

Unconventional superconducting phase diagram of monolayer WTe₂

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The existence of a quantum critical point (QCP) and fluctuations around it are believed to be important for understanding the phase diagram in unconventional superconductors such as cuprates, iron pnictides, and heavy fermion superconductors. However, the QCP is usually buried deep within the superconducting dome and is difficult to investigate. The connection between quantum critical fluctuations and superconductivity remains an outstanding problem in condensed matter. Here combining both electrical transport and Nernst experiments, we explicitly demonstrate the onset of superconductivity at an unconventional QCP in gate-tuned monolayer tungsten ditelluride (WTe₂), with features incompatible with the conventional Bardeen-Cooper-Schrieffer scenario. The results lead to a superconducting phase diagram that is distinguished from other known superconductors. Two distinct gate-tuned quantum phase transitions are observed at the ends of the superconducting dome. We find that quantum fluctuations around the QCP of the underdoped regime are essential for understanding how the monolayer superconductivity is established. The unconventional phase diagram we report here illustrates a previously unknown relation between superconductivity and QCP.

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I. INTRODUCTION

While the application of general arguments based on symmetry, topology, and strong correlations has greatly advanced our knowledge of quantum phases of matter, much remains to be understood regarding the role of quantum fluctuations. At zero temperature, a quantum phase transition (QPT) occurs when quantum fluctuations are large enough to destroy a phase with long-range order [1–3]. The experimental characterization of a QPT and the associated quantum critical point (QCP) is a challenging task. Notable examples are QPTs in superconductors [4–6]. While the Landau-Ginzburg theory provides a general framework of superconducting transitions in conventional bulk superconductors, phase transitions in two-dimensional (2D) thin films are described by the Berezinskii-Kosterlitz-Thouless (BKT) theory, in which unbinding of 2D vortices and antivortices plays a critical role. However, the BKT theory [7] usually describes a transition driven by thermal rather than quantum fluctuations. Despite extensive efforts and progress, the understanding of QPTs from a superconducting to a resistive state in two dimensions,

observed in various systems [4–6] tuned by, e.g., magnetic field, disorder strength, and electron doping [Fig. 1(a)], remains unsatisfactory (see, for example, a recent review [5]).

In addition to 2D superconductors, the physics of a QCP could be important to understand the phase diagram and superconductivity in several unconventional superconductors, such as cuprates [8], iron pnictides [9], and heavy fermion materials [10]. In these cases, quantum fluctuations near a QCP, at which the nearby ordered phase is no longer stable, are believed to play a key role in defining the superconducting dome centered at the QCP [Fig. 1(b)]. The QCPs here are typically buried in the dome [8–10], which hampers experiments. In general, the underlying physics of QCPs and the connection to the unconventional superconductivity in these intriguing situations remain to be understood.

In this work, based on a series of findings in monolayer tungsten ditelluride (WTe₂), we report an unconventional phase diagram that reveals a relation between a QCP and superconductivity. We summarize the key result—an unconventional phase diagram of monolayer WTe₂—in Fig. 1(c). In varying the gate-induced electron doping (n_g), we have uncovered the existence of a superconducting dome that was not apparent in the previous works [11–13]. A recent vortex Nernst experiment [13] provided a hint of its existence from the steep decrease in the pairing potential with increasing n_g . The direct observation of the superconducting dome here allows us to directly access two n_g -tuned QPTs from the superconducting state in the same device: one defines the

