

## Structural and Optical Analysis of Cu–Sb–Chalcogenides with Insights into Laser-Triggered Phase Behaviour

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**Abstract:** This study provides a comprehensive investigation into the synthesis and characterization of Cu–Sb–chalcogenide thin films, namely Cu<sub>3</sub>SbS<sub>4</sub>, Cu<sub>3</sub>SbSe<sub>4</sub>, and CuSbTe<sub>2</sub>. The materials were fabricated using a two-step process: deposition of metallic precursor layers, followed by controlled chalcogenation to form the desired compounds. Detailed structural, optical, and microstructural analyses were performed using grazing-incidence X-ray diffraction, Raman spectroscopy, UV–VIS–NIR spectroscopy, scanning and transmission electron microscopy, and spectroscopic ellipsometry. These techniques confirmed phase formation, crystallinity, and uniform film morphology. The laser-induced phase transition behaviour of Cu<sub>3</sub>SbS<sub>4</sub> and Cu<sub>3</sub>SbSe<sub>4</sub> was systematically examined to assess their suitability for optical switching applications. Thin films were exposed to continuous-wave 532 nm laser irradiation at varying power levels, and structural changes were tracked through Raman spectroscopy. Complementary COMSOL Multiphysics simulations were conducted to estimate surface temperature distributions during laser exposure by solving the heat diffusion equation. Power-dependent and time-resolved Raman measurements on Cu<sub>3</sub>SbS<sub>4</sub> enabled analysis of phase transformation kinetics. The activation energy of the laser-driven transition was determined using Arrhenius analysis of the temporal evolution of Raman features. Overall, the findings advance understanding of the photo-induced phase behaviour of Cu–Sb–chalcogenides and highlight their potential for tunable optical and phase-change applications.

**Keywords:** Thin-Film Synthesis; Phase Transition; Synthesis and Characterization; Chalcogenation Process; Chalcogenide-Based Semiconductors; Renewable-Energy Technologies.

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### 1. Introduction

Chalcogenide-based semiconductors composed of copper (Cu), antimony (Sb), and chalcogen elements—sulfur (S), selenium (Se), and tellurium (Te)—have emerged as a vital class of materials in modern electronics, optoelectronics, and renewable-energy technologies [11]. Their widespread interest stems mainly from the abundance of their constituent elements, their eco-friendly chemical nature, and the wide range of functionalities they offer. As global research efforts intensify to identify low-

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cost, high-performance alternatives to conventional semiconductor materials, Cu–Sb–chalcogenides are gaining prominence for their favourable optical absorption, tunable band gaps, and inherent p-type conductivity [12]. These attributes make them competitive candidates for photovoltaic absorbers, thermoelectric converters, photodetectors, and phase-change memory devices. Despite their potential, many Cu–Sb–chalcogenide phases remain insufficiently explored, particularly in thin-film form, where structural stability, phase purity, and defect control significantly influence device performance [13]. The Cu–Sb–chalcogenide family encompasses a range of phases with distinct compositions and crystal structures. Among them, compounds such as  $\text{CuSbS}_2$ ,  $\text{Cu}_3\text{SbS}_3$ ,  $\text{Cu}_3\text{SbS}_4$ ,  $\text{CuSbSe}_2$ ,  $\text{Cu}_3\text{SbSe}_4$ , and  $\text{CuSbTe}_2$  exhibit unique optical and electrical transport properties that open opportunities in diverse applications. While phases like  $\text{CuSbS}_2$  and  $\text{CuSbSe}_2$  have been investigated for solar-energy harvesting, others, including  $\text{Cu}_3\text{SbS}_4$ ,  $\text{Cu}_3\text{SbSe}_4$ , and  $\text{CuSbTe}_2$ , have attracted attention only recently due to emerging theoretical predictions and promising experimental observations [14]. These lesser-studied materials demonstrate excellent absorption coefficients, relatively narrow to intermediate band gaps, and low lattice thermal conductivity—features essential for high-efficiency thermoelectric and optoelectronic devices.

