Imaging quantum fluctuations near criticality

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A quantum phase transition (QPT) occurs between two competing phases of matter at zero temperature, driven by quantum fluctuations. Although the presence of these fluctuations is well established, they have not been locally imaged in space, and their local dynamics has not been studied so far. We use a scanning superconducting quantum interference device to image quantum fluctuations near the QPT from a superconductor to an insulator. We find fluctuations of the diamagnetic response in both space and time that survive well below the transition temperature, demonstrating their quantum nature. The fluctuations appear as telegraph-like noise with a range of characteristic times and a non-monotonic temperature dependence, revealing unexpected quantum granularity. The lateral dimension of these fluctuations grows towards criticality, offering a new measurable length scale. Our results provide physical insight into the reorganization of phases across a QPT, with implications for any theoretical description. This paves a new route for future quantum information applications.

Quantum fluctuations that stem from the uncertainty principle play a critical role in various physical phenomena. They are crucial for understanding why the structure of our Universe is not homogeneous, having left an imprint in the temperature fluctuations of the cosmic microwave background. Similarly, quantum fluctuations drive transitions between two competing phases of matter as a function of a non-thermal tuning parameter g at zero temperature. QPTs can be found in a variety of physical systems, such as magnets, superconductors and cold atomic gases. Their presence has usually been reported in macroscopic measurements such as transport and susceptibility. One of the hallmarks of such QPTs is a divergence of a spatial length scale ξ ≈ (g − g_c)^−ν with a critical exponent ν as the transition point g = g_c is approached, as well as the softening of an energy scale Ω ≈ (g − g_c)^−z where z is a dynamical exponent. This vanishing energy scale translates into a diverging timescale. The dynamics in a QPT represents quantum tunnelling events between different states. Although quantum fluctuations are well established theoretically and leave an imprint in experiments over an extended range in temperatures, they have never been locally imaged. Our aim here is to reveal details of the QPT by imaging the length and timescales of the quantum fluctuations near a quantum critical point (QCP).

For this we need a suitable material system and an imaging technique that is sensitive to the desired length and timescales. Additionally, we need to be able to identify the contribution of quantum fluctuations over and above thermal fluctuations. The system that we choose is a 2D film of an s-wave superconductor (NbTiN) that can be tuned through a QPT from a superconductor to an insulator by decreasing the thickness, without the complication of competing orders. We present data on a set of NbTiN films with thicknesses in the range 15–25 nm, approaching the QPT from the superconducting side (Fig. 1a).

Superconductivity is described by a complex-valued order parameter, with an amplitude and a phase. As the QPT is approached, the amplitude remains non-zero but the phase of the order parameter fluctuates, thus driving the superfluid density, which is measured locally proportional to the superfluid density, which is measured locally on the scale of micrometres (Fig. 1c and Methods). This enables identification of the spatial extent of superconducting fluctuations, λ_d, that represents phase correlations among many ξ pairs and diverges towards the critical point.

With the scanning SQUID, we study quantum fluctuation in two ways. The first is by rastering the SQUID over the sample and generating maps of the local diamagnetic susceptibility, revealing emergent puddles or regions where superconductivity is suppressed (Fig. 1d). The images reveal small streaks in the scan direction, which reflect dynamic events that can be tracked by time traces. Hence, the second way to probe fluctuations is by parking the SQUID at a specific location and measuring χ as a function of time. We continuously measure the diamagnetic currents that shield the applied magnetic field at a chosen frequency. We emphasize that this method is different from a continuous d.c. measurement of magnetism that detects flux flow of mobile vortices. The fluctuations manifest themselves as telegraph-like noise in the time traces (Fig. 1e).

In the following, we show that the observed temporal and spatial fluctuations are of quantum nature. As the system is pushed towards the QCP, the fluctuations are found to survive well below the superconducting transition temperature T_c, persisting in a regime in which thermal fluctuations are expected to be considerably suppressed. We identify a characteristic length scale (micrometres) of the fluctuations that grows towards the QCP. We demonstrate that the fluctuations occur at random locations and are therefore not linked to sample inhomogeneities. Investigation of these
Fig. 1 | Scanning SQUID measurements near the quantum phase transition show fluctuations in space and time. a, A schematic diagram of a QPT as a function of a non-thermal tuning parameter $g$ (here, the resistance per square at a temperature just above $T_c$). b, An illustration showing many $\lambda_{\text{pair}}$ grains. Red arrows represent the phase degree of freedom. Phase fluctuations weaken the Josephson coupling between the grains and create weak superconducting regions on a micrometre scale which we note as $\lambda_{\text{dia}}$, marked in red. c, An optical image of the SQUID susceptometer, showing the sensing area (the pick-up loop) and the field coil. d, A scanning SQUID susceptibility image in sample S1, showing weaker and stronger areas of superconductivity. The weaker areas appear as streaks in the scan direction marked by arrows. The scan is taken at 0.6$T_c$, far below the superconducting transition (scale bar, 5$\mu$m). The susceptibility signal is defined as the flux measured by the SQUID divided by current in the field coil, $\Phi_0/A$. The image contains 100 thousand (100k) pixels with a pixel size of 120 $\times$ 120 nm$^2$. Data acquisition per pixel is 40 ms. e, The streaks are dynamic. A SQUID parked in a specific location records the susceptibility as a function of time. The streaks manifest themselves as telegraph-like noise in the susceptibility signal with discrete jumps over a wide range of timescales.

