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Ultrafast Charge Carrier Dynamics in Two Dimensional Materials

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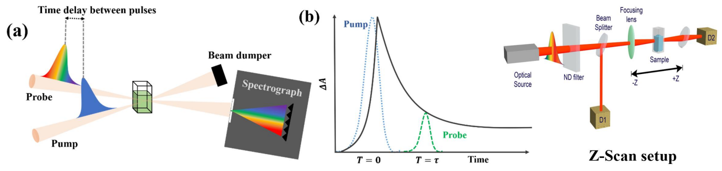
- **2D SnS is a member of emerging layered metal monochalcogenide. Theoretical investigations revealed a sizeable band gap, quantum confinement effect, high carrier mobility and large absorption coefficient in few-layer SnS.**
- **In ultrathin SnS understanding of elementary quasiparticles and their interaction or dynamics in condensed matter physics system is critical for better energy conversion and photonic applications.**
- **SnS layers are environmentally, thermally and dynamically stable. Our findings highlight the prospects for the synthesis of ultrathin anisotropic SnS towards the betterment of thermoelectric and photonic devices.**

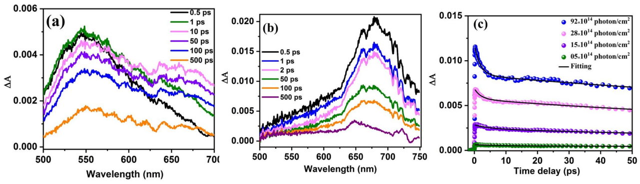
Experimental techniques



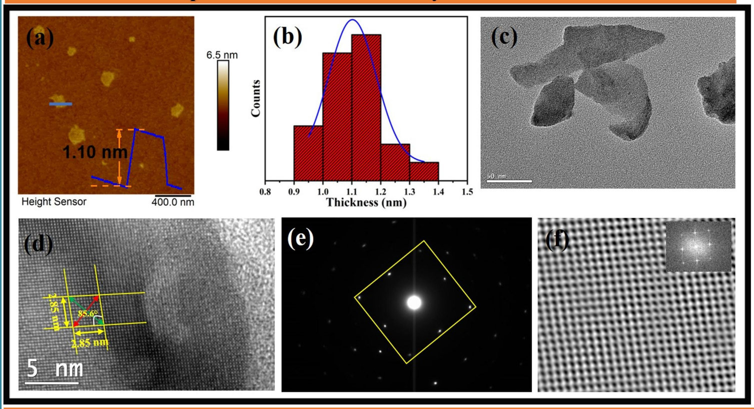
Transient absorption spectroscopy (TAS) setup

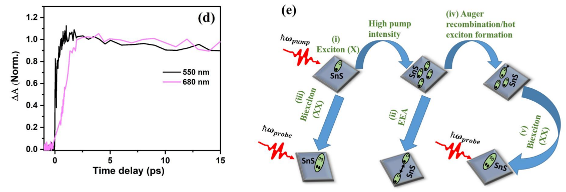
Z-Scan setup

Charge carrier dynamics



Microscopic characterization of layered SnS nanosheets





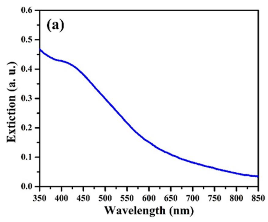
- The rate of annihilation of excitons is proportional to the square of the exciton density, N_x

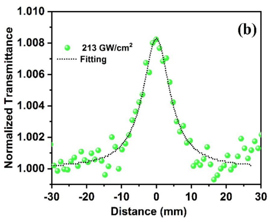
$$\frac{dN_x}{dt} = -kN_x^2$$

$$N_x(t) = \frac{N_0}{1 + kN_0 t} + C$$

- Annihilation rate $4.34 \times 10^{-4} \mathrm{cm^2/s}$

Nonlinear optics in 2D SnS nanosheets, using Z-Scan



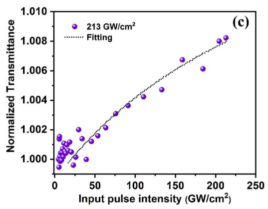


$$P(t) = \varepsilon_0\left(\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^{(3)}(t) + \cdots\right)$$

Here, ε_0 is the dielectric permittivity of vacuum, coefficients χ^n are the nth order susceptibilities of the medium

$$T = \frac{1}{\sqrt{\pi q_0}}\int_{-\infty}^{\infty} ln(1 + q_0 e^{-x^2})dx$$

For $|q_0| < 1$, the transmittance in terms of peak irradiance can be expressed in summation form as



$$T = 1 - \frac{\alpha_s}{1 + \frac{I_z}{I_{sat}}} - \alpha_{ns}$$

Where I_{sat} is the saturation intensity

$$T = 1 - \frac{\alpha_{NL} I_0 L_{eff}}{2\sqrt{2}\left[1 + \left(\frac{z}{z_0}\right)^2\right]}$$

Here $q_0 = \alpha_{NL} I_z L_{eff}$, α_{NL} is NLO absorption coefficient

(a) Absorption spectra of SnS nanosheets (b) Open aperture (OA) Z-scan curve (c) Normalized transmittance as a function of input pulse intensity.

Conclusions

- **We successfully conduct liquid phase exfoliation of SnS to obtain ultrathin (~ 1.15 nm) nanosheets.**
- **Exfoliated ultrathin SnS nanosheets are analyzed by TEM and AFM.**
- **High nonlinear absorption coefficient (-0.5 $\times 10^{-3}$ cm/GW) and low saturable intensity (570 GW/cm²) of SnS nanosheets due to strong EEA and absorption of delayed photon make them suitable for ultrafast Q-switching**
- **Our Transient absorption (TA) results reveal rich exciton-exciton and coherent exciton–photon interactions arising from many-particle excited effects in ultrathin SnS.**
- **At higher exciton density, Auger process leads to the formation of hot exciton, which is converted to a biexciton by absorbing another photon of low energy.**
- **This work sheds light on synthesis, nature of the phonon dynamics and nonlinear optical phenomena in 2D materials for futuristic energy conversion and photonic applications**

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