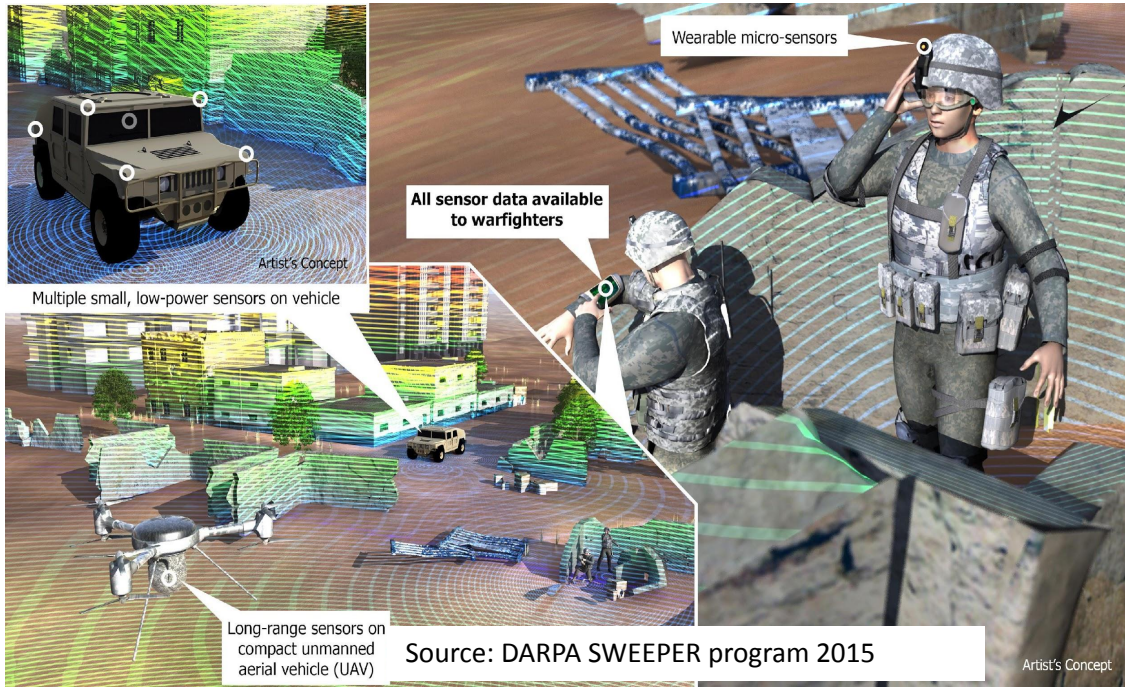
Nanophotonics – poised to revolutionize displays and imaging!

RICE

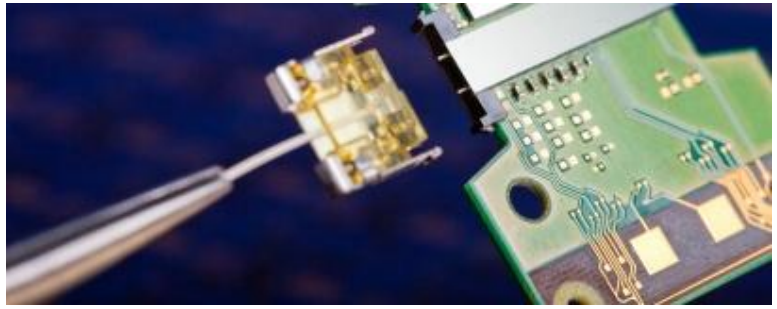
LiDAR



Virtual reality

Artificial vision

3D displays



Optical computing

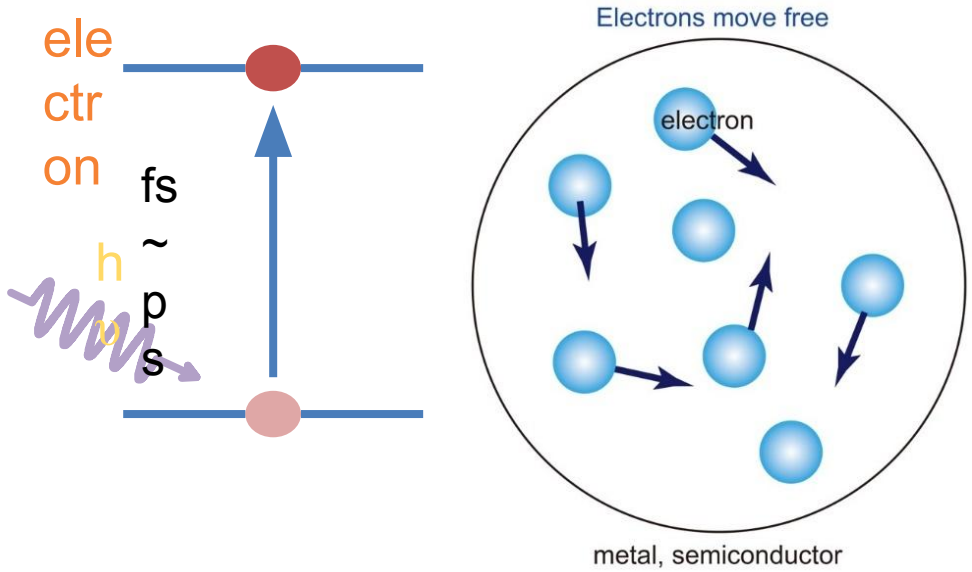
Quantum materials exhibit fast and large tunability!

Either small or slow tuning speed limit the application

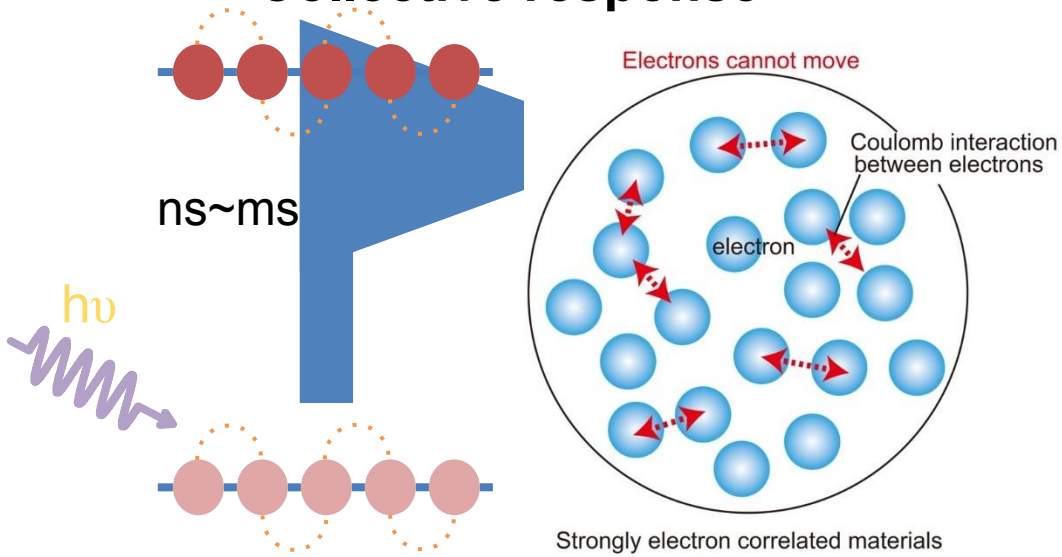
- Mechanical and thermal tuning can be slow
- Small tunability due to a single mechanism involved in light-matter interaction

