# Quantum Phase Transition in Few-Layer $NbSe_2$ Probed through Quantized Conductance Fluctuations

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We present the first observation of dynamically modulated quantum phase transition between two distinct charge density wave (CDW) phases in two-dimensional $2H$-$NbSe_2$. There is recent spectroscopic evidence for the presence of these two quantum phases, but its evidence in bulk measurements remained elusive. We studied suspended, ultrathin $2H$-$NbSe_2$ devices fabricated on piezoelectric substrates—with tunable flakes thickness, disorder level, and strain. We find a surprising evolution of the conductance fluctuation spectra across the CDW temperature: the conductance fluctuates between two precise values, separated by a quantum of conductance. These quantized fluctuations disappear for disordered and on-substrate devices. With the help of mean-field calculations, these observations can be explained as to arise from dynamical phase transition between the two CDW states. To affirm this idea, we vary the lateral strain across the device via piezoelectric medium and map out the phase diagram near the quantum critical point. The results resolve a long-standing mystery of the anomalously large spectroscopic gap in $NbSe_2$.

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Despite intensive research over several decades, charge density waves (CDW) continue to remain at the forefront of modern condensed matter physics [1–3]. CDW in quasi-one dimension is understood to arise from the Peierls mechanism—an inherent instability of a coupled electron-phonon system which creates a gap in the single-particle excitation spectrum leading to the emergence of a collective mode formed of electron-hole pairs [4].

In higher dimensions this electron-phonon interaction induced renormalization of the lattice wave vectors is often not enough to give rise to CDW [5–14]. One of the best known examples is $2H$-$NbSe_2$, where the mechanism of CDW is still widely debated [15–20]. It has been suggested that the origin may lie in the strong momentum and orbital dependence of the electron-phonon coupling [18,19]. A natural consequence of this is the sensitivity of the CDW order to lattice perturbations. This has recently been verified by scanning tunneling microscopy (STM) measurements, which find the existence of 1Q striped quantum phase competing with the standard 3Q phase in locally strained regions [21]. The tridirectional 3Q phase respects the threefold lattice symmetry and has a periodicity $Q \simeq 0.328G_0$, where $G_0$ is the reciprocal lattice vector. The 1Q is a linear phase with a periodicity $Q \simeq (2/7)G_0$ [21]. Calculations indicate that, for $T \ll T_{CDW}$, the system is very close to a quantum critical point separating these two phases and any small perturbation, like local strain, can induce a quantum phase transition (QPT) between these two [20,22,23]. There are, however, no direct experimental evidences of this QPT.

We probe for the possible existence of QPT in ultrathin, suspended $2H$-$NbSe_2$ devices through time dependent conductance fluctuation spectroscopy [24]. We find that, for devices where the strain is dynamic, the electrical conductance fluctuates between two precise values separated by a quantum of conductance, with a well-defined time scale. These fluctuations can be quenched either by damping out the strain fluctuations or by introducing lattice disorder into the system. We can control the transition between the two distinct quantum states by modulating the strain in devices fabricated on piezoelectric substrates. Through detailed calculations and analysis, we show that our observations are consistent with strain induced dynamic fluctuations between 3Q and 1Q quantum phases in $2H$-$NbSe_2$. We also establish that the energy scale of ~35 meV, often seen in spectroscopy studies in $2H$-$NbSe_2$, is associated with the energy barrier separating the two CDW phases.

We study two classes of devices. The first class, which we call "on substrate," is prepared on $SiO_2/Si^{++}$ substrates by mechanical exfoliation from bulk $2H$-$NbSe_2$ followed by standard electron beam lithography [25]. The second class of devices is suspended—few-layer $2H$-$NbSe_2$ flakes were mechanically exfoliated from bulk single crystals on silicone elastomer polydimethylsiloxane and transferred onto Au electrodes prefabricated on either $SiO_2/Si^{++}$ or $BaTiO_3/SrTiO_3$ substrates. The aspect ratios (width/length) of the samples were close to an integer, ranging from two to six. To study the effect of disorder, both these classes of devices are fabricated from multiple bulk $2H$-$NbSe_2$ crystals having a range of superconducting

 