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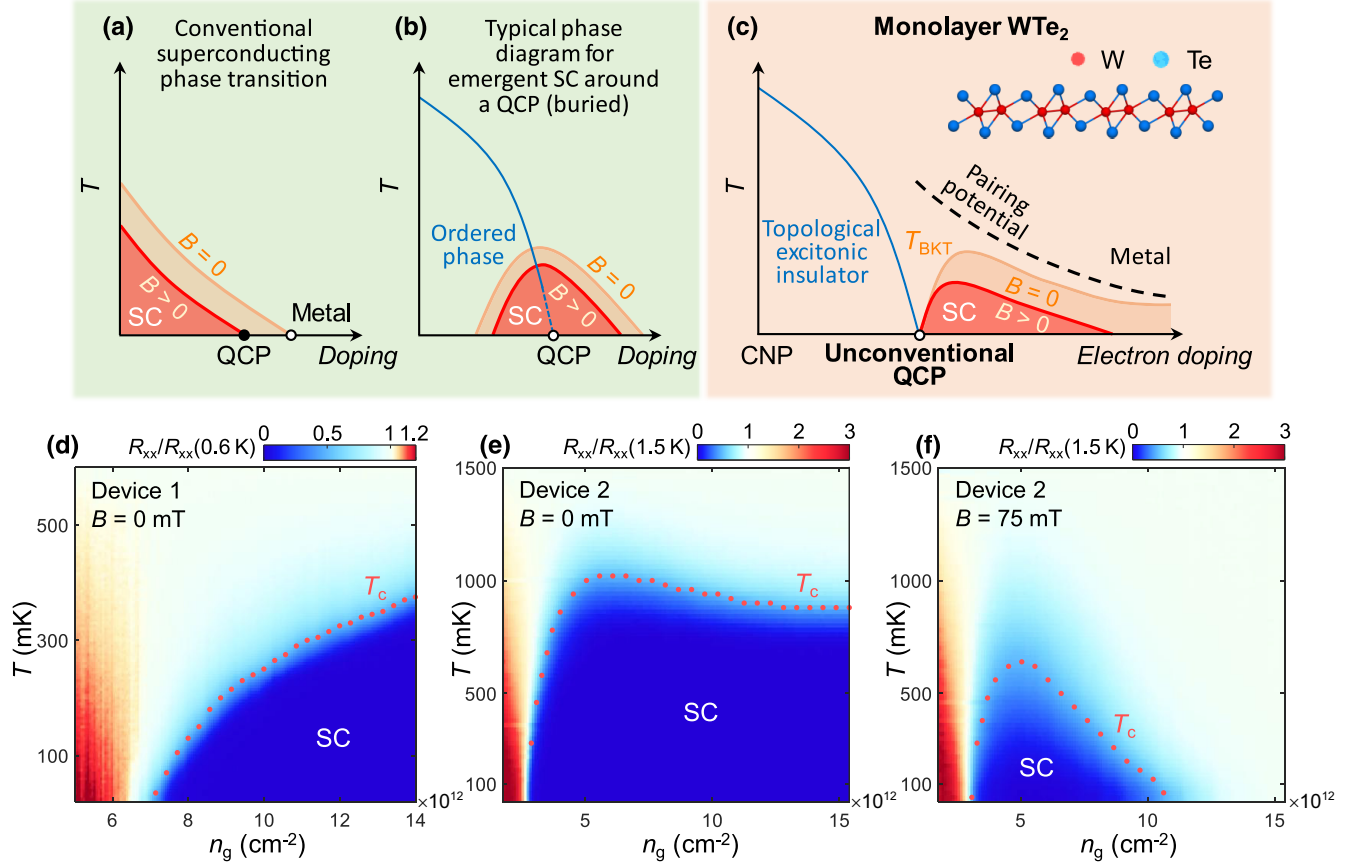


FIG. 1. Phase diagram of the superconducting monolayer WTe₂. (a) A schematic phase diagram of conventional superconducting transitions as a function of a tuning parameter, such as doping. (b) A typical schematic of the phase diagram for several unconventional superconductors, where QCP is buried in the superconducting dome. (c) The phase diagram of monolayer WTe₂, as summarized in this work. The pairing potential increases with decreasing electron doping, and superconductivity sharply emerges only on one side of an unconventional QCP. (d) Four-probe resistance (R_{xx}) as a function of n_g and T , measured on the device 1. R_{xx} is normalized to its value at 600 mK for each n_g to highlight the temperature dependence. The red dots represent the transport critical temperature (T_c), defined as the temperature at which R_{xx} drops to 50% from its normal state. (e) Normalized R_{xx} up to 1.5 K measured on a different device (device 2). (f) Normalized R_{xx} measured on the same device (device 2) at a finite B of 75 mT, which clearly displays a superconducting dome. SC denotes superconductor.

