µV peak to peak). c, Series of differential conductance spectra obtained along the blue line in b, multiplied by the normal-state resistance around $E_F$. d, Conductance map at $V_b = +3.6$ mV. Areas with strong quasiparticle interference patterns are marked by red contours, which are obtained by Fourier filtering (see Extended Data Fig. 8) of the QPI data using the filter shown in the inset (red circles). Inset, Fourier transform; crosses indicate Bragg peak locations. e, Pair-breaking gap map, with $\Delta = \Delta_{\text{Bragg}} - \Delta_{\text{QPI}}$, the inset shows a typical spectrum. f, Coherence peak-height map (QPS), extracted simultaneously with the pair-breaking gap, as indicated in the inset. All maps in b–f were obtained in the same field of view as that used for the topograph in a, and the images are aligned to each other at each point using the simultaneously acquired topographs. The setup conditions for d–f are $V_{\text{set}} = -6$ mV, $I_{\text{set}} = 0.3$ nA and $V_{\text{mod}} = 400$ µV peak to peak.
of the maxima. Figure 3e shows the gap map for the same field of view as that used for the Josephson map. The gap variations agree with previous reports\textsuperscript{24}. It is clear that the pair-breaking gap is independent of the superfluid density; instead, we find a correlation to the quasiparticle character, as described below.

In unconventional superconductors, there is a recurring theme that connects quasiparticle excitation line shapes with the presence of superconductivity: photoemission demonstrates that incoherent quasiparticles in the normal state become coherent below the critical temperature\textsuperscript{18,25}, $T_C$. Previous STM measurements showed Bogoliubov QPI patterns at low energies that were even sharper than theory would predict but vanished well below the gap energy\textsuperscript{26}. Those measurements suggested a remarkable relation between the average quasiparticle excitation spectrum and superconductivity, but did not address the inhomogeneous character of unconventional superconductors.

Although recently a relation between superfluid density and quasiparticle character has been conjectured to hold also locally for single-layer superconductors\textsuperscript{29,30}, there also exist indications for pairing in the BEC or superfluids\textsuperscript{29,30}. There also exist indications for pairing in the BEC or superfluids\textsuperscript{29,30}. The length scales of the superfluid inhomogeneity and of its correlation to the QPS (Fig. 4b, inset) are of the same order as the average inter-pocket distances of the maxima. Figure 3e shows the gap map for the same field of view, with atomic resolution. Indeed, we find a striking correlation between the superfluid density and QPS, QPI, $\Delta_C$, and the topographic height (see Extended Data Fig. 9).

In summary, we detected and directly imaged a strongly inhomogeneous superfluid and simultaneously measured the electronic and topographic properties in the same field of view, with atomic resolution. We found that the inhomogeneity of the superfluid is not caused by structural disorder resulting from the Se/Te alloying, by inter-pocket scattering or by variations of the pair-breaking gap energy (Fig. 4b, inset). Instead, the superfluid density shows strong positive correlation with the sharpness of the quasiparticle peak: superconductivity appears to be needed for coherent quasiparticles locally, on the length scale of Cooper pairing. It will be instructive to use the techniques described here to investigate the superfluid density in other materials, including superconductor–insulator transitions, disordered conventional superconductors or twisted bilayer graphene\textsuperscript{21,33}. Lastly, we anticipate that future temperature-dependent superfluid density and gap measurements will elucidate what local and global mechanisms limit $T_C$ in unconventional superconductors.