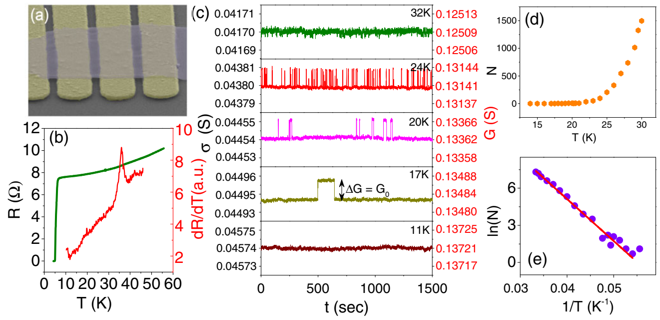


FIG. 1. (a) False color SEM image of a typical suspended ultrathin $2H$-$NbSe_2$ device. (b) Sample resistance $R$ (left axis) and its temperature derivative (right axis) as a function $T$ for device S1. (c) Plot of conductance per square, $\sigma$ (left axis) and conductance (right axis) measured at different $T$ for the same device. Note that with decreasing temperature, the frequency of the jumps reduce, although their amplitude remains unchanged. The length of the double headed arrow corresponds to $\Delta G = e^2/h$. (d) Plot of the number of conductance switches, $N(T)$ over a 30 min period versus temperature. (e) Fit of $N(T)$ to the Arrhenius equation.

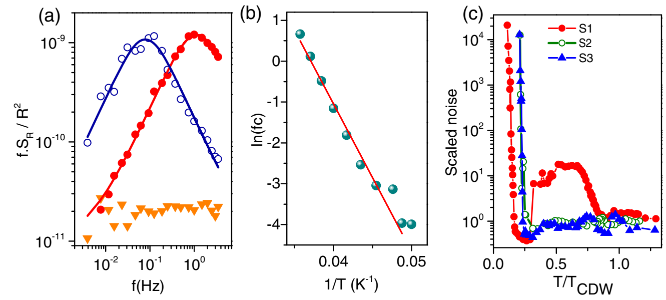


FIG. 2. (a) Scaled PSD of resistance fluctuations, $fS_R(f)/R^2$ vs $f$ at a few representative $T$ (open blue circle, filled red circle, and inverted orange triangles correspond to data at 27, 23, 11 K, respectively). The solid lines are fits to Eq. (1). (b) Plot of $f_C$ as a function of inverse temperature on a semi-log scale, the straight line is an Arrhenius fit to the data. (c) Plots of the relative variance of resistance fluctuations $\langle\delta R^2\rangle/\langle R\rangle^2$ (scaled by the value of $\langle\delta R^2\rangle/\langle R\rangle^2$ at $T = T_{\mathrm{CDW}}$) vs $T/T_{\mathrm{CDW}}$ for different classes of devices, S1: clean trilayer suspended device (red filled circle); S2: clean, approximately 25 nm thick substrated device (green open circle); and S3: Co-doped approximately 10 layer thick suspended device (blue triangle).

$T_c$ and residual resistivity ratios $\{[R(300\,\mathrm{K})]/[R(15\,\mathrm{K})]\}$ [25]. Devices are also fabricated from bulk $2H$-$NbSe_2$ doped with cobalt to introduce disorder in a controlled manner. The devices range in thickness from bilayer to about 50 nm, as obtained both from optical contrast and AFM measurements (Supplemental Material [26]). A SEM image of a typical suspended device is shown in Fig. 1(a).

Figure 1(b) shows the evolution of the resistance, $R$ with temperature, $T$ of a suspended trilayer device S1. The onset of CDW at $T_{\mathrm{CDW}} \sim 35$ K is indicated by a peak in the $dR/dT$ plot. The high value of the residual resistivity ratio, 8.5 and the relatively high superconducting $T_C$, 6 K indicate the defect free nature of the device. The time series of conductance fluctuations at different $T$ is plotted in Fig. 1(c). For $T$ very close to $T_{\mathrm{CDW}}$, the time series consists of random fluctuations about the average value, arising from the generic $1/f$ noise in the device. Below 30 K, we find the appearance of random telegraphic noise (RTN) with the conductance fluctuating between two well-defined levels separated by the quantum of conductance, $e^2/h$. The RTN persists right down to about 12 K below which superconducting fluctuations become dominant. The measurements are repeated on clean, suspended devices of different flake thicknesses. It was seen that with increasing thickness, the magnitude of the conductance jumps increased, remaining in all cases close to an integer multiple of $e^2/h$ (Supplemental Material [26], Fig. S5). Figure 1(d) shows a plot of the total number of switches over a period of 30 min at different temperatures. The switching statistics could be well described by an Arrhenius function [Fig. 1(e)]. The magnitude of the activation energy was found to be lie in the range $32 \pm 3$ meV in all such suspended, clean devices.

