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Imaging quantum fluctuations near criticality

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Abstract

A quantum phase transition (QPT) occurs between two competing phases of matter at zero temperature, driven by quantum fluctuations. Though 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 in the vicinity of 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 about 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 a variety of 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 [1]. Similarly, quantum fluctuations drive quantum phase transitions (QPT) between two competing phases of matter as a function of a non-thermal tuning parameter g at zero temperature [2]. QPTs can be found in a variety of physical systems, such as magnets [3], superconductors, and cold atomic gases [4, 5]. Their presence has usually been reported in macroscopic measurements such as transport and susceptibility.

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One of the hall marks of such QPTs is a divergence of a spatial length scale $\xi \sim (g - g_c)^{-\nu}$ with a critical exponent ν as the transition point as $g = g_c$ is approached, as well as the softening of an energy scale $\Omega \sim (g - g_c)^{-\nu z}$ where z is a dynamical exponent. This vanishing energy scale translates into a diverging time scale. The dynamics in a QPT represents quantum tunneling events between different states. Though quantum fluctuations are well established theoretically, and leave an imprint in experiments [6–14] over an extended range in temperatures they have never been locally imaged. Our aim in this paper is to make the QPT come alive by imaging the length and time scales of the quantum fluctuations near a quantum critical point.

For this we need a suitable material system and an imaging technique that is sensitive to the desired length and time scales. Additionally, we need to be able to identify the contribution of quantum fluctuations over and above thermal fluctuations. The system 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 of 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 nonzero but the phase of the order parameter fluctuates thus driving the superfluid density (phase stiffness) down to zero [15–17]. Transport measurements reveal inhomogeneities in the superconducting properties on a scale of tens of microns [18]. However, to date, no local measurements have been performed on this length scale. Fluctuations of the amplitude of the superconducting order parameter have been observed using scanning tunneling microscopy [19–23]. These measurements have identified superconducting grains whose size is on the order of the coherence length, $\xi_{\text{pair}} \sim 10$ nm, related to the pair size. In accordance with theoretical predictions, the amplitude of the superconducting order parameter remains finite at the QPT. Thus in order to visualize the growing fluctuations near a QPT we need a probe that is sensitive to the phase of the order parameter (Fig. 1b). A suitable candidate is the scanning superconducting quantum interference device (SQUID). Scanning SQUID measures the strength of superconductivity via the diamagnetic response, χ , proportional to the superfluid density, which is measured locally on the scale of microns [24] (Figure 1c and Methods). This enables identification of the spatial extent of superconducting fluctuations, λ_{dia} , that represents phase correlations among many ξ_{pair} grains and diverges towards the critical point.

Utilizing 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 for probing 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 quantum critical point (QCP), the fluctuations are found to survive well below T_c , persisting in a regime where thermal fluctuations are expected to be considerably suppressed. We identify a characteristic length scale of the fluctuations, found to be on a micron scale, that grows towards the quantum critical point. We demonstrate that the fluctuations occur at random locations and are therefore not linked to sample inhomogeneities. Investigation of these fluctuations reveals unexpected behavior such as non-monotonic temperature evolution of local superconductivity and a wide range of time-scales 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 for explorations on other novel materials such as cuprates and pnictides, where the analysis is more complicated because of competing charge ordered and magnetic phases.

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, Φ_0 . Presumably, the noise arises from states with different susceptibility levels, 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, $t_{\max}$, found to span orders of magnitude. In our measurements, we have tracked $t_{\max}$ between a few tens of milliseconds to tens of minutes. This behavior 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 χ . 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 within 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. Fig. 3a depicts maps at different temperatures in S1 showing the development of such islands. The characteristic scale of the islands is on the order of tens of microns, much larger than the coherence length, ξ_{pair} ($\sim 10\text{nm}$). 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 ξ_{pair} when the Josephson coupling, arising from the tunneling 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 tunneling, forming regions of weaker superconductivity, on the scale of λ_{dia} (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 [25]. 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. S1). In the film S1, the temperature dependence of χ -maps reveals the presence of fluctuations well below T_c , indicating their quantum nature. The presence of puddles in a χ -map is manifested through χ -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, $\text{STD} = \sqrt{(\langle \chi(x,y)^2 \rangle - \langle \chi \rangle^2) / \text{area}}$.

We find that the width of the histograms and the STD depends 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 [26]. As the critical point is approached, the susceptibility signals become smaller, reflecting the fact that the Pearl penetration length, $\Lambda = \lambda_L^2/d$, where d is the film thickness, grows towards criticality (see Fig 3c and Supplementary Fig. S2). In this region, the resistive transition widens as expected, but the change in the magnetic behaviour is much more dramatic. 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 χ -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. S3).

It is important to 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 we cycle the temperature far above T_c (Fig. 4a and Supplementary Fig. S4). 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.

