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Peer Review File

Electron-phonon-dominated charge-density-wave fluctuations in TiSe2 accessed by ultrafast nonequilibrium dynamics

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The manuscript entitled "Electron-phonon dominated charge density wave fluctuations in TiSe2 accessed by ultrafast nonequilibrium dynamics" by Fragkos et al. investigates the microscopic origin of charge density wave (CDW) fluctuations in 1T-TiSe2 at room temperature using time-resolved extreme ultraviolet momentum microscopy in combination with density functional theory. The origin of the CDW phase in TiSe2 remains a central and controversial problem in condensed matter and the authors' work is timely. The authors demonstrate that the CDW fluctuations persist well above a transition temperature, which is known from the prior literature. What is surprising, however, is the observation of coherent phonon dynamics in this temperature regime. From this observation, the authors argue that the fluctuations are dominated by electron-phonon coupling rather than excitonic correlations. The authors' experimental investigations and high data quality are to be complimented. However, there are still some issues with the interpretation of the experiments. If the authors can address the questions I raise below adequately, I would support publication in Communications Physics.

Major issues:
1) The authors make clear that electron-phonon coupling is important in TiSe2. This is not really a disputed point in the literature. The main question is the extent to which excitonic contributions contribute, which is a little more difficult to say. Since all measurements were conducted at room temperature in this study, could it be that CDW fluctuations above Tc are dominated by electron-phonon coupling but excitonic correlations play a larger role near Tc? The referenced paper by Kogar et al would seem to suggest that the "electronic soft mode" only comes into play at lower temperatures.

2) Given the 171 fs pump-probe cross correlation and the use of Fermi-Dirac fits to extract the electronic temperature, how reliable are the reported electronic temperatures at the sub-100fs time delays? Also, not being in expert in ARPES, it would be helpful for the authors to spell out a little more how the extraction of the electronic temperature was carried out in the supplementary materials.

3) It seems like Fig.4(e) would imply that near room temperature (1.45*T_CDW = 290K) there should no longer be a bimodal distribution of intensity in the line cut of the Se 4p replica. However, the data in Fig. 2(d) and 2(e) does show a bimodal distribution. What is the reason for this inconsistency?

4) The plots of the electronic temperature in Fig. 3(e) show a positive slope before the arrival of the pump pulse. Why are these curves not flat lines before t0?

5) The authors suggest that if the CDW fluctuations were excitonic, their suppression should coincide with the peak of the electronic temperature. I do not understand this point. Wouldn't the excitonic condensate, if present, be destroyed immediately upon excitation, before electronic thermalization, while the ionic CDW component would decay on the slower phonon timescale? Does this undermine their argument that the delay relative to the electronic temperature rules on excitonic order?

6) Unless I missed it, the authors do not present the spot size of the probe. It is important to compare the probe spot size to the fluctuation length scale at room temperature, which is presented in the reference by Guo et al. Is the observation of the

coherent phonon only possible with the fluctuation length scale is greater than the spot size? Is the fluctuation length scale even relevant? We cannot yet answer this question without the appropriate information.

Reviewer #2

(Remarks to the Author)
This manuscript employs time-resolved extreme ultraviolet momentum microscopy and DFT calculations to investigate the time-dependent evolution of the Se 4p orbital backfolded bands in 1T-TiSe2, aiming to explain the origin of CDW fluctuations. The experimental data are clear, showing distinct changes in the CDW backfolded bands before and after photoexcitation, providing solid evidence for the observation of fluctuations. The subsequent recovery process associated with phonons is also clearly demonstrated. The theoretical approach, utilizing Fan-Migdal electron self-energy corrections to address the CDW problem, is novel and insightful. The disappearance of the backfolded band with increasing temperature effectively illustrates the CDW phase transition.

However, I have several questions regarding the connection between the theoretical and experimental results that require clarification:

1. The electronic temperature in Fig.3(e) is derived from the electron occupation numbers, bypassing the processes of electron thermalization and part of the electron-phonon coupling, which typically occur around 100 fs. Consequently, the " delay" might not fully correspond to the coherent suppression of CDW-fluctuation melting as proposed.

2. In the theoretical simulations, only the temperature is varied while the crystal structure remains fixed. This makes it difficult to directly compare the results with the UED experimental findings reported in Reference 21, where structural changes are likely involved.

3. From an experimental perspective, the melting of the backfolded band appears to be more directly linked to electronic excitation rather than electron-phonon coupling. This is suggested by the coincidence of the occupation number curve and the backfolded band intensity curve, indicating a stronger correlation between them, which seems to differ somewhat from the theoretical interpretation provided.

4. The authors attribute the behavior of the experimental backfolded band largely to phonons, noting that its periodic oscillations match the frequency of the A1g phonon mode. This suggests that atomic displacement is a direct factor influencing the backfolded band. To strengthen this argument, it would be helpful to compare the band structure of the CDW phase directly. Such a comparison might be more persuasive for illustrating the role of atomic displacement than the Fan-Migdal electron self-energy results alone.

Reviewer #3

(Remarks to the Author)
Fragkos and coworkers present a study of the charge-density-wave (CDW) material 1T-TiSe2 using time-resolved ARPES and DFT simulations. This material continues to be of interest to the community because of the debate surrounding its CDW phase, and particularly about the role of exciton correlations, inspired by the excitonic insulator scenario. This scenario has however lost favour in recent years, and instead there is a growing consensus about the dominant role of lattice (electron-phonon) interactions, which is similar to the conclusions of the present work.

One of the most influential papers on the topic reported “CDW fluctuations” in TiSe2 which persist to room temperature, i.e. above the expected transition at 200 K, as evidence for excitonic correlations, with signatures in ARPES spectra [Ref. 6]. Here, Fragkos and coworkers also observe similar signatures at room temperature (295 K) and then focus on the femtosecond dynamics of these so-called room temperature CDW fluctuations (RT-CDW).

It should be noted that room temperature fluctuations have been investigated already using similar techniques [Ref. 17], and in general there are numerous tr-ARPES studies of this material. Regardless, the authors possibly present some new interesting findings. Firstly, they report that the melting dynamics of the RT-CDW is delayed with respect to the hot electron population, and that it survives even under relatively intense perturbations up to ~1 mJ cm-2 fluence, which is generally thought to be too high for any purely excitonic phase to withstand. This forms their main arguments in favour of electron-phonon interactions (and against excitons). Secondly, they also observe coherent oscillations of the 3.5 THz CDW mode, despite being in the room temperature phase, again as evidence for fluctuations above TCDW, but previously not observed as far as I am aware. This is perhaps the strongest experimental result of the paper in my opinion. I do not see the benefits of “momentum microscopy” in this work, as the presented data analysis could also be obtained using traditional XUV tr-ARPES – although the authors do not oversell this aspect.

The paper is reasonably well written and presented, and it has the potential for publication in Communication Physics. It’s main conclusions challenge some previous beliefs [Ref. 6] and is generally more aligned with the growing consensus in the literature. However, I include some comments that I would like the authors to address before recommending the work.