A new class of tunable optical materials: **Quantum materials!**

Conventional Materials:
Single particle response

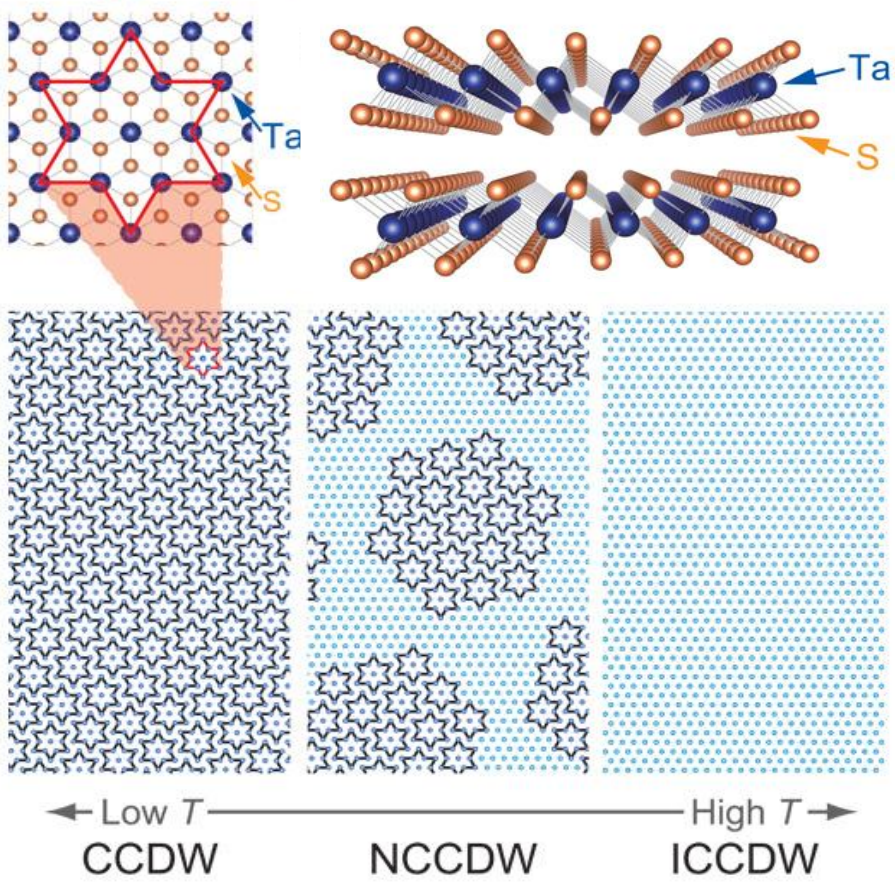


Quantum Materials:
Collective response

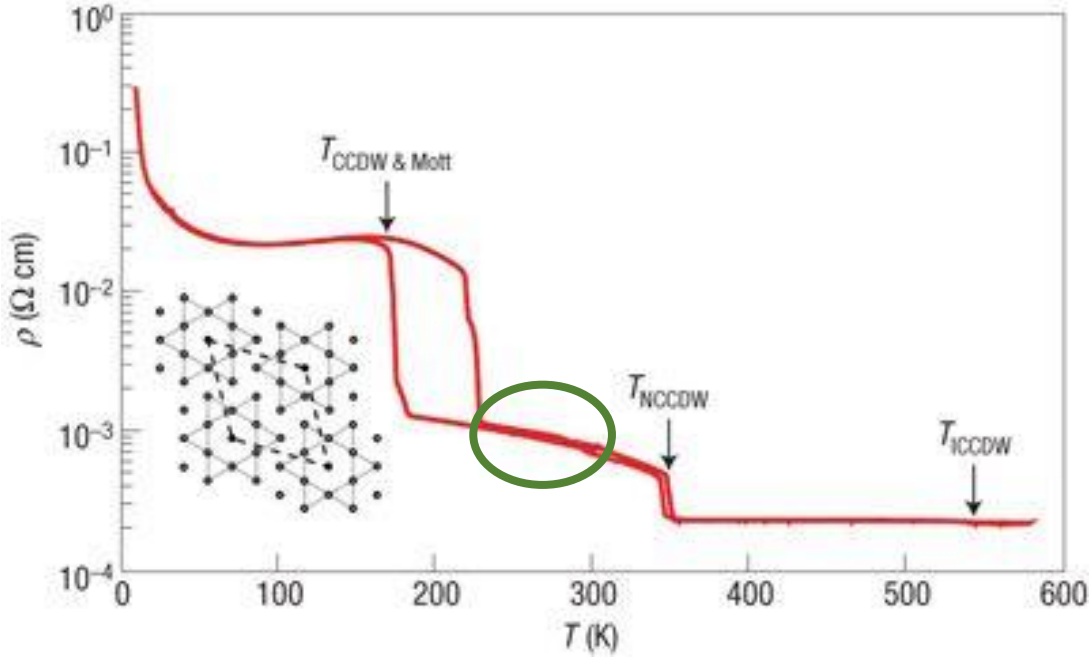


P. Gopalan, B. Sensale-Rodriguez, *Adv. Opt. Mater*. 2019. K. Hallman, et al., *Adv. Opt. Mater.* (2020).

1T-TaS_2 exhibits charge density waves at room temperature



- CCDW: commensurate CDW (λ_{CDW} is an integer multiple of the lattice constant)
- NCCDW: nearly commensurate CDW (nearly an integer)
- ICCDW: incommensurate CDW (not an integer)