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Fig. 4 | Correlation between ($I_C$-$R_N$)$^2$ and coherence peak height. a, Sorted spectra of the coherence peak height ($V_{set} = −6\, \text{mV}$, $I_{set} = 0.3\, \text{nA}$) and the zero-bias Josephson peak (inset; $V_{set} = −6\, \text{mV}$, $I_{set} = 5\, \text{nA}$). The spectra were sorted by binning of the superfluid density map shown in Fig. 3b. The colours correspond to the quasiparticle strength indicated by the colour bar in Fig. 3f. b, Correlation between the coherence peak height and the superfluid density extracted from ($I_C$-$R_N$)$^2$ as discussed in the text, yielding a correlation factor of 0.58 (dashed line). The inset shows the distance dependence of the correlation factors between the superfluid density and QPS, QPI, $\Delta_C$, and the topographic height (see Extended Data Fig. 9).
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METHODS

Sample preparation and measurement. FeTe₂Se₁₋ₓ single crystals with an optimal concentration of x = 0.55 are grown by the Bridgman method. The magnetic susceptibility curve as a function of temperature shows a sharp transition at TC ≈ 14.5 K. To perform STM and scanning tunnelling spectroscopy measurements, we use a modified commercial scanning tunnelling microscope (USSM-1500, Unisoku Co., Ltd). The samples are cleaved in ultrahigh vacuum (base pressure, Pbase = 1 × 10⁻¹⁰ mbar) at low temperature (T ≈ 30 K) and are immediately mounted in the pre-cooled STM head (T ≈ 2.2 K) to prevent surface reconstruction and contamination. Mechanically grinded Pt–Ir wires are used as STM tips.

To make a superconducting tip, we indent the metallic tip into a Pb(111) surface cleaned by repeated sputtering and annealing. The STM topographs in Fig. 2a (Fig. 3a) are acquired by a constant current mode with Vset = −10 mV (+6 mV) and Iset = 5.0 nA (0.12 nA). A standard lock-in technique is employed for the tunnelling spectrum measurements. We apply a voltage modulation of 0.1–0.2 mV for the quasiparticle tunnelling spectra (Figs. 3d–f) and 0.01–0.02 mV for the Josephson tunnelling spectra (Figs. 2c, 3b) with a frequency of 887 Hz. The former is obtained with Vset = −6 mV and Iset = 0.3 nA and the latter with Vset = −6 mV and Iset = 5 nA. All measurements reported here are performed at 2.2 K.

Determined the superfluid density with Josephson STM. In Josephson STM, the Josephson junction is formed between an STM tip and a sample (both superconducting) that are separated by a vacuum barrier (Extended Data Fig. 1). The tunnelling current of Cooper pairs contains information about the superfluid density in the sample or tip. Starting from the wavefunctions ψsf of the superconductors on the tip (t) and the sample (s),

\[ \psi_{s(t)} = \sqrt{\Psi_{s(t)}} \exp \left(-i \frac{\phi_{s(t)}}{\hbar} \right) \]

where \( \Psi_{s(t)} \) is the superfluid density and \( \phi_{s(t)} \) is the phase of the condensate in the sample or tip, it can be shown that the supercurrent follows the Josephson relation

\[ I = I_c \sin (\phi_t - \phi_s) \]

with

\[ I_c = \sqrt{\Psi_{s(t)} \Psi_{s(t)}} \]

(\( \kappa \) is a coupling constant) being the maximum (critical) supercurrent that the junction can sustain. Assuming the superfluid density in the tip to be constant, one can treat the critical supercurrent as a measure for probing the superfluid density in the sample. We note that although the relation between critical current and superfluid density is straightforward in single-band s-wave superconductors, it can become more complicated in multiband systems, where the critical current is related to effective superpositions of the superfluid densities. Because the ratios between tunnelling elements into different orbitals is spatially constant, and because the gap sign is only weakly coupled to the different orbitals, the changes in \( I_c \) that we measure reflect changes of the superfluid density in the sample (see the main text). We also note that the Josephson effect and its phenomenology in an ultra-small STM junction is somewhat different from planar junctions, as we will describe in the following sections.

Energy scales relevant in ultra-small junctions. We start with a description of the measurement circuit shown in Fig. 1d. Our Josephson tunnelling junction consists of a Pb-coated tip and an atomically flat cleaved Fe-based superconductor (inset of Extended Data Fig. 1). The corresponding circuit diagram of this Josephson junction involves the following key components: the critical supercurrent \( I_c \), the normal-state resistance \( R_N \) and the junction capacitance \( C_J \) biased by a voltage source at \( V_b \). In addition, the junction is coupled with a complex impedance \( Z_C \), which corresponds to the electromagnetic environment of the junction and accounts for any form of dissipation that may be present.

