Kinetic Spinodal Instabilities in the Mott Transition in V_2O_3: Evidence from Hysteresis Scaling and Dissipative Phase Ordering

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We present the first systematic observation of scaling of thermal hysteresis with the temperature scanning rate around an abrupt thermodynamic transition in correlated electron systems. We show that the depth of supercooling and superheating in vanadium sesquioxide (V_2O_3) shifts with the temperature quench rates. The dynamic scaling exponent is close to the mean field prediction of $2/3$. These observations, combined with the purely dissipative continuous ordering seen in "quench-and-hold" experiments, indicate departures from classical nucleation theory toward a barrier-free phase ordering associated with critical dynamics. Observation of critical-like features and scaling in a thermally induced abrupt phase transition suggests that the presence of a spinodal-like instability is not just an artifact of the mean field theories but can also exist in the transformation kinetics of real systems, surviving fluctuations.

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Metastable states do not exist in equilibrium statistical mechanics, as any legitimate free energy must be convex in the thermodynamic limit [1]. But many real systems do spontaneously fall out of equilibrium in a window of thermal hysteresis around the abrupt phase transition (APT) [2]. The accompanying nonergodic behavior—arrested kinetics [3,4], spatial inhomogeneity [5,6] and phase coexistence [7–9], and rate dependence [10–12]—is well documented.

Within the mean field (MF) picture, this metastable phase is predicted to abruptly terminate at the spinodals, the two values of field or temperature where the barrier against nucleation vanishes [2,7,13–18]. The analogy between the MF spinodals and the critical point in the power law divergence of susceptibility [2,13,18–20] and their being fixed points under renormalization group transformation [17,21] has long been discussed [2,18,22]. Except for the case of strictly athermal systems [7,14,24], these ideas were never taken seriously because one would expect this singularity to be physically inaccessible; fluctuations accompanying any finite-temperature phase transformation would necessarily yield pathways involving nucleation before the spinodal is experimentally reached [1,13,20,25].

Nevertheless, long-ranged forces arising, for example, due to the accompanying structural transition [26] may to an extent [27] suppress fluctuations. This will naturally lead to deep supersaturation and thermal hysteresis in the phase transformation and thus take the system beyond the regime of the classical nucleation theory [28–32]. As the nucleation barriers get smaller, simulations show spatially diffused and continuous ordering mechanisms [29], where the dynamic limit of metastability can extend to the critical nucleus shrinking to less than one molecule [30]. Hence, operationally, the phase ordering may actually mimic the MF spinodal behavior, with fluctuations only making quantitative corrections. In fact, there is increasing theoretical evidence that the essence of this MF picture, i.e., the existence of singular fixed points, is retained in the dynamical behavior even for model systems with short-ranged interactions at finite temperature [21,33].

Focusing on the APT in V_2O_3 [5,34–36], in this Letter, we report the first experimental observation of such dressed MF behavior in phase ordering via the study of dynamic hysteresis.

Experiments.—Figures 1(a) and 1(b) show quasiequilibrium differential thermal analysis (DTA) and transport measurements done at temperature ramp rates of < 0.5 K/min using polycrystalline V_2O_3 (see Supplemental Material [37]); the transition is strongly hysteretic and the abrupt change in resistance is accompanied by latent heat $L \approx 2$ kJ/mole ($\sim 15RT_c$) [53]. This APT is known to arise due to three simultaneous—electronic, structural, and magnetic—transformations [Fig. 1(b), inset] [34,35]. That the window of thermal hysteresis around ~ 158 K does correspond to the metastable region where the physical observables are no longer state variables is seen in the multivaluedness of the sample resistance in Fig. 1(c). These minor hysteresis loops were drawn using the temperature protocol shown in Fig. 1(c) (inset).