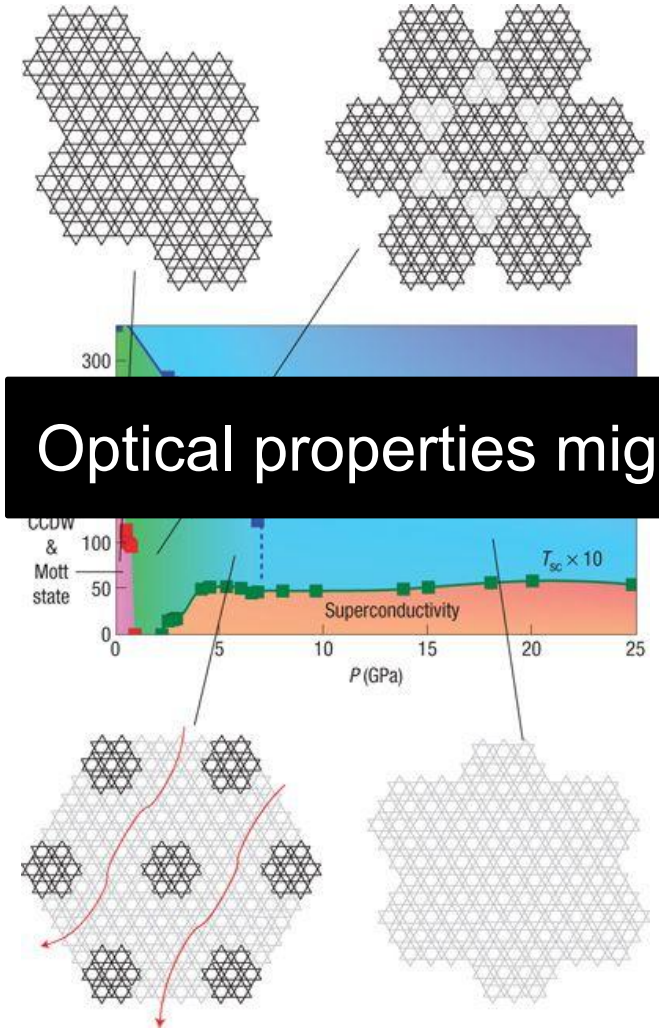


Charge density waves (CDW) result in a large change in electrical resistance with temperature.

Journal of Electron Spectroscopy and Related Phenomena, Strongly correlated systems, 117–118 (2001): 433–49.

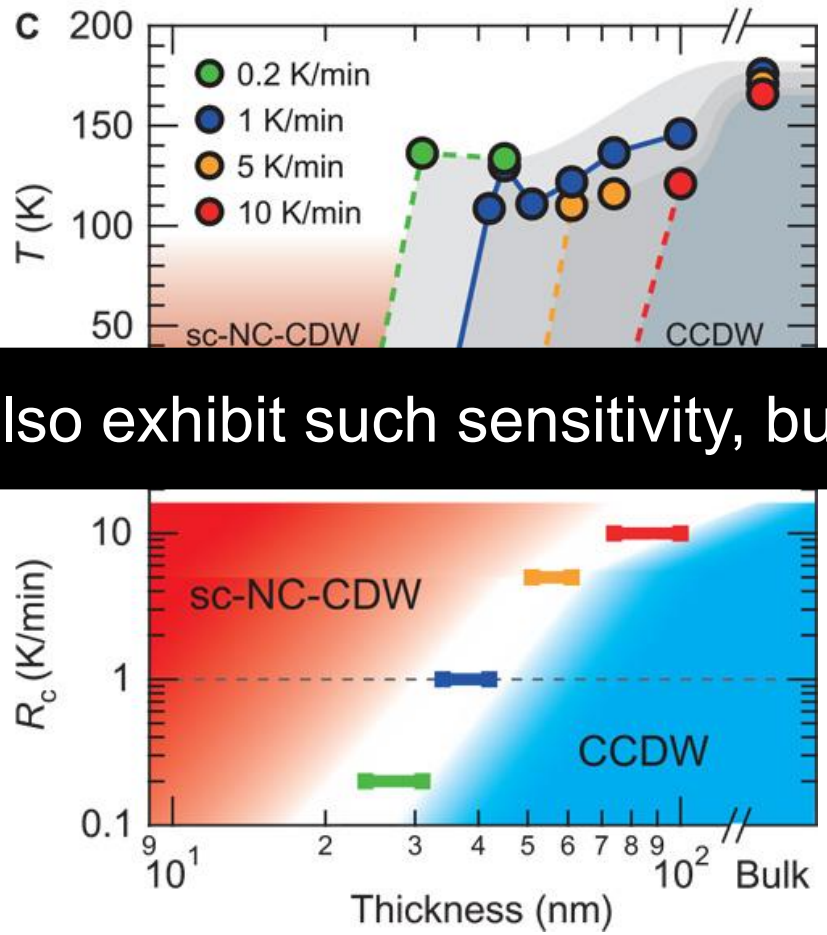
Physical properties of 1T-TaS_2 are sensitive to stimuli

Pressure tuning phase



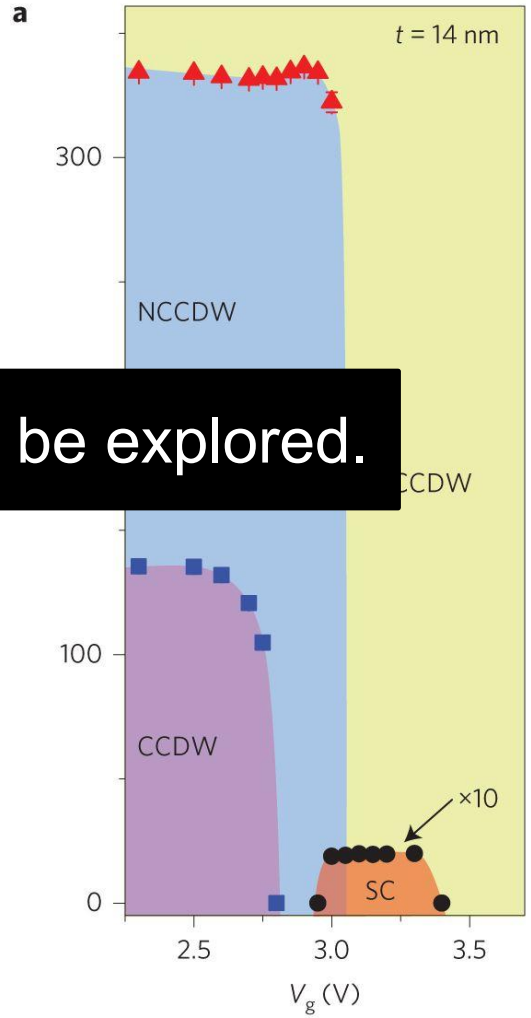
Journal of Electron Spectroscopy and Related Phenomena, Strongly correlated systems, 117–118 (2001): 433–49.

Thickness tuning phase



Yoshida, Masaro, et al. *Science advances* 1.9 (2015): e1500606.