Nevertheless, their synthesis often presents challenges, particularly the formation of competing secondary phases and compositional deviations, which can compromise their performance. Achieving phase-pure thin films, therefore, remains a crucial scientific goal [15]. A variety of deposition and growth techniques—including spray pyrolysis, electrodeposition, thermal evaporation, spin coating, chemical bath deposition, and solvothermal routes—have been employed to synthesize Cu–Sb–chalcogenide materials. Each method offers unique benefits, such as scalability, cost efficiency, or precise control of film thickness. At the same time, several limitations persist, including non-uniform film morphologies and inadvertent formation of impurity phases. For this reason, multi-step or hybrid growth strategies have gained acceptance, particularly those involving the deposition of metallic precursors followed by controlled chalcogenation. Two-stage growth approaches are especially effective because they allow independent control over precursor stoichiometry and chalcogen incorporation, enabling improved crystallinity, enhanced phase purity, and better-tuned optical properties. Beyond conventional synthesis, the dynamic behaviour of these materials under external stimuli—particularly light—has opened another promising research dimension. Several chalcogenides undergo structural transformations upon exposure to laser irradiation, making them suitable for memory devices, programmable optics, and thermal sensing technologies. Laser-induced phase transitions can occur within microseconds or faster, allowing rapid switching between different structural states. Such transformations often proceed through local heating, bond rearrangements, and defect reorganisation, all of which can be monitored in real time using Raman spectroscopy. Raman-based approaches not only reveal the vibrational signature of a material but also provide insights into the kinetics and energetics of phase evolution.

Though many binary and ternary chalcogenides have been studied under laser irradiation, systematic investigations on Cu–Sb–chalcogenides—especially the least explored phases—remain sparse. This research aims to address these gaps by focusing on three significant but understudied Cu–Sb–chalcogenide materials:  $\text{Cu}_3\text{SbS}_4$ ,  $\text{Cu}_3\text{SbSe}_4$ , and  $\text{CuSbTe}_2$ . These compounds possess structural and optical characteristics that are promising for photovoltaic and thermoelectric applications, yet comprehensive experimental studies are limited. For instance,  $\text{Cu}_3\text{SbS}_4$  and  $\text{Cu}_3\text{SbSe}_4$  are known to crystallize in the farnatinite structure, enabling strong light absorption and favourable charge transport. Likewise,  $\text{CuSbTe}_2$  has been theoretically predicted to exhibit exceptionally high absorption coefficients and a suitable bandgap for ultra-thin solar absorbers. Still, its thin-film synthesis and experimental characterisation have been scarcely reported. By synthesizing and analysing these materials in thin-film form, this study seeks to expand the available knowledge base and assess their suitability for next-generation energy devices. This study presents an in-depth exploration of the synthesis and detailed characterization of Cu–Sb–chalcogenide-based materials, specifically  $\text{Cu}_3\text{SbS}_4$ ,  $\text{Cu}_3\text{SbSe}_4$ , and  $\text{CuSbTe}_2$ . These compounds were fabricated using a two-step method in which metallic layers were first deposited onto suitable substrates and subsequently converted into the target chalcogenides through a controlled chalcogenation process. The resulting thin films were examined using a wide range of analytical techniques, including grazing-incidence X-ray diffraction, Raman spectroscopy, UV–VIS–NIR spectroscopy, scanning electron microscopy (SEM), transmission electron microscopy (TEM), and spectroscopic ellipsometry, enabling a comprehensive assessment of their structural, optical, and microstructural properties.