The relevant energy scales in such ultra-small junctions are the charging energy \( E_C \), the Josephson energy \( E_J \) and the thermal energy \( E_T \). The charging energy is related to the Coulomb energy change that occurs when a Cooper pair tunnels across an ultra-small junction. The charging energy for Cooper pairs is \( E_C = (2e)^2 / (2C) \), because they carry charge of 2e. The Josephson energy is a measure of how strongly the two superconducting condensates forming the junction are coupled to each other (for this reason, it is also called coupling energy) and is proportional to the critical supercurrent, \( E_J = hC_J / (2e) \), where \( h \) is the reduced Planck constant. The thermal energy (also known as thermal noise) corresponds to the kinetic energy due to non-zero temperature, \( E_T = k_B T \), where \( k_B \) is the Boltzmann constant.

In contrast to planar junctions, ultra-small STM junctions have larger Coulomb gaps. We estimate \( C_J \approx 1 \) fF for the STM junction and thus \( E_C = 276 \mu eV \) (about 3.2 K). In addition, a typical tunnelling junction has a relatively large normal-state resistance \( R_N \) (>0.1 MΩ). Using the formula suggested by Ambegaokar and Baratoff, }

\[ E_J = \frac{\pi h}{4e} \left( \Delta_{CP} \tan \left( \frac{\Delta_{CP}}{2k_B T} \right) \right) \]

the Josephson energy \( E_J \) corresponds to 3.5 μeV (about 40 mK) for a symmetric Josephson junction with normal-state resistance \( R_N = 1 \) MΩ and pair-breaking gap \( \Delta_{CP} = 1 \) meV. It is important to compare these relevant energy scales to be able to choose the applicable model to describe the current–voltage \((I–V)\) characteristics of the junction, and to eventually extract the superconducting order parameter.

In our experiment, \( E_T \) is smaller than both the measurement temperature (2.2 K) and \( E_J \) (from \( E_J > k_B T \)).

RCSJ model, IZ model and \( P(E) \) theory. There exist different frameworks to describe the \( I–V \) characteristics of a Josephson junction. The three most commonly used ones are the resistively and capacitively shunted Josephson junction (RCSJ) model, the IZ model and \( P(E) \) theory. We briefly discuss the most important details of these three frameworks below.

The standard RCSJ model\(^{44}\) is applicable when the junction is isolated from its electromagnetic environment \( Z_{env}(\omega) = 0 \), and temperature fluctuations are usually neglected \( (E_T < E_J) \). In this model one solves the equation the temporal evolution of the phase difference \( \Delta \phi = \phi_s - \phi_t \) for each current–bias condition. The voltage drop across the junction is then calculated from the time derivative of the phase difference according to the relation\(^{46} \)

\[ \frac{\Delta \phi}{\Delta t} = \frac{\Delta \phi}{\Delta t} \]

The evolution of the phase is similar to that of a classical particle moving in a tilted-washboard potential landscape, as illustrated in Extended Data Fig. 2a. When the bias current is \( I_{bias} \leq I_c \), the particle is trapped inside a potential minimum, resulting in zero voltage drop across the junction because \( \Delta \phi / \Delta t = 0 \). For \( I_{bias} \geq I_c \), the potential minima are not deep enough to trap the particle. In this case, the phase evolves in time and a voltage drop proportional to \( I_{bias} \) is observed.

As shown in Extended Data Fig. 2b, a typical \( I–V \) curve calculated using the RCSJ model clearly exhibits a current equal to \( I_c \) at zero bias.

When temperature fluctuations become important \((E_T > E_J ; \text{see Extended Data Fig. 2b})\) and for the simplest case of an Ohmic environment \( Z_{env}(\omega) = R \), the RCSJ model can be solved analytically. In this phase-diffusive limit, Ivanenko and Zilberman calculated the \( I–V \) characteristics\(^{48} \) to find that the effect of the environment is introduced by a finite slope at zero bias.