Dynamic hysteresis.—Figures 2(a) and 2(b) show the DTA and the resistance data for linear temperature ramp rates, between 0.2 and 60 K/min. We observe a systematic

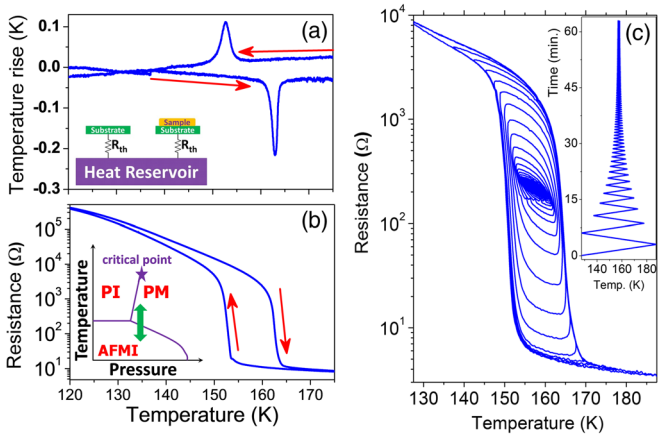

FIG. 1. (a) Exothermic (∼153 K) and endothermic (∼162 K) latent heat peaks in the DTA experiment. (Inset) Schematic of the setup [37]. (b) The resistance changes over many orders of magnitude around the same temperature. (a),(b) Quasiequilibrium properties measured at slow temperature ramp rates ($\lesssim 0.5$ K/ min). (Inset) Phase diagram of V_2O_3 in the pressure-temperature plane with antiferromagnetic insulator (AFMI), paramagnetic insulator (PI), and paramagnetic metal (PM) phases separated by first-order lines [34]. The arrow corresponds to the temperature-dependent transition (at ambient pressure) studied in this Letter. This transition also corresponds to a structural change from a rhombohedral (PM) to monoclinic (AFMI) phase. (c) Metastability of the hysteretic region is seen in the multivaluedness of the sample resistance. The fraction of the phase-transformed material within this region can be controlled by the sample's thermal history. (Inset) The corresponding time dependence of the temperature sweep.

delay in the onset temperature that is dependent on the temperature ramp rate R. A model-independent way to depict this dynamic shift is to plot the rate-dependent depths of supercooling and superheating. This is done in

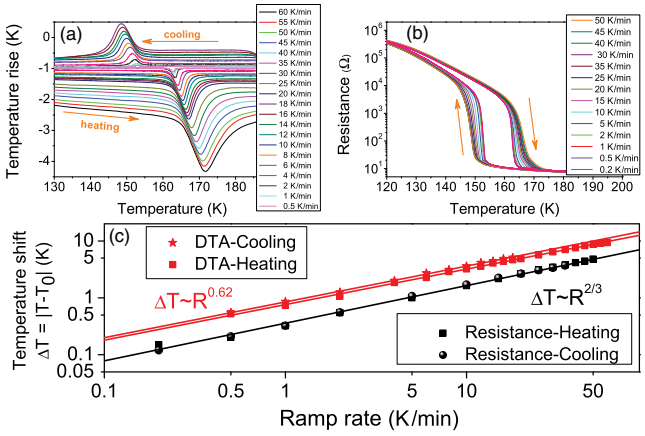

FIG. 2. (a) DTA signal and (b) the sample resistance as a function of temperature for different linear ramp rates. (c) Shift in the transition temperature with temperature scanning rate $\Delta T(R)$. The temperature shifts inferred from the DTA extrema are best fitted to $\Upsilon = 0.62$ [37]. The transport data also approximately obey $\Delta T(R) \propto R^{2/3}$.

Fig. 2(c) where the dynamically renormalized shifts $\Delta T(R)$ in the observed transition temperature $T^{\rm heat}_{\rm obs}(R)$ and $T^{\rm cool}_{\rm obs}(R)$ are seen to obey the scaling relationship $\Delta T(R) = |T^i_0 - T^i_{\rm obs}(R)| \propto R^{\Upsilon}$, $i =$ heat (heating) or cool (cooling) over two decades of ramp rates. $T^{\rm heat}_0$ and $T^{\rm cool}_0$ were used as free parameters, varied to yield the best straight lines in the log-log graph [37]. $T^{\rm heat}_0 \approx 162.77$ K and $T^{\rm cool}_0 \approx 153.35$ K thus correspond to the transition temperatures under quasistatic heating and cooling, respectively. The fact that $T^{\rm heat}_0$ and $T^{\rm cool}_0$ are not known *a priori* make the estimation of Υ difficult. The Supplemental Material [37] discusses this further. The values of T^i_0, which minimize the error in the straight line fits (on log-log scale), yield $\Upsilon \approx 0.62$ for both cooling and heating. Another independent estimate yields $\Upsilon = 0.62 \pm 0.06$ for heating and $\Upsilon = 0.64 \pm 0.09$ (cooling) [37]. It is significant that one should observe this symmetry. The above analysis was performed for the DTA data. It can be seen that the resistance data, where there is a greater ambiguity in extracting the actual transition temperatures, also nevertheless suggest that $\Upsilon \approx 2/3$.