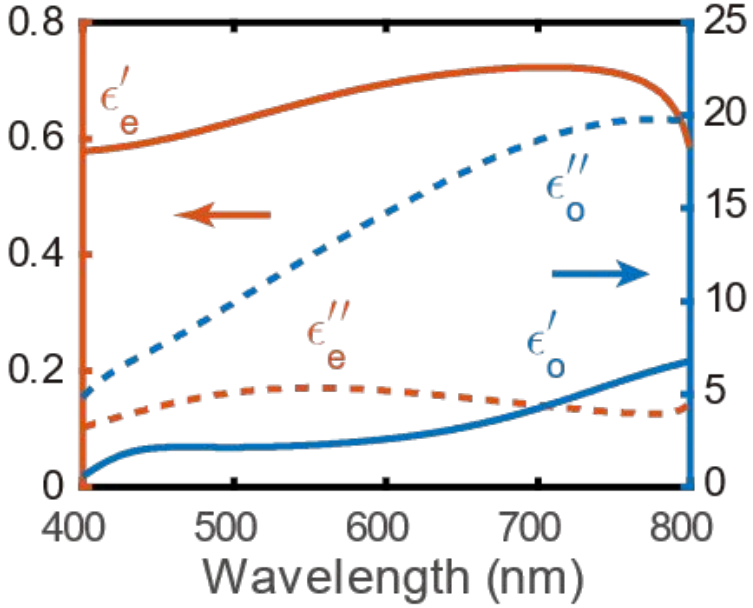
Bias tuning phase

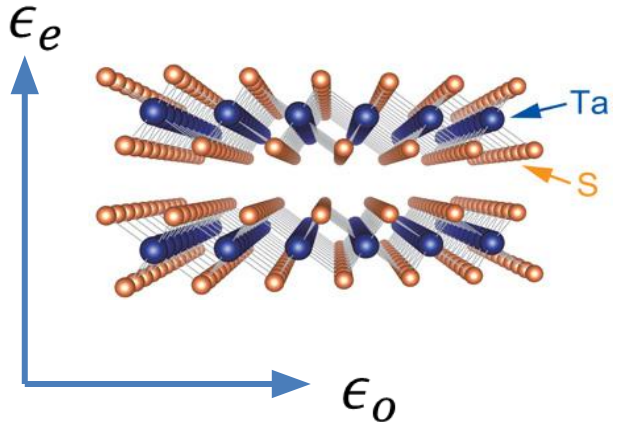


Yu, Y., Yang, F., Lu, X. *et al. Nature Nanotech* **10,** 270–276 (2015).

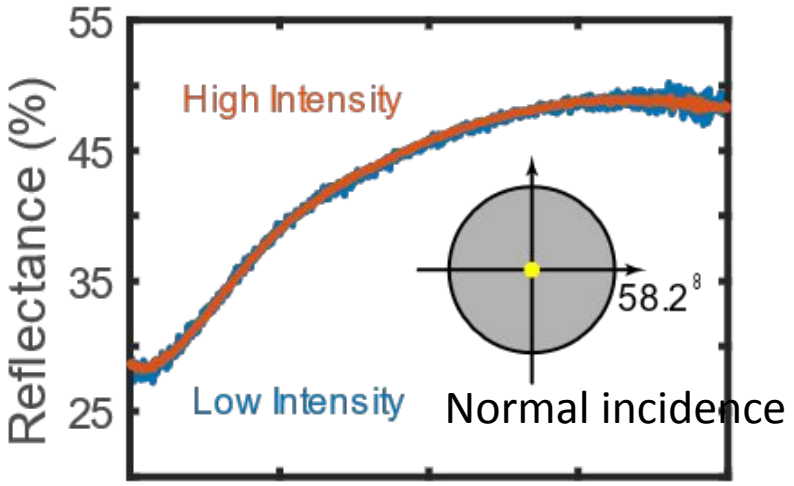
Optical properties might also exhibit such sensitivity, but yet to be explored.

Low intensity light tunes out-of-plane permittivity of 1T-TaS_2 by unity order!

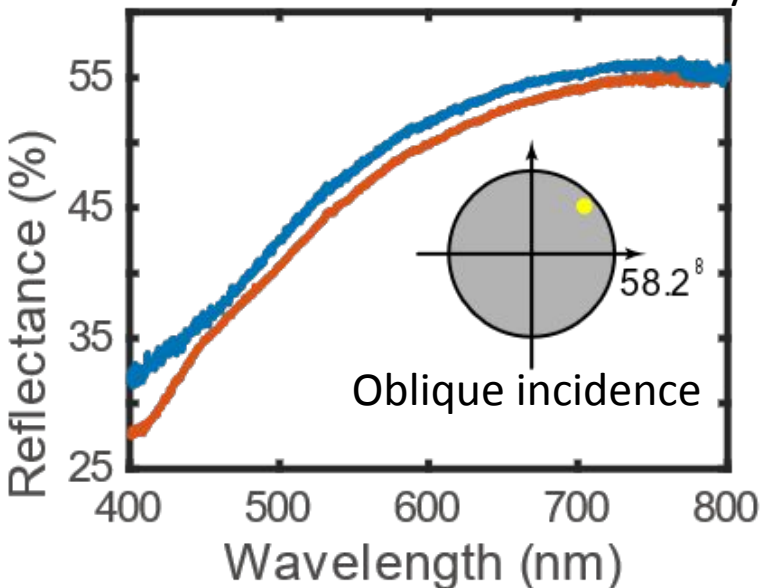


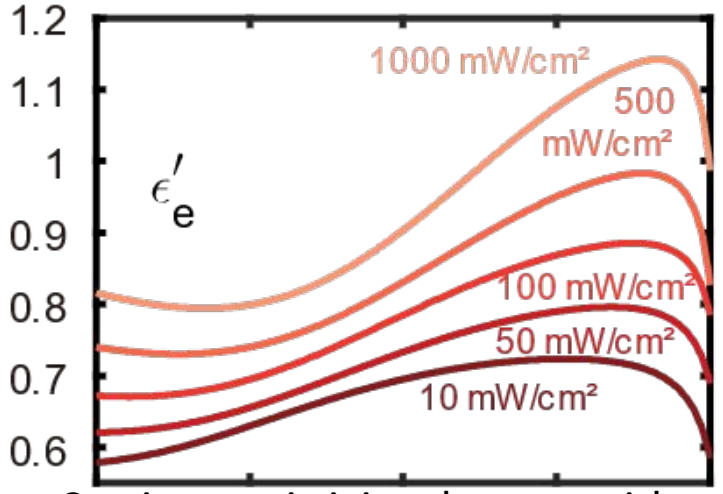


Li, and Naik, *Nano Lett.* (2020)

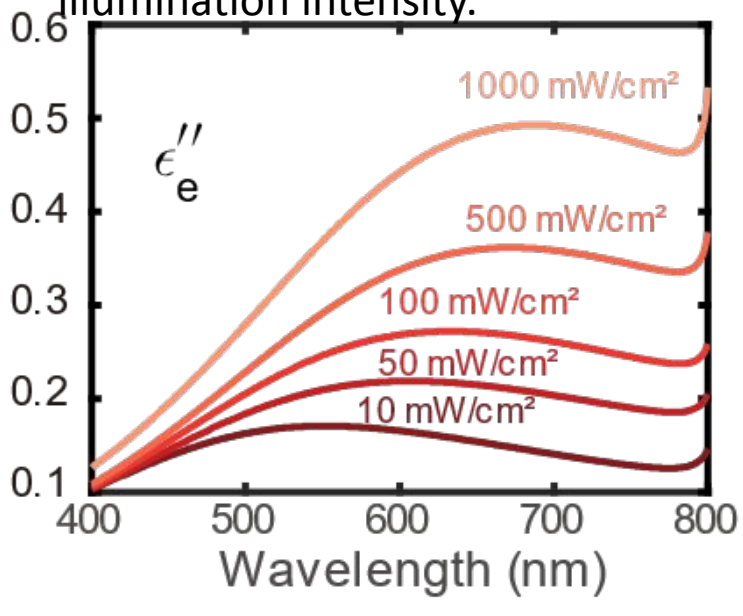


Oblique incidence reflectance is sensitive to illumination intensity.

