Revealing Collective Modes in an Electronic Crystal using Structured Light

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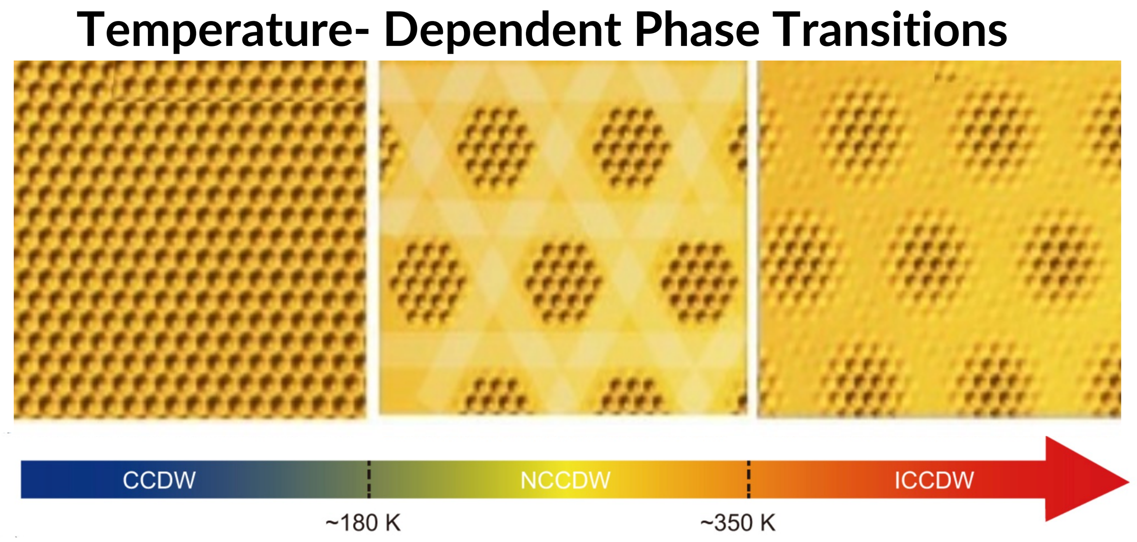
INTRODUCTION

- **Electronic crystals** form when electrons organize into ordered patterns within a material.
- **Transition metal dichalcogenides** are especially interesting because their electrons interact strongly with the vibrations of the crystal lattice.
- Among them, **1T- tantalum disulfide (1T – TaS2)** has remarkable properties due to multiple transitions between charge density wave states.
- Using ***structured light pulses***, we excite and probe collective oscillations in 1T-TaS_2, enabling selective detection of specific modes including a ***hidden mode***, not visible at equilibrium state.

