Exceptionally Slow, Long-Range, and Non-Gaussian Critical Fluctuations Dominate the Charge Density Wave Transition

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$(TaSe_4)_2I$ is a well-studied quasi-one-dimensional compound long-known to have a charge-density wave (CDW) transition around 263 K. We argue that the critical fluctuations of the pinned CDW order parameter near the transition can be inferred from the resistance noise on account of their coupling to the dissipative normal carriers. Remarkably, the critical fluctuations of the CDW order parameter are slow enough to survive the thermodynamic limit and dominate the low-frequency resistance noise. The noise variance and relaxation time show rapid growth (critical opalescence and critical slowing down) within a temperature window of $\varepsilon \approx \pm 0.1$, where ε is the reduced temperature. This is very wide but consistent with the Ginzburg criterion. We further show that this resistance noise can be quantitatively used to extract the associated critical exponents. Below $|\varepsilon| \lesssim 0.02$, we observe a crossover from mean-field to a fluctuation-dominated regime with the critical exponents taking anomalously low values. The distribution of fluctuations in the critical transition region is skewed and strongly non-Gaussian. This non-Gaussianity is interpreted as the breakdown of the validity of the central limit theorem as the diverging coherence volume becomes comparable to the macroscopic sample size. The large magnitude critical fluctuations observed over an extended temperature range, as well as the crossover from the mean-field to the fluctuation-dominated regime highlight the role of the quasi-one-dimensional character in controlling the phase transition.

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Charge density waves (CDWs) are fascinating cooperative phenomena characterized by periodic modulation of the conduction electrons accompanied by lattice distortion [1–4]. Within the framework of the Ginzburg-Landau theory, the CDW state is represented by a superconductor-like two-component order parameter, $\Psi(\vec{r},t) = \Delta(\vec{r},t)e^{i\phi(\vec{r},t)}$ [2,3,5], where $\Delta(\vec{r},t)$ is the amplitude of the spatiotemporal modulated charge density and $\phi(\vec{r},t)$ its phase.

CDWs have been extensively investigated for their crucial role in Fermi surface instability [13–15], metal-insulator transitions [16,17], and superconductivity [18,19]. Over the years, the dynamics of collective CDW modes have been studied [2–4] using techniques as varied as neutron scattering [20,21], Raman spectroscopy [22–24], ultrafast time-domain THz spectroscopy [25,26], optical pump-probe experiments [27–29], ultrafast x-ray scattering [30], and time-resolved photoemission spectroscopy [31,32]. There has also been an upsurge of interest due to recent claims of axial anomaly associated with the collective CDW modes [33–35].

The CDW phase is often observed in low-dimensional materials where we expect no long-range order at finite temperature [36]. This apparent violation of the Hohenberg-Mermin-Wagner theorem is prevented by the fact that the chainlike CDW materials such as TTF-TCNQ, $NbTe_4$, $TaTe_4$, and $(TaSe_4)_2I$ are not strictly one-dimensional but merely anisotropic, with non-negligible interchain interactions. Nevertheless, the vestiges of the quasi-one-dimensional nature remain in the strong role that fluctuations play in significantly lowering the transition temperature $T_{\mathrm{CDW}}^{\mathrm{3D}}$ much below what is predicted by the mean-field theory, $T_{\mathrm{CDW}}^{\mathrm{MF}} = 2\Delta/(3.53k_B)$ [4]. Fluctuations are also known to modify the density of states and other physical properties [4,37,38]. Yet, direct measurements of the fluctuations themselves have been elusive.

In this Letter, we investigate the temperature-dependent resistance noise spectra in $(TaSe_4)_2I$ and carefully map out the critical scaling of the fluctuations over two decades of the reduced temperature on both sides of the CDW transition, which is found to be 263.03 K. $(TaSe_4)_2I$ belongs to the quasi-1D series of transition-metal-tetra-chalcogenides, $(MX_4)_nI$ ($M = \mathrm{Nb, Ta}$; $X = \mathrm{S, Se}$; $n = 2,\ 3,\ 10/3$) which has recently garnered attention for intriguing electronic properties [33,35,39–41], including unusual phase coexistence of superconductivity and ferromagnetism [42].

