Critical Slowing Down at the Abrupt Mott Transition: When the First-Order Phase Transition Becomes Zeroth Order and Looks Like Second Order

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We report that the thermally induced Mott transition in vanadium sesquioxide shows critical slowing down and enhanced variance ("critical opalescence") of the order parameter fluctuations measured through low-frequency resistance-noise spectroscopy. Coupled with the observed increase of the phase-ordering time, these features suggest that the strong abrupt transition is controlled by a critical-like singularity in the hysteretic metastable phase. The singularity is identified with the spinodal point and is a likely consequence of the strain-induced long-range interaction.

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Despite their ubiquity across systems and scales [1–16], first-order or abrupt phase transitions typically get only a passing reference in a traditional statistical physics course [17]. This is because they lack many of the remarkable features of the second-order or continuous transitions that arise from the diverging susceptibility [17–19]. The singularity leads to a power-law divergence of the correlation length, thereby leading to a description that is universal and largely independent of the microscopic details [17–19]. Experimentally, this singularity manifests in the sharp enhancement of the dynamical timescales (critical slowing down) [18,20] and the variance of fluctuations (critical opalescence) [18,21,22].

In this Letter, we experimentally demonstrate the slowing down and enhancement of fluctuations at an abrupt phase transition (APT) and argue that a certain class of APTs is also controlled by critical-like singularities. Specifically, we have studied the celebrated Mott transition in V_2O_3 [3,23–26]. As the scope of this work transcends the microscopic and material details of the system under investigation, here, we only highlight the two essential characteristics [27]. First, that this is an APT is unambiguously inferred from the large latent heat [Fig. 1(a) (inset)]. Second is a curious feature—V_2O_3 shares with some other vanadates [4–7], nickelates [8–11], manganites [12], intermetallic alloys [13,14], other charge-ordered materials [15,16], and spin-transition polymers [28–30])—that the transition is always hysteretic.

Hysteresis indicates that this "mixed-order" behavior (viz., the observation of critical slowing down as well as a latent heat at the transition) [31,32] may originate from the spinodal singularity [2,33–45]. This would then be a manifestation of the classic mean-field physics, which is already contained in van der Waal's equation (without Maxwell correction) [2,34,45]. Within equilibrium thermodynamics, an APT occurs at the binodal, which is the point where the free energy minima for the two phases have the same value [Fig. 1(c)]. But, nucleation barriers can lead to supersaturation into a metastable phase [46]. The depth of supersaturation is thus a function of the efficacy of the fluctuations in affecting first passage to the lower energy equilibrium phase [47]. It is believed that only in the (zero-temperature or infinite-range interaction) mean-field limit can the hysteretic passage though the metastable phase extend up to the stability limits: the spinodals [2,45]. As $\chi_T^{-1} = \delta^2 F/\delta\phi^2 = 0$ at the spinodals (where $F[\varphi]$ is the analytically continued free energy density, χ_T is the susceptibility [19], and ϕ is the spatially averaged order parameter), the correlation length and relaxation times diverge at the spinodals for the same reason that they do at the critical point [2,33,35]. Consequently, the spinodals are fixed points under the renormalization group transformation and should display critical behavior and universality [41–43].

Although this instability has been studied in a variety of zero-temperature noise-free systems [35,36,40,48,49], spinodals have not been experimentally established in thermodynamic systems where fluctuations are present [50]. From the scaling behavior of the dynamic hysteresis and the average qualitative nature of the phase ordering, we have recently proposed that the Mott transition in V_2O_3 does indeed occur around such bifurcation points [23]. The present work, by focussing on fluctuations, transcends the mean-field picture to establish that the transition has a genuine thermodynamic character. Our observations of critical slowing down [20] and enhancement [37] of the order parameter fluctuations indicate that one can at least get close enough to the spinodal such that the singularity controls many of the features of the transition, even if there are no divergences [33,51].

On account of the criticality and the essential role of hysteresis, it is meaningful to distinguish these transitions from the usual first-order transitions [2]. Following the Ehrenfest criterion, these may be called "zeroth order" because the free energy is itself discontinuous [23,47] [Fig. 1(c)].