To understand these observations, note that, in the MF picture, the order parameter ϕ would evolve by the same equation that is used for *critical dynamics*. For nonconserved ϕ, this is the dissipative time-dependent Landau (TDL) equation or model A [38,39]

$$\frac{\partial \phi}{\partial t} = -\lambda \frac{\delta}{\delta \phi} f(\phi, T) + \zeta(t). \tag{1}$$

Here $f(\phi, T)$ is the free energy, and λ is a kinetic parameter. The stochastic force $\zeta(t)$ is zero under MF approximation. As a consequence of the above dynamics, critical-like slowing down would be observed around the transition if (and only if) the system approaches a genuine bifurcation point where the dynamic susceptibility is singular. Under the sweep of field or temperature with time, a systematic delay in the onset of phase switching is predicted with a definite scaling form. The change in the area $A(R)$ of the hysteresis loop (or, equivalently, the shift in the transition point) must dynamically scale with R, the rate of change of field H, or temperature T, as a power law [39,40,54–58]

$$A(R) = A_0 + aR^{\Upsilon}, \tag{2}$$

where A_0 is the area of the quasistatic hysteresis loop. Under the deterministic evolution demanded by the MF theory, the instabilities are the spinodals $T^{\rm heat}_0$ and $T^{\rm cool}_0$ determined above. Remarkably, numerical calculations of the different (spatially averaged) free energies describing field- or temperature-induced APT all yield $\Upsilon = 2/3$ [37,54,56,57,59,60], rather close to what we have experimentally observed.

This universality has been justified by dynamic scaling arguments [39]. $\Upsilon = 2/3$ can indeed be recovered under the conditions of the linear ramp of the field [40] or

temperature [40] if the other critical exponents are chosen to be those belonging to Fisher's ϕ^3 theory with imaginary coupling that describes the Yang-Lee-edge singularity [41]. This is reasonable because of the known equivalence within the MF Ising model of the Yang-Lee edge (imaginary fields, $T > T_c$) with the spinodal (real field, $T < T_c$) through analytic continuation [65].

Free energy and order parameter.—Because of the interplay of lattice, spin, and orbital degrees of freedom that gives rise to three simultaneous transitions in V_2O_3, the temperature-induced APT is more complicated than the Ising-like transition at the Mott critical point [67]. A phenomenological extension to the Ising model that will make it a temperature-driven APT and also, albeit in a rather simplistic way, capture the accompanying structural transition is the compressible Ising model [37,42,43]. Here the lattice compressibility is coupled to the spin via the exchange coupling constant J of the Ising model [37]. In the mean field approximation, the resulting dimensionless free energy per spin can be written as [37]

$$f = \frac{T}{2T_c}[(1+\phi)\ln(1+\phi) + (1-\phi)\ln(1-\phi)] - \frac{\phi^2}{2} - \xi\phi^4. \tag{3}$$

The scalar nonconserved order parameter ϕ (the average "magnetization" per site) is identified with the fraction of the insulating phase. $|\phi| < 1$ at any nonzero temperature and T_c is the critical temperature. For $[T/(12T_c)] - \xi < 0$, one would observe an APT during thermal cycling.

Figure 3 shows a method to experimentally estimate ϕ at the given temperature using DTA. ϕ is taken to be proportional to the integrated area around the DTA dip (peak) for the cooling (heating) curves in Figs. 3(a) and 3(b), respectively, as a function of the temperature of approach and is plotted in Fig. 4. A 1000 s wait at the temperature of approach ensures quasistatic conditions. Also in Fig. 4, ϕ is independently estimated from the resistance data using McLachlan's effective medium theory [66] to approximately handle the percolative nature of the transport [37].