1. The authors observe a ~140 fs offset between the perturbation of the backfolded band intensity and the onset of the elevated electronic temperature which is used to argue in favour of electron-phonon interactions because it matches the half period of the AM. Such an observation was made recently in related compound 1T-TaSe2 [PRL 130, 156401 (2023)] by tr-ARPES, and is generally considered a signature of the electron-phonon (lattice) interaction time. While the authors are

probably correct, the relatively short timescale of the higher frequency 3.5 THz AM (280fs/2 = 140fs) makes this claim more challenging – and so they should be extra careful to show that the analysis is robust.

Currently in Fig. 3(e), it is difficult to assess whether there is really a temporal offset, as this depends on the normalization method of the three curves. It seems that the "0" y-axis scale of the blue curve is set at t = 0, while the "0" y-axis of the orange and yellow curves is set to their final value at t = 3 ps. What is the reason for this choice?

What is the origin of the increasing electronic temperature (orange) and excited state population (yellow) before time zero? In addition, it seems that both the temperature and the population is greater at -0.5 ps than at 3 ps. How can this be?

2. Another argument for the electron-phonon mediated fluctuations is based on its robustness to photoexcitation up to ~1 mJ cm-2. However, a better gauge of the strength of photoexcitation is the transient electronic temperature: Here, we see a maximum of only ~800 K (from 300 K) which is rather modest, as several thousands of Kelvin are often seen for mJ cm-2 fluences in the literature. In fact, it is comparable to (or slightly less than) the electronic temperature (~1000 K) reported in Ref. 17 for only half the pump fluence ~0.5 mJ cm-2 at 1300 nm. Therefore, I wonder if the authors are overestimating their pump fluence.

Aside from this fact, a much more useful test would be a fluence dependence to understand the critical threshold for supressing the fluctuations. With only 1 data point, it is difficult to argue conclusively about its robustness.

3. Minor comment about Lines 357-377: I don't understand the relevance of the comparison with Ref. 58 in this context. What are the authors trying to say in this section? Some clarification is needed.

4. The temperature dependent (Fermi-Dirac smearing) DFT simulations including electron-phonon interactions are interesting. However this section needs significant clarification in my opinion.

Presumably T* is the temperature for suppression of the RT-CDW, but it is not defined anywhere in the main text.

In Fig 4 there is a typo in the caption: "but close the temperature T_*".

In Fig. 4(b) and (d): add temperature labels.

In addition, the FD effective temperatures are not written explicitly anywhere. We have some clues only by inspecting Fig. 4(e) where we see "1.01TCDW" and "1.45TCDW" – this should be written in the caption and main text in my opinion. The labels in Fig. 4(e) are too small. Related to this, what temperature do the authors believe the RT-CDW to be melted from the simulations? It's not written anywhere. In panel (e), the 4th curve seems to have some shoulders of the folded VB, while fluctuations are only absent in the 5th curve i.e. at 1.45TCDW (around 290 K) – this suggests the predicted CDW is significantly weaker than the experiment shows (persists even at Tel = 800 K)… However, after reading the supplementary, we discover that the scale is actually set by the simulated TCDW within the Harmonic approximation (TCDW = 1100 K) and not the experimental one TCDW = 200K. This should be explicitly stated near the beginning of the section when discussing Figure 4 to avoid confusion. The method for determining TCDW (harmonic) should also be described – for example does it come from (negative) phonon instabilities? Supplementary Figure 2 panels (b) and (e) might be useful to show in the main text.

If Fig. 4(f) what are the shaded grey areas? The temperature label is too small.

In caption Fig. 4(f) it says "The right panel shows the phonon energy at the M point" but this the intensity profile of the phonon energy.

5. Finally, the authors include data measured by STM in the Supplementary. They do not find evidence for the fluctuations (RT-CDW) and conclude "…that the fluctuations probably occur at timescales below the scanning rate of the STM" and that this "supports our result that the CDW fluctuations in TiSe2 are dynamical."

What do they mean by "dynamical" here? In this context, it sounds like it is transient or somehow visible only in the time-domain as a metastable state induced by the photoexcitation in tr-ARPES. Yet, we know that the RT-CDW exists in equilibrium (Ref. 6 and this work Fig. 2(a)). This terminology appears throughout the paper, particularly related to the DFT simulations. Some further clarification would be helpful.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors have addressed my concerns and I now recommend publication.

Reviewer #2

(Remarks to the Author)
The authors have improved their manuscript based on my first-round suggestions. I recommend its publication.

Reviewer #3

(Remarks to the Author)
I have now seen the revised manuscript and reply to reviewers by Fragkos and coworkers. The authors have clearly made great efforts to address the main points of the reviewers and I believe that the manuscript has improved. Most of the answers are satisfactory.

The explanation for the nonequilibrium (and rising) electronic temperature at negative time delays is rather worrying. The effect is not insignificant as the authors claim because the increasing electronic temperature is directly visible in Fig. 3(e), i.e. the pre-pulse drives a sizeable portion of the total variation on the scale 300 to 800 K. I am not aware of other tr-ARPES setups with such issues. It means that at t = 0, we are not observing the true effect of the pump-induced change of the system from its equilibrium state, but from some initially excited state in which there is a small transient electronic population generated by the pre-pulse up to -1 ps. This may have implications for the interpretation of the results in the specific case of TiSe2 because of the possibility of the excitonic component to the CDW, which would be sensitive to weak electronic perturbations. However, we can expect that this will not have an effect on the RT-CDW fluctuations which are seemingly robust, and hopefully it does not effect the main conclusion of the work. The authors now report the issue on page 4. They may also consider to show the cross-correlation (Fig R2) in the supplementary for full transparency.

Despite these issues, I believe that the work deserves to be published. The observation of the 3.5 THz mode in the RT dynamics is an important result and will trigger further investigation in the community. I now recommend publication in Communications Physics.

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Reply to reviewers

November 7, 2025

In this rebuttal, we address all the comments and remarks of the reviewers. The comments of reviewers are in **blue**, our answers are in **black**, and new text added to the manuscript is in **red**.

Disclaimer: We have copied reviewers' comments and reformatted them to be compatible with LaTeX. Nothing else has been modified.

Reviewer 1:

The manuscript entitled "Electron-phonon dominated charge density wave fluctuations in TiSe2 accessed by ultrafast nonequilibrium dynamics" by Fragkos et al. investigates the microscopic origin of charge density wave (CDW) fluctuations in 1T-TiSe2 at room temperature using time-resolved extreme ultraviolet momentum microscopy in combination with density functional theory. The origin of the CDW phase in TiSe2 remains a central and controversial problem in condensed matter and the authors' work is timely. The authors demonstrate that the CDW fluctuations persist well above a transition temperature, which is known from the prior literature. What is surprising, however, is the observation of coherent phonon dynamics in this temperature regime. From this observation, the authors argue that the fluctuations are dominated by electron-phonon coupling rather than excitonic correlations. The authors' experimental investigations and high data quality are to be complimented. However, there are still some issues with the interpretation of the experiments. If the authors can address the questions I raise below adequately, I would support publication in Communications Physics.

Firstly, we thank Reviewer 1 for their positive feedback on our work and for recommending our paper for publication in Communications Physics once the questions are adequately addressed.