The phase-transition behaviour of  $\text{Cu}_3\text{SbS}_4$  and  $\text{Cu}_3\text{SbSe}_4$  under laser exposure was further investigated to evaluate their suitability for laser-induced switching applications. Thin films of both materials were subjected to continuous-wave 532 nm laser irradiation at varying power levels, and the resulting structural modifications were monitored through Raman spectroscopic analysis. To support these observations, COMSOL-based simulations were performed to estimate the temperature distribution on the film surfaces during irradiation by solving the heat diffusion equation. Additionally, power-dependent, time-resolved Raman measurements were performed on  $\text{Cu}_3\text{SbS}_4$  to characterize the kinetics of its laser-driven phase transformation. By analyzing the temporal evolution of Raman features and applying Arrhenius modeling, the activation energy for the transition was determined. Overall, this work enhances the fundamental understanding of Cu–Sb–chalcogenides, particularly their photo-induced phase behaviour, and highlights their potential for applications requiring tunable optical or structural functionality. An additional objective of this work is to examine how these materials respond to controlled laser irradiation. Understanding laser-induced phase transitions is essential for future applications in optical storage, non-volatile memory, and

reconfigurable photonic components. Using continuous-wave lasers and Raman spectroscopy, researchers investigate structural changes in  $\text{Cu}_3\text{SbS}_4$  and  $\text{Cu}_3\text{SbSe}_4$  thin films under varying irradiation conditions. Complementary numerical modelling, based on heat-diffusion simulations, provides estimates of temperature evolution during exposure, helping to correlate experimental observations with physical mechanisms. Time-resolved Raman measurements further allow the extraction of kinetic parameters, including activation energy, which is crucial for describing the rate and nature of the phase transitions.

## 2. Literature Review

Extensive research over the past decade has highlighted the technological relevance of multinary chalcogenide semiconductors, particularly copper-based systems incorporating antimony and chalcogen elements. Cu–Sb–chalcogenides have been explored as alternatives to conventional absorbers and phase-change materials because they combine earth-abundant constituents with favourable optical absorption, moderate band gaps, and intrinsically low thermal conductivity [1]. Early studies primarily focused on binary copper chalcogenides and ternary Cu–Sb–S systems such as  $\text{CuSbS}_2$ , motivated by their strong light absorption and compatibility with low-cost deposition techniques. These investigations established that antimony incorporation plays a critical role in stabilizing the crystal structure and tuning electronic properties [2]. Subsequent work focused on higher-order phases, such as  $\text{Cu}_3\text{SbS}_4$  and  $\text{Cu}_3\text{SbSe}_4$ , which crystallize in the famatinite structure. Researchers reported that these compounds exhibit absorption coefficients exceeding  $10^4$ – $10^5$   $\text{cm}^{-1}$  in the visible region, making them suitable for thin absorber layers. Experimental studies using sputtering, thermal evaporation, and solution-based routes demonstrated that post-deposition chalcogenation significantly improves crystallinity and phase purity. However, several reports also highlighted persistent challenges, including sulfur or selenium deficiency, formation of Cu-rich secondary phases, and grain-boundary-induced recombination losses [3]. Parallel theoretical investigations predicted that  $\text{Cu}_3\text{SbS}_4$  and  $\text{Cu}_3\text{SbSe}_4$  possess direct or quasi-direct band gaps and favourable effective masses for charge transport. Density functional theory calculations indicated that antisite defects and vacancies dominate the defect chemistry, influencing both the conductivity type and the carrier concentration. These predictions motivated experimental efforts to control stoichiometry through precursor design and annealing conditions. Despite this progress, systematic experimental validation of defect-driven phase stability in thin films remains limited [4].

Compared to sulfide and selenide systems, Cu–Sb–Te compounds such as  $\text{CuSbTe}_2$  have received relatively little experimental attention. Available studies largely focus on bulk or nanocrystalline forms, in which high optical absorption and narrow band gaps have been reported. Thin-film investigations are scarce, mainly due to difficulties in incorporating tellurium and controlling its volatility during growth. Nevertheless, recent reports indicate that telluride phases may offer superior absorption in ultra-thin configurations, positioning them as promising candidates for next-generation optoelectronic and thermoelectric devices [5]. Another rapidly developing research direction concerns the response of chalcogenide materials to external stimuli, especially laser irradiation. Laser-induced phase transitions have been widely studied in classical phase-change materials, but their exploration in Cu–Sb–chalcogenides is still emerging. Previous studies on related copper chalcogenides have shown that localised laser heating can induce compositional redistribution, defect migration, and phase segregation. Raman spectroscopy has been identified as a particularly effective tool for tracking these transformations due to its sensitivity to local bonding environments [6]. Only a limited number of works have addressed laser-triggered transformations in ternary Cu–Sb–chalcogenides. Existing reports suggest that these materials undergo sequential decomposition pathways rather than abrupt amorphous–crystalline switching. However, detailed kinetic analysis, spatial mapping of transformed regions, and correlation with thermal modelling have rarely been combined in a single study. As a result, the fundamental mechanisms governing photo-induced phase evolution in these systems are not yet fully understood [7]. Overall, the literature reveals clear opportunities for further research: comprehensive thin-film studies of underexplored Cu–Sb–chalcogenide phases, a deeper understanding of laser-induced structural dynamics, and integration of experimental observations with numerical modelling. The present manuscript addresses these gaps by providing a unified experimental and analytical framework for investigating synthesis, characterisation, and laser-driven phase behaviour [8].