upper end of the dome in the overdoped regime, while the other defines the lower end in the underdoped regime. The overdoped QPT appears to follow conventional expectations, but the underdoped QPT features an unconventional QCP at the doping n_{c1} (identified in Ref. [13]). Remarkably, as we change the magnetic field (B), the latter remains fixed. At large B , superconductivity emerges at a singular value of n_g right above n_{c1} . As B is lowered to zero, this point spawns the entire regime of $n_g > n_{c1}$, but the superconductivity is strictly forbidden at $n_g < n_{c1}$. Using electrical transport and Nernst measurements, we report and investigate this highly asymmetric superconducting dome on one side of the QCP.

II. RESULTS

A. Superconducting dome of the monolayer WTe₂

The monolayer WTe₂ devices and measurement schemes used in this work follow our earlier report [13]. Figure 1(d) plots the electronic phase diagram of the monolayer WTe₂ observed in the four-probe resistance (R_{xx}) measured in device 1. It displays a critical electron density (n_{c1}) of $\sim 6.5 \times 10^{12} \text{ cm}^{-2}$ and a monotonically increasing transport critical temperature

(T_c) when the carrier density (n_g) increases, consistent with previous results [11–13]. The vortex Nernst experiment [13] revealed that R_{xx} of a 2D superconductor is sensitive to the BKT transition (at T_{BKT}) but not the pairing potential (or the superconducting gap Δ). Interestingly the pairing potential of monolayer WTe₂ is stronger at lower n_g [13], opposite to the trend of T_c vs n_g . The usual Bardeen-Cooper-Schrieffer (BCS) prediction of $\Delta \sim 1.76 k_B T_c$ is thus no longer valid here. The exact form of T_c as a function of n_g is strongly influenced by vortex pinning and hence influenced by disorder in the sample. To demonstrate this, we show the phase diagram of a different device (device 2) in Fig. 1(e), fabricated following the same procedure (see Fig. S1 of the Supplemental Material [14] for a comparison of the two devices). While the appearance of gate-tuned superconductivity is qualitatively consistent with device 1, n_{c1} is now a much lower value, $2.7 \times 10^{12} \text{ cm}^{-2}$, and more importantly, T_c is no longer a monotonic function of n_g . Instead, with increasing n_g above n_{c1} , T_c first rapidly increases to about 1 K and then decreases. We attribute the different transition temperatures observed in these devices to a varied amount of disorders and impurities, to which T_c appears to be very sensitive. In our current fabrication process, we aim to