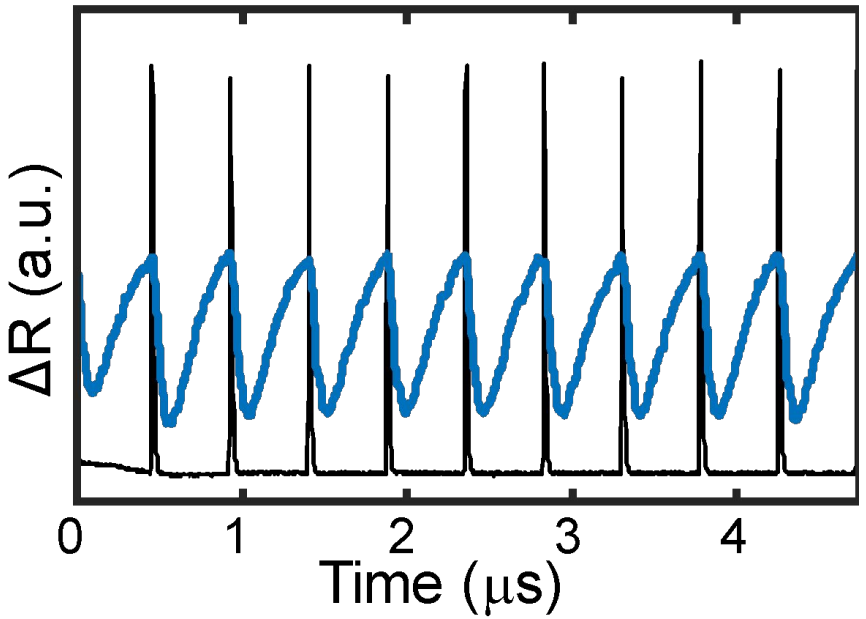


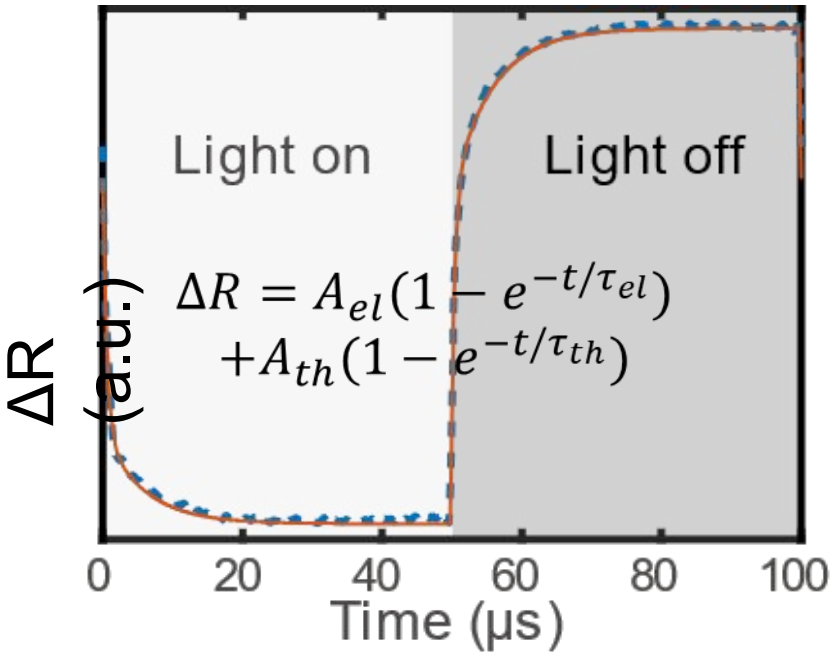


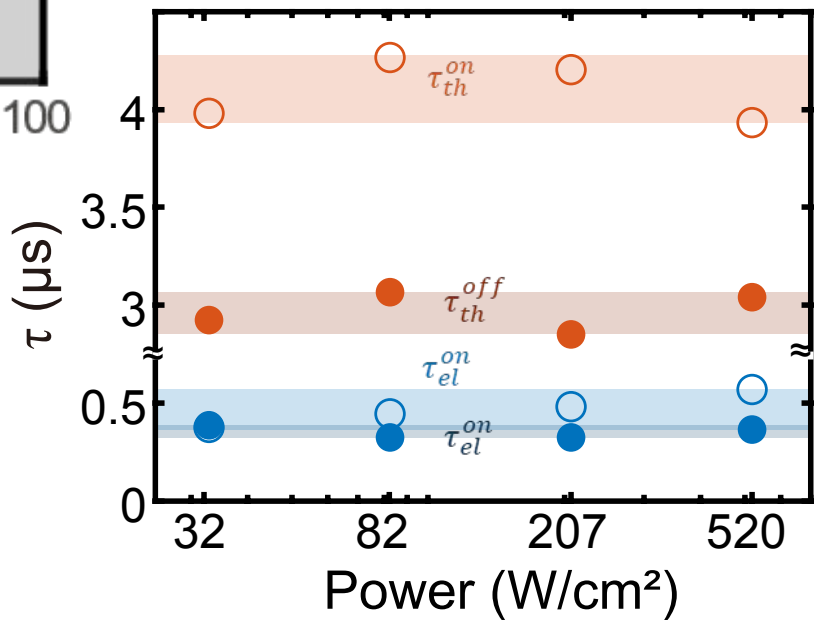
C-axis permittivity changes with illumination intensity.



Photorefractive effect of 1T-TaS_2 in z-axis is fast!





