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Dynamical slowing down in an ultrafast photo-induced phase transition

Alfred Zong,1 Pavel E. Dolgirev,2,3 Anshul Kogar,1 Emre Ergeçen,1 Mehmet B. Yilmaz,1 Ya-Qing Bie,1, * Timm Rohwer,1,4 I-Cheng Tung,4 Joshua Straquadine,5,6,7 Xirui Wang,1 Yafang Yang,1 Xiaoze Shen,4 Renkai Li,8 Jie Yang,8 Suji Park,8,9 Matthias C. Hoffmann,10 Benjamin K. Ofori-Okai,8 Michael E. Kozina,8 Haidan Wen,4 Xijie Wang,8 Ian R. Fisher,5,6,7 Pablo Jarillo-Herrero,1 and Nuh Gedik1,4

1Massachusetts Institute of Technology, Department of Physics, Cambridge, Massachusetts 02139, USA.
2Skolkovo Institute of Science and Technology, Skolkovo Innovation Center, 3 Nobel Street, Moscow, 143026, Russia.
3Department of Physics, Harvard University, Cambridge, Massachusetts, 02138, USA.
4Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois, 60439, USA.
5Geballe Laboratory for Advanced Materials, Stanford University, Stanford, California 94305, USA.
6Department of Applied Physics, Stanford University, Stanford, California 94305, USA.
7SIMES, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA.
8SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA.
9Department of Materials Science and Engineering, Stanford University, Stanford, California 94305, USA.
10Linac Coherent Light Source, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA.

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Complex systems, which consist of a large number of interacting constituents, often exhibit universal behavior near a phase transition. A slowdown of certain dynamical observables is one such recurring feature found in a vast array of contexts. This phenomenon, known as critical slowing down, is well studied mostly in thermodynamic phase transitions. However, it is less understood in highly nonequilibrium settings, where the time it takes to traverse the phase boundary becomes comparable to the timescale of dynamical fluctuations. Using transient optical spectroscopy and femtosecond electron diffraction, we studied a photo-induced transition of a model charge-density-wave (CDW) compound, LaTe$_3$. We observed that it takes the longest time to suppress the order parameter at the threshold photoexcitation density, where the CDW transiently vanishes. This finding can be captured by generalizing the time-dependent Landau theory to a system far from equilibrium. The experimental observation and theoretical understanding of dynamical slowing down may offer insight into other general principles behind nonequilibrium phase transitions in many-body systems.

In a second-order symmetry-breaking phase transition, the spatial extent of fluctuating regions diverges close to the critical temperature, $T_c$. Correspondingly, the relaxation time of these fluctuations tends to infinity, a phenomenon known as critical slowing down [1, 2]. The phenomenology of slowing dynamics near a critical point is much more general: it has been observed in first-order transitions [3, 4], glasses [5, 6], dynamical systems [7], and even microbial communities [8]. Its common occurrence makes it a robust signature of phase transitions in a vast array of complex systems [9].

Close to equilibrium, critical slowing down has been well characterized in condensed matter systems. Theoretically, it is described by a dynamical critical exponent, whose value depends on the dynamic universality class [2]. Experimentally, the evidence comes from a vanishing rate of change in the order parameter close to $T_c$, with early reports in Refs. [3, 10, 11]. While these measurements probe the slowing dynamics in the time domain, it can be observed in the frequency domain as well. For example, inelastic neutron scattering has revealed a narrowing quasi-elastic peak along the energy axis as $T_c$ is approached, indicating a suppressed relaxation rate of critical fluctuations [12–14]. Moreover, if there is a collective mode associated with the phase transition, the mode softening in the vicinity of $T_c$ is also taken as a signature of critical slowing down [15].