To probe in detail the statistics of the RTN, we performed low frequency resistance fluctuation spectroscopy at different temperature using a digital signal processing (DSP) based ac technique (Supplemental Material [26]). At each temperature the resistance fluctuations were recorded for 30 min. The resultant time series of resistance fluctuations were digitally decimated and anti-aliased filtered. The power spectral density (PSD) of resistance fluctuations, $S_R(f)$ was calculated from this filtered time series using the method of Welch periodogram [24]. The $S_R(f)$ was subsequently integrated over the bandwidth of measurement to obtain the relative variance of resistance fluctuations: $\langle\delta R^2\rangle/\langle R\rangle^2 = \int S_R(f)df/\langle R\rangle^2$. Figure 2(a) shows the measured PSD at a few representative temperatures. We find that the PSD over the temperature window 12 K <T < 30 K deviate significantly from $1/f$ nature, this $T$ range coinciding with that over which RTN was seen [Fig. 1(c)]. The PSD of an RTN is a Lorentzian of corner frequency $f_C = 1/\tau$, where $\tau$ is the time scale of the resistance switches between the two levels. Motivated by this, we analyzed the PSD data using the relation

$$\frac{S_R(f)}{R^2} = \frac{A}{f} + \frac{Bf_C}{f^2 + f_C^2} \tag{1}$$

The first term in Eq. (1) represents the generic $1/f$ noise in the device, while the second term quantifies the contribution from a Lorentzian [27]. Constants $A$ and $B$ measure the relative strengths of the two terms and are derived from the fits to the experimental data [Fig. 2(a)]. We find $f_C$ to be thermally activated, $f_C = f_0 e^{-E_a/k_B T}$ [Fig. 2(b)]. The value of the energy barrier is found to be $E_a = 35 \pm 3$ meV, which matches very well with that obtained from an analysis of the RTN jump statistics.

The relative variance of resistance fluctuations $\langle\delta R^2\rangle/\langle R\rangle^2$, normalized by its value at 60 K, is plotted in Fig. 2(c). The noise shows a broad peak over the temperature range $0.3 < T/T_{\mathrm{CDW}} < 0.9$, where RTN are present. We have verified that the additional contribution to the noise in this temperature range arises from the Lorentzian component in the PSD. We find that over the temperature range where RTN are absent, the distribution of resistance fluctuations is Gaussian, as is expected for uncorrelated fluctuations. With decreasing $T$, $\langle \delta R^2 \rangle / \langle R \rangle^2$ shoots up because of the onset of superconducting fluctuations. This has been seen before in many different superconducting systems and will not be discussed further in this Letter [28–30].

Turning now to the origin of these RTNs, we note that these can possibly arise, in CDW systems which have a single-particle energy gap at the Fermi level (e.g., $NbSe_3$ and $TaS_3$) [31–33], due to the switching of the ground state of the system between pinned and sliding states. In some of these systems sharp noise peaks were observed even at values of electric fields lower than the threshold field for slippage of the CDW [34]. However, unlike $NbSe_3$ and $TaS_3$, the CDW in $2H$-$NbSe_2$ does not slide. This is consistent with our observation that the RTN in $2H$-$NbSe_2$ were independent of electric field. This suggests that RTN in $2H$-$NbSe_2$ must have an origin distinct from those seen in gapped CDW systems like $NbSe_3$ and $TaS_3$. There is a due concern that the observed RTN may arise due to the interplay of superconducting fluctuations above $T_c$ and CDW order. Measurements performed under perpendicular magnetic fields much higher than $H_{c2}$ of bulk $2H$-$NbSe_2$ do not have any effect on either the frequency or the amplitude of these two level fluctuations, ruling out this interpretation (Supplemental Material [26]). We also considered the possibility that the RTN can arise due to the quantization of the number of density waves along the perpendicular direction, as seen in some systems [35,36]. We ruled this out by noting that in $2H$-$NbSe_2$ the weak interlayer van der Waals interaction precludes the formation of any long range density waves perpendicular to the planes. This is supported by spectroscopic studies.

