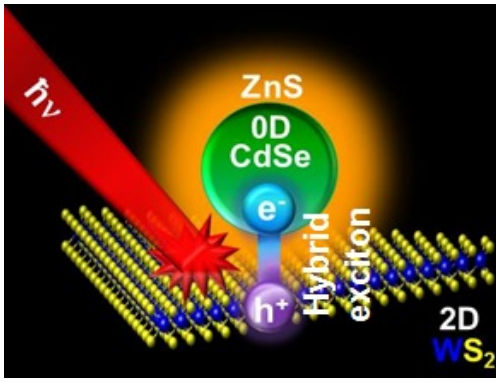
Engineering Materials for Ultrafast Charge Transfer

Achievement: Efficient transformation of photexcitation facilitated by hybrid excitons engineered from a two-dimensional layer (2D)/zero-dimension (0D) quantum dot nanostructure.



Photoexcitation of a 2D WS_2 monolayer creates excitons which rapidly (sub-45 fs) dissociate and transfer electrons to 0D ZnS-capped CdSe quantum dots, as revealed by ultrafast pump-probe laser spectroscopy. The resulting 0D/2D hybrid exciton, with an electron in the quantum dot and a hole in the 2D layer, is more weakly bound than the original 2D exciton.

Significance and Impact: The hybrid excitons are several times more weakly bound than in parent 2D materials, enabling dissociation that can improve the conversion efficiency of light into electricity

Research Details:

- A hybrid nanostructure composed of a monolayer of WS_2 covered with a monolayer of CdSe-ZnS quantum dots
- Photoluminescence and micro-absorption spectroscopies characterized the hybrid nanoscale system and its individual building block.
- Femtosecond pump-probe spectroscopy revealed ultrafast formation (~45 fs) of weakly-bound excitons following electron transfer from WS_2 monolayer to the quantum dots.

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Overview:

In a synthesized hybrid nanostructure composed of a monolayer of tungsten disulfide (2D-WS_2) and a single layer of core-shell cadmium selenide (CdSe)-zinc sulfide (ZnS) quantum dots (0D-QDs), we show that following selective photo-excitation of the 2D-WS_2, electron transfer to the 0D-QDs takes place on an ultrafast sub-45 fs time-scale. The electron now located at the 0D-QDs is bound to the hole left at the 2D-WS_2 forms a hybrid exciton that is several times more weakly bound than the tightly bound excitons in 2D-WS_2 MLs. Such photoinduced interfacial charge transfer is at the heart of many applications, including photovoltaics, photocatalysis, and photodetection and thus may open new opportunities for optoelectronic applications of 2D and 0D nanomaterials.

Monolayer two-dimensional transition metal dichalcogenide (2D-TMD) semiconductors recently emerged as new candidates for atomically-thin optoelectronics. Unfortunately, excitons in these materials are tightly bound (>500 meV), which makes their dissociation challenging and constrains 2D-TMD application in photovoltaic and photodetection applications. In this work, a novel approach was advanced to mediate this problem by demonstrating how strongly-bound excitons of 2D tungsten disulfide (WS_2) monolayers, covered with a single layer of semiconductor quantum dots (0D-QDs) of core/shell CdSe/ZnS, are converted into hybrid excitons (HXs) that are several times more weakly bound. Femtosecond pump-probe spectroscopy measurements indicated that HXs are formed through electron transfer to the 0D-QDs on a sub-45 fs time-scale. The conversion of strongly bound excitons in 2D

semiconductors into easily dissociable HXs may open new opportunities for optoelectronic applications, notably in photovoltaics where exciton dissociation is of prime importance.