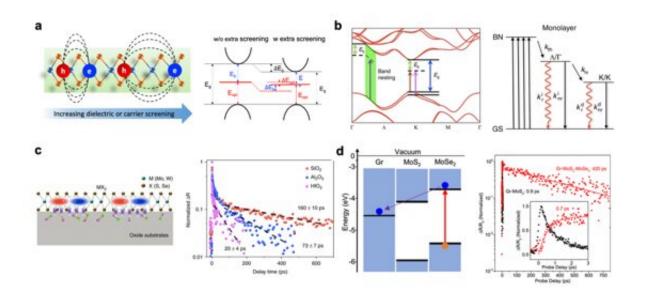


## Modulation of photocarrier relaxation dynamics in two-dimensional semiconductors

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a, Modulation on the Coulomb interaction. (left) Illustration of increased screening of Coulomb interactions in 2D semiconductors. (right) Schematic illustration showing the impact of increased screening of Coulomb interactions on the electronic bandgap (Eg), exciton binding energy (Eb) and optical bandgap (Eopt) of 2D semiconductors. b, Modulation through initial distribution of photocarriers in electronic band structures (left) The electronic band structure of monolayer TMDs by DFT calculation. The green area shows the band nesting region. (right) Relaxation pathways of photocarriers in monolayer TMDs, where the excitation is from ground state (GS) to the band nesting (BN) region. c, Modulation through interfacial electron-phonon coupling. (left) Illustration of interfacial electron-phonon (e-ph) coupling. (right) Photocarrier dynamics of monolayer MoSe2 on different substrates. d, Modulation through engineering the band alignment of vdW heterostructures. (left) Band alignment of the graphene/MoS2/MoSe2 trilayer sample. (right) Electron transfer from MoSe2 to graphene and its lifetime in the trilayer. Credit: Yuhan Wang, Zhonghui Nie,



Fengqiu Wang

Two-dimensional (2D) semiconductors can host a rich set of excitonic species because of the greatly enhanced Coulomb interactions. The excitonic states can exhibit large oscillator strengths and strong light-matter interactions, and dominate the optical properties of 2D semiconductors. In addition, because of the low dimensionality, excitonic dynamics of 2D semiconductors can be more susceptible to various external stimuli, enriching the possible tailoring methods that can be exploited.

Understanding the factors that can influence the dynamics of the optically-generated <u>excited states</u> represents an important aspect of excitonic physics in 2D semiconductors, and is also crucial for practical application as excited state lifetimes are linked to the key figures of merit of multiple optoelectronic and photonic devices. While certain experiences have been accumulated for bulk semiconductors, the atomic nature of 2D semiconductors might makes these approaches less effective or difficult to be adapted. On the other hand, the unique properties of 2D semiconductors, such as the robust excitonic states, the sensitivity to external environmental factors and flexibility in constructing vdW heterostructures, promise modulation strategies different from conventional materials.

In a new review article published in *Light: Science & Applications*, a team of researchers, led by Professor Fengqiu Wang from Nanjing University, China summarize the so far obtained knowledge and progresses on the modulation of photocarrier <u>relaxation</u> dynamics in 2D semiconductors. After a brief summary on the photocarrier <u>relaxation dynamics</u> in 2D semiconductors, the authors first discuss the modulation of Coulomb interactions and the resulting effects on the transient properties. The



Coulomb interactions in 2D semiconductors can be modulated by introducing additional screening from the external dielectric environment or injected <u>charge carriers</u>, leading to the modification of quasi-particle bandgaps and the exciton binding energy. Then the influencing factors on photocarrier dynamics and the manipulating methods are discussed according to the relaxation pathways or mechanisms they are associated with.

The first discussed factor is the initial distribution of photocarriers in electronic band structures, which can affect their decay processes by enabling different available relaxation pathways in the energy and momentum space. After that defect-assisted and phonon-assisted relaxation are discussed. While the approaches utilizing defect-assisted relaxation such as ion bombardment and encapsulation are similar to those for bulk semiconductors, the modulation on phonon-assisted relaxation for 2D semiconductors can be different.

"On one hand, the coupling between charge carriers and phonons can be enhanced due to the suppressed dielectric screening; on the other hand, the high surface-to-volume ratio make 2D materials more susceptible to the external phononic environment." Moreover, the flexibility in constructing vdW heterostructures and the ultrafast charge transfer across the interfaces enables tailoring the photocarrier dynamics though band alignment engineering.

The transition between different particle species also offers the opportunity to modulate through changing the ratios between different quasiparticles, which can modify the relative portion of different relaxation pathways, and thus the transient optical responses of the whole sample. At last, the modulation of the dynamics of spin/valley polarization in 2D TMDs is discussed, and the discussion is mainly focusing on the methods to increase the lifetime of the spin/valley polarization.



Through this review, the authors aim to provide guidance for developing robust methods tuning the photocarrier relaxation behaviors and strength the physical understanding on this fundamental process in 2D semiconductors. As noted by the authors in conclusion, "Tremendous research efforts are still needed in both fundamental understanding and practical modulation of the photocarrier relaxation in 2D semiconductors."

**More information:** Yuhan Wang et al, Modulation of photocarrier relaxation dynamics in two-dimensional semiconductors, *Light: Science & Applications* (2020). DOI: 10.1038/s41377-020-00430-4

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