Remarkably, the two free parameters of the model, ξ and T_c, are already fixed by the experimentally inferred spinodal temperatures. $T_c \approx 153.5$ K and the value of ξ is numerically determined to be 0.154 from the value of the other spinodal temperature to be 162.5 K. Thus to describe the dynamics [Eq. (1)], the remaining free parameter λ is also fixed by fitting any one of the dynamic hysteresis curves; $\lambda = 3.5\ \mathrm{s}^{-1}$ was obtained by fitting the curve for the inferred order parameter (fraction of the insulator phase) by evolving Eq. (1) for heating with a linear temperature ramp at the rate of 50 K/ min. In the simulation discussed in the Supplemental Material [37], a hysteresis scaling exponent of 2/3 is observed, as is expected from generic arguments given above. The results for the inferred order parameter

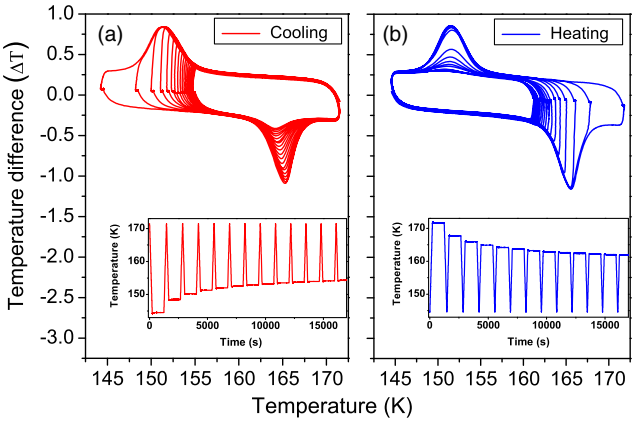

FIG. 3. Reversal curves for (a) cooling and (b) heating measured in DTA to map out the limits of stability and the width of the spinodal region and directly estimate the change in the order parameter across the transition. (Insets) Corresponding time dependence of temperature. (a) Starting with the initial temperature of 171.5 K, the sample is cooled to a set temperature T_i^s, $i = 1$. On reaching this set temperature, the system stays there for 1000 s to reach the equilibrium value. The system is heated back to 170 K and the process is repeated with a slightly higher set temperature, such that $T_2^s > T_1^s$. The area of the latent heat peak at ~165 K is a measure of the amount of material transformed, the order parameter at T_i^s. This allows us map out the cooling spinodal region. (b) Shows the same idea implemented for the heating spinodal. The order parameter extracted from these measurements is shown in Fig. 4(a).

from the compressible Ising model are also shown in Fig. 4. The transition was given a very small but finite width by assuming that the sample is an inhomogeneous ensemble with a Gaussian distribution of $T_c = 153.8$ with a standard deviation of 0.18 K.

Phase ordering in quench-and-hold experiments.—Starting from the initial temperature of 100 K (240 K), the experimental contour plots in Fig. 5 are obtained by rapidly (at the rate of 50 K/ min) heating (cooling) the sample to different target temperatures T_w slightly above (below) the quasistatic transitions temperatures [37]. Once T_w was reached, the temperature was kept constant and the

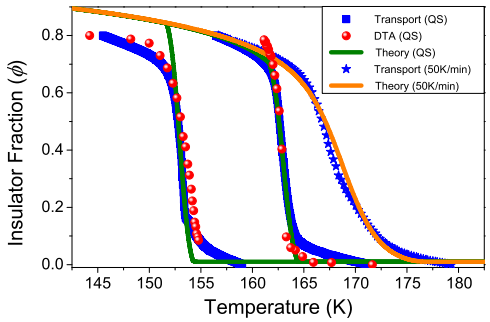

FIG. 4. (○) The quasistatic (QS) order parameter ϕ vs temperature extracted from DTA using data in Fig. 3. (□, ∗) ϕ inferred from resistance measurements. (—) Compressible Ising model.