In this relation, the critical current appears as a scaling factor. Extended Data Fig. 2f shows the \( I–V \), \( I–dI/dV \) curves for inelastic Cooper pair tunnelling in the Coulomb blockade regime with an antenna-mode environmental impedance, which is natural to assume in an STM setup\(^{48,49} \). The formalism used in this section allows us to interpret each Josephson STM spectrum\(^{48,50,51} \), as well as the recently measured Josephson STM maps (see refs \(^{13,81}\)).

Characterizing the superconducting tip on a Pb(111) surface. To form a Josephson junction, we make superconducting STM tips by indenting a sharp metallic Pt–Ir tip into a clean Pb(111) surface. Pb is a conventional superconductor with \( T_C = 7.2 \) K. The indenterations are repeated until the tip shows a pair-breaking gap equal to that of bulk Pb (ref. \(^{19}\)). The bulk-like superconductivity of the tip is verified using tunnelling spectroscopy. The differential conductance spectrum exhibits a finite superconducting gap determined by two sharp coherent peaks. Because of the superconducting tip, these are much sharper than what one would expect from conventional thermal broadening. In the measured spectrum shown in Extended Data Fig. 3b, all quasiparticle states of the sample are shifted by the superconducting gap of the tip, and thus the tunnelling spectrum clearly shows sharp coherent peaks at an energy equal to the sum of the two superconducting gaps, \( \Delta_{CP1} + \Delta_{CP2} = 2.6 \) meV. We fit the spectrum with the formula

\[ I(V) = \frac{e^2 \Delta_{CP1} \Delta_{CP2}}{4e} \left( P(2eV) - P(-2eV) \right) \]
\[
\frac{dI}{dV} = \frac{G_N}{e} \int_0^\infty \frac{d\Delta}{\Delta} \left[ \frac{\partial \Delta}{\partial \Delta} |f(T, \epsilon) - f(T, \epsilon + eV)| - \frac{\partial \epsilon}{\partial \Delta} \right] d\epsilon
\]

where \(G_N\) is the normal-state conductance, \(\epsilon\) is the integration variable for energy and \(f(T, \epsilon)\) is the Fermi–Dirac distribution at temperature \(T\). For the density of states of the tip and sample, \(D_{00}\), we use a modified Dynes formula:

\[
P_{\Delta CP} = \left| \frac{\text{Re} \text{sgn}(\epsilon)}{\sqrt{\epsilon^2 + 2\gamma \epsilon - \Delta_{\Delta CP}^2}} \right|
\]

where \(\gamma\) represents a phenomenological broadening term. We find good agreement between the measured data and the model for \(\Delta_{\Delta CP} = \Delta_{\Delta CP} = 1.3\) meV, \(\gamma = 45\) meV and \(T = 2.2\) K. Here, the effective temperature of 2.2 K is estimated by fitting the spectra acquired with a normal Pt–Ir tip on a Pb(111) superconducting surface (Extended Data Fig. 3a). The parameters \(\Delta_{\Delta CP}\) and \(\gamma\) are also free fitting parameters.

**Critical current as a function of normal-state conductance on FeTe0.55Se0.45**

The superconducting Pb-coated tip is then used on FeTe0.55Se0.45. Extended Data Fig. 4a, b shows the \(I_{CP}\)-dependent \(I-V\) curves. We observe that for decreasing \(G_N\), the current reaches a maximum value at finite bias around the Fermi level, which is an indication of Cooper pair tunnelling. In the differential conductance spectra shown in Extended Data Fig. 4c, d, we can resolve sharp resonances at finite bias originating from the energy exchange between Cooper pairs and the electromagnetic environment of the junction.