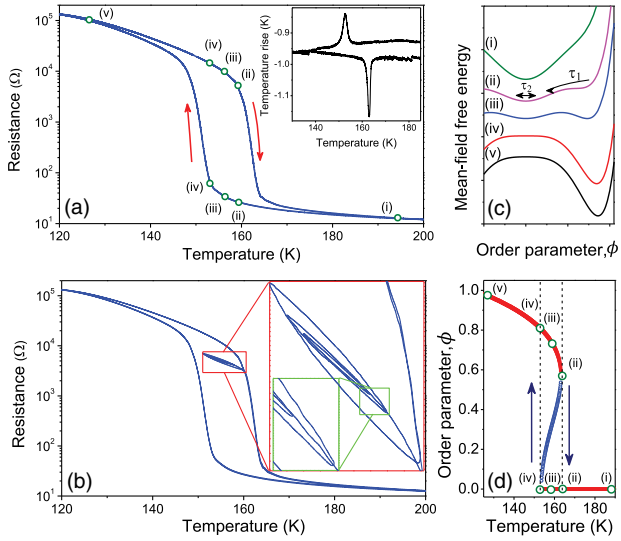

FIG. 1. (a) Temperature dependence of resistance of V_2O_3 shows a sharp and hysteretic transition around 153 K while cooling and 162 K while heating. (Inset) A large latent heat is also measured in a differential thermal analysis [23]. (b) Return-point memory [15,52,53]: Although multivalued in the hysteretic region, after any excursion out of a given hysteresis loop in the form of a minor loop, the same value of the resistance is recovered on return. Thus, the memory of the excursion is wiped out. (c) Mean-field free energy as a function of the spatially averaged order parameter ϕ for the compressible Ising model (essentially, the ϕ^6 theory) [23] can be used to capture transition semiquantitatively. The points corresponding to curves (i)-(v) are also roughly marked on the resistance curve in Fig. 1(a). Curve (iii) represents the binodal where the two minima are equal, and curves (ii) and (iv) are the two spinodals where one minimum becomes an inflection point. τ_2 is the relaxation time of fluctuations measured through the autocorrelation of the resistance noise, and τ_1 is the time associated with phase ordering. (d) Location of extrema of the above free energy as a function of temperature with the same points (i)-(v) marked. In the temperature window of ∼153–162 K where hysteresis is seen, the order parameter ϕ has globally stable, metastable, and unstable extrema.

V_2O_3 is our material of choice [23] because of the sharp and definitive APT with a large latent heat over a narrow temperature window [Fig. 1(a)]. Although one cannot define state variables in the metastable phase, the system does show reproducible quasistationary behavior (essential for the fluctuation spectroscopy), as seen in the return-point memory [15,52] of hysteresis [Fig. 1(b)].

The temperature dependence of the resistance of a typical polycrystalline sample V_2O_3 used in the study, after it has been thermally cycled between ∼77 and 250 K a few hundred times, is shown in Fig. 1(a), with the inset showing the latent heat peaks in the differential thermal analysis measurement [23]. The hysteresis window (∼153–162 K) can be seen to contain the metastable phase for which the properties are history dependent, whereas the regions outside this window seem to represent equilibrium states. In rest of this Letter, the results for the heating and the cooling transitions are shown in parallel [54] because the similarity of the two datasets is central to the main conclusions of the work.

Resistance-noise spectroscopy has been a powerful tool for the study of fluctuations close to phase transitions [9,11,14,20]. The experiments reported here involved carefully recording the time series of the fluctuating resistance R with the sample temperature kept precisely fixed at the given value [54].

Figures 2(a) and 2(b) show some of these time series of the normalized resistance fluctuations $\Delta R(t)/R$ at different temperatures after the removal of the very slowly varying smooth background [54]. It is hard to discern any significant departure from their Gaussianity [Figs. 2(c) and 2(d)]. To make contact with the theory of phase transitions (see the discussion below), let us assume that the essence of the transition may be captured by a scalar order parameter $\varphi(\mathbf{x}, t)$ equal to the fraction of the insulating phase in the sample [23,55]. The time series of the sample resistance