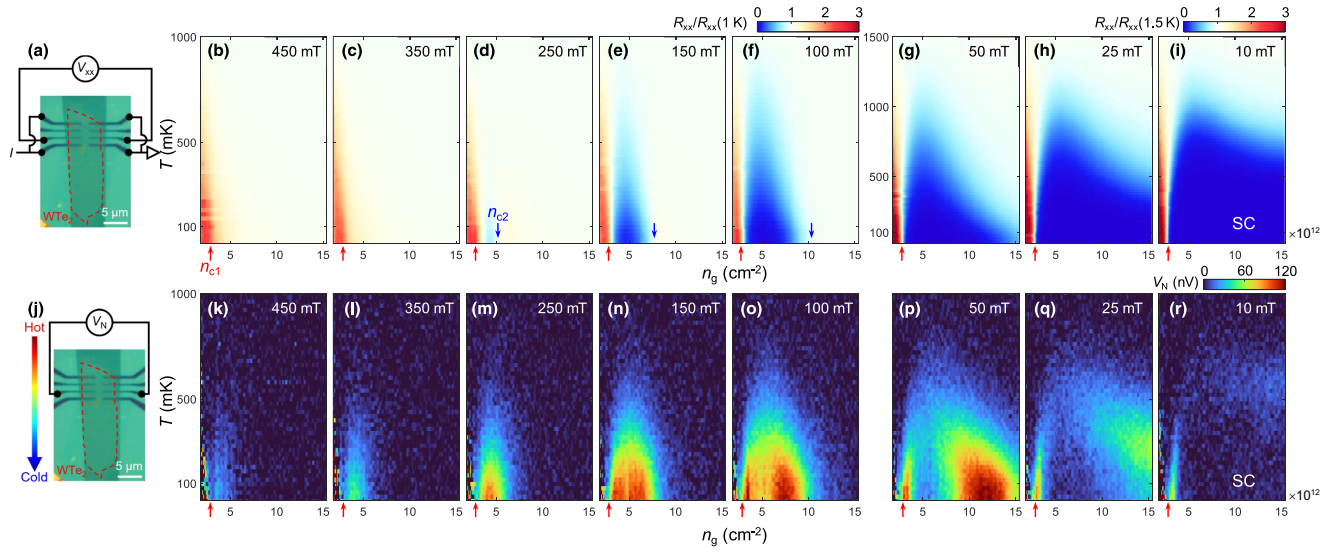


FIG. 2. Onset of superconductivity at the quantum critical point. (a) An optical microscope image of device 2 and the four-probe resistance measurement configuration. (b)–(i) Resistance phase diagrams as a function of n_g and T at selected magnetic fields as indicated. R_{xx} is normalized to its value at 1 K for (b)–(f) and 1.5 K for (g)–(i). (j) An illustration of the Nernst effect measurement configuration. (k)–(r), Maps of Nernst signal (V_N) as a function of n_g and T at the same B as (b)–(i). Red and blue arrows indicate the values of n_{c1} and n_{c2} , respectively.

minimize the level of disorders but they are still present. We hope a future generation of device fabrication that can better control and distinguish the types and amount of disorders could reveal more information on how T_c is influenced by disorders.

The nonmonotonic variation of T_c suggests the presence of a dome-shaped superconducting regime if n_g can be further increased. Unfortunately, in our typical devices, the high- n_g end of the dome at zero B requires a large n_g that is beyond the limit of gating due to the dielectric breakdown of hexagonal boron nitride. However, with a finite B (applied in the direction normal to the 2D flake), the superconductivity at high n_g is suppressed first and then the superconducting dome becomes apparent [Fig. 1(f)]. These observations unambiguously confirm that the pairing potential is indeed weaker at higher n_g . Based on the direct observation of the dome, we next uncover insight into the origin of the monolayer superconductivity.

B. Onset of superconductivity at the QCP

Figures 2(a)–2(i) present the resistance phase diagram spanned by T and n_g , taken from device 2 in various B . At $B > 350$ mT, there is no hint of superconductivity (blue region) in the R_{xx} diagram. When B is lowered below ~ 250 mT, superconductivity emerges at the singular point n_{c1} . As B further decreases to zero, the superconducting region rapidly expands to define the dome. Strikingly, the growth of the dome—strictly confined to the region above n_g —is asymmetric to the extreme.

The sharp asymmetry also applies to fluctuations of the order parameter detected by the vortex Nernst signal [Fig. 2(j)]. The applied temperature gradient $-\nabla T$ drives a current of mobile vortices which generates a Nernst voltage V_N engendered by phase slippage of the condensate order parameter. The Nernst experiment provides a sensitive probe of superconducting fluctuations that cannot be seen in resistance