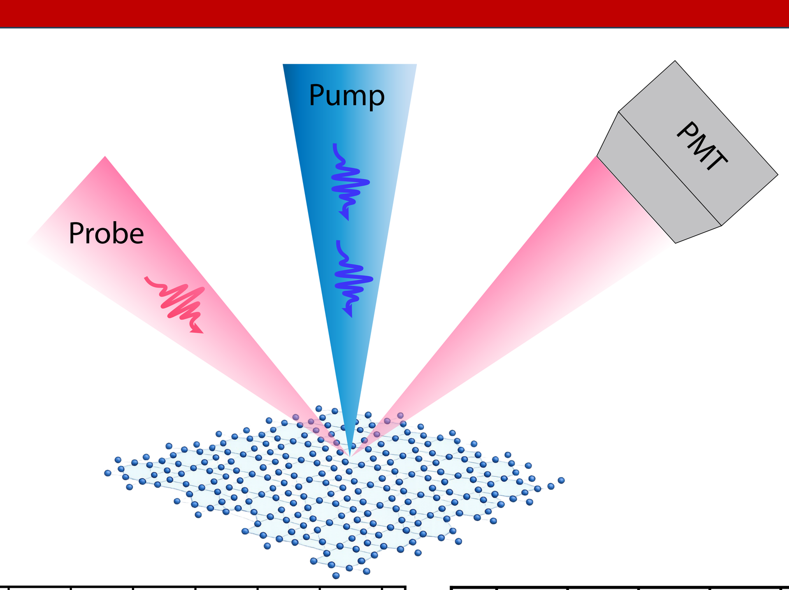
CONCEPTS

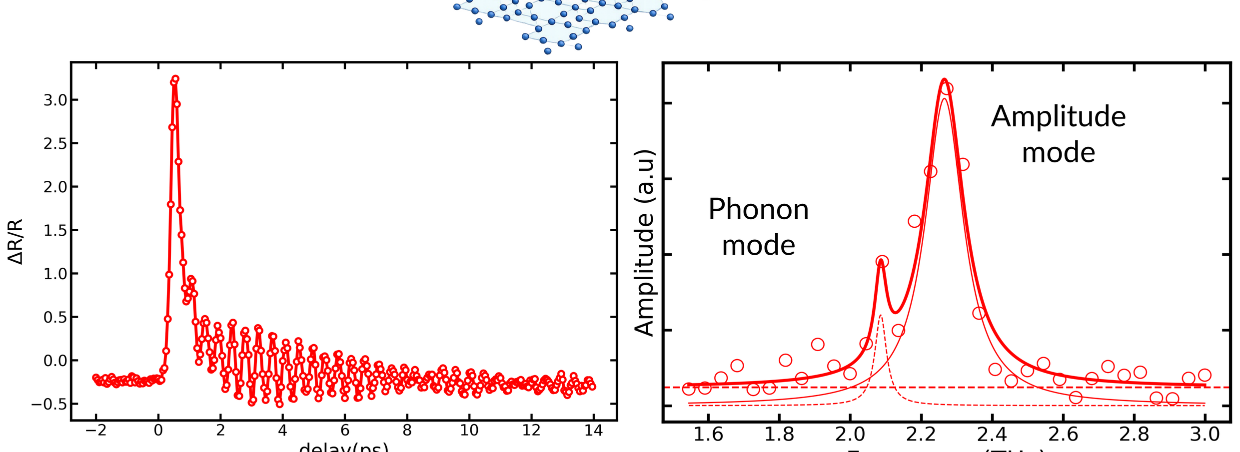
Temperature- Dependent Phase Transitions



1T – TaS_2 transitions from highly ordered commensurate phase to the domain structured nearly commensurate and finally to the incommensurate phase.