Fig. 2 | Spatial and temporal distribution of events. a, Histogram of switching events as a function of event duration, for a time trace taken at 0.78$T_c$ shown in the inset. The measurements are performed by applying a small magnetic field at a frequency of 2.32 kHz. The event duration reflects changes in the diamagnetic response and is not directly related to the excitation frequency. The susceptibility signal varies between two discrete values, with amplitude of 0.05$\Phi_0/A$ and time resolution of 10 ms. b, Map of $\chi$-values taken at 0.78$T_c$. Values span a range of 0.05$\Phi_0/A$. Scale bar, 10$\mu$m (left). The image contains 35k pixels with a pixel size of 100 $\times$ 50 nm$^2$. Data acquisition per pixel is 83 ms. The red crosses represent positions where the SQUID was parked for 300 s to record $\chi$ with time resolution of 10 ms. The map (right) shows the maximal event times for each point (cross). Colour codes appear in c. c, Representative time traces for each of the colours used in b. The vertical axis scales are 0.05$\Phi_0/A$. 
fluctuations reveals unexpected behaviour such as non-monotonic temperature evolution of local superconductivity and a wide range of timescales under nominally the same conditions. The paradigmatic material system NbTiN exhibits a QPT that is not masked by competing orders. By setting standards and benchmarks on this material, we open the door to explorations of other new materials such as cuprates and pnictides, where the analysis is more complicated because of competing effects such as antiferromagnetism and charge density waves.

We begin by discussing our results obtained on sample S1, closest to the QPT. In this sample, we observe telegraph-like noise with an amplitude consistent with a signal from one fluxoid, \( \Phi_0 \). Presumably, the noise arises from states with different susceptibility levels, in which the lower level contains one more fluxoid. A histogram of a time trace at a specific location reveals a large distribution of times (Fig. 2). Furthermore, parking the SQUID at different locations on the sample exhibits very different histograms, which we characterize by the maximal switching time, \( t_{\text{max}} \), found to span orders of magnitude. In our measurements, we have tracked \( t_{\text{max}} \) between a few tens of milliseconds and tens of minutes. This behaviour is not predicted by any of the existing theories.

The switching events can also be seen in the images: as the SQUID is rastered over the sample, it detects many quantized fluctuations of \( \chi \). These appear as streaks in the susceptibility images such as that highlighted in Fig. 1d. A dark grey streak reflects a region of weak superconductivity entering the SQUID’s sensitive area, staying there for a limited period of time and then disappearing. When the event times are beyond the time window of our measurement, we observe puddles of weaker superconductivity (dark islands in Fig. 3a) arising from spatial fluctuations of the order parameter. Figure 3a depicts maps at different temperatures in sample S1 showing the development of such islands. The characteristic scale of the islands is tens of micrometres, much larger than the coherence length, \( \xi_{\text{par}} \approx 10 \text{ nm} \). This is one of the fundamental findings enabled by our SQUID measurements. The superconducting grains can establish superconductivity on a scale larger than a grain with size \( \xi_{\text{par}} \) when the Josephson coupling, arising from the tunnelling of Cooper pairs between grains, is strong enough. Each grain is associated with a phase degree of freedom representing the coherence of the Cooper pairs on a particular grain. Strong Josephson coupling aligns the phases of individual grains, creating large-scale regions of strong superconductivity. Close to the transition, phase fluctuations break the Josephson tunnelling, forming regions of weaker superconductivity on the scale of \( \lambda_{\text{ph}} \) (Fig. 1b) that are detected by our scanning SQUID.

Puddles like the ones shown in Fig. 3a for sample S1, close to quantum criticality, can also be seen in classical phase transitions\(^3\).
However, in a thermally driven transition the fluctuations are limited to a narrow temperature region close to $T_c$ (see results for Nb in Supplementary Fig. 1). In sample S1, the temperature dependence of $\chi$-maps reveals the presence of fluctuations well below $T_c$, indicating their quantum nature. The presence of puddles in a $\chi$-map is manifested through $\chi$-histograms (Fig. 3b), revealing the broadening of the susceptibility distribution as the temperature is reduced below $T_c$. Moreover, the width of the distribution continues to increase as the temperature is reduced for an extended temperature range. We quantify this effect by the standard deviation of the susceptibility values of each image, $\text{STD} = \sqrt{\left(\langle \chi(x,y)^2 \rangle - \langle \chi \rangle^2 \right)/\text{area}}$.