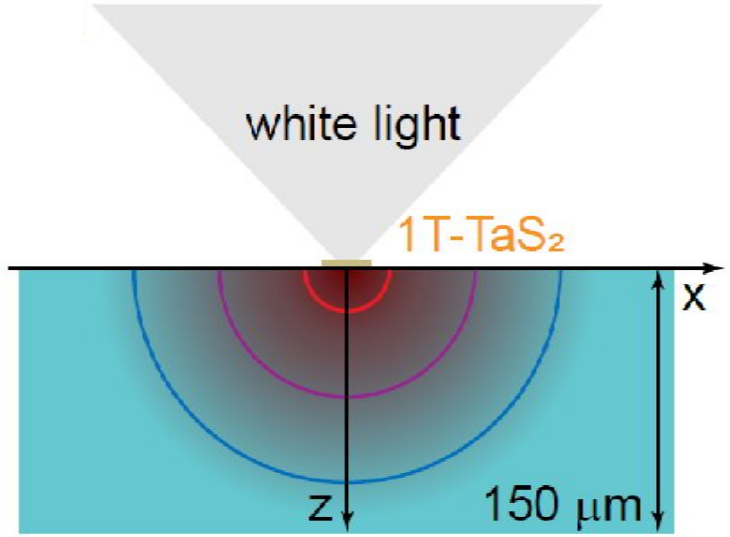


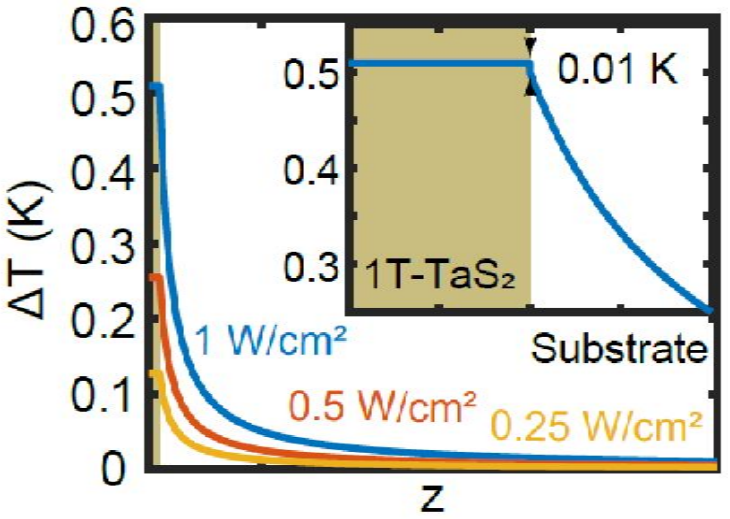
Electronic reconfiguration results in the sub-microsecond relaxation time constant

Li, and Naik, *Nano Lett.* (2020)

Possible mechanisms of optical tunability in 1T-TaS_2

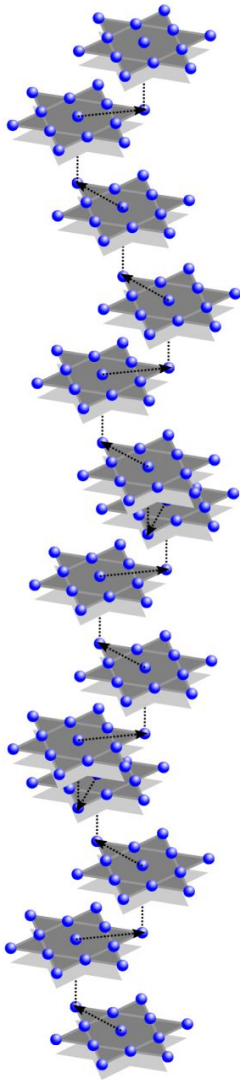
- Free electron generation?
  - 1 W/cm^2 @ 550 nm => 27.7 ns^{-1}·nm^{-2} => with 1μs relaxation time and 100 nm thick film, $\Delta n \approx 3 \times 10^{17} cm^{-3}$
  - Too small Δn for unity order index change in visible
- Nonlinear optics?
  - No higher harmonic generation
- Light induced thermo-optic effect?
  - White light intensity is too small to expect heating
- Collective electronic mode?



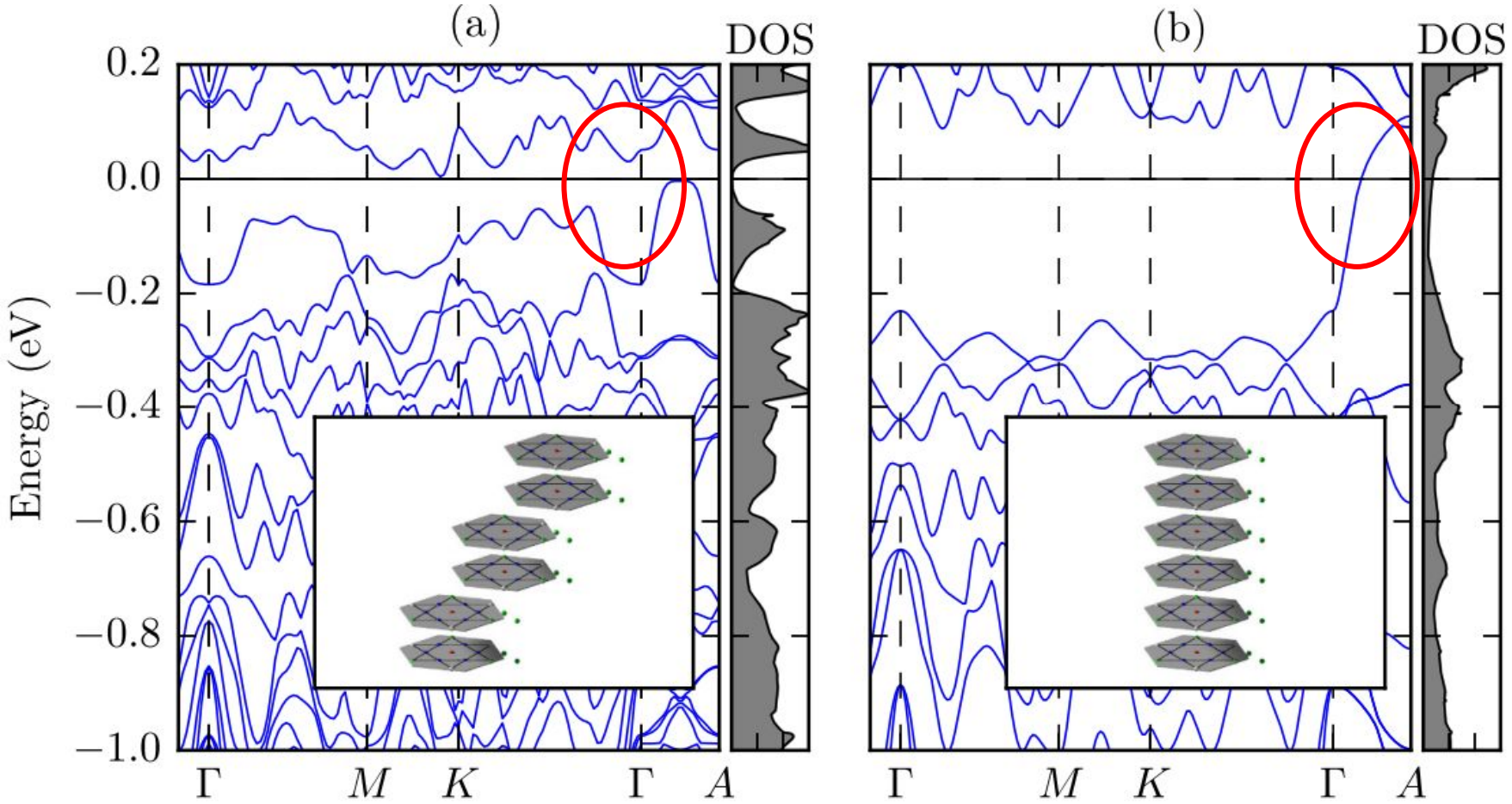


Li, and Naik, *Nano Lett.* (2020)

CDW stacking changes electronic properties dramatically!