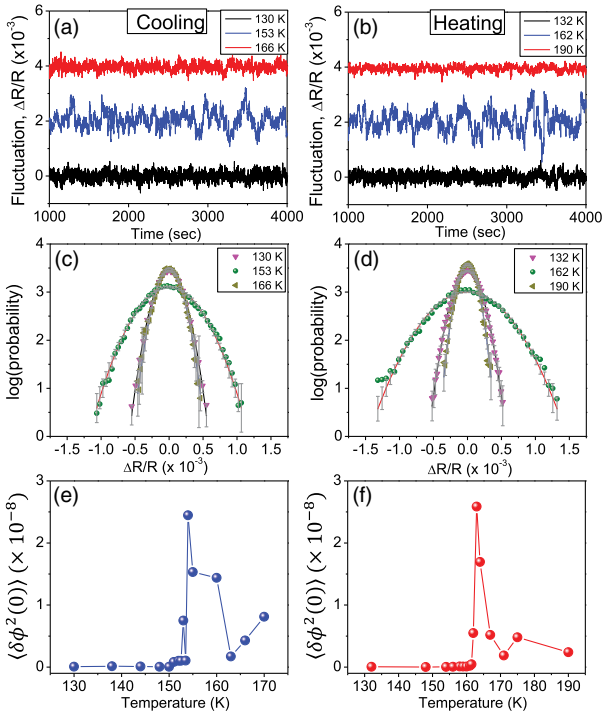

FIG. 2. Time series of the resistance fluctuations at different temperatures. The left column depicts the measurements done under cooling, and the right panel shows them under heating. (a),(b) Normalized resistance fluctuations after the time series were detrended by imposing a low-frequency cutoff of about 10^{-3} Hz [54]. (c),(d) The probability distribution of fluctuations. Solid lines represent the Gaussian distribution with zero mean, and the given variance and the error bars roughly represent 99.7% confidence for this distribution. (e),(f) Variance of the order parameter fluctuations $\langle \delta\phi^2(0) \rangle$ roughly inferred from the resistivity time series. The fluctuations are expected to follow the divergence of χ_T around the spinodals [Eq. (5)].

[Figs. 2(a) and 2(b)] can then be approximately converted to the time series of the fluctuations in the spatial average of this order parameter $\delta\phi(t)$ using results from the percolation theory [54,56]. Enhanced variance of the order parameter fluctuations $\langle\delta\phi^2(0)\rangle$ around the transition [Figs. 2(e) and 2(f)] is the first indication of criticality [21].

Figures 3(a) and 3(b) show the corresponding normalized power spectral density $S_R(f)/R^2$ in the frequency window of $0.3 - 0.003$ Hz [54] at different temperatures. The PSD obeys the characteristic $f^{-\mu}$ behavior [9,20], with an increase in the value to $\mu \approx 1.6$ around the transition, indicating a shift of the spectral weight of fluctuations to lower frequencies. Such slowing down has been previously observed in the resistance-noise data at the Mott critical point [20] and simulations of the Ising model [22]. Noise studies at the APT have also observed qualitatively similar features [9,11,14]; but, the combination of the large hysteresis, a very sharp transition, and the similarity in the behavior along the cooling and the heating runs in bulk V_2O_3 studied here has made the identification with the spinodal singularity more obvious. Our ongoing work on epitaxial $NdNiO_3$ thin films also qualitatively reproduces Figs. 3(c) and 3(d), indicating its generality. Indeed, recent works on a ferrite [16] and VO_2 [5,6] also invoked "pseudocriticality" [38] to explain the slowed dynamics around APT.

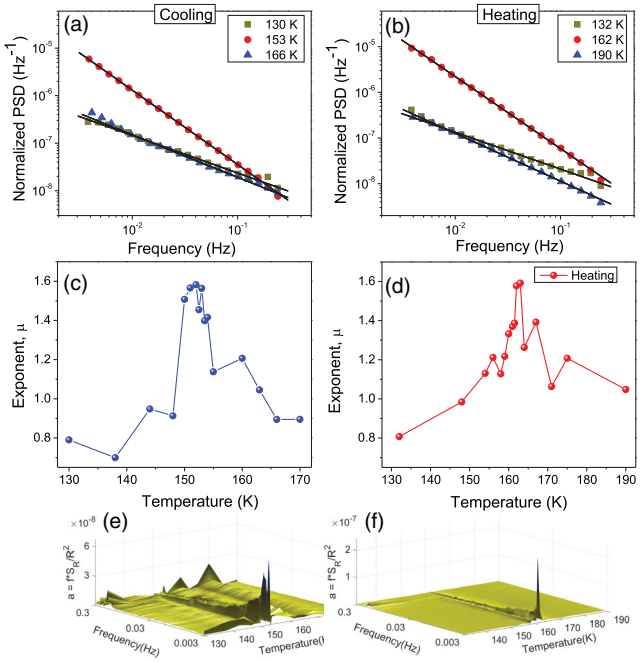

FIG. 3. Critical slowing down of fluctuations around the spinodals. (a),(b) Normalized noise power spectral density (PSD) S_R/R^2 at some representative temperatures. (c),(d) Temperature dependence of exponent μ, where $S_f \sim f^{-\mu}$. (e),(f) A surface plot of $f \times S_R/R^2$ to visualize the non-$1/f$ behavior when the spectral weight shifts to lower frequencies.