We find that the width of the histograms and the STD depend strongly on the proximity to the QPT. In our set of samples, decreasing the thickness transits the system from a superconductor to an insulator through a quantum critical point. As the critical point is approached, the susceptibility signals become smaller, reflecting the fact that the Pearl penetration length, $\lambda = \lambda_L / d$, where $\lambda_L$ is the London penetration depth and $d$ is the film thickness, grows towards criticality (see Fig. 3c and Supplementary Fig. 2). In this region, the resistive transition widens as expected, but the change in the magnetic behaviour is much more striking. Far away from the QPT (sample S3), the peak in the STD appears only in a narrow region around $T_c$. However, as we approach the critical point, the temperature range of the magnetic granularity grows significantly. For S1, closest to the QPT, it is evident that the fluctuations in time and space are no longer limited to a narrow region of temperatures and are present even far below the transition. The $\chi$-maps further show that the length scales of the puddles grow towards the QPT, as expected near the quantum transition (Fig. 3e and Supplementary Fig. 3).

We emphasize that the observed granularity in susceptibility is not linked to inhomogeneities in the system or to local $T_c$ variations. The temperature evolution of the islands in a certain location is different every time that we cycle the temperature far above $T_c$ (Fig. 4a and Supplementary Fig. 4). This results in a variety of STD curves, which are different for every temperature cycle, indicating the electronic nature of the fluctuations (Fig. 4b). Had the puddles been pinned by morphological inhomogeneities, the magnetic granularities would form at the same location in each thermal cycle.

Further support for the electronic nature of the granularity comes from the complex re-entrant behaviour of susceptibility measured at a specific location as a function of temperature. Figure 3c shows that local $\chi(T)$ curves include events that are non-monotonic in temperature, unlike the behaviour expected from nucleation and growth near classical phase transitions. In sample S3, away from the critical point, local $\chi(T)$ curves do not vary between different
locations and are monotonic with temperature. The complexity of the $T$ dependence of the fluctuations is apparent in the temperature evolution of the STD and the characteristic event times (Fig. 4d).

The multi-peak shape is a result of the development of regions with different superconducting properties.

A quantum Josephson junction array with lattice constant $a$ can model the QPT in a film with superconducting grains of size $\xi_{a} = a$ (see Methods). In this model, the competition between the charging energy and the Josephson energy drives a QPT from an ordered arrangement of phases in the superconducting phase to a disordered arrangement in the insulating phase (see Methods).

We obtain $\chi$-maps for different $g$ and $T$ (Fig. 5) using Monte Carlo simulations of a $(2 + 1)$-dimensional model in which the extra dimension reveals quantum tunnelling processes of Cooper pairs from one grain to the next. We find, in analogy to the experimental data, that deep in the superconducting phase (small $g$), thermal fluctuations in a narrow temperature region around $T_c$ destroy the superconducting order, leading to a proliferation of dark patches (weaker diamagnetic response); see Fig. 5a. Close to criticality (Fig. 5b–d), fluctuations in the diamagnetic susceptibility are present even at temperatures well below $T_c$, owing to the presence of quantum fluctuations. This is demonstrated by the broadening of the STD towards $g_c$ (Fig. 5e), clearly reproducing the qualitative behaviour observed in the experiments (Fig. 3).

So far, experiments have concentrated only on the superconducting side of the transition. Similar quantum fluctuations are expected in the insulating phase as well. This is captured in our simulations (Supplementary Fig. 5) that visualize the effect of quantum fluctuations on both sides of the transition by looking at the supercurrents along the imaginary time $\tau$ is revealed by calculating the frequency-dependent connected current–current correlation function $\Delta_{c}(\omega_{c}) = FT(\delta I_{c}(\tau)\delta I_{c}(\tau))$ at zero frequency, plotted as a function of $g$. Here, $FT$ is the Fourier transform and $J_c$ is the integer current in the imaginary time direction. The correlator shows a peak around $g_c \approx 4$, as it picks up the switching between zero and non-zero current states near the QCP due to increased quantum fluctuations.
A non-trivial correlation profile will enable extraction of the entanglement entropy information.

Data availability
The data that support the findings of this study are available from the corresponding author upon request.

Methods
Methods, including statements of data availability and any associated accession codes and references, are available at https://doi.org/10.1038/s41567-018-0264-z.

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Author contributions
A.K. and B.K. designed the experiment and performed the measurements. A.F. initiated the research and participated in experiments. T.I.B. provided the samples and related measurements. H.K., Y.L.L. and N.T. performed the calculations. N.T. and B.K. prepared the manuscript with input from all co-authors.