Another support for the electronic nature of the granularity is the complex reentrant behavior of susceptibility measured at a specific location as a function of temperature. Fig. 3c shows that local $\chi(T)$ curves include events that are non-monotonic in temperature, unlike the behavior 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.

The quantum Josephson junction array with lattice constant a can model the QPT in a film with superconducting grains of size $\xi_{\text{pair}} = a$ (see Methods). In this model, the number - phase uncertainty 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 χ -maps for different T and g (Fig. 5) using Monte Carlo simulations of a (2+1) dimensional model where the extra dimension reveals quantum tunneling 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 behavior 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. S5) that visualize the effect of quantum fluctuations on both sides of the transition by looking at the supercurrents along the imaginary time (τ) axis. These currents depict the tunneling of Cooper pairs between sites. Our results show that a peak in the long timescale correlations of currents in imaginary time coincides with proximity to the quantum critical point (Fig. 5f).

QPTs are expected to show a divergence of the characteristic time scale of fluctuations close to g_c . Our results reveal that the reality is much more complicated. The wide range of characteristic times shown in Fig. 4 can be interpreted as a distribution of g 's in a single sample. The g 's are redistributed every cooldown. In global measurements, such as resistivity, this behavior is averaged and the local complexity does not show up. Scanning tunneling microscopy measurements, on the other end of the scale, are successful in probing *emergent* quantum fluctuations on the scale of ξ_{pair} , but not the phase fluctuations. The scanning SQUID allows access to the relevant window of length scales which contains information on the diverging length (λ_{dia}) and spread of times, thus providing new insight to the dynamics of systems close to a QPT.

The findings presented in this letter suggest that the spatial and temporal complexity of quantum fluctuations should be taken into consideration in any model that attempts to treat the microscopics of a quantum transition. The ability to probe local quantum events opens new perspectives for quantum information applications. For instance we envision using two SQUIDs, one fixed and another scanning, as a means to study quantum entanglement in a solid state system. In such a setup one would measure simultaneously the time trace of fluctuations in two distant locations and search for correlations (uniquely selected by the excitation frequency). A non-trivial correlation profile will enable extraction of the entanglement entropy information.

Methods

Samples

NbTiN films are grown by the atomic layer deposition (ALD) technique that provides atomically smooth surfaces [27]. The gaseous reactants are NbCl_5 , TiCl_4 , and NH_3 , and NH_3 . For achieving optimal superconducting properties we use an AlN buffer layer grown on top of the Si substrate [28]. 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 an approximately 7:3 solid solution of NbN and TiN [29].

Magnetic measurements

We use a cryogenic scanning SQUID to map the local susceptibility in the following way [30, 31]: we place a current-carrying loop close to the surface of the sample, which generates a local magnetic field over a few microns. The SQUID pickup loop ($\sim 1\mu\text{m}$), 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 pickup 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 χ , the change of magnetic flux as a result of a change in the applied field induced by the current in the field coil $\frac{d\Phi}{dH}$ in units of fluxoid per ampere (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 we use and by the time over which the signal is averaged for each pixel. Faster processes (e.g. Josephson dynamics at the GHz) 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 ξ_{pair} . 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. S6). This theoretical tuning parameter can be related to the experimentally tuning parameter, i.e. the normal state sheet resistance R_{sq} . This has been done previously in a related model where the dc conductivity response was calculated using

the Kubo formalism [32]. The experimentally measured R_{sq} is a function of disorder and temperature.

We simulate the quantum XY model using Monte Carlo techniques in which configurations of supercurrents arising from tunneling of Cooper pairs are generated on a (2+1)-dimensional lattice. For each current pattern generated by the simulation, we calculate the local magnetization $M(\mathbf{r})$ according to the discrete analog of $\mathbf{J} = \nabla \times \mathbf{M}$. In the experiments, the magnetic field is applied using a relatively large loop, whereas the diamagnetic response is measured using a smaller SQUID pickup 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: $\chi(\mathbf{r}) = -M(\mathbf{r})/B_g = \langle M(\mathbf{r})M_g \rangle$.

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 [33]. The integer currents depict the Cooper-pair tunneling across a junction (see Supplementary Fig. S6). Explaining telegraph noise with timescales of order of 1000s requires a more detailed modeling of the interplay between disorder, quantum fluctuations, and thermal activation.

We perform quantum Monte Carlo simulations of the ICM using a worm algorithm [34], 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). In order 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.

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

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Fig. 1. Scanning SQUID measurements near the quantum phase transition (QPT) 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 ξ_{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 micron scale which we note as λ_{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 image contains 100k pixels with a pixel size of $120 \times 120 \text{ nm}^2$. Data acquisition per pixel is 40 ms. (e) The streaks are dynamic. The 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 time scales.