According to the IZ model, the maximum (\(I_{\Delta\Delta CP}\)) in the \(I-V\) characteristic curves is related to the critical supercurrent according to the formula

\[
I_{CP} = \frac{8G_{\text{max}}k_B T}{h}
\]

However, we can use the maximum from our \(I-V\) curves and use the above formula for quantifying \(I_{CP}\) (we use \(T = 2.2\) K, which is equal to our measurement temperature). In Extended Data Fig. 4e, we plot \(I_{CP}\) as a function of the normal-state junction conductance, \(G_N = 1 / R_N\). A linear trend is observed, which is consistent with the so-called Ambegaokar–Baratoff formula. A linear fit to our data gives a slope of 1.534 meV, which is used to estimate \(\Delta_{\Delta CP}\) from the formula of an asymmetric junction

\[
I_{CP} = \frac{2}{\epsilon} \left( \frac{\Delta_{CP} \Delta_{CP}'}{\epsilon - \Delta_{CP} + \Delta_{CP}'} \right) K\left( \frac{\Delta_{CP} - \Delta_{CP}'}{\Delta_{CP} + \Delta_{CP}'} \right)
\]

where \(K(x)\) is the elliptic integral of the first kind of the function. Assuming \(\Delta_{CP} = 1.3\) meV, we find \(\Delta_{\Delta CP} = 0.67\) meV. This is to be compared with the gap that we read from our conductance spectra. We find that the coherence peak is located at 3.08 meV. Subtracting \(\Delta_{\Delta CP}\) from the coherence peak location gives \(\Delta_{CP} = 1.68\) meV. We believe that this deviation can be attributed to the unconventional superconducting nature of FeTe0.55Se0.45. It has been predicted theoretically that for Cooper pair tunnelling between a conventional s-wave superconductor and an unconventional s±-multiband superconductor (here FeTe0.55Se0.45), \(I_{CP}\) grows linearly with \(G_N\). However, for that case the slope is expected to be lower than for the single-band, s-wave case. A reduction in the Josephson current was also observed in a multiband superconductor without a sign-changing gap using an s-wave superconducting tip. We expect that better calculations of the orbital decomposition of the gap structure and of the individual tunnelling processes for different orbitals will allow the quantitative interpretation of our data.

**Visualization of superfluid density for samples with inhomogeneous \(G_N\)**

To visualize the spatial variations of the superfluid density, we record differential conductance spectroscopic maps on a grid of points \((r_x, r_y)\). By taking the derivative of the IZ formula with respect to the voltage, we obtain

\[
\frac{dI}{dV} = \frac{I_{\text{Z conv}}^2}{2} \frac{V_c^2 - V^2}{(V^2 + V_c^2)^2}
\]