Competing interests
The authors declare no competing interests.

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Methods

Samples. NbTiN films are grown by the atomic layer deposition (ALD) technique, which provides atomically smooth surfaces\(^2\). The gaseous reactants are NbCl\(_5\), TiCl\(_4\), and NH\(_3\). For optimal superconducting properties, we use an AlN buffer layer grown on top of the Si substrate\(^2\). NbTiN films of thicknesses \(d = 15, 20\) and \(25\) nm are grown, varying only the number of ALD cycles \((240, 420\) and \(768\) cycles, for \(S1, S2\) and \(S3\) respectively), with all other parameters of the ALD process held constant. Further analysis shows that the NbTiN films are approximately a 7:3 solid solution of NbN and TiN (ref. \(^2\)).

Magnetic measurements. We use a cryogenic scanning SQUID to map the local susceptibility in the following way\(^3\): we place a current-carrying loop close to the surface of the sample, which generates a local magnetic field over a few micrometres. The SQUID pick-up loop \((\sim 1\) μm), concentric with this ring, senses the local field lines. Close to a superconducting sample, a portion of the applied field is eliminated by the superconductor, and the pick-up loop detects a weaker flux. This signal carries information about the suppression of the external magnetic field by the local diamagnetic supercurrents in the sample. The measured quantity is \(\chi\), the change of magnetic flux as a result of a change in the applied field induced by the current in the field coil \(\Phi_r\), in units of fluxoid per ampere, \(\Phi_r/A\) (Fig. 1c). By scanning the SQUID sensor at a constant height over the surface of the sample, we obtain maps of the local diamagnetic response, thus providing an XY image of the local superfluid density.

The magnetic susceptibility of a sample is probed by recording the flux response in the pick-up loop to an a.c. current in the field coil, as a function of position or as a function of time. The fields generated by the field coil are in the range of \(0.1–5\) Gauss, at frequencies of \(0.7\) to \(10\) kHz. In this manner, we continuously check how well the sample is able to suppress the applied field. If the response changes with location or with time, we record this small change, reflecting a change in the superfluid density. This measurement is different from a d.c. measurement mode, where the dynamics of vortices moving under the probe is detected. The time resolution is limited by the frequency that we use and by the time over which the signal is averaged for each pixel. Faster processes (such as Josephson dynamics at GHz rates) are not detected by this measurement scheme.

Quantum Monte Carlo simulations. To capture the essence of the experiments, we model the superconducting film as a Josephson junction array, consisting of a square lattice of superconducting grains of size \(d_x, d_y\). The parameters in our model are the charging energy required to add a charge to a single grain, \(E_C\), and the nearest-neighbour Josephson coupling \(E_J\) between superconducting grains (see Supplementary Fig. 6). This theoretical tuning parameter can be related to the experimentally tuning parameter, the normal state sheet resistance \(R_s\). This has been done previously in a related model where the dc conductivity response was calculated using the Kubo formalism\(^5\). The experimentally measured \(R_s\) is a function of disorder and temperature.

We simulate the quantum XY model using Monte Carlo techniques in which configurations of supercurrents arising from tunnelling of Cooper pairs are generated on a \((2 + 1)\)-dimensional lattice. For each current pattern generated by the simulation, we calculate the local magnetization \(M_t(r)\) according to the discrete analogue of \(J = V \times M_t\). In the experiments, the magnetic field is applied using a relatively large loop, whereas the diamagnetic response is measured using a smaller SQUID pick-up loop. To make a sensible comparison with experiments, we define the ‘local diamagnetic susceptibility’ as the local magnetization induced by a global uniform field. We calculate this quantity using a Kubo formula:

\[
\chi(r) = -\langle \delta M_t(r) / \delta B_r \rangle = \langle M_t(r) M_r \rangle.
\]

Our quantum Josephson junction array system is equivalent to a \((2 + 1)D\) XY model with two spatial dimensions and one imaginary time dimension. A duality transformation maps this to an ‘integer current model’ (ICM) in which there is an integer-valued current along every Josephson junction with the constraint that the net current into each grain is zero\(^6\). The integer currents depict the Cooper-pair tunnelling across a junction (see Supplementary Fig. 6). Explaining telegraph noise with timescales of the order of \(1,000\)s requires a more detailed modelling of the interplay between disorder, quantum fluctuations and thermal activation.

We perform quantum Monte Carlo simulations of the ICM using a worm algorithm\(^4\), for various values of the coupling \(g = E_c / E_J\) and the temperature \(T/T_c\). The experiments clearly show that the superconducting granularity is not determined by disorder, since a temperature cycling creates grains in different locations (see Fig. 4a). For the Monte Carlo to yield meaningful results displaying local quantum fluctuations, it was necessary to add a fraction \(p\) of missing bonds that serve to nucleate the superconducting fluctuations and reveal their spatial scales.

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