We establish that the $(TaSe_4)_2I$ undergoes a fluctuation-dominated transition at $T_{\mathrm{CDW}} = 263.03$ K [33,43]. The critical fluctuations associated with this CDW condensation

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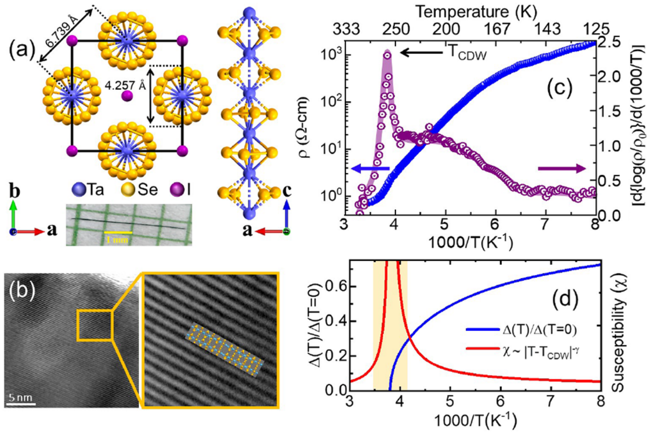

FIG. 1. Characterization of the $(TaSe_4)_2I$ crystal. (a) (top) Schematic of the crystal structure from single crystal x-ray diffraction experiment at $T \sim 300$ K. Projection of the structure of the tetragonal unit cell onto the *ab* and *ac* planes, respectively, to highlight the quasi-1D structure. (bottom) Image of the as-grown single crystal. (b) HRTEM image depicting the crystallinity. (c) Resistivity plotted on a logarithmic scale as a function of inverse temperature reveals the activated behavior at low temperatures. The CDW transition ($T_{\rm CDW} \sim 263$ K), barely discernible in the resistivity plot, is clearly seen in the rapid growth of the logarithmic derivative of the resistivity. (d) Schematic of the temperature variation of the order parameter (amplitude) and susceptibility. The yellow shade highlights the fluctuation-dominated region.

of the electrons survive in the thermodynamic limit and manifest in measurements involving timescales of seconds and length scales comparable to the sample size, over an extremely wide temperature window ($\simeq$52 K). Remarkably, we observe the crossover from the mean-field behavior to a fluctuation-dominated regime [37] with a different set of critical exponents. The anomalously low values of the critical exponents in the fluctuation-dominated regime may even suggest a crossover to a hysteresis-free weak first-order transition. In the process, we establish that resistance noise spectroscopy which has been a very popular tool to study phase transitions [5,44–48], can be used for quantitative study of the critical exponents [47].

Single crystals of $(TaSe_4)_2I$ were grown using chemical vapor transport technique [5]. Schematic of crystal structure, inferred from single-crystal x-ray diffraction, along with a picture of $(TaSe_4)_2I$ crystal are shown in Fig. 1(a). High-resolution transmission electron microscopy (HRTEM) image [Fig. 1(b)] clearly indicates the crystallinity and quasi-1D nature. The temperature-dependent resistivity, shown in Fig. 1(c), is nonhysteretic and shows 4 orders of magnitude increase as the temperature is lowered from 300 to 125 K. Although barely discernible in this resistivity plot, the logarithmic derivative (right axis) exhibits a prominent peak at $T_{\rm CDW} \approx 263$ K, consistent with prior investigations [43]. Below $T_{\rm CDW}$, resistivity exhibits approximately activated behavior as $\rho(T) \approx \rho_0\, \mathrm{e}^{(E_g/k_BT)}$ which is indicative of the growth in $n_{\rm CDW}$ forming the insulating CDW state with decreasing temperature. The conduction process can be approximately thought of as being due to the normal carriers which are created via single-particle excitation across the CDW gap E_g, like in a simple semiconductor. The estimated activation energy over the linear region of the plot is $E_g \simeq 216 \pm 2$ meV, in very good agreement with previous reports [15,33]. The experimental $d[\log \rho_{\rm dc}(T)/{\rm d}(1000/{\rm T})]$ exhibits a maximum at $T_{\rm CDW}$ instead of the cusp singularity predicted by the mean-field theory [4]. This already highlights the importance of order parameter fluctuations. The fluctuation region around $T_{\rm CDW}$ can be estimated by the Ginzburg criterion ($\Delta T_{\rm GL}$). The estimated $\Delta T_{\rm GL} = [k_B^2 T_{\rm CDW}/32\pi^2(\Delta C V_\xi)^2] \approx 52$ K, where $\Delta C = 0.83\ {\rm J\ K^{-1}\ mol^{-1}}$ is specific heat anomaly [49,50] and $V_\xi = 2111\ {\rm nm}^3$ is the coherence volume of $(TaSe_4)_2I$.