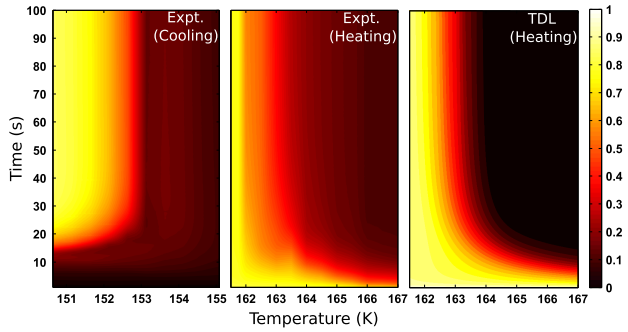

FIG. 5. Critical-like quench-and-hold dynamics of the order parameter (insulator fraction) around the cooling and heating spinodals. The data show the first 100 s of the time evolution after the target temperature T_w (denoted by the abscissa) has been reached [37]. The calculation involves evolving the compressive Ising model using model A [Eq. (1)] with no free parameters [37].

time evolution of the resistance at different T_w is plotted in terms of the insulator fraction [37,66]. Figure 5 demonstrates that (in a qualitative sense) the phase transformation proceeds symmetrically, with similar timescales. Given that the metastable phase is bounded also on the low-temperature side makes the physics of arrested kinetics of the metastable phase qualitatively different from that observed in glasses.

The corresponding calculation [using Eqs. (1) and (3)] for the noise-free evolution of the order parameter after shock heating is also shown in Fig. 5. With T_c, ξ, and λ already fixed, the entire contour plot has no free parameter. The qualitative match with the highly constrained MF calculation supports the picture of phase transformation occurring via barrier-free continuous ordering. Calculation for the cooling quench, due to its sensitivity on the initial conditions, is not discussed.

Conclusions.—The unreasonable efficacy of the MF theory in capturing the essence of the transformation in V_2O_3 gives credence to the idea that—and this is the key result of our work—spinodal-like instabilities can be present in a real material exhibiting a finite-temperature abrupt phase transition. Scaling of dynamic hysteresis with the observed exponent and barrier-free phase ordering are both manifestations of these instabilities.

At least for phase transformation under such deep supersaturation, recent work on very different aspects of the problem suggests that fluctuations may not fundamentally affect these qualitative aspects. Within the Ising model in thermal *equilibrium* (and $T < T_c$), the MF spinodal corresponds to the two values of magnetic field demarcating the limit of metastability. Rigorous mathematical analysis shows that the effect of fluctuations (or, equivalently, making the range of interactions finite) is simply to rotate this spinodal magnetic field in the complex plane, giving it a nonzero imaginary value [32,65]. Thus, in analogy with the Yang-Lee argument that the complex zeros of the partition function only touch the real axis in the thermodynamic limit, fluctuations essentially mimic finite-size effect [32]. Remnants of this singularity should be discernable in the broadened transition if the range of the potential is large enough, as it might be for V_2O_3 due to the deep supersaturation. Furthermore, for a dynamically changing control parameter, the system may get too sluggish in the vicinity of the transition (because of the critical-like slowing down) to turn on these fluctuations before the transition has occurred. Numerical solutions of model A [Eq. (1)], now also including the stochastic term ($\zeta \neq 0$), do indeed show that fluctuations only slightly change the value of the exponent [39,68] describing this dynamic overshoot, still not far from our observations.

While these arguments make the experimental observations plausible, it is emphasized that the metastable phase is properly only to be defined in a dynamical sense. Dynamically emerging spinodal-like singularities have been seen in simulations of Lennard-Jones fluids [29–31], binary alloys [69], elementary models [21,33], and perhaps even in some other experiments [70]. Thus, while the precise nature of the kinetic spinodals is yet to be determined, their existence in specific contexts seems real enough. These instabilities have been variously interpreted—for example, as the boundary between the regions of homogeneous and heterogenous nucleation [69].

Such strongly hysteretic "zeroth-order" transitions [71] thus form a new class of transitions that may potentially be observed in many other systems, including similar oxides undergoing metal-insulator transition [6,9], manganites [10], intermetallic shape-memory alloys, and magnetocaloric materials [72]. A better understanding of the phase transformation kinetics in such systems should help chart the uncertain territory of metastable states in the language of critical phenomena.

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