measurements [13,15]. As reported [13], we derive maps of V_N [Figs. 2(k)–2(r)] under conditions corresponding to the R_{xx} maps of Figs. 2(b)–2(i), respectively. One finds that at low B no superconducting fluctuation is observed inside the dome [Fig. 2(r)], whereas, at the transition (i.e., the boundary of the dome), a strong Nernst signal develops. With increasing B , the dome shrinks, accompanied by strong fluctuations visible on the map. At high B [e.g., at 250 mT; Fig. 2(m)], the fluctuations are concentrated near n_{c1} (more precisely slightly above n_{c1} , i.e., $n_g \rightarrow n_{c1}^+$). The V_N signal is observable at fields as high as 450 mT [Fig. 2(k)], well above the magnetic field needed to fully suppress the resistance signature of superconductivity in the R_{xx} maps. The first appearance of superconducting fluctuations at $n_g \rightarrow n_{c1}^+$, confirmed in both R_{xx} and V_N , implies the onset of superconductivity at n_{c1} . The superconducting fluctuations are large above the critical point at n_{c1} , but unresolvable below.

C. Two distinct QPTs

As mentioned, a striking feature in Fig. 2 is that, whereas the superconducting region expands rapidly to the high- n_g end as B is lowered to zero, it never expands to the low-density region ($n_g < n_{c1}$). The Nernst signal (i.e., superconducting fluctuations) is strictly absent below n_{c1} ; this is the sudden death phenomenon of the fluctuations pointed out earlier [13]. With the dome-shaped superconductivity, we now can directly access two independent gate-tuned superconducting QPTs in one device, induced by either decreasing or increasing n_g respectively from the same superconducting state. The sudden death feature only occurs at the underdoped QPT (at n_{c1}), but not at the QPT in the overdoped region (at n_{c2}). This aspect is evident from the sharply distinct fluctuation patterns around the two QPTs shown in Fig. 2(r). Whereas V_N in the vicinity of the overdoped QPT extends over a very broad region, it is narrowly confined to the critical point in the underdoped QPT