Major issues: 1) The authors make clear that electron-phonon coupling is important in TiSe2. This is not really a disputed point in the literature. The main question is the extent to which excitonic contributions contribute, which is a little more difficult to say. Since all measurements were conducted at room temperature in this study, could it be that CDW fluctuations above Tc are dominated by electron-phonon coupling but excitonic correlations play a larger role near Tc? The referenced paper by Kogar et al would seem to suggest that the "electronic soft mode" only comes into play at lower temperatures.

We thank Reviewer 1 for this interesting point. It is indeed true that our work studies the CDW fluctuations at room temperature and, therefore, the obtained conclusions and evidence on the origin apply only to the phase diagram above T_{CDW}, i.e., in the fluctuating phase. In fact, in our present paper, we do not provide any conclusive arguments on the microscopic mechanisms below T_{CDW}. We only suggest that the present conclusions and a set of

arguments on the origin of the CDW fluctuations might have an impact on understanding the CDW phase. Namely, previous experiments observed the creation of coherent phonons also in the CDW phase, with stronger intensity of the oscillations compared to what we found here for the CDW fluctuating phase. Having that in mind, we view the CDW fluctuating phase as a precursor to the CDW phase, with the same creation mechanism.

As for the soft electronic mode observed in Kogar et al. 2017 [R1], recent EELS experiment [R2] convincingly argues that this soft mode at the edge of the Brillouin zone is actually the CDW-related phonon, while plasmons are strongly damped away from the BZ center. Recent theoretical studies done with time-dependent density functional theory (TDDFT) [R3] and ab-initio Bethe-Salpeter equations (BSE) [R4] have reached a similar conclusion, i.e., that there is no soft excitonic mode above T_{CDW}. This is a prerequisite to have an excitonic insulator phase. In fact, there is only a low-energy excitonic mode below T_{CDW} when the lattice is distorted by electron-phonon coupling [R4]. On the other hand, there is much evidence on the existence of the CDW-related phonon below, above, and around T_{CDW}.

Changes in the manuscript: We have slightly soften our claims regarding the mechanism of the low-temperature CDW phase in the abstract and conclusions. For instance, in the abstract we modified the last sentence to read: could be useful in understanding the nature of the CDW phase transition in 1T-$TiSe_2$ and similar quantum materials..

2) Given the 171 fs pump-probe cross correlation and the use of Fermi-Dirac fits to extract the electronic temperature, how reliable are the reported electronic temperatures at the sub-100fs time delays?

Indeed, the finite temporal resolution in our experiments imposes a fundamental limit on our ability to observe processes occurring on timescales much shorter than this resolution. Nevertheless, it remains possible to reliably extract the delay between two processes that occur on a relatively shorter timescale than the cross-correlation time, provided the signal-to-noise ratio is sufficiently high. While this concept is well understood within the time-resolved spectroscopy community, we aim to illustrate it here in a simple, intuitive, and pedagogical manner.

To investigate this, we computed two Gaussian error functions separated by a delay of 30 fs, with variable full-width at half-maximum (FWHM) rise times of 170 fs and 300 fs, and with different levels of random amplitude noise (5%, 20%, and 60%). These functions can be conceptually compared to two light-induced processes, e.g., excited-state population and electronic temperature, assuming no relaxation or dissipation following photoexcitation in this simplified model. Our goal is to extract the delay between the initial rise times of these processes. We then applied a fitting procedure to determine the central time position and its associated uncertainty, based on the 95% confidence level, in a manner analogous to the analysis of experimental data. The delay between the two functions (30 fs) was intentionally chosen to be much smaller than the FWHM rise times (170 fs and 300 fs), which in this simple model corresponds to the temporal resolution. As shown in Fig. R1(a), with a 170 fs temporal resolution and with a high signal-to-noise ratio, the 30 fs delay between the two functions can be retrieved with good precision and accuracy. When keeping the same temporal resolution but increasing the amplitude noise (Fig. R1(b)-(c)), the delay can still be determined with satisfactory accuracy, though with reduced precision. Repeating the analysis with a FWHM rise time of 300 fs, i.e., an order of magnitude larger than the delay between the curves, shows that both accuracy and precision deteriorate as temporal resolution and noise increase.

In conclusion, thanks to the high-statistics data enabled by the high-repetition-rate beamline, and the long acquisition times afforded by its high stability, we can reliably extract meaningful relative timing of nonequilibrium processes on the sub-100 fs timescale.

Also, not being in expert in ARPES, it would be helpful for the authors to spell out a little more how the extraction of the electronic temperature was carried out in the supplementary materials.

Extracting the time-dependent electronic temperature from time-resolved angle-resolved photoemission spectroscopy (trARPES) data in metallic and semimetallic systems is a well-established procedure. The process begins with acquiring high-quality energy- and momentum-resolved photoemission intensity as a function of pump-probe delay times, which constitutes the standard trARPES data collection. For each delay, the photoemission intensity is integrated over all momenta to produce time-resolved energy distribution curves (EDCs). These EDCs are then

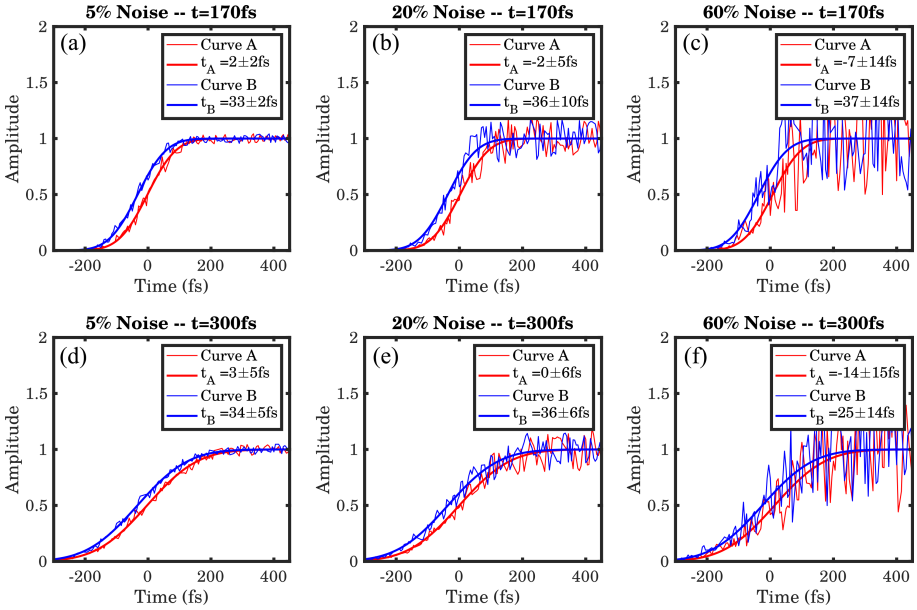

Figure R1: **The effect of amplitude noise and pulse duration on the extraction of small delays between two processes.** We simulated two Gaussian error functions separated by a delay of 30 fs, with variable full-width at half-maximum (FWHM) rise times of 170 fs [**(a)-(c)**] and 300 fs [**(d)-(f)**], and with different levels of random amplitude noise, i.e. 5% [**(a)** and **(d)**], 20% [**(b)** and **(e)**], and 60% [**(c)** and **(f)**]. The fitted central time positions and associated uncertainties, based on the 95% confidence level, are shown in the legend.

analyzed to extract the electronic temperature by fitting them to a Fermi-Dirac distribution convolved with the experimental energy resolution, typically modeled as a Gaussian broadening. Importantly, only the edge of the EDCs, near the Fermi level, is used in the fitting procedure to minimize the influence of strong energy-dependent modulation of the photoemission intensity due to variations in the density of states at higher binding energies. Repeating this fitting for each delay allows one to construct a time-dependent profile of the electronic temperature. Advanced analyses can also include modeling of non-thermal distributions shortly after excitation, as electrons may initially deviate from a Fermi-Dirac distribution before fully thermalizing. However, given the finite time resolution of our measurements and the scope of this study, we did not pursue the detection of subtle non-thermal effects immediately following excitation.