Γ – A: out-of-plane direction

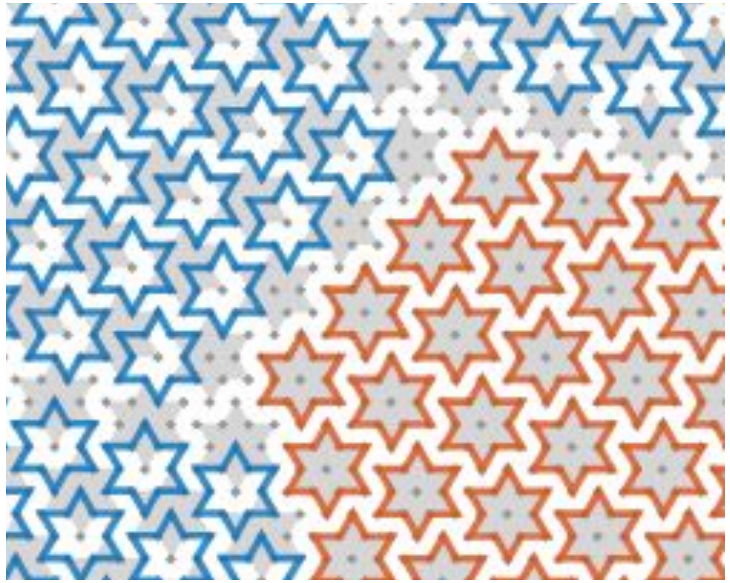


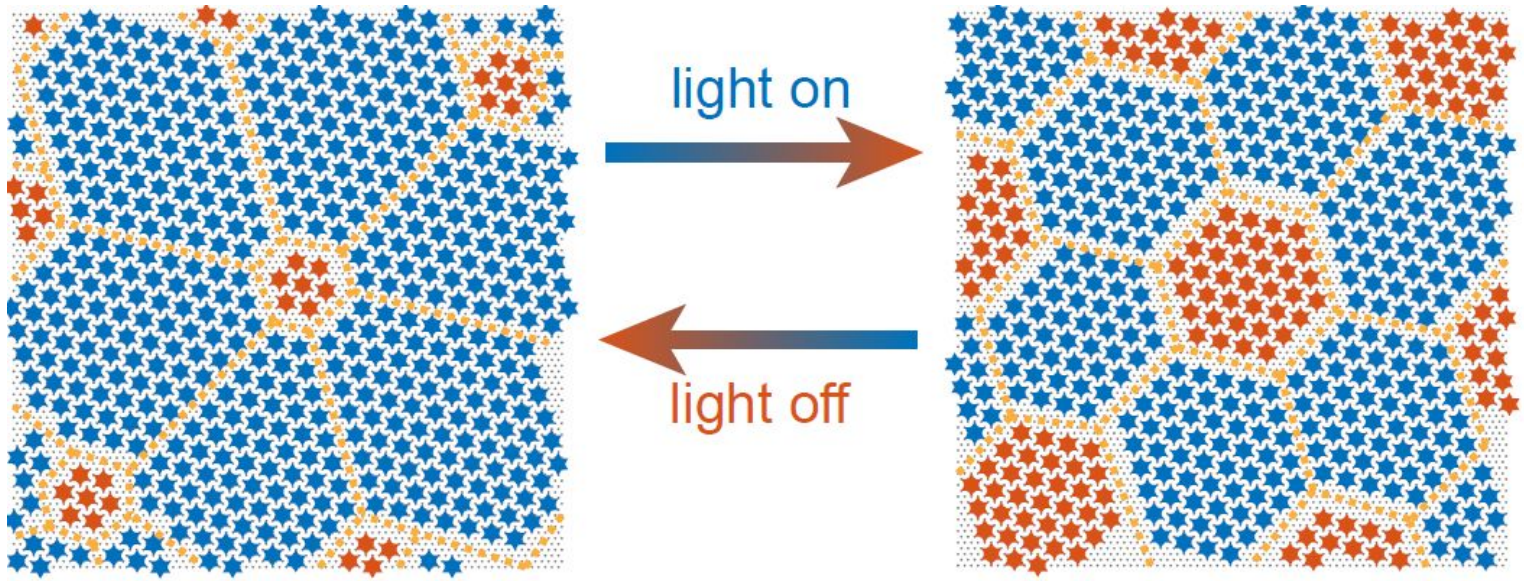
Different charge domain stacking leads to very different electronic (and optical) properties.

Nature communications 11, no. 1 (2020): 1-7. *Physical Review B* 98, no. 19 (2018): 195134. *Phys. Rev. Lett.* ***122****, 106404 (2019).*

Light reorganizes stacking of CDW domains across layers

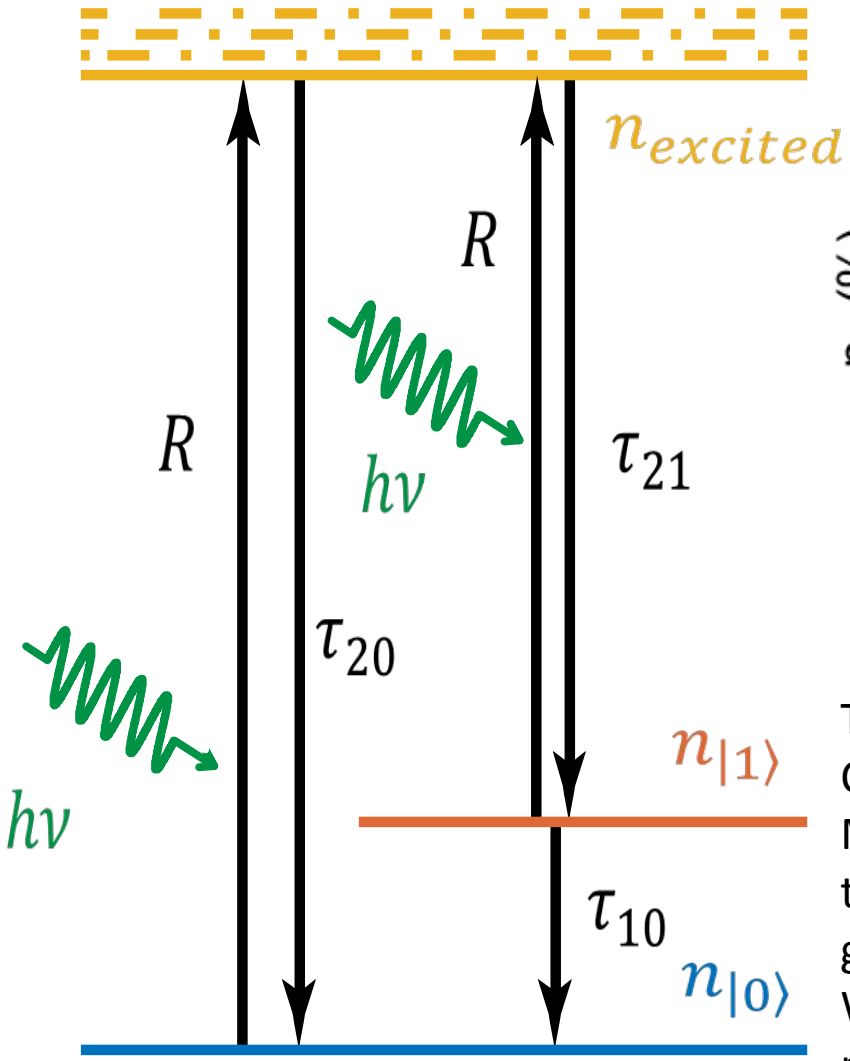
Hypothesis: Light reorganizes the CDW stacking order and thus changes the refractive index of 1T-TaS2. Illumination causes the center-to-corner stacking domains to shrink and the center-to-center stacking domains to grow.