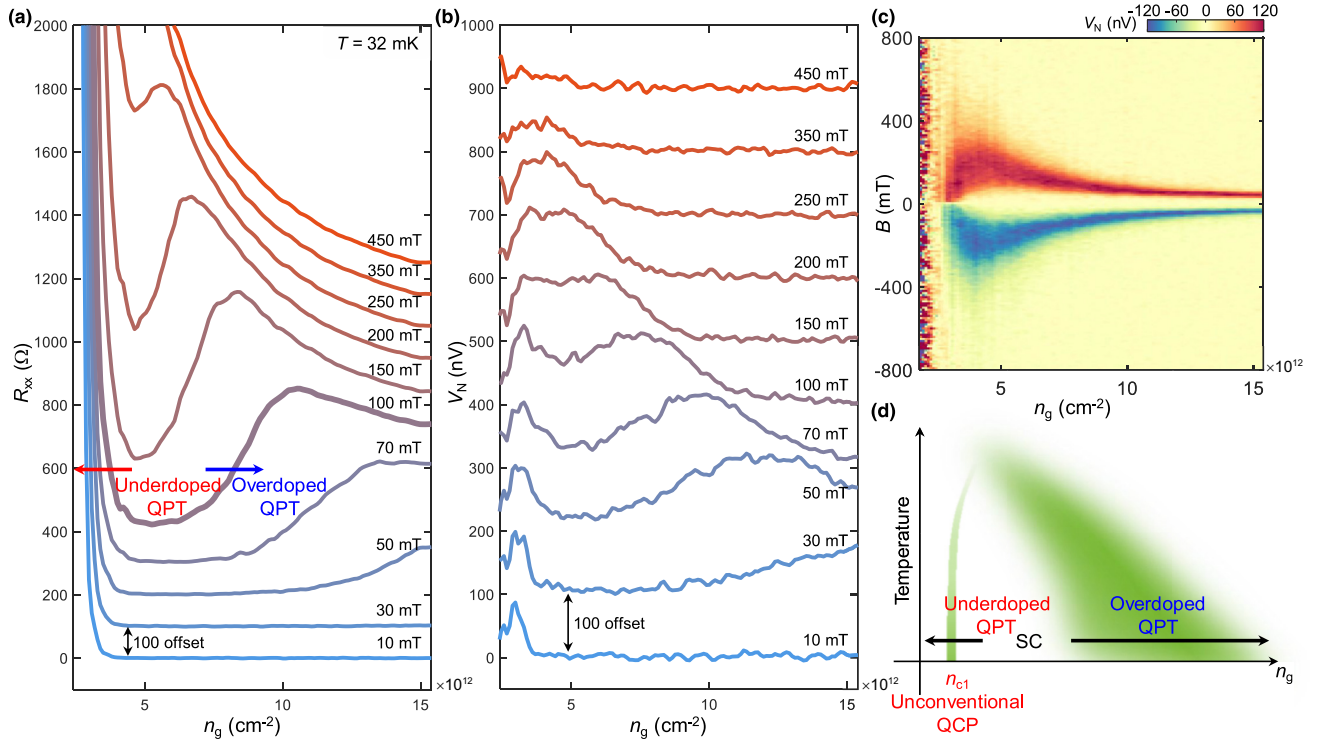


FIG. 3. Two distinct quantum phase transitions. (a) R_{xx} as a function of n_g measured at selected magnetic fields. The red (blue) arrow indicates the QPT in the underdoped (overdoped) regime. For clarity, the curves are offset from each other by 100 Ω . (b) The Nernst signal (V_N) as a function of n_g measured at the same selected magnetic fields in (a). A sharp (broad) peak of Nernst signal is observed in the underdoped (overdoped) regime. For clarity, the curves are offset from each other by 100 nV. (c) A map of the Nernst signal as a function of n_g and B , taken at base T . (d) A schematic phase diagram of superconducting fluctuations revealing the sharp QPT in the underdoped regime and the broad QPT in the overdoped regime.

at n_{c1} . In Supplemental Material Fig. S2 [14], we also show that the two transitions are independent of the displacement electric field applied by the two gates.

We further emphasize this distinction in Fig. 3. The two QPTs can be clearly observed in R_{xx} tuned by n_g , measured at the base T [Fig. 3(a)]. If B is fixed at ~ 100 mT, R_{xx} vanishes abruptly at n_{c1} once it enters the superconducting state. This is followed by a subsequent increase in R_{xx} when n_g exceeds n_{c2} . A distinctive feature is that n_{c2} is field sensitive, unlike n_{c1} ($n_{c2} \sim 10.4 \times 10^{12} \text{ cm}^{-2}$ at $B \sim 100$ mT). The corresponding Nernst signals measured at fixed B are displayed in Fig. 3(b), as well as in the color map in Fig. 3(c). The data confirm that n_{c1} remains fixed as B is varied, but n_{c2} is highly sensitive to B . The overdoped QPT occurs over a much broader density range with a fluctuation tail that extends deep into the normal state. We summarize in Fig. 3(d) the fluctuation pattern inferred from the Nernst data (extrapolated to the zero B limit). We conclude that the overdoped QPT is closer to conventional 2D superconductor-to-metal/insulator transitions observed in other systems including superconducting thin films [4–6] [as illustrated in Fig. 1(a)], whereas the underdoped QPT at n_{c1} is unconventional.

D. Unconventional pairing mechanism

An interesting feature of the superconductivity in monolayer WTe_2 is that it is unrelated—and unconnected—to a 3D parent superconducting state (WTe_2 in the bulk is not a superconductor). This distinction is shared with the super-

conductivity in magic-angle twisted graphene [16]. The fact that next to the superconducting state there is a topological insulator phase at charge neutrality suggests that the pairing nature of the superconductivity deserves careful studies. Topological aspects of unconventional superconductivity in monolayer WTe_2 are discussed in, e.g., Refs. [17,18].