### 2.1. Probing the Dynamics of Laser-Induced Phase Transformations in $\text{Cu}_3\text{SbS}_4$ Thin Films

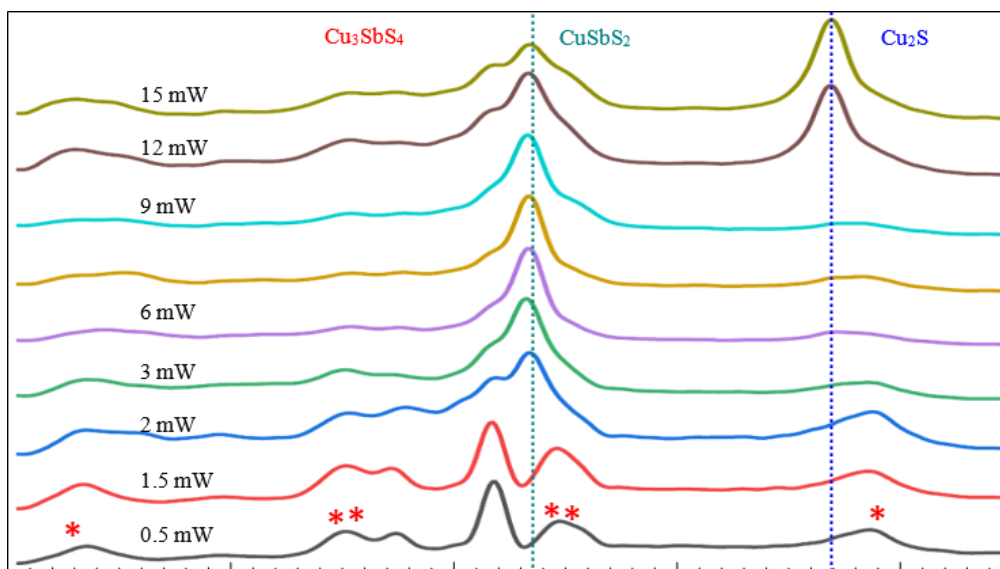
Copper–antimony–sulfide (CAS) compounds exhibit rich structural behaviour in response to external stimuli, such as temperature, pressure, or light. Among them,  $\text{Cu}_3\text{SbS}_4$  has recently attracted attention for its tunable optical and electrical properties and its ability to undergo structural transitions under controlled laser irradiation. Such transformations are of particular interest because they offer the opportunity to tailor material properties at micro- and nanoscale dimensions, enabling applications in optoelectronics, photonic devices, and phase-change memory elements [9]. Laser-driven phase transitions occur when incident photons deliver enough energy to heat the material above a transition threshold locally. Under these conditions, atoms may reorganize within the lattice, leading to the formation of new phases with different symmetries, vibrational modes, and electronic properties. Probing these dynamic structural changes requires sensitive characterization tools, among which Raman spectroscopy is especially powerful. Raman scattering directly reflects a crystal's vibrational properties, enabling real-time monitoring of phase evolution [10]. This paper presents a detailed examination of the structural transformations in  $\text{Cu}_3\text{SbS}_4$

thin films upon laser exposure. By combining power-dependent Raman measurements, time-resolved spectroscopy, Raman mapping, surface and compositional analysis, and TEM investigations, researchers elucidate the sequential transition pathway.