The most compelling explanation of the RTN we observe in $2H$-$NbSe_2$ is phase fluctuations between 1Q and 3Q phases. Earlier calculations [22], supported by the STM measurements [21], demonstrated that the crossover between 3Q and a 1Q CDW phases at a given temperature can be induced by a strain as small as 0.1%. Experiments show that suspended $2H$-$NbSe_2$ devices in contact with Au pads experience an average strain of about 0.1% at low temperatures [37] which is sufficient to drive the system close to the boundary separating these two quantum phases [22]. In such a suspended mesoscopic device, at a finite temperature, the strain dynamically fluctuates due to thermally enhanced mechanical vibrations. This fluctuating strain can lead to a dynamical phase transition from 3Q to 1Q and vice versa in $2H$-$NbSe_2$ at a fixed temperature. This would cause the conductance of the system to fluctuate between two well defined values if the conductivity of the two phases are different. We validate this conjecture through detailed density-functional theory (DFT) based band structure calculations of the conductance in the two distinct quantum phases of $2H$-$NbSe_2$.

We calculate the dc conductivity $\sigma$ in both the 3Q and 1Q CDW phases using a two-band model, relevant for this compound [38]. The noninteracting dispersions $\xi_{1k,2k}$ are directly deduced from the DFT calculations (Supplemental Material [26]) [39]. The CDW order parameters are introduced within the mean-field approximation:

$$H = \sum_{i,\mathbf{k}} \Big[ \xi_{i,\mathbf{k}} c^{\dagger}_{i,\mathbf{k}} c_{i,\mathbf{k}} + \sum_{\nu} (\xi_{i,\mathbf{k}+\mathbf{Q}_\nu} c^{\dagger}_{i,\mathbf{k}+\mathbf{Q}_\nu} c_{i,\mathbf{k}+\mathbf{Q}_\nu}, + \Delta_{i,\nu} c^{\dagger}_{i,\mathbf{k}} c_{j,\mathbf{k}+\mathbf{Q}_\nu}) \Big] + \mathrm{H.c.} \quad (2)$$

Here the band index $i = 1, 2$, and the nesting index $\nu$ takes 3 values in the 3Q phase and 1 value in the 1Q phase. $c_{i,\mathbf{k}}$ is the annihilation operator for the electron in the $i$th-band at momentum $\mathbf{k}$. The mean-field CDW gap $\Delta_{i,\nu}$ is defined between the two bands. We obtain the quasiparticle energies $E_{i,\mathbf{k}}$ by exact diagonalization of the Hamiltonian in Eq. (2), and there are four, and eight quasiparticle states in the 1Q and 3Q phases, respectively.

The conductivity of the two phases primarily depends on the CDW gap values $\Delta_{i,\nu}$, which are related to the CDW potential $V_\nu$ by $\Delta_{i,\nu} = V_\nu \sum_{i,\mathbf{k}} (\Delta_{i,\nu}/2E_{i,\mathbf{k}}) \tanh(\beta E_{i,\mathbf{k}}/2)$, with $\beta = 1/k_B T$. The interaction $V_\nu$ arises from the electron-phonon coupling [22] and is directly related to strain, and, therefore, it becomes directional dependent. A CDW phase arises along a direction $\nu$ when the corresponding strain induced potential exceeds the critical potential $V_\nu > V_c \sim 2W$, where $W \sim 1.21$ eV is the bandwidth. Since the present system reside in the vicinity of the critical point, $V_\nu \sim V_c$, and the phase diagram is very sensitive to strain. In the 3Q phase, all three $V_\nu > V_c$, while in the 1Q phase, only $V_1 > V_c$, and the rest are $< V_c$.