3) It seems like Fig.4(e) would imply that near room temperature ($1.45\times T_{CDW} = 290$K) there should no longer be a bimodal distribution of intensity in the line cut of the Se 4p replica. However, the data in Fig. 2(d) and 2(e) does show a bimodal distribution. What is the reason for this inconsistency?

This inconsistency is a consequence of the fact that the experimental and theoretical values of T_{CDW} are different. Performing harmonic DFPT calculations, where only static electron-phonon coupling contributions are included with no anharmonic corrections, we get that T_{CDW} is 1105 K. In order to get a result for T_{CDW} that is closer to the experiment, one would need to include anharmonic effects [R5] and probably higher-order electron-phonon coupling [R6], which is numerically demanding. For instance, with the inclusion of anharmonic effects, one gets $T_{\mathrm{CDW}} = 400 - 500$ K. However, note that the inclusion of these effects will not modify the conclusions of the present paper, since phonon-phonon corrections will not impact the electronic structure directly. The impact will be only indirect, where the softening of the CDW phonon will occur at temperatures between $400 - 500$ K, instead of at 1105 K, and therefore the CDW fluctuating phase would be present for $T > 400$ K instead of for $T > 1105$ K.

Since the theoretical and experimental temperature scales differ, it is not so straightforward to determine the actual temperature at which the CDW fluctuations are quenched, i.e., a bimodal distribution of intensity in the line cut of the Se 4p replica diminishes, and $TiSe_2$ enters normal (metallic or semimetallic) phase. From the present harmonic DFPT theory, the transition to the normal phase is around $T^* = 1600$ K, which is roughly 500 K from theoretical T_{CDW} or at $1.5 \times T_{CDW}$. Regarding the actual experimental value of this temperature T^*, we are not aware of any recent discussion on this matter. We are only aware of resistivity measurement done in Ref. [R7], which shows a peak below 200 K, and then a decrease of resistivity up until around 400 K, where the resistivity starts to increase again. A decrease could be a consequence of gapped and semi-gapped states, as in the CDW and CDW fluctuating phase, while an upturn in resistivity at 400 K hints at the formation of the normal metallic or semimetallic state. Therefore, the experimental value for T^* might be around 400 K, which is around 200 K above the T_{CDW} or at $2 \times T_{CDW}$. However, the discussion on the value of T^* goes beyond the scope of the present paper.

Changes in the manuscript: Added two sentences in the section on 'Theoretical Analysis of Dynamical Electron Phonon Interactions':

Temperatures used in the theoretical simulations are scaled with respect to the T_{CDW} value from density functional perturbation theory (DFPT), which is 1105 K.

Note that the bimodal intensity profile in our simulations disappears continuously, while it is completely vanished above T^*=1600 K, which is 500 K above the theoretical T_{CDW}.

4) The plots of the electronic temperature in Fig. 3(e) show a positive slope before the arrival of the pump pulse. Why are these curves not flat lines before t0?

We thank the reviewer for pointing this out. Another reviewer also asked a similar question. This effect can be readily explained by the specific temporal profile of the pump pulse used in our experiment. We are so accustomed to observing a similar behavior in all our pump–probe scans, regardless of the investigated material, that we neglected to explain the origin of the positive slope observed before the arrival of the main pump pulse in the time-dependent electronic temperature curves.

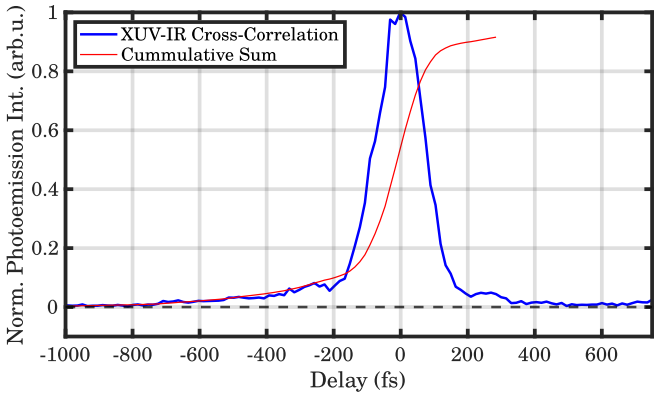

Figure R2: **Cross-correlation between p-polarized IR pump and p-polarized XUV probe** In blue, the normalized photoemission intensity as a function of pump-probe delay, extracted for laser-assisted photoemission (LAPE) valence band replica in GeS semiconductor.

Our laser system is based on a Yb-fiber amplifier optimized to deliver relatively short pulses (sub-150 fs). However, residual high-order spectral phase distortions lead to the presence of a weak pre-pulse extending up to nearly one picosecond before the main pulse. This is clearly illustrated in the cross-correlation between the IR pump and XUV probe pulses, extracted from a non-resonant two-photon photoemission process (here, laser-assisted photoemission

in GeS), as shown in Fig. R2. The blue curve reveals the existence of a pre-pulse extending several hundred femtoseconds before the main pulse, while its cumulative sum (red curve) phenomenologically represents how such a temporal profile would heat the electrons, neglecting thermalization and dissipation effects. The leading edge of this pre-pulse is therefore responsible for the positive slope observed before time zero in the electronic temperature traces. Nevertheless, because the associated temperature increase is very small compared to that induced by the main pulse, it does not significantly affect the physical conclusions discussed in the paper.

Changes in the manuscript: Based on review comments, we have added this sentence to the manuscript: The increase in electronic temperature and excited-state population observed at negative time delays is attributed to a weak prepulse extending up to a few hundred femtoseconds in the temporal profile of the infrared pump pulses.

5) The authors suggest that if the CDW fluctuations were excitonic, their suppression should coincide with the peak of the electronic temperature. I do not understand this point. Wouldn't the excitonic condensate, if present, be destroyed immediately upon excitation, before electronic thermalization, while the ionic CDW component would decay on the slower phonon timescale? Does this undermine their argument that the delay relative to the electronic temperature rules on excitonic order?