### 3. Materials and Experimental Methods

#### 3.1. Film Preparation

$\text{Cu}_3\text{SbS}_4$  thin films were synthesized using a two-step growth approach. In the first step, metallic Cu and Sb layers were deposited onto c-plane sapphire substrates using RF magnetron co-sputtering. The Cu/Sb ratio was optimized to approximately 1:1 to ensure the correct precursor stoichiometry (Figure 1).



**Figure 1:** XRD patterns of Cu–Sb samples at different milling times showing the formation of  $\text{Cu}_2\text{Sb}$  and  $\text{CuSb}$  phases

These precursor films were then sealed in a vacuum-tight quartz ampoule together with elemental sulfur. In the second step, the ampoule was placed inside a tubular furnace. The temperature was ramped to  $425^\circ\text{C}$  at  $10^\circ\text{C min}^{-1}$  and maintained for one hour to allow sulfur to react with the metal films. The system was subsequently cooled to room temperature under ambient conditions. This controlled sulfurization ensured the formation of phase-pure  $\text{Cu}_3\text{SbS}_4$  films.

#### 3.2. Laser Irradiation Procedure

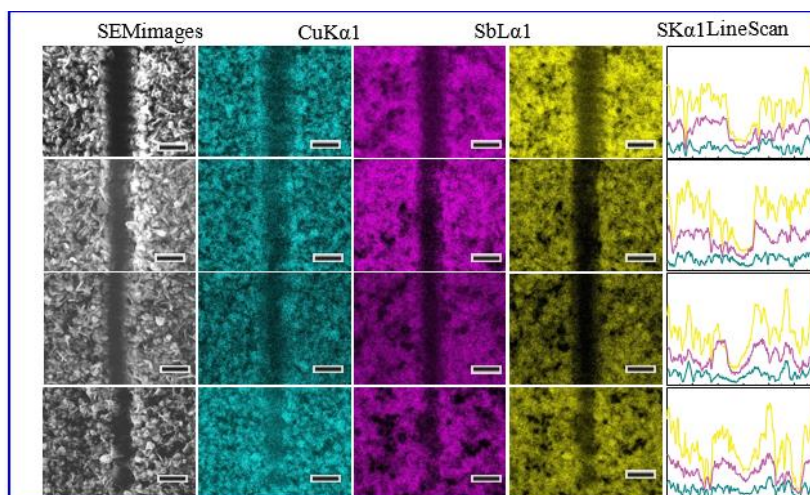
Laser-induced transformations were studied using a 532 nm continuous-wave Nd: YAG laser integrated into a confocal Raman setup. The laser beam, focused to a  $\sim 1\ \mu\text{m}$  spot, was scanned on the sample surface using a precision piezoelectric stage. Incident laser powers ranged from 0.5 to 15 mW, allowing the systematic observation of structural changes as a function of energy input.

#### 3.3. Raman Spectroscopy

Raman spectra were collected in a backscattering geometry using a WITec alpha-300R system equipped with an 1800 lines/mm grating. Time-resolved Raman studies involved acquiring sequences of spectral frames at fixed intervals to track phase evolution. Additional Raman mapping experiments were conducted to visualize spatial variations in phase composition following laser exposure.

#### 3.4. Microscopy and Elemental Analysis

Surface morphology was imaged using field-emission scanning electron microscopy (FE-SEM). Energy-dispersive X-ray (EDX) spectroscopy provided elemental distribution maps in laser-modified regions. High-resolution transmission electron microscopy (HRTEM) and selected-area electron diffraction (SAED) were employed to reveal nanoscale structural changes in irradiated flakes (Figure 2).