In the mean-field state, we find a substantial suppression of the density of states (DOS) at $E_F$ in the 3Q phase, with a gap which is calculated to be $\Delta_0 \sim 35$ meV [Fig. 3(a)]. However, in the 1Q phase, the spectral weight loss at $E_F$ is significantly less. These results are consistent with the STM data [21]. Therefore, we anticipate that the conductivity in the 3Q phase will be lower than that in the 1Q phase.

We first calculate the conductivity $\sigma$ using the standard Kubo formula. We then obtain the conductance $G$ by normalizing the value of $\sigma$ with the dimensions of the present device [$G = \sigma \times (\mathrm{width}/\mathrm{length})$]. We assume band independent gap values. For the ratio of $\Delta_{3\mathrm{Q}}/\Delta_{1\mathrm{Q}} = 1.06$, we find that the difference in conductance between the two CDW phases, $\Delta G = G_{1\mathrm{Q}} - G_{3\mathrm{Q}} \sim e^2/h$, as seen experimentally. We also notice that over the temperature range $T = 17$–$24$ K, $\Delta G$ changes very little as the self-consistent gap remains essentially unchanged over this narrow

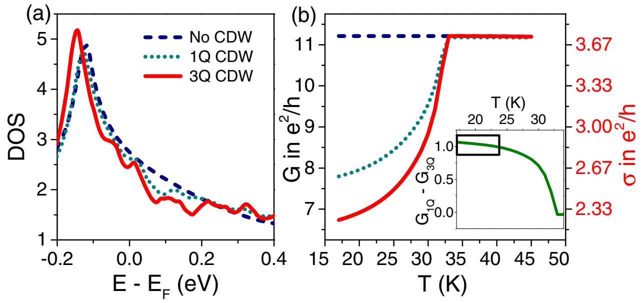


FIG. 3. (a) Plot of the calculated DOS in the two CDW phases. Also plotted for comparison is the DOS in the absence of CDW. (b) Temperature dependence of the computed conductance per square (right axis) and conductance (left axis) for the three cases. The inset shows the difference in conductance between the 3Q and the 1Q CDW phases over the temperature range where the RTN were observed. The box region is where the conductance fluctuations were experimentally found to be $e^2/h$.

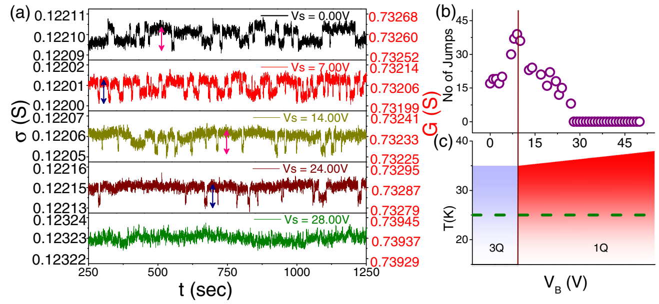


FIG. 4. (a) Plot of the time series of conductance per square (left axis) and conductance (right axis) measured at $T = 25$ K for the suspended 10 layer thick $2H$-$NbSe_2$ device (S5) fabricated on BTO substrate. The numbers in the legend refer to the voltage $V_B$ across the substrate while the length of the double headed arrows correspond to $\Delta G = 2e^2/h$. (b) Dependence of the number of conductance jumps, measured over a period of 30 min on the voltage $V_B$. (c) Schematic phase diagram of the system in temperature-strain plane. The blue shaded region is in the 3Q phase while the pink shaded region is in the 1Q phase. The green dotted line represents the isotherm at 25 K along which the data presented in panels (a) and (b) were collected.

temperature window [Fig. 3(b)]. This result is consistent with our experimental observations. We do not have a microscopic understanding of why this quantity should be an integral multiple of $e^2/h$. This may require the inclusion of topological terms in the calculation which is beyond the scope of the present work.