For symmetry-breaking phase transitions in a highly nonequilibrium setting, the dynamics are much less understood. Recent studies have found important features in nonequilibrium transitions, such as topological defects, which are absent in their equilibrium counterparts [16–18]. Despite the differences, a slowdown in dynamics is thought to carry over to systems far from equilibrium. For example, in a rapid quench into a broken-symmetry state, the Kibble-Zurek theory suggests that critical slowing down plays a central role in domain formation: as the phase boundary is traversed at a faster rate than the system can respond, spatially disconnected regions may adopt distinct configurations of the same degenerate ground state [19]. Characteristic domain structures in liquid crystals have indeed been observed [20, 21], providing indirect evidence for the slowdown.

To study the dynamics in a nonequilibrium setting, charge-density-wave (CDW) transitions instigated by an intense femtosecond laser pulse provide an accessible platform with well-controlled tuning parameters. A suite of time-resolved probes can track the evolution of electronic and lattice orders after strong photoexcitation [18], offering insights into the critical behavior, if present, during the phase transition. Immediately after photoexcitation, a coherently-excited CDW amplitude mode was ob-
served to soften transiently [16], hinting at critical slowing down. Right below $T_c$, a diverging relaxation time back to equilibrium was interpreted as another signature [4, 22]. However, observables in previous studies, such as amplitude mode frequency or quasiparticle relaxation time across the spectroscopic gap, are only well defined in the broken-symmetry state [23–25]. To demonstrate slowing dynamics in the vicinity of a nonequilibrium phase transition, ideally one would measure an increased timescale near the phase boundary compared to both ordered and disordered states.

In this work, we circumvent this obstacle by focusing on a different observable during the photo-induced melting of a CDW: the time taken to suppress the condensate. With increasing photoexcitation densities, the perturbed system will enter one of the two transient states, where the CDW is either partially or completely suppressed [18]. The two states are separated by the threshold excitation density, $F_{\text{th}}$, where the condensate first vanishes completely. Through transient reflectivity and time-resolved diffraction measurements at different excitation densities, we observed that it takes the maximum time to suppress the CDW right at $F_{\text{th}}$, indicating dynamical slowing down near the boundary between the two transient states. Here, we use dynamical slowing down to emphasize the highly nonequilibrium nature of the system and to distinguish it from critical slowing down commonly defined in a second-order phase transition in equilibrium [1, 2].

The material of interest is a paradigmatic CDW system, LaTe$_3$ [26]. Like other rare-earth tritellurides, LaTe$_3$ possesses a quasi-2D structure (Fig. 1(a)) and develops a unidirectional CDW with wavevector $q_{\text{CDW}}$ along the $c$-axis below $T_c \approx 670$ K [27]. In equilibrium, the CDW transition is characterized by the appearance of satellite peaks in a diffraction pattern (Fig. 1(b)) as well as gap openings at certain parts of the Fermi surface connected by $q_{\text{CDW}}$ [28].

Upon the arrival of a strong femtosecond laser pulse, the CDW order is transiently suppressed [18, 29, 30]. We first establish the timescale for this process by performing ultrafast electron diffraction (UED) and transient optical spectroscopy (TOS), which reveal how the lattice and electrons respond to intense photoexcitation, respectively (Fig. 1(a)). Previous measurements from time- and angle-resolved photoemission spectroscopy (trARPES) [18] and time-resolved X-ray diffraction (trXRD) [29] are also included to obtain a comprehensive and consistent view of the ultrafast melting process.

While UED and trXRD track the evolution of CDW satellite peaks at characteristic wavevector $q_{\text{CDW}}$ (Fig. 1(b)), TOS and trARPES probe the change in the spectroscopic gap (Fig. 1(c)) [18]. Despite the different observables, the initial response that corresponds to CDW melting proceeds with a similar timescale, denoted by $\tau$ (Fig. 1(d)). The rising edges across the four techniques in Fig. 1(d) all span a time interval of $\tau \approx 400$ fs, with variations arising from the different temporal resolutions in each setup [31] and different photoexcitation densities used (Fig. 2(d)). The agreement among structural and electronic probes suggests the presence of strong electron-phonon coupling in this system. Notably, the value of 400 fs is on the same scale as the period of the 2.2 THz CDW amplitude mode [25], further indicating the vital role of lattice vibrations in the formation of the charge order [32].