Actually, this does not undermine our argument, but it strengthens it. If it exists, the exciton relevant to the excitonic condensation would consist of electrons in Ti-d pocket at the M point, and holes in the Se-p states at the Γ point. So the relevant photo-excited carrier density for the quenching of this mode is the total photo-induced density of the system, which can be described by the effective temperature. Also, one expects that the quenching dynamics of the exciton are fastest. In Fig. 3(e) we show the thermalization of the total density via effective electron temperature (red symbols) and the dynamics of the photo-excited carriers around the M point and above E_F (yellow symbols). If the CDW fluctuations are driven by the exciton, then its quenching dynamics would correspond to the dynamics of hot electrons, but that is not the case. Instead, we provide several convincing arguments that link the dynamics of the CDW fluctuations to the soft CDW-related phonon, and therefore to the electron-phonon coupling.

First of all, as mentioned here, the dynamics of the CDW fluctuation suppression and recovery does not coincide with the fast dynamics of hot electrons. The latter is peaked almost at the pump-probe overlap at $\Delta t = 0$ ps, while the partial quench of the CDW fluctuation is offset by approximately 140 fs, which corresponds to the half period of the CDW amplitude mode. This shows the correlation between the CDW amplitude mode and CDW fluctuations. Also, the CDW fluctuation signals (replica bands) are not fully quenched upon photo-excitation, even though we use relatively high fluences for which one expects the melting of excitons. Next, the partial suppression and recovery times of the CDW fluctuation dynamics are around 100 fs and 700 fs, which exactly corresponds to the time dynamics of CDW-related phonon hardening as observed in ultrafast electron diffraction done under similar conditions. Further, the signal of the CDW fluctuations contain the coherent oscillations, which we attribute to the CDW amplitude mode, and which we consider a very direct evidence of the involvement of the phonons and electron-phonon coupling in the formation of the CDW fluctuations. Finally, all these observations can be explained with the ab initio theory that invokes only the dynamical electron-phonon coupling and soft phonons.

Changes in the manuscript: The following sentence was added on page 5: Namely, exciton, if it exists, would be formed from the holes in Se 4p and electrons in Ti 3d bands, and, therefore, its quenching should be correlated with the total photo-excited carrier density, which can be described with the effective temperature. Furthermore, it is expected that the suppression of the excitonic mode is very fast and that it should correspond to the fastest signal, however, the RT-CDW signal suppression is offset from $\Delta t = 0$ by approximately 140 fs.

6) Unless I missed it, the authors do not present the spot size of the probe. It is important to compare the probe spot size to the fluctuation length scale at room temperature, which is presented in the reference by Guo et al. Is the observation of the coherent phonon only possible with the fluctuation length scale is greater than the spot size? Is the fluctuation length scale even relevant? We cannot yet answer this question without the appropriate information.

We thank the reviewer for this very interesting question. Our probe spot footprint on the sample is roughly $33\mu m$ by $45\mu m$). The in-plane lattice parameter (unit cell dimensions) in real space is approximately 3.54 Å. This means that our probe photon sees more than 90,000 unit cells. In Guo et al., the authors reported that characteristic length

scales between CDW domain walls are about 22 unit cells perpendicular to the in-plane wave vector of the CDW component, 7 unit cells parallel to the in-plane wave vector, and 2 unit cells in the out-of-plane direction. This means that our probe spot size is 3-4 orders of magnitude larger than the CDW fluctuation length scale at room temperature.

We have added the beam size in the paper: The time-resolved momentum microscopy setup featuring a polarization-tunable infrared (1.2 eV, sub-150μm) pump and XUV probe (21.6 eV, sub-50μm) pulses

More details about the beam size characterization are given in the references cited in the paper.

Reviewer 2:

This manuscript employs time-resolved extreme ultraviolet momentum microscopy and DFT calculations to investigate the time-dependent evolution of the Se 4p orbital backfolded bands in 1T-TiSe2, aiming to explain the origin of CDW fluctuations. The experimental data are clear, showing distinct changes in the CDW backfolded bands before and after photoexcitation, providing solid evidence for the observation of fluctuations. The subsequent recovery process associated with phonons is also clearly demonstrated. The theoretical approach, utilizing Fan-Migdal electron self-energy corrections to address the CDW problem, is novel and insightful. The disappearance of the backfolded band with increasing temperature effectively illustrates the CDW phase transition.

We thank Reviewer 2 for positive and constructive comments and for recognizing our work as novel and interesting.

However, I have several questions regarding the connection between the theoretical and experimental results that require clarification:

1. The electronic temperature in Fig.3(e) is derived from the electron occupation numbers, bypassing the processes of electron thermalization and part of the electron-phonon coupling, which typically occur around 100 fs. Consequently, the " delay" might not fully correspond to the coherent suppression of CDW-fluctuation melting as proposed.

We thank the referee for this important point regarding the derivation of electronic temperature from the occupation numbers. We agree that before full thermalization (tens of femtoseconds), the system might not be described precisely by the Fermi-Dirac distribution and finds itself in a quasi-thermalized state, characterized by an effective temperature. Nevertheless, since the system progressively approaches the Fermi-Dirac distribution after the excitation, and considering that this effective temperature reaches its maximum well before the maximum suppression of the backfolded band (near coincidence with the peak in the excited-state population above the Fermi level). Both of which are delayed relative to the peak in electronic temperature by approximately 140 fs, which, as mentioned, nearly perfectly corresponds to half the period of the amplitude mode oscillation.

Changes in the manuscript:

2. In the theoretical simulations, only the temperature is varied while the crystal structure remains fixed. This makes it difficult to directly compare the results with the UED experimental findings reported in Reference 21, where structural changes are likely involved.

It is true that in our DFT and DFPT calculations we only account for the temperature variations, while the crystal structure is kept fixed, i.e., we do not apply periodic lattice distortions (PLDs). In fact, for temperatures $T \geq T_{\mathrm{CDW}}$, relevant for the present study, the adiabatic DFT and DFPT are not able to capture the presence of the PLDs in 2×2 cell of $TiSe_2$. Above T_{CDW} the ground state structure of $TiSe_2$ in DFT is 1×1 hexagonal cell without PLDs, while below T_{CDW} one gets the PLDs. As such, the DFT and DFPT alone are not able to capture the CDW fluctuating phase above T_{CDW}. By performing the dynamical electron-phonon calculations and by exploring its impact on the electronic structure (via Fan-Migdal self-energy), we found out that the CDW fluctuating phase (along with the replicated bands and pseudogaps) can be captured for $T \geq T_{\mathrm{CDW}}$ when the M phonon mode is soft but positive. As we discuss in the main text, these results on the correction to the electronic structure then suggests that the fluctuating PLDs are also present above T_{CDW}. This could be in principle accounted for by implementing these spectral functions containing dynamical electron-phonon corrections into the dynamical phonon self-energy, which could be then used in the dynamical matrix calculations for the phonon spectra. This self-consistent treatment would then lead to different ground state and would be able to account for finite PLDs beyond the adiabatic value of T_{CDW}. In other words, this treatment would provide finite PLDs up until $T = T^*$. But this is a very demanding task and goes beyond the scope of the present paper.

Regarding Ref. [21], what they report in their UED measurements are the time dynamics of the M soft phonon signal in the diffraction. By using an appropriate equation they can link these signal modifications to the phonon population and frequency modification. Finally, using this analysis they've been able to extract the frequency shift

(hardening) of the M phonon mode as a function of pump-probe delay time. And in our calculations we account for softening and hardening of the M phonon mode in the undistorted 1×1 unit cell.