Our results provide direct experimental evidence for an unconventional pairing mechanism in monolayer WTe_2 . (i) The fact that the pairing potential increases with decreasing n_g [13] suggests that electron interaction is important for pair formation. With lower n_g , the screening effect is reduced and hence stronger electron interactions. (ii) The abrupt appearance of superconductivity at n_{c1} is incompatible with the phonon-mediated BCS scenario. In this scenario, the BCS gap [19] is $\Delta \sim \hbar \omega_D \exp(-1/NV)$, where N is the density of state (DOS) near the Fermi energy, V is the electron-phonon coupling strength, ω_D is the Debye cutoff frequency, and $\hbar$ is the Planck constant. For a small window of n_g near n_{c1} , we do not expect a dramatic change in V or ω_D . Thus in the BCS framework, one would need to assign the onset of superconductivity near n_{c1} [Fig. 2(d)] to a dramatic enhancement of N , such as the presence of a Van Hove singularity (VHS), near this specific doping. We next demonstrate that this is not the case here.

Figures 4(a) and 4(b) display the R_{xx} maps tuned by B and n_g , taken at base T and 1 K respectively, both again demonstrating the onset of superconductivity right above n_{c1} . Figure 4(c) plots the four-probe conductance ($G_{xx} \equiv 1/R_{xx}$)