Among the four techniques discussed, TOS possesses the best temporal resolution and signal-to-noise ratio [31]. Hence, it enables us to more quantitatively investigate the timescale of CDW suppression, $\tau$, as we vary the laser excitation density, $F$, quoted in terms of absorbed photon number per unit volume [18]. Figure 2(a) shows the temporal evolution of the transient reflectivity, $\Delta R/R$, across a large range of $F$. The trace from Fig. 1(d) is overlaid at the corresponding $F$. The data presented was taken using a probe photon energy of 1.80 eV (690 nm), which is selected among the white light super-continuum because it is the energy most sensitive to the dynamics of the CDW gap [31].
The non-monotonic trend of \( \tau \) in the UED experiments suggests the same non-monotonic behavior compared to the TOS measurements, the initial timescale in signal-to-noise ratio and poorer temporal resolution compared to the TOS measurements, the initial timescale in signal-to-noise ratio and poorer temporal resolution.

In Fig. 2(b), we present an example cut (red curve) at \( F = 5.81 \times 10^{20} \text{ cm}^{-3} \). To quantitatively evaluate the initial response time \( \tau \), we performed a global fit for traces at all excitation densities using a two-component phenomenological model with minimal parameters. An example fit is presented in Fig. 2(b), showing excellent agreement. We attribute the two components to quasiparticle excitations in different parts of the Brillouin zone (see Ref. [31] for details of the fitting model and its interpretation), and we extract \( \tau \) from the rise time of the first component (Fig. 2(b), blue dashed curve).

Remarkably, the rise time \( \tau \) displays a non-monotonic trend as a function of excitation density (Fig. 2(d)), with a maximum at \( \approx 2 \times 10^{20} \text{ cm}^{-3} \) (black arrow). The non-monotonic trend of \( \tau \) is independent of the fitting model, and is clearly observed in the raw data (Fig. 2(a)). To confirm this observation, we similarly track the suppression of superlattice peaks using UED at various excitation densities (Fig. 2(d), blue diamonds and Fig. 2(c)).

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Significantly larger errors due to lower signal-to-noise ratio and poorer temporal resolution compared to the TOS measurements, the initial timescale in the UED experiments suggests the same non-monotonic behavior in \( \tau \). We further note a recent measurement on SnTe3 [33], a CDW compound in the same family as LaTe3, which demonstrates a similar trend in the initial response.

To associate this non-monotonic behavior in \( \tau \) with dynamical slowing down during photo-induced CDW melting, we next establish that the melting proceeds the slowest precisely at the threshold excitation density when the CDW in the illuminated sample volume is just fully destroyed, namely, \( F_{\text{melt}} \approx 2 \times 10^{20} \text{ cm}^{-3} \). We make three observations in this regard. First, the value \( 2 \times 10^{20} \text{ cm}^{-3} \) corresponds to the point where the superlattice peak is observed to completely disappear in UED measurements [18], suggesting that \( \tau \) indeed peaks at the threshold excitation density. Second, the time for the initial fast relaxation in transient reflectivity displays a steeply-increasing trend at \( F_{\text{melt}} \) (Fig. 2(c)). This is attributed to a vanishing energy gap at the Fermi level when the CDW is completely suppressed, which limits the decay rate of excited quasiparticles [23–25]. Third, the maximum reflectivity change, \( I_{\text{peak},1} \), also displays distinct behavior below and above \( F_{\text{melt}} \) (Fig. 2(c)). Below \( F_{\text{melt}} \), the presence of a CDW gap modifies the transient population of excited quasiparticles, resulting in a super-linear \( I_{\text{peak},1} \) as a function of excitation density (Fig. 2(c), blue curve; see [31]). Beyond \( F_{\text{melt}} \), the excited quasiparticle population is directly proportional to the excitation density (Fig. 2(c), black line). Above an even higher value \( F_{\text{bleach}} \), the peak reflectivity \( I_{\text{peak},1} \) plateaus (Fig. 2(c), vertical dashed line) due to quasiparticle bleaching [31]. It is worth emphasizing that at \( F_{\text{melt}} \), the lattice temperature stays below \( T_c \) at all time delays after photoexcitation [18], reaffirming that the observed CDW melting is nonthermal in nature and not a result of transient lattice heating above the transition temperature.