**Figure 2:** Laser power–dependent Raman analysis

Raman spectra were first collected at increasing laser powers to examine how  $\text{Cu}_3\text{SbS}_4$  responds to rising thermal input. At low powers ( $\leq 1$  mW), the spectrum displayed the characteristic vibrational modes of tetragonal  $\text{Cu}_3\text{SbS}_4$ , dominated by the intense  $A_1$  mode near  $319\text{ cm}^{-1}$ . Additional external vibrational modes were observed near  $135$ ,  $247$ ,  $276$ ,  $347$ ,  $358$ , and  $490\text{ cm}^{-1}$ , all consistent with the spectra of phase-pure  $\text{Cu}_3\text{SbS}_4$ .

### 3.5. Transition to $\text{CuSbS}_2$

When the laser power exceeded  $1.5$  mW, a new Raman feature emerged at  $333\text{ cm}^{-1}$ , signalling the formation of  $\text{CuSbS}_2$ , an orthorhombic phase. This peak originates from symmetric Sb–S stretching vibrations and is the most intense mode of  $\text{CuSbS}_2$ . Its appearance indicated that the laser energy had raised the local temperature enough to induce atomic reorganisation within the film.

### 3.6. Transition to $\text{Cu}_2\text{S}$

At powers above  $12$  mW, a clear Raman peak appeared near  $472\text{ cm}^{-1}$ , corresponding to cubic  $\text{Cu}_2\text{S}$ . The presence of  $\text{Cu}_2\text{S}$  indicates substantial decomposition driven by extensive heating. Under these high-energy conditions, the  $\text{CuSbS}_2$  phase further transforms, demonstrating that the film undergoes a two-step phase cascade under intense optical excitation.

### 3.7. Summary of Observed Phase Pathway

The laser-induced evolution can be summarized as:

- $\leq 1$  mW:  $\text{Cu}_3\text{SbS}_4$  stable
- $1.5$ – $9$  mW: Formation and Stabilization of  $\text{CuSbS}_2$
- $\geq 12$  mW: Decomposition into  $\text{Cu}_2\text{S}$

This clearly illustrates that laser power dictates the film's structural identity. Raman Mapping of Laser-Transformed Regions. To visualize the spatial distribution of phases after exposure, 2D Raman mapping was performed on areas irradiated with  $12$  mW. Three spectral windows were used:

- $309$ – $329\text{ cm}^{-1}$  →  $\text{Cu}_3\text{SbS}_4$
- $323$ – $343\text{ cm}^{-1}$  →  $\text{CuSbS}_2$
- $462$ – $482\text{ cm}^{-1}$  →  $\text{Cu}_2\text{S}$

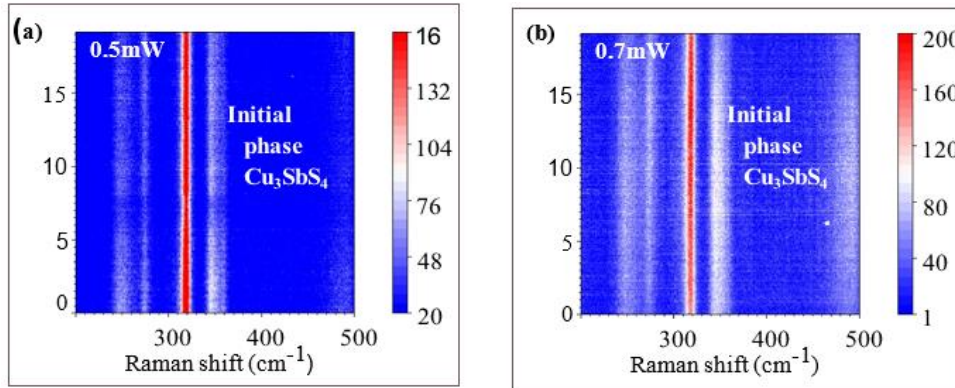
By assigning red, green, and blue colours to these windows, respectively, a composite RGB map was created.

### 3.8. Spatial Distribution of Phases

The map revealed a distinct concentric pattern:

- **Center (Blue):**  $\text{Cu}_2\text{S}$  dominates at the highest temperatures.
- **Intermediate Annulus (Green):**  $\text{CuSbS}_2$  forms in moderately heated regions.
- **Outer Rim (Red):** Residual  $\text{Cu}_3\text{SbS}_4$  persists where heating is minimal.