Nevertheless, since we agree that the fluctuating PLDs are present also for $T > T_{\mathrm{CDW}}$, we have also performed the calculations that include the distortions of the atomic structure along the CDW phonon eigenvectors as in the CDW phase below T_{CDW}. We have placed this figure in SI as a new Supplementary Figure 4. Here we also reproduce this figure for the sake of clarity. In Figs. R3(a) and R3(b) we show the results for undistorted 1×1 cell with Fan-Migdal self-energy corrections included in the presence of the M soft phonon. The rest of the figures are done in 2×2 unit cell with various values of the PLDs, and then unfolded to the 1×1 Brillouin zone. It is clear that both FM self-energy corrections and PLDs induce the same modifications to the electronic structure. We believe that both of these effects are connected and both should be included in our calculations, but DFT is not able to capture PLDs above T_{CDW}, while the self-consistent treatment described above is too demanding to perform at the present moment.

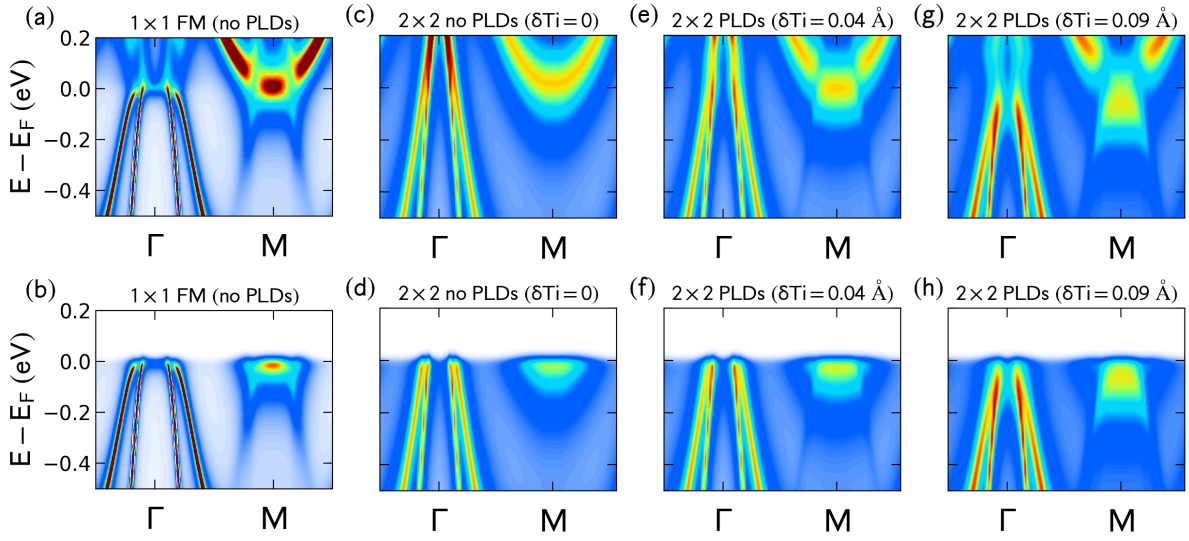

Figure R3: (a)-(b) Electron spectral function of 1T-$TiSe_2$ with dynamical electron-phonon interaction included via Fan-Migdal self-energy (FM) for temperature just above the CDW transition temperature $T \gtrsim T_{\mathrm{CDW}}$. These calculations are done for 1×1 unit cell with no periodic lattice distortions (PLDs). (c)-(h) The band structures of the 2×2 CDW phase with various values of PLDs mapped to the 1×1 unit cell. In these calculations we do not include the FM self-energy. The lower panels are equivalent to the plots in the upper panels, but weighted with the Fermi-Dirac distribution function. The comparison between the results of 1×1 band structure with FM self-energy and 2×2 result with PLDs shows that the dynamical electron-phonon scattering introduces very similar modifications to the band structure as the symmetry-breaking CDW PLDs.

Changes in the manuscript:

We have placed the new Supplementary Figure 4 in SI, and corresponding short discussion on page 7 of the main text:

In Supplementary Figure 4 we demonstrate how the dynamical electron-phonon scattering introduces very similar modification to the electronic structure as the symmetry-breaking periodic lattice distortions in the CDW state.

3. From an experimental perspective, the melting of the backfolded band appears to be more directly linked to electronic excitation rather than electron-phonon coupling. This is suggested by the coincidence of the occupation number curve and the backfolded band intensity curve, indicating a stronger correlation between them, which seems to differ somewhat from the theoretical interpretation provided.

Despite the slight discrepancy from the electronic and effective temperature, as discussed in point 1, the nearly perfect time delay corresponding to a half period of phonon oscillation also experimentally hints at the employment of the phonon-mediated process. Both coherent and incoherent mechanisms likely contribute to suppression; however, the characteristic time delay—corresponding to half an amplitude mode cycle—suggests that coherent suppression plays a dominant role. In addition, the integrated excited-state population appears to evolve similarly to the suppression of the backfolded band, this correlation should be interpreted cautiously. The occupation number curve, which sums over a broad energy range, hides any energy-dependent dynamics within the excited states. In contrast, the backfolded band is localized at a specific binding energy and momentum. Fig. 3(c) then indicates that the suppression dynamics of the backfolded band differ from the excited states closer to the binding energies of the backfolded band. This additionally suggests that the melting of the CDW-related spectral weight involves more than just electronic excitation or carrier redistribution (as discussed in point 1).

Nevertheless, we would also like to stress out that all of the electron-phonon-related mechanisms will be directly impacted by the electron excitations. For instance, photo-excited modifications to the screening will modify the phonon frequency and electron-phonon coupling strengths. Also, both of these quantities will be directly sensitive to the electron/hole occupation numbers, and thus to the electronic excitations. This can be directly observed by examining the phonon self-energy formula.

4. The authors attribute the behavior of the experimental backfolded band largely to phonons, noting that its periodic oscillations match the frequency of the A1g phonon mode. This suggests that atomic displacement is a direct factor influencing the backfolded band. To strengthen this argument, it would be helpful to compare the band structure of the CDW phase directly. Such a comparison might be more persuasive for illustrating the role of atomic displacement than the Fan-Migdal electron self-energy results alone.

Our results indeed indicate that the fluctuating band replica and fluctuating periodic lattice distortions are interconnected. This issue was already addressed in our response to point 2 above. To address this issue in the manuscript, we have included the new Supplementary Figure 4 in SI.

Reviewer 3:

Fragkos and coworkers present a study of the charge-density-wave (CDW) material 1T-TiSe2 using time-resolved ARPES and DFT simulations. This material continues to be of interest to the community because of the debate surrounding its CDW phase, and particularly about the role of exciton correlations, inspired by the excitonic insulator scenario. This scenario has however lost favour in recent years, and instead there is a growing consensus about the dominant role of lattice (electron-phonon) interactions, which is similar to the conclusions of the present work.

One of the most influential papers on the topic reported "CDW fluctuations" in TiSe2 which persist to room temperature, i.e. above the expected transition at 200 K, as evidence for excitonic correlations, with signatures in ARPES spectra [Ref. 6]. Here, Fragkos and coworkers also observe similar signatures at room temperature (295 K) and then focus on the femtosecond dynamics of these so-called room temperature CDW fluctuations (RT-CDW).