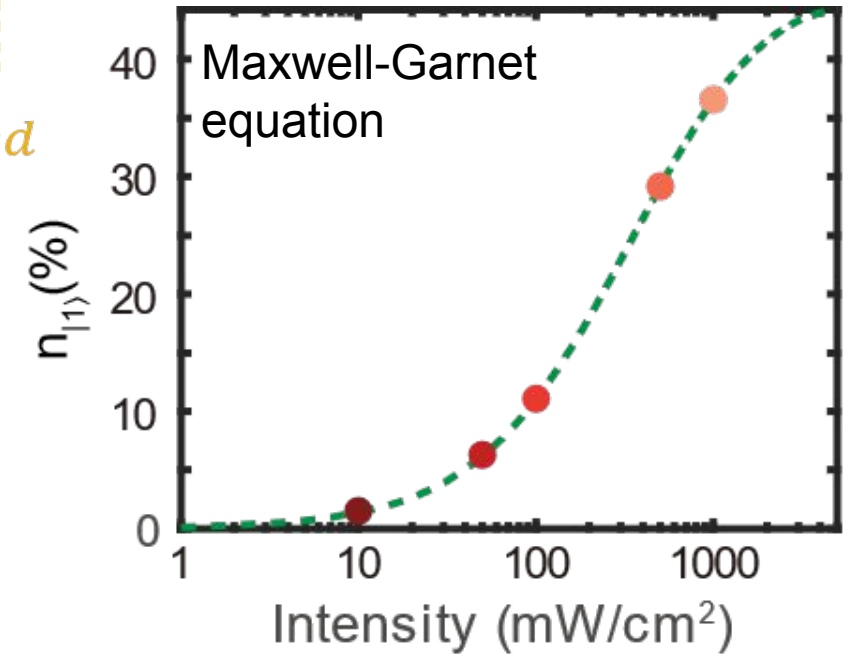


Li, and Naik, *Appl. Phys. Lett*. 2021

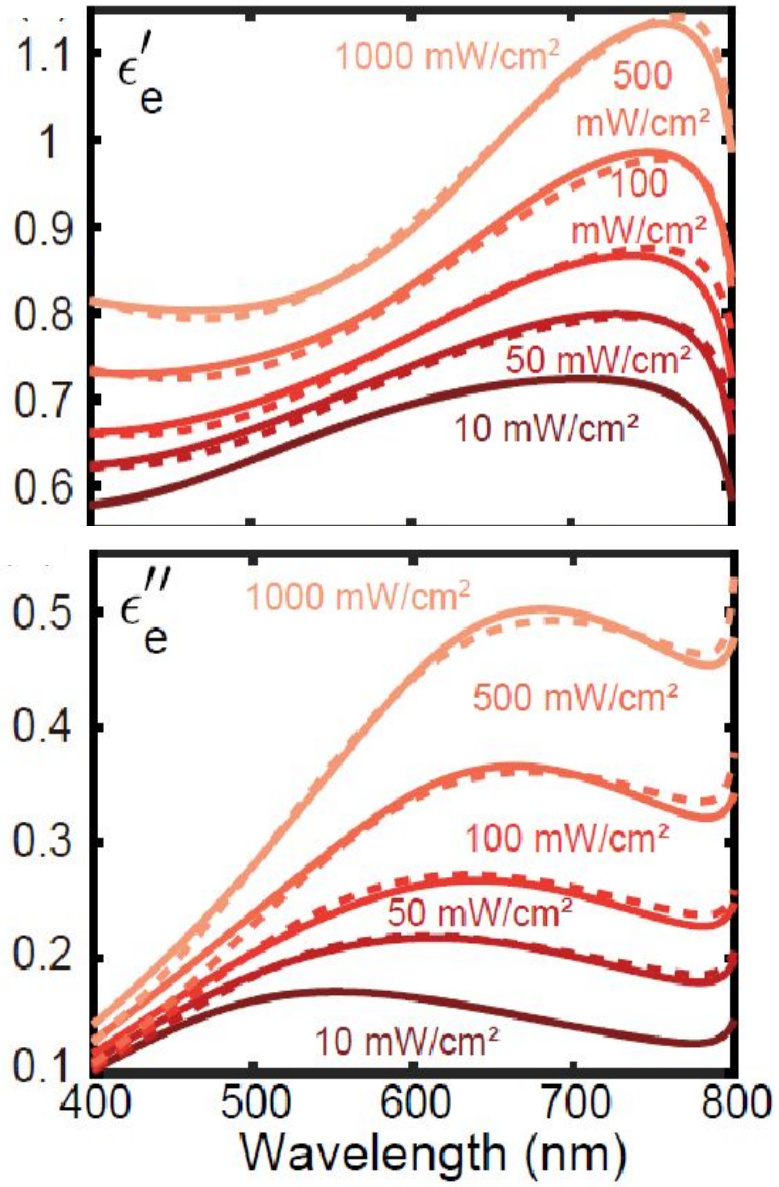
3-level model & Maxwell-Garnet equation describe 1T-TaS_2 optical properties



Li, and Naik, *Nano Lett.* (2020)



The 3-level model predicts the fraction of CDW domains in the two stacking orders. Maxwell-Garnett's effective medium theory predicts the optical constants for a given stacking domain composition. With no free-fitting parameter, our model matches the experimental results well. This proves our hypothesis.



1T-TaS_2 exhibit light tunable optical properties at room T

We show that 1T-TaS_2 is

- An anisotropic dielectric in visible.
- Shows large index tunability with AC and DC biases.
- Exhibits a light tunable ϵ_c
- Light reorganizes CDW domain stacking.
- 3-level system and Maxwell-Garnett model predicting ϵ_c.

- Light is an important tool to control the energy landscape of strong correlations.
- Possible light-induced superconductivity
- 1T-TaS_2 is promising for fast tunable nanophotonics!