Here we investigate the critical nature of the order parameter fluctuations across the CDW transition by measuring the low-frequency resistance noise [5,44–48,51]. The time series of the four-probe resistance fluctuations were measured using a lock-in-based phase-sensitive detection under a very small bias [5,48,51] over a period of 80 min with the sample temperature kept fixed to within ± 2 mK [5]. Figure 2(a) represents the mean-subtracted time series of the resistance fluctuations, $\delta R(t) = R(t) - \langle R(t)\rangle$ at given temperatures.

The normalized power spectral density (PSD) of these representative resistance fluctuations between $f_{\rm min} = 10^{-3}$ Hz to $f_{\rm max} = 1$ Hz, is shown in Fig. 2(b). PSD exhibits the typical $1/f^\alpha$ behavior [48]. The noise exponent (α), relative variance $[(\langle \delta R^2\rangle/\langle R^2\rangle) = \int_{f_{\rm min}}^{f_{\rm max}} (S_R(f)/R^2) df]$ and relaxation time (τ), plotted with respect to common temperature axis in Figs. 2(c)–2(e). The fluctuations reveal three distinct regimes (also see Fig. 4 below): (I) uncorrelated Gaussian fluctuations in the high-temperature normal state, (II) a strongly correlated fluctuation regime (critical regime) around the CDW transition, and (III) suppressed resistance fluctuations in the long-range ordered CDW state at lower temperatures. The noise observed in regions I (normal state) and III (CDW ordered state) exhibits a moderate magnitude and nearly Gaussian $1/f^{1.00\pm0.05}$ type spectrum as shown in Fig. 2(c). This is clearly the usual $1/f$ flicker noise [52,53] and defines the *noise floor.*

We now focus on region II where the critical thermodynamic fluctuations emerge above the aforementioned noise floor and dominate the resistance noise $\delta R(t)$. The critical regime extends over a broad temperature window [38], $\varepsilon = (T - T_{\rm CDW})/T_{\rm CDW} \approx [-0.1, +0.1]$, which agrees rather well with a naive estimate based on Ginzburg criterion, $\Delta T_{\rm GL} \approx 52$ K. The noise exponent α increases to $\alpha \sim 1.25$ indicating a substantial shift in the spectral weight of fluctuations to lower frequencies in the vicinity of $T_{\rm CDW}$. The relative variance also grows by an order of

