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# Charge-density-wave in 1T-$TiSe_2$: exciton-phonon separation by femtosecond valence band dynamics

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**Abstract.** We investigate the driving mechanism leading to charge-density-wave transition in 1T-$TiSe_2$ single crystals. Our results show that both exciton instability and phonons cooperate to develop the charge ordered phase below 202 K.

## 1 Introduction

The mechanism behind the charge-density-wave (CDW) phase transition in 1T-$TiSe_2$ is a well-known scientific puzzle in the field of strongly correlated materials. Decades of theoretical and experimental investigations have not yet led to a complete agreement on the observed phenomena. In early studies, purely electronic models were proposed involving electron-hole Coulomb interactions [1] in the context of excitonic insulator instability [2]. On the other hand, some models highlighted the role of the lattice, based on different mechanisms e.g. Fermi Surface (FS) nesting [3] or Jahn-Teller (JT) lattice distortion [4]. In the 2000s, further investigations have shown the importance of excitonic effects on the CDW transition and the electronic-based model were favored within the scientific community [5]. Yet a couple of experiments supported the JT type distortion scenario [6]. More recently, a hybrid model proposed the co-existence of JT and excitonic effects in a single picture [7,8]. Supporting the latter hypothesis, our results show the femtosecond dynamics of the valence band (VB) as a measurable parameter of CDW after an intense optical excitation. We aim at disclosing the underlying physics of the CDW phase formation, but also at separating the weight of each contribution in the whole process.

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## 2 Results and discussion

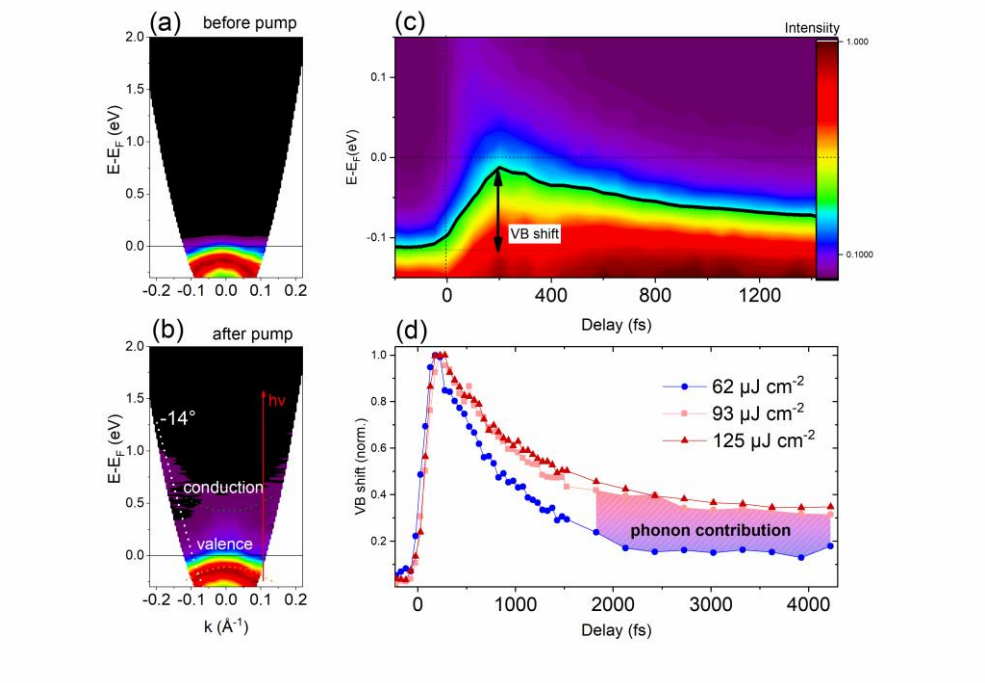


**Fig. 1.** trARPES maps of 1T-$TiSe_2$ before (a) and at 50 fs after (b) photo-excitation. In (b) the red arrow shows the direct optical transition, the conduction band is clearly populated and the -14° angle (white dashed line) represents where the VB dynamics is extracted. (c) Valence band (VB) dynamics as a function of the pump-probe delay. The solid black curve shows the transient variations of VB maximum in the first 1.5 ps, a clear evidence of the photo-induced CDW phase melting. (d) Normalized VB dynamics as a function of the pump-probe delay for different pump fluences from 62 to 125 μJ/$cm^2$.

In the layered single crystal 1T-$TiSe_2$, the CDW phase occurs below 202 K, resulting in a semimetal to semiconductor transition and opening a band gap between the VB at Γ-point and the conduction band (CB) at the M point of the first Brillouin Zone. Another sign of the CDW phase development are the replicas of the valence band appearing at the M points, accompanied by the folding of the CB back into the Γ point [9-11]. A direct measure of both parameters is feasible using angle resolved photoemission spectroscopy (ARPES). By employing the standard pump-probe scheme with ultrashort laser pulses, femtosecond time-resolution can be achieved (trARPES). Here we use pump pulses at 1.85 eV, to melt the CDW, and probe pulses at 6 eV, to photo-emit electrons generating the ARPES images of 1T-TiSe2 at 77 K (see Fig. 1(a) and 1(b)). The temporal resolution of the setup is 70 fs [12]. We focus on the VB at Γ, and use trARPES to study the VB shift and spectral weight dynamics revealing a dissimilar ultrafast behavior (see Fig. 1(c)).

We also measured the transient variations of valence band upon changing the pump fluence in the range of 61 to 125 μJ/$cm^2$ (see Fig. 1(d)). We observed a non-linear recovery of the VB shift on the picosecond timescale, showing larger time constants for higher pump

fluences. This was interpreted as the effect of CDW phonons. On the other hand, just after pumping, the femtosecond VB dynamics was dominated by the excitonic instabilities. We propose a model based on exciton-phonon contributions which fits the trARPES data. Using the model, we can disentangle the relative effect of electrons and lattice in the formation of CDW phase.

## 3 Conclusions

Our study revealed that the exciton and lattice distortion jointly drive the CWD phase transition in 1T-$TiSe_2$. By a direct observation of the VB dynamics, obviously linked to the CDW gap, we disentangled both contributions in non-equilibrium condition.

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