It should be noted that room temperature fluctuations have been investigated already using similar techniques [Ref. 17], and in general there are numerous tr-ARPES studies of this material. Regardless, the authors possibly present some new interesting findings. Firstly, they report that the melting dynamics of the RT-CDW is delayed with respect to the hot electron population, and that it survives even under relatively intense perturbations up to 1 mJ cm-2 fluence, which is generally thought to be too high for any purely excitonic phase to withstand. This forms their main arguments in favour of electron-phonon interactions (and against excitons). Secondly, they also observe coherent oscillations of the 3.5 THz CDW mode, despite being in the room temperature phase, again as evidence for fluctuations above TCDW, but previously not observed as far as I am aware. This is perhaps the strongest experimental result of the paper in my opinion. I do not see the benefits of "momentum microscopy" in this work, as the presented data analysis could also be obtained using traditional XUV tr-ARPES – although the authors do not oversell this aspect.

The paper is reasonably well written and presented, and it has the potential for publication in Communication Physics. It's main conclusions challenge some previous beliefs [Ref. 6] and is generally more aligned with the growing consensus in the literature. However, I include some comments that I would like the authors to address before recommending the work.

We are grateful to Reviewer 3 for the positive evaluation of our work and for providing some very useful comments that help improve our manuscript.

1. The authors observe a 140 fs offset between the perturbation of the backfolded band intensity and the onset of the elevated electronic temperature which is used to argue in favour of electron-phonon interactions because it matches the half period of the AM. Such an observation was made recently in related compound 1T-TaSe2 [PRL 130, 156401 (2023)] by tr-ARPES, and is generally considered a signature of the electron-phonon (lattice) interaction time. While the authors are probably correct, the relatively short timescale of the higher frequency 3.5 THz AM (280fs/2 = 140fs) makes this claim more challenging – and so they should be extra careful to show that the analysis is robust.

Currently in Fig. 3(e), it is difficult to assess whether there is really a temporal offset, as this depends on the normalization method of the three curves. It seems that the "0" y-axis scale of the blue curve is set at t = 0, while the "0" y-axis of the orange and yellow curves is set to their final value at t = 3 ps. What is the reason for this choice?

All these quantities, i.e., total excited state population, backfolded band intensity, and electronic temperature, are obviously extracted from the same dataset. Their relative timing is thus inherently accessible in our data. However, the question about how to extract the relative timing from the data, raised by the Review, is an important point. In fact, the "0" y-axis of the orange and yellow curves seems to be 'set' to their final value at t = 3 ps since there is a weak pre-pulse in the pump, as discussed extensively above in an answer to Review 1, and below in the answer to the next question. This weak pre-pulse slightly heats the electrons before the arrival of the main pulse, and as discussed before, its effect is small compared to the main pulse and does not affect our conclusions. Concerning

the statement that "*"0" y-axis scale of the blue curve is set at t = 0*, in fact, the intensity of the backfolded band is very similar before time-zero and at t= 3 ps. The reported temporal offset is thus not affected by such subtle normalization consideration here.

What is the origin of the increasing electronic temperature (orange) and excited state population (yellow) before time zero? In addition, it seems that both the temperature and the population is greater at -0.5 ps than at 3 ps. How can this be?

The same question was raised by Reviewer 1, to whom we already provided a detailed response supported by Fig. R2.

In brief, the pre-time-zero increase in electronic temperature (orange) and excited-state population (yellow) arises from a weak pre-pulse in the pump. This pre-pulse, originating from residual spectral phase distortions in our Yb-fiber amplifier, precedes the main pulse by several hundred femtoseconds and is clearly visible in the IR–XUV cross-correlation (Fig. R2). Although it causes slight pre-heating of the electrons, its effect is negligible compared to the main pulse and does not affect our conclusions. For further details, please refer to our response to Reviewer 1.

2. Another argument for the electron-phonon mediated fluctuations is based on its robustness to photoexcitation up to 1 mJ cm-2. However, a better gauge of the strength of photoexcitation is the transient electronic temperature: Here, we see a maximum of only 800 K (from 300 K) which is rather modest, as several thousands of Kelvin are often seen for mJ cm-2 fluences in the literature. In fact, it is comparable to (or slightly less than) the electronic temperature (1000 K) reported in Ref. 17 for only half the pump fluence 0.5 mJ cm-2 at 1300 nm. Therefore, I wonder if the authors are overestimating their pump fluence.

The Photoemission Electron Microscopy (PEEM) mode of our time-of-flight momentum microscope enables the acquisition of real-space images of photoelectrons emitted from the sample surface. We employed PEEM to determine the spatial footprints of both the pump and probe beams on the sample. Since photoemission induced by the IR pump (1.2 eV) occurs via a multiphoton process, the PEEM image does not directly correspond to the IR beam profile; the effective nonlinearity of the photoemission process must be considered. To quantify this nonlinearity, we measured the total photoemission intensity as a function of the IR pump power. The true IR beam footprint can then be obtained by scaling the measured multiphoton PEEM footprint by the square root of the effective nonlinearity. This procedure provides a highly accurate determination of the IR spot size, and consequently, of the incident fluence on the sample. Therefore, we are confident that our fluence estimates are not overestimated. The differences in maximum electronic temperatures reported in previous studies, i.e., 1000 K at 0.5 mJ/cm2 for 1300 nm (Ref. 17) and 800 K at 0.95 mJ/cm2 for 1030 nm, likely arise from variations in the sample's absorption coefficient at these wavelengths.

Aside from this fact, a much more useful test would be a fluence dependence to understand the critical threshold for supressing the fluctuations. With only 1 data point, it is difficult to argue conclusively about its robustness.

We fully agree with the reviewer that systematic fluence-dependent measurements would be extremely valuable for elucidating the critical threshold required to suppress the fluctuations. In the present study, however, our primary focus is on the first experimental demonstration of the existence of this coherent phonon mode within the fluctuating CDW regime, as well as on its microscopic origin, which we investigate using advanced theoretical modeling. A detailed investigation of fluence dependence, while highly interesting, lies beyond the scope of the current work and is therefore reserved for future studies.

3. Minor comment about Lines 357-377: I don't understand the relevance of the comparison with Ref. 58 in this context. What are the authors trying to say in this section? Some clarification is needed.

We completely agree with the reviewer and have revised the paragraph for clarity.

4. The temperature dependent (Fermi-Dirac smearing) DFT simulations including electron-phonon interactions are interesting. However this section needs significant clarification in my opinion.

Presumably T* is the temperature for suppression of the RT-CDW, but it is not defined anywhere in the main text.

In Fig 4 there is a typo in the caption: “but close the temperature T*”.

We thank the Reviewer for noticing the typo in the caption of Fig.4 that is now fixed. Yes, T^* is the approximate temperature when the CDW fluctuations are suppressed and the system enters into the normal state, i.e., semimetallic phase. We say approximate because this transition is not sharp, but rather continuous. We estimate in our theoretical simulations that above $T^* = 1600\,\mathrm{K}$ the CDW fluctuations are diminished. As discussed in our answer to point 3) of Reviewer 1, we think that the corresponding experimental value is $T^* \approx 400\,\mathrm{K}$. However, this is beyond the scope of the present work.