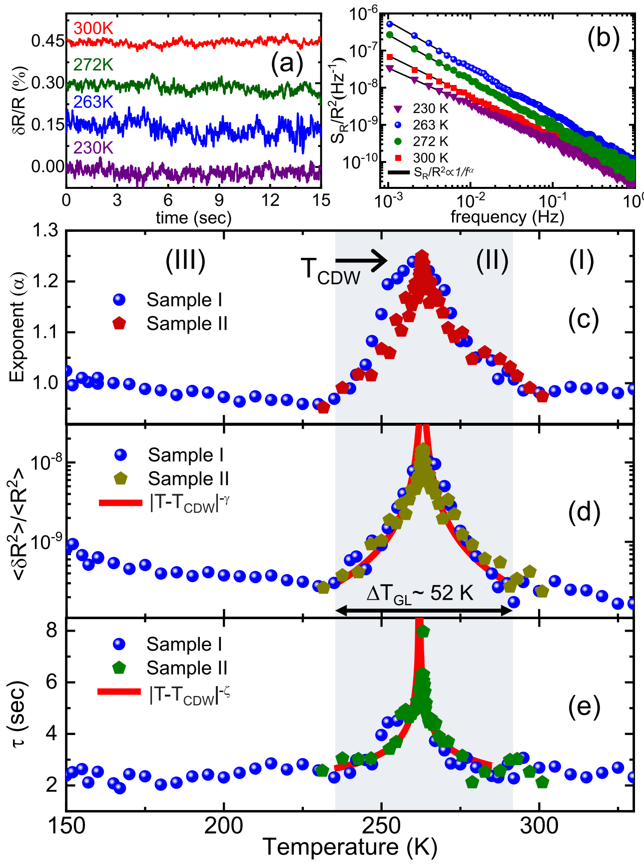

FIG. 2. Departure from $1/f$ behavior and critical slowing-down of fluctuations. (a) Representative time series of resistance fluctuations around T_{CDW} with a vertical shift of 0.15% for clarity. (b) Normalized PSD $[S_R(\mathrm{f})/R^2]$ of corresponding resistance fluctuations. (c) Temperature dependence of PSD exponent α, where $S_R(\mathrm{f})/R^2 \propto 1/f^\alpha$. (d) Temperature variation of the relaxation time (τ) which is estimated from the autocorrelation of resistance fluctuations. (e) The relative variance of resistance fluctuations for two samples. The solid lines (red) in (d)–(e) show the diverging trend at $T = T_{\mathrm{CDW}}$.

magnitude around T_{CDW} [Fig. 2(d)]. The rapid growth in the variance (critical opalescence) around T_{CDW} clearly indicates that these are critical fluctuations [44]. The corresponding critical slowing down is directly observed in Fig. 2(d) where the relaxation time τ for the decay of the autocorrelation, $C(t) = \langle \delta R(t') \cdot \delta R(t'+t) \rangle_{t'}$ is plotted as the function of temperature [5,46].

Critical opalescence and critical slowing around phase transitions continue to be of interest across a range of systems spanning different length scales—from quark-gluon matter [54–56], to jamming transitions [57,58], dynamical systems [59,60], ecology and biology [61,62], and, of course, a diverse array of condensed matter systems [29,44,63].

The low-frequency resistance fluctuations can arise from fluctuations in either the carrier mobility $\delta\mu$ or in the density of electrons δn, viz., $\delta R(t) = (\partial R/\partial n)\delta n(t) + (\partial R/\partial \mu)\delta\mu(t)$ [64]. Within the two-fluid model of CDW ground state [4,5,65], the total electron density is the conserved sum of normal n_{normal} and the condensed fraction, n_{CDW}, i.e., $n = n_{\mathrm{normal}} + n_{\mathrm{CDW}}$. At low bias, the n_{CDW} is pinned and therefore insulating. The temporal changes in mobility are also negligible because they are noncritical and a result of the scattering events occurring on the microscopic time scales of 10^{-12}–10^{-9} sec. They would thus average out on our timescale of interest.

Thus $\delta R(t) \simeq (\partial R/\partial n_{\mathrm{normal}})\delta n_{\mathrm{normal}}(t)$. Since the total density n is conserved [4,5,65,66], $\delta n_{\mathrm{CDW}}(t) \simeq -\delta n_{\mathrm{normal}}(t)$. Thus $\delta R(t) \simeq -(\partial R/\partial n_{\mathrm{normal}})\delta n_{\mathrm{CDW}}(t)$ [5]. Furthermore, as $\delta n_{\mathrm{CDW}} = \delta(\mathrm{Re}[\Psi])$ [5], the critical fluctuations in the order parameter directly translate into the experimentally measured resistance fluctuations.

We can thus understand our experimental results within the thermodynamics of critical transitions. The resistance fluctuations [Fig. 2(d)] essentially reflect the order parameter fluctuations, viz., $(\langle \delta R^2 \rangle / \langle R^2 \rangle) \propto \langle \delta n^2_{\mathrm{CDW}} \rangle \simeq \langle \delta \Delta^2 \rangle \simeq \int d^3 r G(r) = k_B T \chi_T$. The fluctuation-dissipation theorem has been used in the last term and $G(r)$ is the equal-time two-point correlation function of the order parameter [67]. Since the isothermal susceptibility χ_T should also diverge as $\chi_T \propto |T - T_{\mathrm{CDW}}|^{-\gamma}$, we expect $(\langle \delta R^2 \rangle / \langle R^2 \rangle) \propto |T - T_{\mathrm{CDW}}|^{-\gamma}$ with a mean-field value of $\gamma = 1$. This rapid growth of the variance is the manifestation of critical opalescence and is indeed seen in Figs. 2(d) and 3(a).