We fit our spectrum with the above formula using the pre-factor \(I_{\text{Z conv}}^2/2\) and \(V_c\) as free parameters. A typical IZ fit of the conductance spectrum is shown in Extended Data Fig. 4f. This allows us to construct atomic-scale \(I_{CP}(r)\) maps, which express the magnitude of the critical supercurrent as a function of location \(r\). Extended Data Fig. 5a, b shows examples of such maps, obtained in the same 25 × 25 nm² field of view and using opposite setup bias (−10 mV in Extended Data Fig. 5a and +10 mV in Extended Data Fig. 5b). These maps reveal spatial variations of the critical supercurrent on a small length scale of a few nanometres.
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Extended Data Fig. 1 | Schematic of a Josephson tunnelling junction. The Josephson junction consists of a Pb-coated platinum–iridium (Pt-Ir) tip and an atomically flat FeTe$_{0.55}$Se$_{0.45}$ surface, separated by a thin vacuum barrier. Inset, the Fe lattice is encapsulated by the chalcogen atoms selenium (Se) and tellurium (Te).
Extended Data Fig. 2 | Various models used to describe Josephson tunnelling and their $I$–$V$ characteristic curves. a, Tilted-washboard potential $U(\Delta \phi)$ for $I_{\text{bias}} < I_C$. b, $I$–$V$ curve calculated using the RCSJ model for $Z(\omega) = 0$ and $T = 0$ K. The voltage drop is zero (dissipationless supercurrent) until $I_{\text{bias}} > I_C$. c, Thermal phase fluctuations (illustrated by yellow dashed arrows) in the tilted-washboard potential. d, $I$–$V$ and corresponding differential conductance $dI/dV$, calculated using the IZ model for $Z(\omega) = R$ and the phase-diffusive regime ($E_T > E_C$). For the simulation we used $R_N = 1$ kΩ and $T = 2$ K. e, Energy diagram of sequential inelastic Cooper pair tunnelling. f, $I$–$V$ and differential conductance $dI/dV$, calculated using $P(E)$ theory for a $Z(\omega)$ corresponding to the tip-induced antenna mode of energy $h\nu = 200 \mu$eV and the Coulomb blockade regime ($E_T < E_C$). The $I$–$V$ and $dI/dV$ curves in d and f correspond to a voltage of $V_B$ inside the gap.
Extended Data Fig. 3 | Tunnelling spectra of Pt–Ir/Pb and Pb/Pb junctions. a, Differential tunnelling spectra of a Pt–Ir/Pb junction with (blue dots) and without (red dots) electronic filtering using home-built lumped-element low-pass filters in series with commercial 1.9-MHz low-pass filters and grounding for all non-essential lines ($V_{\text{set}} = +5$ mV, $I_{\text{set}} = 0.10$ nA). We use a modified Dynes formula to fit our spectra, and the results give effective temperatures of 2.38 K (green line) and 2.20 K (yellow line). The other parameters are the same in both cases ($\Delta_{CP} = 1.30$ meV and $\gamma = 50$ μeV). b, Normalized conductance spectrum (blue curve) of a Pb/Pb junction, acquired with a junction resistance of 5 MΩ ($V_{\text{set}} = +5$ mV, $I_{\text{set}} = 1.0$ nA). The fit (red dashed curve) is consistent with the quasiparticle spectrum of a symmetric Josephson junction with a pair-breaking gap of 1.3 meV at 2.2 K.
Extended Data Fig. 4 | Josephson tunnelling spectroscopy in a Pb/FeTe$_{0.55}$Se$_{0.45}$ junction. a, b, $R_N$-dependent $I$–$V$ curves (see key in e). c, d, Corresponding $R_N$-dependent $dI/dV$ curves, multiplied by $R_N$. The datasets are offset for clarity. For decreasing $R_N$, the zero-bias peak and the small modulations (with a period of 0.1 meV) induced by Cooper pair tunnelling become more pronounced. e, Linear relation (black dashed line) between the critical Josephson current $I_C$ and the normal-state conductance, $G_N$. Each point is the average of 20 points extracted from 20 $I$–$V$ curves, as described in the main text. For each $G_N$, we also extract the standard deviation of $I_{max}$ and obtain the error bars shown in the figure by performing error propagation via the IZ formula. The red dashed line corresponds to the AB formula for a asymmetry Josephson junction in which two superconductor electrodes have the same s-wave symmetry with different pair-breaking gaps ($\Delta_{CP,t} = 1.30$ meV and $\Delta_{CP,s} = 1.68$ meV). f, Fitting (black curve) of a normalized conductance spectrum (red circles) using the IZ model. All $dI/dV$ spectra are acquired with $V_{set} = -10$ mV and a lock-in modulation of $V_{mod} = 20$ $\mu$V peak to peak.
Extended Data Fig. 5 | Superfluid density maps with inhomogeneous normal-state resistance. a, b, Maps of the critical Josephson current $I_C$. Spatial variations of the normal-state resistance, $R_N$. c, d, Maps of $(I_C R_N)^2$ associated with the superfluid density. The images in the left (right) column were acquired with a setup bias of $-10$ mV ($+10$ mV) and a setup current of 10 nA. To map the intrinsic superfluid density, it is necessary to normalize the measured $I_C$ by multiplying with $R_N$. Topographs were acquired simultaneously with these measurements and used to align the different maps.
Extended Data Fig. 6 | Determination of normal-state junction resistance, $R_N$. a, $R_N$-dependent differential conductance curves, $dI/dV$ (see key in c). For the normal-state conductance $G_N$, we use average values of $dI/dV$ over the full energy range. The $G_N$ values are indicated with the dashed lines and labels. b, $R_N$-dependent $I-V$ curves acquired simultaneously with $dI/dV$. c, The $I-V$ curves multiplied with $R_N$. All of the curves coincide. All spectra were acquired with $V_{set} = -10 \text{ mV}$ and a lock-in modulation of $V_{mod} = 100 \mu \text{V}$ peak to peak.
Extended Data Fig. 7 | Quasiparticle interference induced by inter-pocket scattering in FeTe$_{0.55}$Se$_{0.45}$.  