To interpret the non-monotonic trend of the initial response time \( \tau \) measured in TOS, we need to understand what physical quantity is probed by transient reflectivity. Unlike the superlattice peak intensity in diffraction measurements or in-gap spectral weight in trARPES, optical reflectivity is not a direct gauge of the CDW order parameter. Typically, in a gapped system, the value of transient reflectivity is taken to be proportional to the excited quasiparticle density [23–25], which in turn is sensitive to the gap size. For example, clear oscillations are present in \( \Delta R \) traces (Figs. 1(d), 2(a), and 2(c)), with a dominant contribution from the CDW amplitude mode [25] – the modulation of the gap magnitude. Based on this sensitivity of \( \Delta R \) to the gap size as well as the consistency of the initial timescale in Figs. 1(d) and 2(d) across techniques, we take the initial rise time \( \tau \) in transient reflectivity as the time needed for the amplitude of the CDW order parameter to be maximally suppressed. The value of \( \tau \) is well separated from any electron-electron scattering timescale \( \lesssim 100 \text{ fs} \) [23, 34], and represents a simultaneous population of excited quasiparticles and renormalization of the gap, which occur self-consistently.

Having established the precise meaning of \( \tau \), we draw...
some parallels between the present nonequilibrium study and its equilibrium counterparts to interpret the observation in Fig. 2(d). At equilibrium, when the temperature is close to $T_c$, time-domain measurements of the order parameter indicate a reduced rate of change, which signifies critical slowing down [3, 10, 11]. Here, we use photoexcitation density in lieu of temperature, and we extend the timescale to the femtosecond regime. Similarly, we interpret the maximum value of $\tau$ at exactly the threshold excitation density as a signature of dynamical slowing down in this ultrafast phase transition.

To understand how a slowdown in dynamics can be extended to a regime far from equilibrium, we again make reference to the established framework of symmetry-breaking transition in equilibrium, which is parameterized by an order parameter $\psi$. On a phenomenological level, we consider the standard Landau potential, $W(\psi) = -\alpha |\psi|^2 + \frac{\beta}{4} |\psi|^4$, which gives the simplest description of the second-order CDW transition in LaTe$_3$ [26]; here, $\alpha$, $\beta$ are the usual Landau coefficients. To see the slowdown near $T_c$, the typical treatment is to solve the time-dependent Landau equation [2], \[\frac{\partial \psi}{\partial t} = -\Gamma \frac{\delta W}{\delta \psi} + \eta(t),\]

where $\Gamma$ is a phenomenological parameter and $\eta(t)$ is the Langevin noise term representing fluctuations. Close to $T_c$ where the order parameter $\psi$ is small and the free energy $W(\psi)$ develops a flat bottom, the relaxation time of long-wavelength fluctuations in $\psi$ can be shown to approach infinity, which is the origin of critical slowing down [2].

We now generalize this treatment to a highly nonequilibrium regime. To account for the different responses by the electronic and phononic subsystems, we consider two components, one for the electrons ($\psi_e$) and the other for the lattice ($\psi_l$). The Landau free-energy functional is [2, 31, 35–37]:

\[W = -\alpha_e |\psi_e|^2 + \frac{\beta}{2} |\psi_e|^4 - \zeta (\psi_e \psi_e^* + \psi_l \psi_l^*) + \alpha_l |\psi_l|^2. \quad (1)\]