Initial studies using a single pump for Pump probe

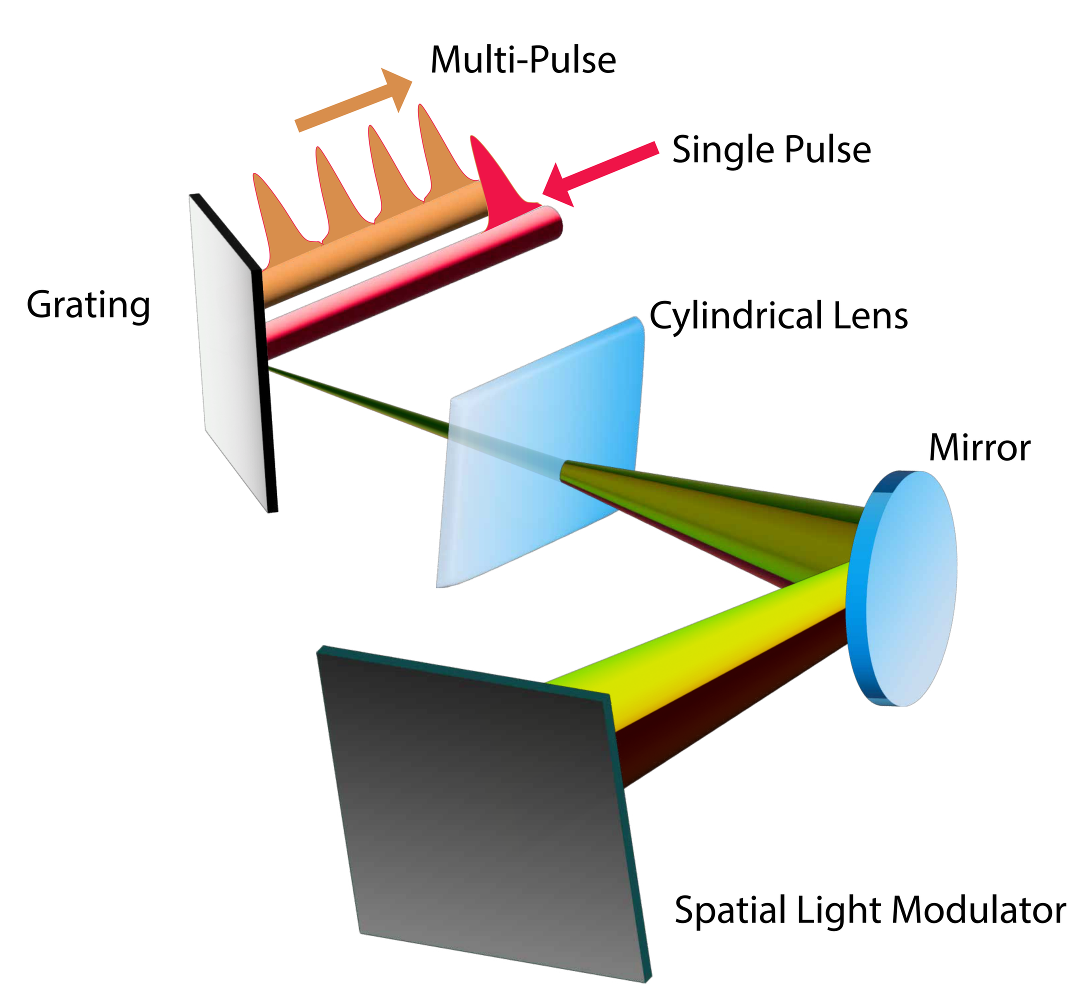




Preliminary data taken at 100 K at 1.7 mJ/cm²

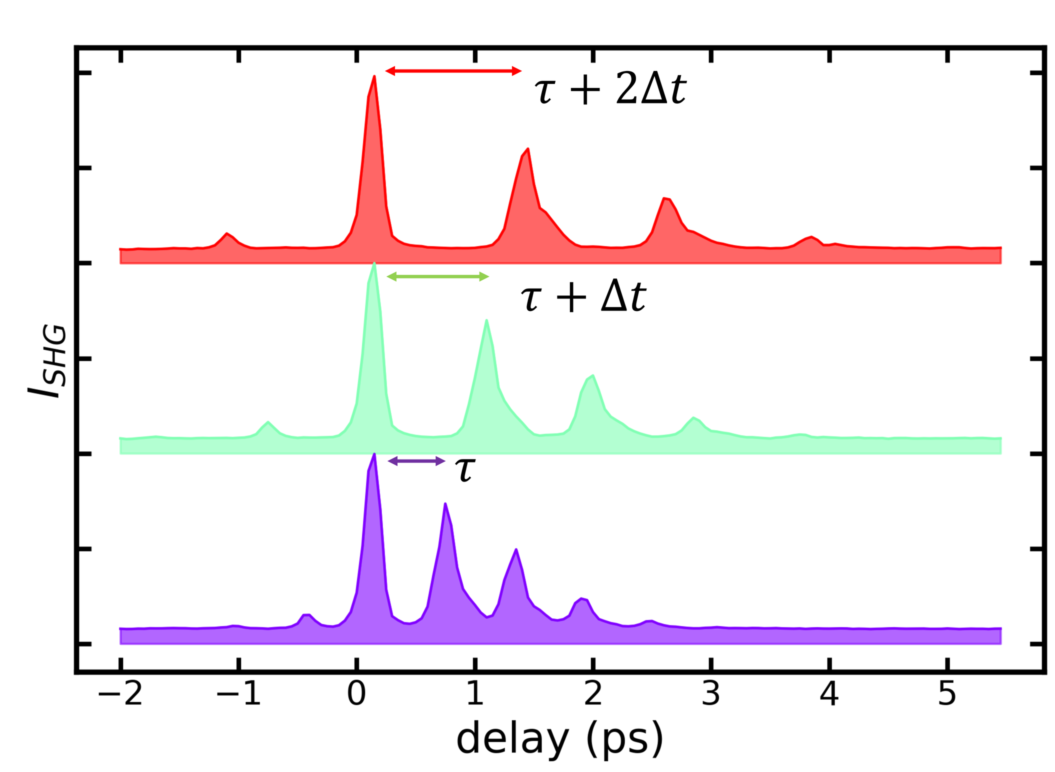
EXPERIMENTAL DESIGN

Generation of multi-pulse using a 2D- shaper



A programmable pulse shaper generates tailored pulse sequences to control excitation and probe the resulting dynamics in the material.