This distribution corresponds well with the Gaussian power profile of the laser beam. The transformation zone extended roughly  $1\ \mu\text{m}$  from the beam center (Figure 3).



**Figure 3:** Surface morphology and elemental redistribution (a) & (b)

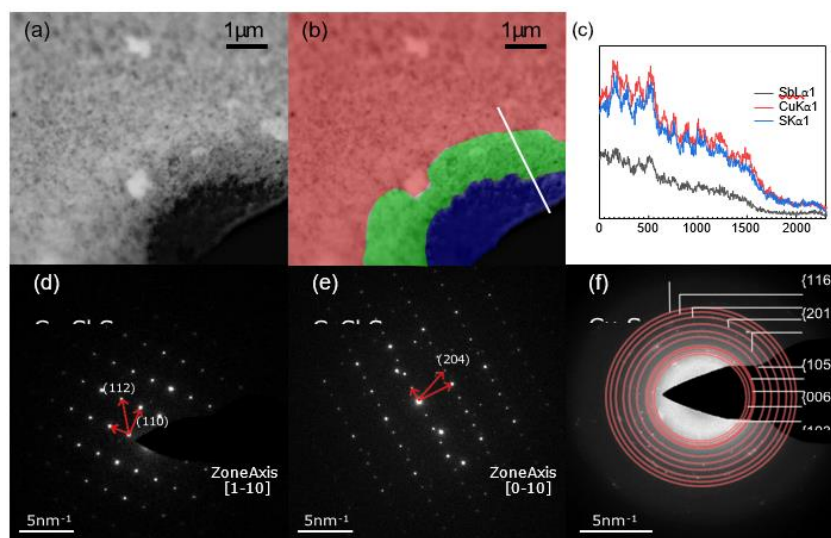
SEM images of laser-scanned tracks indicated that the diameter of the irradiated zone increased with laser power. The morphological changes suggested material loss due to thermal evaporation. EDX mapping confirmed notable elemental redistribution:

- Sulfur loss occurred first, consistent with its higher volatility.
- Antimony loss followed at higher power.
- Copper remained relatively stable but still showed depletion at extreme powers.

Line-scan EDX profiles revealed clear compositional dips at the center of laser tracks, indicating localized evaporation and rearrangement. This aligns with the formation of  $\text{Cu}_2\text{S}$  in highly irradiated regions.

### 3.9. Stability at Low Power

At 0.5–0.7 mW, the  $\text{Cu}_3\text{SbS}_4$  peak remained unchanged throughout the measurement, demonstrating long-term stability at low thermal loads (Figure 4).



**Figure 4:** TEM analysis of laser-transformed regions (a) to (f)

To reveal nanoscale structural details, laser-irradiated flakes were examined via TEM.

### 3.10. Phase Segregation Across Beam Profile

Low-resolution TEM images displayed three distinct regions:

- **Central Zone:** Dominated by small grains and polycrystalline rings →  $\text{Cu}_2\text{S}$
- **Intermediate Zone:** Mixture of  $\text{Cu}_3\text{SbS}_4$ ,  $\text{CuSbS}_2$ , and  $\text{Cu}_2\text{S}$
- **Peripheral Zone:** Mostly intact  $\text{Cu}_3\text{SbS}_4$  lattice

These spatial variations directly correspond to the thermal gradient produced by the laser.

### 3.11. Electron Diffraction Signatures

SAED patterns further confirmed:

- Clean, well-defined spots in outer regions → tetragonal  $\text{Cu}_3\text{SbS}_4$
- Mixed diffraction rings in the middle region → coexistence of multiple phases
- Uniform diffraction rings in the center → polycrystalline  $\text{Cu}_2\text{S}$

EDX line-scan analysis again indicated material loss and stoichiometric shifts consistent with Raman results. This comprehensive investigation demonstrates how  $\text{Cu}_3\text{SbS}_4$  thin films respond to controlled laser irradiation, revealing a clear, sequential transformation pathway.