Here, $\alpha_e$, $\alpha_l$, and $\beta$ are model parameters; the $\zeta(\cdot)$ term represents electron-phonon coupling. The equations of motion are given by:

\[
\frac{d\psi_e}{dt} = -\frac{\delta W}{\delta \psi_e} \quad \text{and} \quad \frac{d^2\psi_l}{dt^2} = -\frac{\delta W}{\delta \psi_l}, \quad (2)
\]

where the electronic dynamics is similar to that in the equilibrium treatment and heavy lattice ions are assumed to behave like classical oscillators. Detailed discussion of the model and the numerical solution to Eq. (2) are described in Ref. [31]; here, we only highlight the physical picture summarized in Fig. 3(a). Unlike the equilibrium case where microscopic fluctuations, captured by $\eta(t)$, initiate the dynamics, the temporal evolution in the nonequilibrium case is driven by a femtosecond laser pulse, where a coherently excited CDW amplitude mode plays an instrumental role [32]. The pulse modifies the free-energy functional ($\alpha_e$ in Eq. (1)) and sets off the order parameter to seek a new global minimum. Though we draw the free energy as fixed curves, it should be noted that it evolves dynamically according to Eq. (1). As the dynamics of $\psi_e$ and $\psi_l$ closely follow each other [31], we use a single circle to denote the order parameter in Fig. 3(a). At the critical excitation density, $F_{\text{melt}}$, beyond which the order parameter vanishes transiently, the time taken to suppress the order is the longest ($\tau_2 \sim \tau_1 \sim \tau_3$ in Fig. 3(a)). Similar to the equilibrium situation, the slow evolution reflects a transiently flat potential landscape when the order parameter is close to zero, which leads to its reduced rate of change.

Using our experimental parameters for the time-dependent Landau equation, the calculated CDW suppression time, $\tau$, is shown in Fig. 3(b). There is no adjustable parameter except a constant that converts a dimensionfull $F$ in the experiment to a dimensionless quantity in the computation. Here, $\tau$ is defined as the time spanned between the arrival of the laser pulse and the transient minimum position of order parameter amplitude. It shows a distinct peak at the critical point, $F_{\text{melt}}$, which captures the experimental observation in Fig. 2(d). Notably, the absolute value of the calculated $\tau$ falls under a similar range of magnitudes as observed in the experiment. This timescale is determined by the period of the CDW amplitude mode in the simulation [31], indicating the instrumental role of phonons in mediating the ultrafast transition.

There is one key difference between the calculated and measured trend of $\tau$: the latter lacks a sharp divergence.
at the threshold excitation density. We attribute this rounding of the divergence to the presence of temporally or spatially varying perturbations on the system, such as photo-induced topological defects [16–18] or additional phonons coupled to the CDW order [25, 29, 38], which are not considered in our minimal model. In the Landau picture, they disrupt the flat potential energy landscape when the order parameter approaches zero, which is required for the diverging behavior. Furthermore, the divergence only happens in a very narrow window of excitation densities (Fig. 3(b)), which makes experimental detection challenging as any small uncertainties or fluctuations in the pulse energy can smear the singularity.

In conclusion, two different time-resolved probes are used to systematically study the ultrafast melting of a CDW instigated by an intense laser pulse. We have experimentally demonstrated the phenomenon of dynamical slowing down, manifested as the longest time it takes to suppress the CDW at the threshold excitation density in the nonequilibrium phase transition. The agreement in timescale across techniques and with theoretical simulation by time-dependent Landau equations highlights the important role of phonons in this photo-induced transition. Despite complexities involved in phase transitions far from equilibrium, the observation of slowing dynamics in this setting pinpoints a robust commonality for us to understand nonequilibrium phenomena of more intricate systems.

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* Present address: School of Electronics and Information Technology, Sun Yat-sen University, Guangzhou, Guangdong 510006, China.
† Correspondence to: gedik@mit.edu
‡ Present address: Center for Free-Electron Laser Science, DESY, Notkestraße 85, 22607 Hamburg, Germany.

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