In Fig. 4(a), this signature of slowing down is further evident from the relaxation time τ_2 of the autocorrelation of the fluctuations [54]. In Fig. 4(b), we have also independently estimated the phase-ordering time τ_1 where the sample was shock heated and quench cooled, respectively, at 50 K/ min to the desired target temperature marked on the abscissa [54]. τ_2 also shows a distinct enhancement around the transition [54]. Such an increase in the lifetime with deeper supersaturation (and consequent reduction in the energy barrier) within the metastable region would seem counterintuitive for a thermally activated process like nucleation, but it is the expected behavior at the spinodal [2].

The spinodal singularities.—Let $F[\varphi]$ be the free energy functional of the system with an order parameter $\varphi(\mathbf{x}, t)$ that denotes the fraction of the insulating phase in the material [23,55]. The dynamical behavior of this stochastic variable $\varphi(\mathbf{x}, t)$, which is a nonconserved scalar, is then given by the dissipative model A [18]

$$\frac{\partial}{\partial t}\varphi(\mathbf{x}, t) = -\lambda\frac{\delta F[\varphi]}{\delta\varphi(\mathbf{x})} + \zeta(\mathbf{x}, t). \tag{1}$$

Here, λ is a kinetic parameter (that was previously determined to be 3.5 s^{-1} [23]), and the thermal noise $\zeta(\mathbf{x}, t)$ is (as usual) assumed to be δ correlated, i.e., $\langle\zeta(\mathbf{x}, t)\zeta(\mathbf{x}', t')\rangle = 2\lambda k_B T\delta(\mathbf{x} - \mathbf{x}')\delta(t - t')$, with zero mean [18]. The angular brackets denote canonical ensemble averaging.

If $\varphi = \varphi_0$ at the boundary of the metastable phase (spinodal) in the mean-field approximation, we can Taylor expand $F[\varphi]$ around this saddle point, keeping only Gaussian fluctuations, viz.,

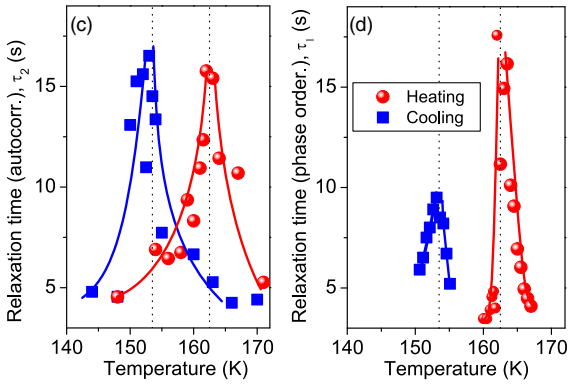

FIG. 4. (a) The temperature dependence of the relaxation time τ_2 for the autocorrelation of the detrended resistance fluctuations, and (b) the phase ordering time τ_1 during heating and cooling. See Supplemental Material [54] for the details of the phase-ordering experiments. Solid lines are to guide the eye. The region between $\sim 153 - 162$ K is the metastable phase. Critical slowing down reflected in both τ_1 and τ_2. In the linear approximation, they are equal to $\tau_{k=0}$ and diverge with the susceptibility exponent.

$$F[\varphi] \approx F[\varphi_0] + \frac{1}{2}\int d\mathbf{x}d\mathbf{x}'\delta\varphi(\mathbf{x})[\beta G(\mathbf{x}-\mathbf{x}')]^{-1}\delta\varphi(\mathbf{x}'),$$

where

$$[\beta G(\mathbf{x}-\mathbf{x}')]^{-1} = \frac{\delta^2 F[\varphi_0]}{\delta\varphi(\mathbf{x})\delta\varphi(\mathbf{x}')}$$

and $\beta^{-1} = k_B T$. $G(\mathbf{x}-\mathbf{x}')$ is the correlation function evaluated at φ_0, and let $\tilde{G}_{\mathbf{k}}$ be its Fourier transform [18]. If $\delta\tilde{\varphi}_{\mathbf{k}}(t)$ is the Fourier transform of $\delta\varphi(\mathbf{x},t) \equiv \varphi(\mathbf{x},t) - \varphi_0$, the first two moments of $\delta\tilde{\varphi}_{\mathbf{k}}(t)$ evolve as

$$\langle\delta\tilde{\varphi}_{\mathbf{k}}(t)\rangle = \langle\delta\tilde{\varphi}_{\mathbf{k}}(0)\rangle \exp(-t/\tau_{\mathbf{k}}), \tag{2}$$

$$\langle\delta\tilde{\varphi}_{\mathbf{k}}(t)\delta\tilde{\varphi}_{\mathbf{k}}(0)\rangle = V\tilde{G}_{\mathbf{k}} \exp(-t/\tau_{\mathbf{k}}). \tag{3}$$

$\tau_{\mathbf{k}} = \beta\tilde{G}_{\mathbf{k}}/\lambda$ is the relaxation time for the $\mathbf{k}$th mode.