## 4. Conclusion

This work presents a comprehensive investigation into the synthesis, structural evolution, and optical behaviour of Cu–Sb–chalcogenide thin films, with particular focus on  $\text{Cu}_3\text{SbS}_4$ ,  $\text{Cu}_3\text{SbSe}_4$ , and  $\text{CuSbTe}_2$ . Through a carefully controlled two-stage fabrication route involving metal co-sputtering followed by chalcogenation, high-quality and phase-pure thin films were successfully obtained. Structural analyses confirmed the formation of tetragonal  $\text{Cu}_3\text{SbS}_4$  and  $\text{Cu}_3\text{SbSe}_4$ , as well as hexagonal  $\text{CuSbTe}_2$ , each exhibiting strong visible-light absorption and band gaps within the desirable range for photovoltaic applications. These findings highlight the suitability of the synthesized compounds as promising absorber materials for future optoelectronic devices. A significant part of this work focused on understanding how these materials respond to laser irradiation. Using a 532 nm continuous-wave laser,  $\text{Cu}_3\text{SbS}_4$  and  $\text{Cu}_3\text{SbSe}_4$  films were subjected to controlled optical excitation, enabling real-time monitoring of their phase behaviour.

Raman spectroscopy played a crucial role in revealing the sequential nature of the transformations— $\text{Cu}_3\text{SbS}_4$  transitioning to  $\text{CuSbS}_2$  and eventually to  $\text{Cu}_2\text{S}$ , and  $\text{Cu}_3\text{SbSe}_4$  converting to  $\text{CuSbSe}_2$  before decomposing into  $\text{CuSe}$  at higher powers. Time-resolved Raman measurements further provided insights into the kinetics of these transitions, enabling the extraction of rate constants and the activation energy for the  $\text{Cu}_3\text{SbS}_4 \rightarrow \text{CuSbS}_2$  transformation. Complementary TEM and EDX analyses validated the structural and compositional changes at the microscale during laser exposure. Overall, this study deepens the understanding of chalcogenide thin-film behaviour under both thermal and optical stimuli. The ability to precisely control phase transitions via laser irradiation opens new avenues for engineering lateral heterostructures, designing optically tunable components, and exploring device concepts that leverage localised phase modification. The outcomes of this work contribute valuable knowledge to the fields of semiconductor materials, phase-change phenomena, and optoelectronic device engineering, while also laying a solid foundation for future investigations into the Cu–Sb–Te system and laser-patterned functional materials.

### 4.1. Future Work and Research Directions

Future research can build upon the outcomes of this study in several promising directions. One immediate extension is to expand laser-irradiation experiments to include different wavelengths, pulse durations, and repetition rates. Such studies would clarify the relative roles of thermal and non-thermal effects in driving phase transitions and may reveal faster or more reversible switching mechanisms. Another important avenue is the integration of electrical and optoelectronic measurements with in-situ laser studies. Monitoring changes in conductivity, carrier concentration, or photoresponse during phase evolution would provide direct links between structural transformation and device-relevant properties. This approach would be particularly valuable for assessing the feasibility of Cu–Sb–chalcogenide materials for memory or sensing applications. Further work should also explore compositional engineering through controlled doping or alloying. Introducing small amounts of additional elements could stabilize desired phases, suppress decomposition, or lower activation barriers for switching. From a modelling perspective, coupling thermal simulations with phase-field or kinetic Monte Carlo methods could yield deeper insight into nucleation and

growth mechanisms under localized heating. Finally, translating laser-induced phase control into scalable device architectures represents a long-term objective. Demonstrating patterned heterostructures, rewritable optical elements, or prototype memory cells would significantly enhance the technological relevance of Cu–Sb–chalcogenide materials. The present study provides a strong platform for pursuing these future research directions.

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**Data Availability Statement:** The data supporting the findings of this study, including datasets related to the structural and optical investigation of Cu–Sb–chalcogenide materials and their laser-induced phase behaviour, are available from the corresponding author upon reasonable request.

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