Similarly, the autocorrelations of the resistance fluctuations capture the autocorrelation of the CDW order parameter fluctuations. Growth of their relaxation time τ [Fig. 2(e)] is the signature of critical slowing down. While such critical slowing down is usually understood within the dissipative time-dependent Landau-Ginzburg dynamics [28,46,67,68], for a CDW system both the amplitude and phase modes are inertial; the free energy has a kinetic energy term that supports wavelike solutions [4]. In such systems, criticality is instead manifested via mode softening [69], i.e., $\omega_\Delta \propto (T - T_C)^{-\zeta}$ [4], where $\zeta = \frac{1}{4}$ from mean field. Here, ζ represents the exponent of the characteristic timescale associated with the propagating amplitude mode. Consequently, the characteristic timescale ($\tau \sim 2\pi/\omega_\Delta$) for the propagation of the fluctuations diverges as the rigidity of the amplitude mode collapses.

For such critical exponent analysis of power law divergence, Figs. 2(c)–2(e) also show measurements on a second sample with closely spaced (linearly spaced on a logarithmic scale) data points within the ΔT_{GL}. The values of the critical exponents γ and ζ, inferred by an error minimization algorithm (see Supplemental Material [5] for details) are shown in Fig. 3. This analysis [5] also yields a more precise $T_{\mathrm{CDW}} = (263.03 \pm 0.05)$ K and that the transition is nonhysteretic to 50 mK and thus a second-order or a weak first-order transition, unlike most CDW compounds [70,71].

While we had inferred the value of the Ginzburg temperature to be around 52 K, in Figs. 2 and 3 shows

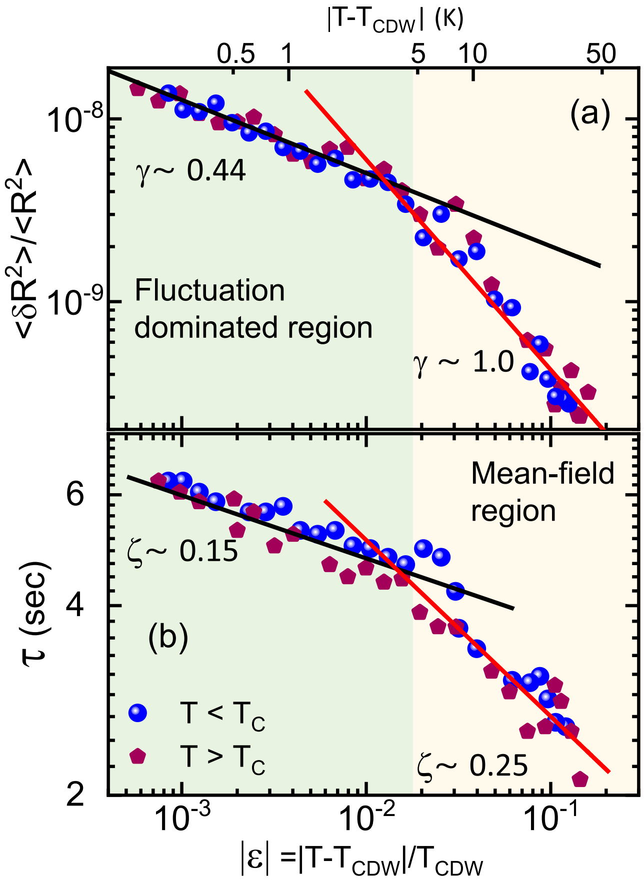

FIG. 3. Power-law scaling analysis: Crossover from the mean field to the critical fluctuation-dominated regime. The fluctuation data from region II shown in Figs. 2(d)–2(e) are replotted as a function of the reduced temperature (ε) in logarithmic scale. (a) The normalized variance $\langle \delta R^2 \rangle / \langle R^2 \rangle$, being proportional to the isothermal susceptibility has the scaling $\chi_T \sim |\varepsilon|^{-\gamma}$, and (b) the relaxation time $\tau \sim |\varepsilon|^{-\zeta}$, where ζ is the mode-softening exponent [4]. Solid lines (black and red) are guides to the eye. Note that the mean-field theory predicts $\gamma = 1$ and $\zeta = 1/4$ [4]. There is a clear crossover to different exponent values as one enters the critical fluctuation-dominated regime.

