Light-Driven Structural and Optical Tuning of a van der Waals Semiconductor

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Van der Waals (vdW) arsenic trisulfide (As_2S_3) has emerged as a highly promising material for advanced nanophotonic applications due to its exceptional refractive index (>3) and substantial in-plane optical anisotropy ($\Delta n \approx 0.4$) [1]. Although the photorefractive properties of amorphous As_2S_3 have been extensively studied [2], its crystalline form has remained largely unexplored, primarily due to limitations in precise and scalable nanostructuring techniques.
In this work, we demonstrate giant photorefractive effects ($\Delta n \approx 0.1$) in crystalline As_2S_3 achievable entirely through optical illumination. Concurrently, atomic force microscopy revealed controlled thickness modulation behaviors, including precise thinning at moderate illumination intensities and substantial photoexpansion of up to 6–7% at higher intensities. These phenomena provide robust mechanisms for optically-driven structural reshaping of the material (Figure 1).
Leveraging these effects, we have successfully implemented high-resolution direct-write nanopatterning using continuous-wave (CW) laser illumination, achieving dot densities approaching 50,000 DPI without employing complex femtosecond lasers or ablative processing. This straightforward and scalable optical approach significantly enhances the versatility and applicability of crystalline As_2S_3 for integrated photonics, adaptive optical systems, and advanced optoelectronic devices.

References

[1] Slavich A. S. et al., Light Sci. Appl., 13 (2024) 68.
[2] Ozols A., Saharovs Dm., Reinfelde M., J. Non-Cryst. Solids, 352 (2006) 2652–2656.

Figures

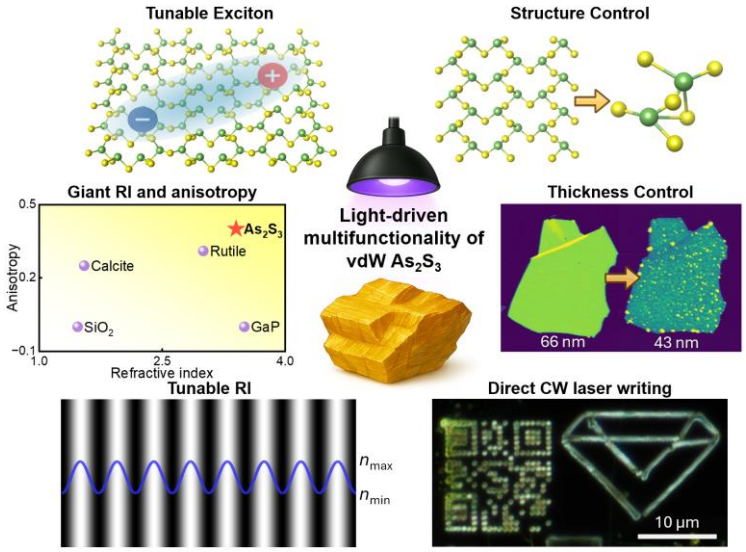

Figure 1: Optically induced multifunctional behavior of crystalline van der Waals As_2S_3.
The unique combination of high refractive index, pronounced optical anisotropy, and strong photosensitivity enables diverse light-driven effects in this material. Upon illumination, crystalline As_2S_3 exhibits tunable excitonic resonances (top left), photoinduced structural transitions (top right), and precise thickness modification (center right). Its anisotropic optical properties (center left) and large refractive index shift under light exposure (bottom left) enable efficient modulation of photonic behavior. Moreover, direct laser patterning with a continuous-wave source facilitates submicron-resolution structuring (bottom right).