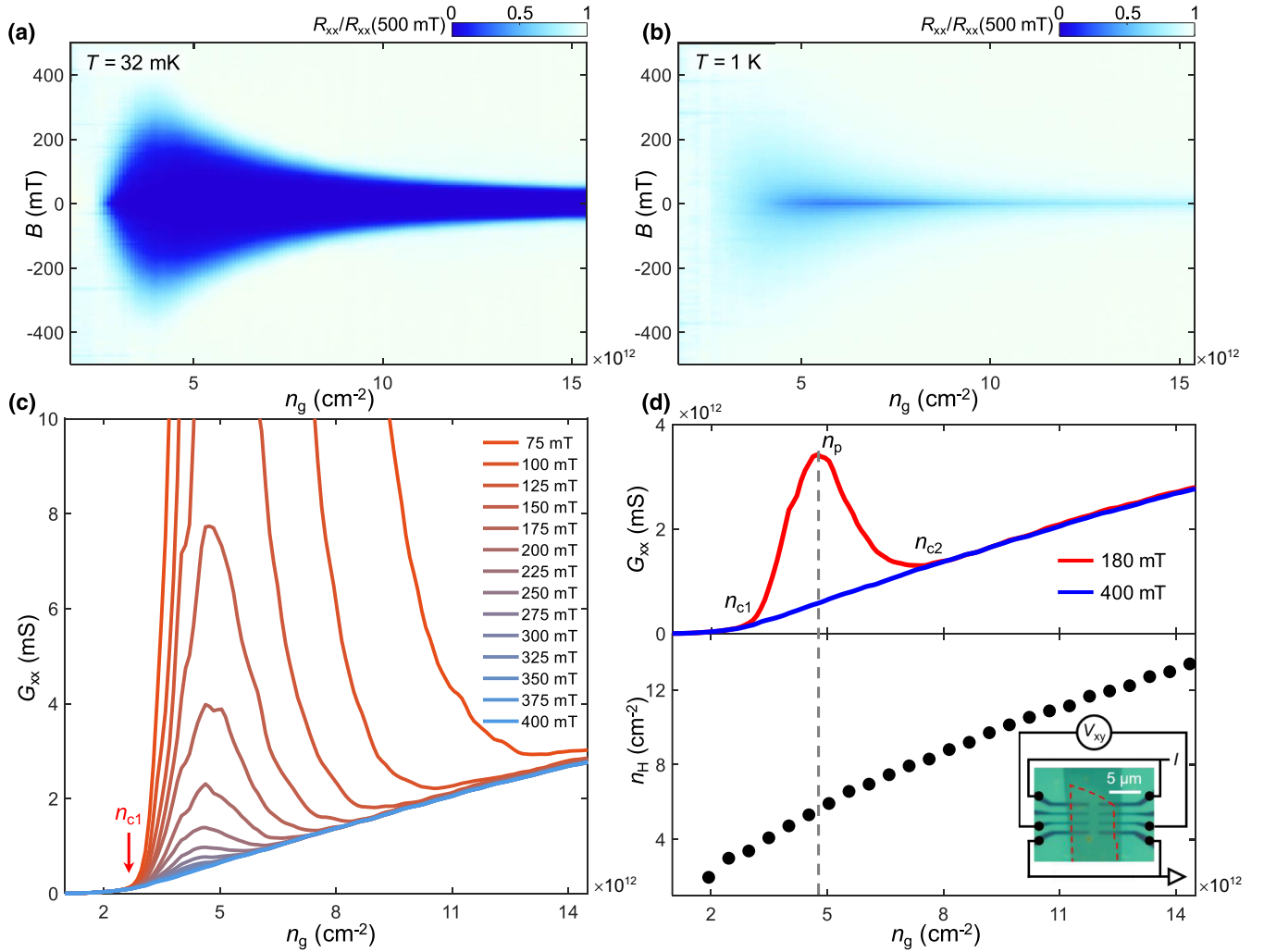


FIG. 4. Non-BCS superconducting behaviors. (a) Normalized R_{xx} as a function of n_g and B measured at base T . For each n_g , R_{xx} is normalized to its value at 500 mT to highlight its B dependence. (b) The same normalized R_{xx} map measured at 1 K. (c) Four-probe conductance (G_{xx}) as a function of n_g measured at selected B , revealing the onset of superconductivity above n_{c1} . (d) Lower panel: Extracted Hall density (n_H) as a function of n_g reveals a nearly linear dependence, consistent with the capacitance model. The inset shows the Hall effect measurement configuration. Upper panel: G_{xx} as a function of n_g taken at 400 mT (blue) and 180 mT (red), respectively.

as a function of n_g at selected B , where the peaks correspond to the growth of superconducting regions. One finds that when superconductivity is fully suppressed at high B , G_{xx} depends on n_g linearly as expected from the Drude model. The same linear relation can be found at high T (Fig. S3 [14]). Interestingly, the conductance peak of superconductivity emerges in the middle of the linear regime [at n_p as indicated in Fig. 4(d) upper panel], where no features in G_{xx} are observed, indicating the absence of VHS or other types of anomalous enhancement in the DOS at this doping. In addition, the presence of VHS would also lead to a sign change in the Hall signal. In Fig. 4(d) (lower panel) and Fig. S4 [14], we present the Hall effect measurements which reveal a nearly linear dependence of the Hall density vs n_g . Clearly no sign change is observed near the onset of superconductivity. The Hall data again confirm the absence of any anomalous features in the DOS near n_{c1} in the normal state. Similar behaviors are consistently observed in device 1 (Fig. S5), where n_{c1} is at a much higher density. The fact that n_{c1} depends sensitively on the sample details (e.g., disorder strength) may be

regarded as another manifestation of unconventional superconductivity since the conventional BCS gap is expected to be robust against disorder as per Anderson's argument [20]. We conclude that the sharp onset of superconductivity at n_{c1} in monolayer WTe_2 cannot be attributed to the phonon-mediated BCS mechanism; instead, it suggests that superconductivity stems from the quantum fluctuations near the unconventional QCP at n_{c1} , where a pre-existing ordered phase below n_{c1} is suppressed. This ordered phase could be the topological excitonic insulator state [21–29]. Unlike other unconventional superconductors such as high- T_c and heavy fermion systems [8–10], in the case of monolayer WTe_2 , the fluctuations near the QCP only give rise to superconductivity on one side of the QCP ($n_g > n_{c1}$), whereas superconductivity is strictly forbidden on the other side.

III. SUMMARY

In this work, we provide insights into the origin of the surprising superconductivity in monolayer WTe_2 , a state arising

from a doped excitonic quantum spin Hall insulator [21–29]. The results reveal an intriguing relation between superconductivity and quantum criticality not seen previously. The asymmetry suggests a type of quantum critical phenomena. Theoretically, a deconfined QCP [30–33] has been proposed in a model describing a transition between a quantum spin Hall insulator and a superconductor. The experimental phenomenology observed here is consistent with the expectation of a deconfined QCP, although further studies are needed to clarify their connections and establish a concrete theory behind the phenomena.

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