**a**, Top-view of the atomic structure of FeTe$_{0.55}$Se$_{0.45}$. The blue and red spheres denote the chalcogen and the transition metal atoms, respectively. 

**b**, Fermi surface of FeTe$_{0.55}$Se$_{0.45}$. Two hole pockets are at the $\Gamma$ point and one electron pocket at the M point, marked by black circles and ellipses. The red (blue) dashed lines correspond to the reciprocal lattice of Fe (chalcogen) layer. Because the unit cell includes two Fe atoms, the Brillouin zone of Fe (red square) atoms is two times larger than that of the full crystal structure (blue square). The alternate vertical positions of the chalcogen atoms above and below the Fe layer lead to a folded band (grey ellipse centred at the M point) composed of out-of-plane $d$ orbitals. Here we highlight the inter-pocket scattering wavevectors with black, yellow and red solid arrows.  

**c**, Sketch of scattering wavevectors in the reciprocal lattice. The colour coding is identical to that in **b**.  

**d**, Stripy patterns in a $dl/dV$ map at +3.9 meV induced by inter-pocket scattering ($V_{set} = -10$ mV, $I_{set} = 20$ nA and $V_{mod} = 200$ $\mu$V peak to peak).  

**e**, FFT spectrum of quasiparticle interference patterns. The circles correspond to the inter-pocket scattering wavevectors in **b** with the same colour coding.
Extended Data Fig. 8 | Determination of quasiparticle interference strength. a, $dl/dV$ map at $V_B = +3.6$ meV (shown in Fig. 3d). b, Bandpass-filtered FFT (not symmetrized) spectrum. c, Inverse FFT map of the filtered spectrum in b. d, Amplitude map of inter-pocket scattering pattern. The amplitude is determined by smoothing the filtered image in c. The red contours correspond to the mean amplitude of the map.
Extended Data Fig. 9 | Correlation plots between superfluid density and various parameters. a–d. Correlation between superfluid density and pair-breaking gap (a); strength of quasiparticle interference (b); topographic height (c) and quasiparticle strength minus the normal-state conductance (d; see inset), which gives similar results to those obtained without subtracting the normal conductance, as in the main text.
Extended Data Fig. 10 | Suppression of superfluid density at the atomic scale. a, b, Topograph (a) and superfluid density map (b). To highlight the atomic-scale variation of the superfluid density, we show the magnified map centred at the atomic defect (marked by a red and white arrow). Same data as in Fig. 3a, b.
### Extended Data Table 1 | Variables and symbols used in the paper

| Variables / Symbols | Meaning |
|---------------------|---------|
| $\Psi_{s(t)}$       | Wave function of the superconducting state of sample (s) or tip (t) |
| $n_{SF,s(t)}$       | Superfluid density of sample (s) or tip (t) |
| $\varphi_{s,t}$     | Phase of the superconducting state of sample (s) or tip (t) |
| $\Delta_{CP,s(t)}$  | Pair-breaking gap of sample (s) or tip (t) |
| $\Delta \phi$       | Phase difference between the sample and the tip |
| $I_C$               | Critical (maximum) supercurrent of the junction |
| $E_J$               | Josephson energy |
| $C_J$               | Junction capacitance |
| $V_B$               | Bias voltage |
| $Z_{env}(\omega)$   | Impedance of the electromagnetic environment |
| $R_N$               | Normal state resistance |
| $G_N$               | Normal state conductance |
| $J(t)$              | Phase-phase correlator |
| $P(E)$              | Tunneling probability function of Cooper-pairs |
| $I_{max}$           | Maximum current at finite bias in the IZ model |