that the fluctuations play a more subtle role in such low dimensional systems [4,37]. Relatively far from the critical point ($0.02 \lesssim |\varepsilon| \lesssim 0.12$), the critical exponents start with a mean-field value ($\gamma = 1.06 \pm 0.08$ for T^+_{CDW}; $\gamma = 1.05 \pm 0.09$ for T^-_{CDW}) and for the autocorrelation time τ, $\zeta = 0.25 \pm 0.02$ for T^+_{CDW}; $\zeta = 0.26 \pm 0.03$ for T^-_{CDW}) consistent with the mean-field predictions of 1 and 1/4, respectively. This is observed on both sides of the transition as expected from conventional phase transition theory. But close to the critical point ($|\varepsilon| \lesssim 0.02$) we observe a crossover to a regime where the exponents are distinctly different; the system exhibits non-mean field behavior with exponents $\gamma = 0.44 \pm 0.09$ and $\zeta = 0.15 \pm 0.03$. Note that the value of γ is anomalously low compared to $\gamma \approx 1.3$, the value expected for the three-dimensional *XY* model [72].

Note that this could also indicate a very weak first-order transition, which may be experimentally indistinguishable from a genuine critical transition. While such a change in the order of the transition can arise on account of the changes in the lattice structure at the transition, previous studies have concluded that the symmetry arguments do not necessitate it [73]. Also, the observation of large fluctuations, scaling of the physical quantities, and the absence of hysteresis is striking. Alternatively, the thermal fluctuations themselves [74,75], given the similarity of the CDW order parameter to the superconductor and liquid crystal order parameter, could yield a very weak first-order transition via the Halperin-Lubensky-Ma mechanism [74]. We tentatively note that the low energy band structure of $(\mathrm{TaSe}_4)_2\mathrm{I}$ is predicted to have Weyl fermions [33–35]. In these systems, fluctuating pseudo-gauge fields (e.g., on the account of strain) [76] could give such a pathway.

Figure 4 summarizes the statistics of the fluctuations at different temperatures. Figure 4(a) shows the Fourier transform of the four-point correlation function, the second spectrum [77] $\mathrm{S}^{(2)}(\mathrm{f}) = \int_0^\infty \langle \delta R^2(t) \delta R^2(t+\varsigma) \rangle \cos(2\pi f \varsigma) d\varsigma$ [5,78–80]. The departures of the normalized "second-variance" $\sigma^{(2)} = \int_0^{f_H - f_L} S^{(2)}(f) df / | \int_{f_L}^{f_H} S_R(f) df |^2$ from

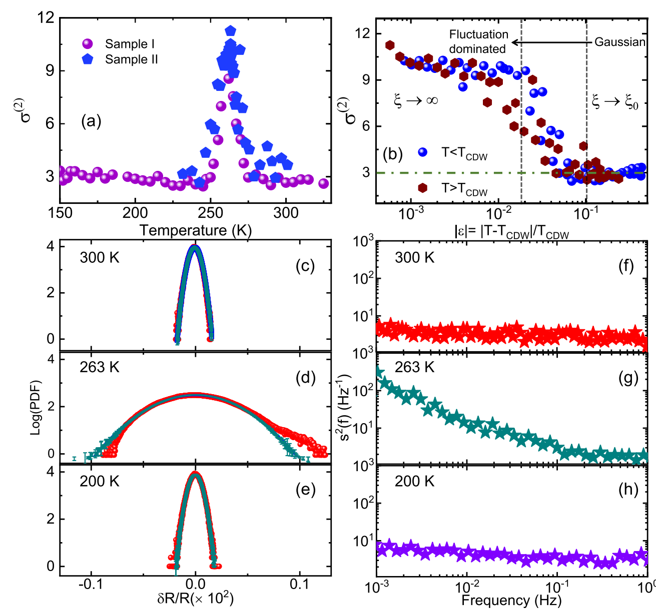

FIG. 4. Correlated fluctuations and non-Gaussianity at the critical point. (a) The normalized second-variance $\sigma^{(2)}$ as a function of temperature. $\sigma^{(2)} \simeq 3$ is expected for the uncorrelated Gaussian fluctuations. (b) $\sigma^{(2)}$, now plotted as the function of $|\varepsilon|$. A build-up of non-Gaussianity is observed as the correlation length grows in the vicinity of the critical point. Compare with the observations in Fig. 3. (c)–(e) The PDF of the normalized resistance fluctuations plotted along with the Gaussian distribution with zero mean, and (f)–(h) frequency-resolved second spectra $S^2(f)$ above, below, and at the critical temperature. Note that the qualitative difference at 263 K where the fluctuations have a much larger variance and the PDF is clearly non-Gaussian.

