

Fabrication and structural reconfiguration of nanostructured As-S chalcogenide films via gold nanoparticles and laser processing

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Abstract: Chalcogenide glasses and nanostructured thin films are promising materials for infrared photonics, optical data storage, and reconfigurable devices due to their high optical nonlinearity and pronounced photo-induced structural response [1,2]. Arsenic sulfide (As-S) systems are widely used as a model platform for studying structure-property relationships under external stimuli. However, the mechanisms governing crystallization, light-induced transformations, and mass transport remain incompletely understood [1,3].

Here, we report the fabrication and laser-induced structural reconfiguration of nanostructured As-S films via gold nanoparticle-assisted growth and laser processing, supported by experimental spectroscopy and atomistic modelling. Gold nanoparticle-assisted chemical vapor deposition on Si substrates leads to selective growth of dendrite-like As-S microcrystallites, in contrast to conventional deposition methods. SEM, XRD, EDX, and Raman spectroscopy confirm their crystalline nature [1]. Depending on synthesis conditions, the fabricated structures are composed of either As₄S₅ or As₄S₄ cage-like molecular units, as confirmed by Raman spectroscopy and DFT calculations. Laser illumination of crystallites consisting of As₄S₄ cages induces structural rearrangements and light-induced transformations, whereas uzonite-like structures built from As₄S₅ molecules are found to be resistant to light-induced degradation. In parallel, *in situ* prepared As₄₀S₆₀ and As₄₅S₅₅ nanolayers were studied under super-bandgap laser irradiation. Synchrotron photoelectron spectroscopy reveals reversible compositional changes during irradiation-annealing cycles, including arsenic surface enrichment. These effect is fully reversible upon thermal annealing [3]. Molecular modelling and DFT calculations suggest that the observed behaviour is governed by realgar-pararealgar-like transitions involving bond breaking, atomic rearrangement, and anisotropic diffusion of As-rich clusters.

Overall, the results demonstrate two complementary routes for tailoring As-S photonic media: nanoparticle-assisted crystallization of metastable phases and reversible laser-driven structural reconfiguration. These findings provide new insight into nanoscale structural dynamics in chalcogenide systems and enable routes toward adaptive infrared photonic materials.

Keywords: As-S chalcogenide, gold NPs, laser induced transformations, Raman spectroscopy, DFT calculations

References

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