Changes in the manuscript: Changed the caption of Fig. 4(c-d) to "Same as **(a)-(b)** but close to the temperature T^*, i.e. when CDW fluctuations are diminished." We also added the definition of temperature T^* into the main text.

In Fig. 4(b) and (d): add temperature labels.

Temperature labels are placed in panels (b) and (d).

In addition, the FD effective temperatures are not written explicitly anywhere. We have some clues only by inspecting Fig. 4(e) where we see “1.01TCDW” and “1.45TCDW” – this should be written in the caption and main text in my opinion. The labels in Fig. 4(e) are too small. Related to this, what temperature do the authors believe the RT-CDW to be melted from the simulations? It’s not written anywhere. In panel (e), the 4th curve seems to have some shoulders of the folded VB, while fluctuations are only absent in the 5th curve i.e. at 1.45TCDW (around 290 K) – this suggests the predicted CDW is significantly weaker than the experiment shows (persists even at Tel = 800 K)… However, after reading the supplementary, we discover that the scale is actually set by the simulated TCDW within the Harmonic approximation (TCDW = 1100 K) and not the experimental one TCDW = 200K. This should be explicitly stated near the beginning of the section when discussing Figure 4 to avoid confusion. The method for determining TCDW (harmonic) should also be described – for example does it come from (negative) phonon instabilities? Supplementary Figure 2 panels (b) and (e) might be useful to show in the main text.

We apologize for the confusion. Our calculations are limited by the instability that occurs in the DFPT phonon dispersion at a quite large electronic temperature (1105 K). DFPT uses a harmonic approximation as well as treats electron-phonon coupling as a static perturbation. For this reason, the CDW transition temperature is overestimated and the theoretical and experimental temperature scales are not directly comparable. In the simulations the RT-CDW is melted at 1600 K, which is 500 K above the theoretical T_{CDW}. We have introduced these information in the main text and the caption of Fig. 4.

Regarding a method to determine the harmonic T_{CDW} a previous version of the manuscript already had this information in the Methods section, where it is written "The soft acoustic phonon mode goes to zero at $T_{\mathrm{el}} = 1105\,\mathrm{K}$. In order to get a more accurate transition temperature T_{CDW} one would need to include the anharmonic corrections [82].". In order to make this even more transparent, we have placed this information also in the main text on page 6.

Changes in the manuscript: Added two sentences in the section on ’Theoretical Analysis of Dynamical Electron-Phonon Interactions’:

Temperatures used in the theoretical simulations are scaled with respect to the T_{CDW} value from density functional perturbation theory (DFPT), which is 1105 K. The DFPT T_{CDW} is the electronic temperature at which the acoustic mode at the M point becomes soft in the harmonic and Born-Oppenheimer approximations.

Note that the bimodal intensity profile in our simulations disappears continuously, while it is completely vanished above T^*=1600 K, which is 500 K above the theoretical T_{CDW}.

If Fig. 4(f) what are the shaded grey areas? The temperature label is too small.

In Fig. 4(f) with the grey color scale we show the phonon spectral function. The intensity is the strongest for the

darkest color. However, we understand this is a bit unclear from the figure so we explicitly added the colorscale.

Changes in the manuscript: A colorscale is added in Fig.4(f).

In caption Fig. 4(f) it says "The right panel shows the phonon energy at the M point" but this the intensity profile of the phonon energy.

We thank the reviewer for noticing this. We fixed the mistake in the caption.

Changes in the manuscript: Changed the caption text of Fig.4 (f)to: "The right panel shows the intensity profile of the phonon spectral function at the M point (orange dashed line in the left panel)."

5. Finally, the authors include data measured by STM in the Supplementary. They do not find evidence for the fluctuations (RT-CDW) and conclude "...that the fluctuations probably occur at timescales below the scanning rate of the STM" and that this "supports our result that the CDW fluctuations in TiSe2 are dynamical."

What do they mean by "dynamical" here? In this context, it sounds like it is transient or somehow visible only in the time-domain as a metastable state induced by the photoexcitation in tr-ARPES. Yet, we know that the RT-CDW exists in equilibrium (Ref. 6 and this work Fig. 2(a)). This terminology appears throughout the paper, particularly related to the DFT simulations. Some further clarification would be helpful.

We appreciate the reviewer for highlighting this potentially unclear formulation. By „dynamical," we mean that the fluctuations, due to their nature, vary in space while the system finds itself constantly in the CDW fluctuation phase. Since we do not observe any localized charge density wave (CDW) signature in the measured STM dataset, we want to clarify that the statement merely suggested that the localization of each CDW modulation at a given position is inherently dynamical, occurring somewhere in a range of 10^{-11} to 10^{-2} seconds. We stressed this in the SM.

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Reply to reviewers

December 3, 2025

In this rebuttal, we address all the comments and remarks of the reviewers. The comments of reviewers are in **blue**, our answers are in **black**, and new text added to the manuscript is in **red**.

Disclaimer: We have copied reviewers' comments and reformatted them to be compatible with LaTeX. Nothing else has been modified.

Reviewer 1:

The authors have addressed my concerns and I now recommend publication.

We thank the reviewer 1 for being fully satisfied with our answers and for recommending the manuscript for publication.

Reviewer 2:

The authors have improved their manuscript based on my first-round suggestions. I recommend its publication.

We also thank the reviewer 2 for being fully satisfied with our answers and for recommending the manuscript for publication.

Reviewer 3:

I have now seen the revised manuscript and reply to reviewers by Fragkos and coworkers. The authors have clearly made great efforts to address the main points of the reviewers and I believe that the manuscript has improved. Most of the answers are satisfactory. The explanation for the nonequilibrium (and rising) electronic temperature at negative time delays is rather worrying. The effect is not insignificant as the authors claim because the increasing electronic temperature is directly visible in Fig. 3(e), i.e. the pre-pulse drives a sizeable portion of the total variation on the scale 300 to 800 K. I am not aware of other tr-ARPES setups with such issues. It means that at t = 0, we are not observing the true effect of the pump-induced change of the system from its equilibrium state, but from some initially excited state in which there is a small transient electronic population generated by the pre-pulse up

to -1 ps. This may have implications for the interpretation of the results in the specific case of TiSe2 because of the possibility of the excitonic component to the CDW, which would be sensitive to weak electronic perturbations. However, we can expect that this will not have an effect on the RT-CDW fluctuations which are seemingly robust, and hopefully it does not effect the main conclusion of the work. The authors now report the issue on page 4. They may also consider to show the cross-correlation (Fig R2) in the supplementary for full transparency. Despite these issues, I believe that the work deserves to be published. The observation of the 3.5 THz mode in the RT dynamics is an important result and will trigger further investigation in the community. I now recommend publication in Communications Physics.

We also thank the reviewer 3 for recommending the manuscript for publication. As for the comment about the cross-correlation, as referee states, we have now acknowledged this at page 4 of the manuscript. To be fully transparent, we have now also included the Fig R2 from the rebuttal to the supplementary information.