3, is a convenient measure of the non-Gaussianity of the fluctuations [44,79,81] within the frequency band (f_L, f_H). The temperature dependence of $\sigma^{(2)}$, computed with $f_L = 20$ mHz and $f_H = 0.2$ Hz, is shown in Fig. 4(a) and [5]. Note the rapid increase in $\sigma^{(2)}$ from the background value of ~ 3 near T_{CDW}. The non-Gaussian nature of fluctuations signifies the breakdown of the central limit theorem as the coherence volumes approach the sample size on the account of the growth of two correlation lengths $\xi_{||}$ and $\xi_{\perp}$ defined along-the-chain and out-of-the-chain directions, respectively, as they both diverge $\propto |T - T_{CDW}|^{-\nu}$ [67]. In Fig. 4(b), we now plot $\sigma^{(2)}$ as a function of ε on both sides of the transition. We find that the Gaussian to the non-Gaussian crossover (departure of $\sigma^{(2)}$ from 3), which begins at the boundary of region II (of Fig. 2) is complete in the fluctuation-dominated regime. The value of the temperature where $\sigma^{(2)}$ saturates is the same as where the critical exponents changed value in Fig. 3.

The representative plots of the probability density function (PDF) and $S^2(f)$ are shown in Figs. 4(c)–4(h). Both PDF and $S^2(f)$ reveal the non-Gaussian fluctuations in the critical region II. Finally, we note that the PDF of the noise around the transition region is asymmetric [Figs. 4(c)–4(e)]. Such skewed non-Gaussian behavior at criticality has been experimentally seen in disparate contexts and is perhaps indicative of extreme value statistics [82,83]. The non-Gaussian fluctuations and critical opalescence have been posited as possible experimental signatures of the elusive quantum chromodynamics critical point [54–56].

Conclusions.—Noise measurements have in the past been a powerful tool to study phase transitions [44,46,81,84,85]. However, phase transitions often accompanied additional features not simply reducible to the order parameter fluctuations. For example, both the correlation-driven (Mott) as well as the disorder-driven (Anderson), metal-insulator transitions [44,52,84,85] are usually accompanied with kinetic arrest, glassy dynamics, two-level behavior, structural (martensitic) transformation, and/or percolative transitions [79].

In contrast, our findings on the continuous and hysteresis-free CDW transition in $(TaSe_4)_2I$ suggest a purely electronic origin (apart from an accompanying Peierls instability). It is thus remarkable that the manifestations of criticality persist over the scale of the sample size ($\sim$mm). Critical opalescence is evident in the large variance of the fluctuations even in the macroscopic resistivity. Similarly, the critical slowing down of the correlation times stretches from the picoseconds [24,29] to the scale of seconds [Fig. 4(d)]. Moreover, these resistance fluctuations could be directly mapped onto the CDW order parameter fluctuations to quantitatively infer the isothermal susceptibility and the dynamic mode softening critical exponents [Fig. 3]. The beginning of the critical region gave exponents in remarkable agreement with the mean-field values. The growing correlation lengths were indirectly inferred from the growth of the non-Gaussian character of the fluctuations [Fig. 4]. While one expects a dimensional crossover from the regime of weakly coupled quasi-one-dimensional chains to the system effectively becoming three-dimensional in the critical regime on the account of the growing transverse correlation lengths [4], the quasi-one-dimensional character of the system was nevertheless manifest in the dominant role played by fluctuations. We observed a crossover to a non-mean field regime on a closer approach to the critical point ($\varepsilon \approx [-0.02, +0.02]$). While theoretically expected [37], such a crossover has been hard to experimentally observe. The anomalously small values of the exponents in this regime do not rule out a very weak first-order transition. This may be caused by an accompanying structural transition or may itself be fluctuation-induced [74].

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