Illustration using Nonlinear crystal (BBO)

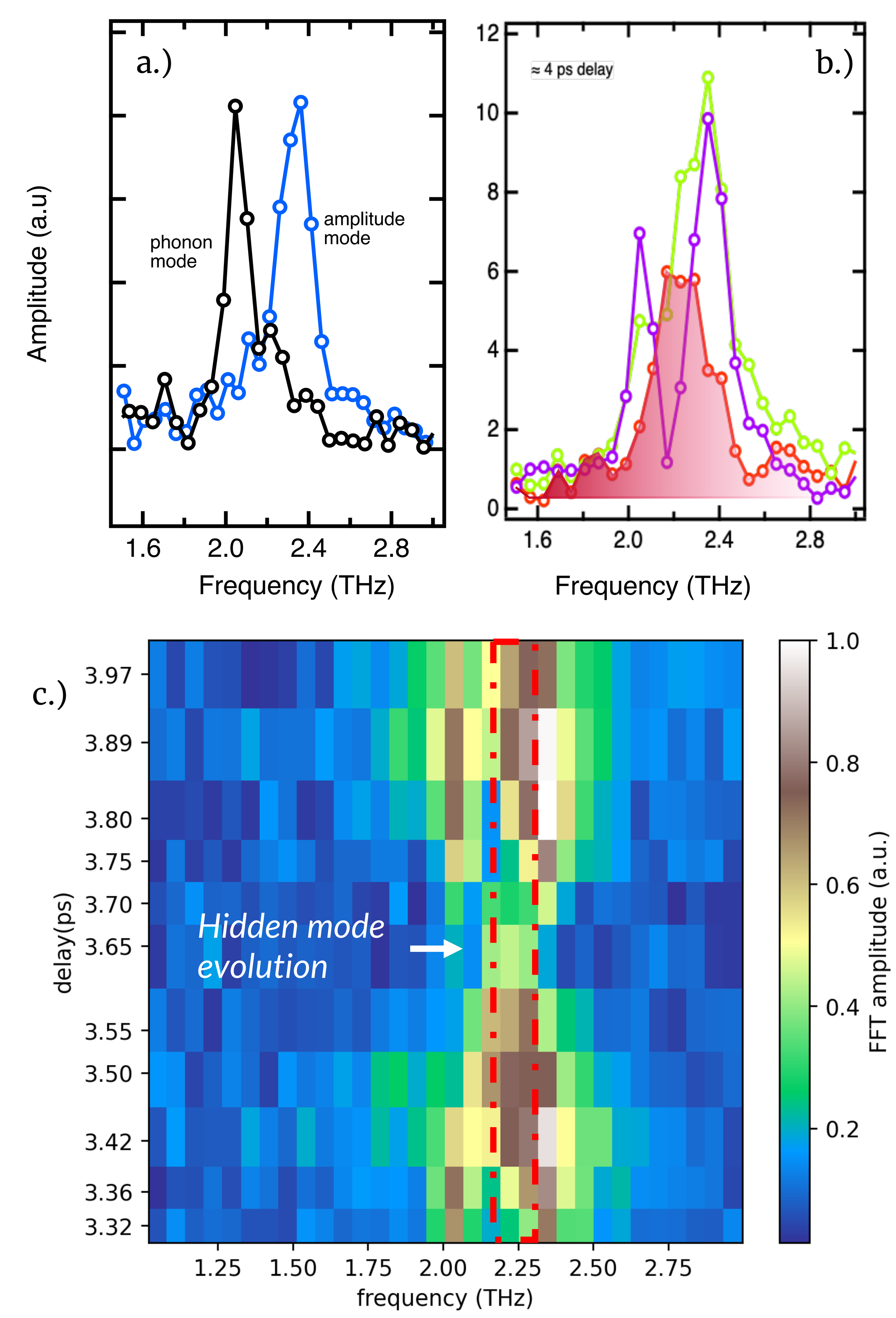


2D SHAPER

- ZEROTH ORDER: better light efficiency
- FIRST ORDER: better range of amplitude modulation

- The maximum pulse spacing achieved is ~10 ps which is due to the limit of the resolution of the spatial light modulator.

RESULTS AND ANALYSIS



- With a multipulse excitation scheme, the timing between pulses can be tuned to selectively drive either the **phonon** modes or the **amplitude** modes in (a).
- The appearance of a photoinduced metastable state – hidden state is also achieved as shown in red trace in (b).
- The evolution of hidden mode is shown in the 2D image map of the Fourier spectra (c) at various multipulse delays.

CONCLUSIONS

Structured light enables selective control of coupled lattice and electronic dynamics in charge density wave materials providing access to hidden, nonequilibrium phases and new routes towards coherent control of quantum materials.

ACKNOWLEDGEMENTS

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REFERENCES

Vaskivskyi, I., et al. "Fast electronic resistance switching involving hidden charge density wave states." *Nature communications* 7.1 (2016): 11442..

Frumker, E., & Silberberg, Y. (2007). Phase and amplitude pulse shaping with two-dimensional phase-only spatial light modulators. *Journal of the Optical Society of America B*, *24*(12), 2940-2947..