The sample resistance, after correcting for percolation effects [54,56], tracks the spatial average of the order parameter [55], i.e., essentially the zero-wave vector Fourier mode $\tilde{\varphi}_{k=0}(t)[= \int d\mathbf{x}\varphi(\mathbf{x},t)] \equiv \phi(t)$. Thus, our measurements reflect Eqs. (2) and (3) with $k = 0$. By the sum rule [19], the isothermal susceptibility $\chi_T = \beta\tilde{G}_{k=0}$. It follows that $\chi_T = [\delta^2 F/\delta\phi^2]^{-1} \to \infty$ at the spinodals, which is in strict analogy with its behavior at the critical point [18]. Phase ordering monitored through resistance should thus evolve as

$$\langle\delta\phi(t)\rangle = \langle\delta\phi(0)\rangle \exp(-t/\tau_0); \quad \tau_0 = \chi_T/\lambda, \tag{4}$$

and the autocorrelation of the resistance noise should be related to

$$\langle\delta\phi(t)\delta\phi(0)\rangle = Vk_BT\chi_T \exp(-t/\tau_0); \quad \tau_0 = \chi_T/\lambda. \tag{5}$$

The phase-ordering time τ_1 and the autocorrelation time τ_2 [Fig. 4] are equal to τ_0 and diverge proportional to χ_T. Equation (4) further shows that the variance $\langle\delta\phi^2(0)\rangle$ should also diverge as χ_T (critical opalescence), which is in agreement with Figs. 2(e) and 2(f) [37]. Regarding the slopes of the PSD (Fig. 3), the linear theory would predict Brownian dynamics ($\mu = 2$) at the spinodals. Theoretical estimates for the critical point of the two-dimensional Ising model under Glauber dynamics suggest $\mu \sim 1.8$ [22].

Discussion.—It had long been an open question whether the spinodal singularity could be seen in finite temperature experiments. Although, for short-ranged interactions, the spinodals are physically meaningless [2], in the opposite limit of infinite-ranged interactions, the mean-field theory becomes exact [45]. Thus, the point of contention is whether the spinodal may be a meaningful concept for long but finite-ranged interactions [50]. Theoretically, the issue is addressed by constructing, as usual [18], a relationship (Ginzburg criterion) between the relative order parameter fluctuations, system dimensionality, and the range of interactions as one approaches the spinodal singularity [2,51,57]. Provided the interactions are sufficiently long-ranged, one can get close enough to the instability that finite temperature effects broaden the transition but may not completely mask it [33,51,58], mimicking a finite-size effect. This is indeed what we see; the singularity still manifests as a very discernable (but finite) growth in the susceptibility. That we have long-range interactions in the system [59] is independently indicated by the observations of broken ergodicity [Fig. 1(b)] and very slow relaxation (Fig. 4), which are otherwise unusual for hard condensed matter systems. Furthermore, the robust presence of hysteresis itself indicates long-range forces on rigorous theoretical grounds [60].

The electronic phase transitions in a number of correlated electron systems are often also accompanied by abrupt structural transitions on account of the large polaronic coupling [10,25]. The resulting strain fields are the sources of long-range interactions [29,58,61]. Indeed, a structural transition does seem like the feature shared across disparate systems undergoing APT with pronounced hysteresis [3–16,23–25,28]. These are all candidates for the zeroth-order transition belonging to the mean-field "universality class" [30,62].

We have not discussed disorder. Although the phase coexistence and ramified fractal-like structures in the transition regions [7] are also explained by the nonclassical nucleation expected at the spinodal [34,58] without invoking disorder, it is perhaps essential for understanding avalanches [40,53] or even the return-point memory [52,53] seen in Fig. 1(b). Although one would naturally expect the smooth evolution suggested by Eq. (1) to be not quite valid [63], the spinodal singularity itself is expected to be robust to disorder for sufficiently long-range interactions [66]. But, this is clearly an issue that requires further systematic investigation.

Finally, catastrophes [47] in natural and social systems—earthquakes, ecosystem collapse, climate change, onset of depression, epilepsy, market crash—are sometimes modeled after such zeroth-order transitions, with the spinodal singularity signifying the loss of "resilience" [67]. Anticipating them is, of course, an important objective of complex systems science. The idea that fluctuations can carry precursory signatures of such an impending event, in the form of critical slowing down accompanied by an enhanced variance of fluctuations [67], is perfectly vindicated by our work.

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