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 measurement 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) χ -map taken at $0.78 T_c$, values span over $0.05 \phi_0/A$ and the scale bar is $10 \mu\text{m}$ (left). The image contains 35k pixels with a pixel size of $100 \times 50 \text{ nm}^2$. Data acquisition, per pixel is 83 ms. The red crosses represent positions where the SQUID was parked for 300s to record the χ with time resolution of 10 ms. The map (right) shows the maximal event times for each point (cross). Color codes appear in c. (c) Representative time traces for each of the colors used in panel b. The vertical axis scales are $0.05 \phi_0/A$.

Fig. 3. The temperature range and length scale of fluctuations grow towards the critical point. (a) Susceptibility maps at different temperatures in S1. For $T > T_c$ no diamagnetic response is observed and the image reflects the SQUID noise. For $0.55 T_c < T < T_c$ the image shows the presence of darker puddles and streaks of weaker superconductivity that survive well below T_c (see the full sequence in the supplementary movie) (b) A histogram of χ values for three images in the bottom row of (a). The wide range of χ values highlights the fluctuation activity in comparison to the narrow range of χ values above T_c . (c) Resistance and susceptibility vs. temperature for a series of NbTiN samples approaching the QPT. The vertical bars represent the span of the susceptibility values in the imaged area. These values are the limits of the color bars in e. (d) Standard deviation (STD) vs. temperature extracted from χ -maps (several are shown in panel (a)). The vertical bars show the range of susceptibility values in each image. The STD is normalized to the highest value in each curve. (e) Susceptibility maps for samples S1, S2 and S3, chosen at the peak of the STD curve (arrows in (d) mark the peak temperature). Each scan contains 13k pixels with a pixel size of $120 \times 120 \text{ nm}^2$. Data acquisition per pixel is 45 ms. Scale bar is $3 \mu\text{m}$ for all images.

Fig. 4. The spatial configuration of fluctuations changes in each cooldown.

(a) Susceptibility maps repeated at the same location on sample S1 at $0.78 T_c$, taken between temperature cycles. The island configuration changes dramatically after a temperature cycle well above T_c (between the two top images, I and II). Successive images at the same conditions (2 bottom images, II and III) are overall similar, but there are visible changes in the position of the streaks around the puddle which indicates their dynamic nature. Each scan contains 13k pixels with pixel size of $120 \times 120 \text{ nm}^2$. Data acquisition per pixel is 45 ms. Scale bars $3 \mu\text{m}$. (b) STD vs. T plots. Both red curves are taken in the same location (extracted from the data in panel (a)) before and after cycling above T_c . The difference in the curves represent the spatial configuration of islands that reorganizes with each cycle above T_c . The green and black curves are taken at different locations on the sample, 150 and 250 μm away from the area presented in (a). The multi-peak curve structure is created by puddles that became active at different temperatures and present complex shapes and non-trivial temperature evolution. (c) χ vs. T plots vary between locations on the sample and reveal a non-monotonic temperature evolution of χ in sample S1. Blue and pink curves were taken at the location marked in (a). The yellow curve was taken in a different location, hundreds of microns away. The black, green and purple curves are taken on different locations in sample S3. Away from the QCP, these curves overlap showing no difference in the χ signal between different locations. Value for S3 multiplied by 0.8 to fit in the same scale as the data

for S1. **(d)** Characteristic time of events vs. T , for the black data set in (b), recorded with the time resolution of 10 ms. Each characteristic time and standard deviation bars are extracted from an individual time trace (inset) or from streaks in a specific image. Event times vary between milliseconds to minutes and display a non-monotonic temperature dependence, with a complex, multi-peak profile. The purple data point is taken from sample S3. In sample S3, away from the critical point these events appear only near T_c .

Fig. 5. Simulations of susceptibility fluctuations.

(a-d) Diamagnetic susceptibility (χ)-maps derived from Quantum Monte Carlo simulations of the quantum XY model at different temperatures for $g = 0.5, 3, 3.6$ and 3.7 respectively. When χ is large in magnitude (white) and uniform, the system has an overall diamagnetic response, indicating a superconducting regime. As temperature increases, fluctuations destroy the superconducting order with the introduction of dark regions that have reduced diamagnetic response. In these maps a small degree of disorder is introduced in order to pin fluctuations (see Methods). As the system approaches the SIT, the range of temperatures over which granularity is observed grows. (e) Standard deviation (STD) of susceptibility fluctuations vs. reduced temperature T/T_c for various values of g . The fluctuation region around T_c is enhanced as the QCP is approached with increasing g . (f) The long time-scale dynamics of currents along imaginary time τ is revealed by calculating the frequency-dependent connected current-current correlation function $\Lambda_{\tau\tau}(\omega_n) = \mathcal{FT}[\langle \delta J_\tau(\tau) \delta J_\tau(\tau') \rangle]$ at zero frequency, plotted as a function of g . 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.