If dynamical phase fluctuations between the two CDW phases is indeed responsible for the observed RTN, it should be possible to modulate the frequency of the conductance jumps by driving the system controllably between the two competing CDW phases. To test this hypothesis, suspended devices of few layer $2H$-$NbSe_2$ are fabricated on piezoelectric $BaTiO_3/SrTiO_3$ (BTO) substrates. In this device, the strain across the device can be modulated by varying the voltage $V_B$ across the substrate. Figure 4(a) shows the evolution of the conductance fluctuations with changing $V_B$ obtained for one such device at 25 K. At very low values of $V_B$ (strain), the frequency of the conductance jumps is low and the system is seen to spend statistically similar amounts of time in both the high and low conductance states. With increasing $V_B$ (and, consequently, increasing strain across the device), the frequency of the conductance jumps initially increases and then decreases rapidly. However, the magnitude of the conductance jumps throughout this process remained quantized in units of $e^2/h$. Eventually, the conductance jumps vanish as the system stabilized in the higher conduction state [Fig. 4(b)]. We note that in different sweep cycles in $V_B$ the RTN are not exactly reproducible. It is difficult at this stage to comment on whether this is due to inherent hysteresis in the piezoelectric response of BTO or if it indicates nonreversibility of the properties of $NbSe_2$.

These results can be understood as follows: with increasing strain via $V_B$, the system approaches the phase boundary separating the 3Q and 1Q phases, leading to an increased probability of switching between the two states. Eventually, the system crosses the phase boundary and, consequently, the switching frequency starts decreasing and finally vanishes as the system settles into the 1Q state. These measurements establish conclusively that, consistent with theoretical calculations, strain can drive the system to the higher conducting 1Q phase from the lower conducting 3Q phase.

As seen from STM measurements on substrated devices, local random strain due to lattice imperfections causes the system to spatially phase separate into an inhomogeneous mixture of 3Q and 1Q phases [21]. This local phase separation cannot cause the measured conductance, which is a macroscopic global averaged property, to fluctuate dynamically between two well-defined conductance levels separated by the quantum of conductance. To validate this conjecture, measurements were performed on $2H$-$NbSe_2$ devices of various thicknesses prepared on $SiO_2/Si^{++}$ substrates (Supplemental Material [26]). Although we observed clear CDW transition in this set of devices from resistivity measurements, no signature of RTN was seen in any of them. The conductance fluctuations in these devices, at all temperatures $T > T_C$, consisted only of generic $1/f$ fluctuations arising from defect dynamics. The magnitude of noise $\langle \delta R^2 \rangle / \langle R \rangle^2$ remained constant over the temperature range $T_{\mathrm{CDW}} > T > T_C$ before showing the sharp rise near superconducting transition [Fig. 2(c)].

To test the effect of disorder, measurements were performed on suspended devices exfoliated from bulk $2H$-$NbSe_2$ crystals having low bulk $T_c$ and low residual resistivity ratio and from bulk $2H$-$NbSe_2$ crystals doped with 0.1% Co. Atomic force microscopy measurements showed that the rms surface roughness of the low $T_c$ flakes was about 3 times higher than that of the high $T_c$ flakes (Supplemental Material [26]). Although we observed a

$dR/dT$ peak at 35 K in these devices indicating the presence of CDW, we did not observe RTN in any of them. The noise in these devices was similar to what was seen for substrated devices indicating the suppression of RTN due to disorder in the system [Fig. 2(c)]. The absence of RTN in all the control experiments involving substrated, Co-doped, and disordered suspended $2H$-$NbSe_2$ devices, as well as the insensitivity of the conductance fluctuations in clean suspended devices to high magnetic fields reinforces our interpretation of the origin of the observed RTN in clean suspended devices as lattice fluctuation mediated.

To conclude, in this Letter we demonstrate controlled, strain induced phase transition between the 1Q and 3Q CDW phases in suspended 2D $2H$-$NbSe_2$. With this, we resolve a long-standing question of finite temperature dynamic phase transition between two quantum phases of the CDW system. We show the energy scale of $\pm 35$ meV, seen repeatedly in spectroscopic measurements [17,21], to be the barrier corresponding to 1Q-3Q phase transition. Our work establishes conductance fluctuation spectroscopy as a technique to probe phase coexistence and phase transitions in nanoscale systems, and can thus be a step forward in the understanding of competing quantum phases in strongly correlated systems.

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