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Publication date: 2015

Document Version
Publisher's PDF, also known as Version of record

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Citation (APA):

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Chalcogenide compounds made by pulsed laser deposition at 355 and 248 nm

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Summary

- Thin films made by pulsed laser deposition may differ depending on the laser wavelength. We compared ZnS, Cu₂SnS₃ and a target enriched with SnS relative to Cu₂SnS₃ using 355 nm and 248 nm lasers.
- Cu₂SnS₃ deposition gives a high density of droplets at 355 nm. The higher UV 248 nm laser was expected to reduce the droplets but did not.
- The SnS enriched Cu-Sn-S films had different morphology and post-annealing composition using the two lasers.

Deposition rate

<table>
<thead>
<tr>
<th>Laser</th>
<th>355 nm</th>
<th>248 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laser type</td>
<td>Nd:YAG</td>
<td>Kr excimer</td>
</tr>
<tr>
<td>Pulse length</td>
<td>5-7 ns</td>
<td>~20 ns</td>
</tr>
<tr>
<td>Spot size</td>
<td>2.5 mm²</td>
<td>2.2 mm²</td>
</tr>
<tr>
<td>Fluence</td>
<td>0.4-2.3 J/cm²</td>
<td>0.4-2.2 J/cm²</td>
</tr>
<tr>
<td>Target-substrate dist</td>
<td>4-4.5 cm</td>
<td>4 cm</td>
</tr>
<tr>
<td>Pulse repetition rate</td>
<td>10 Hz</td>
<td>10 Hz</td>
</tr>
</tbody>
</table>

The bandgap of ZnS (3.5-3.6 eV) exceeds the 3.49 eV photon energy of the 355 nm laser, so as expected, ZnS deposition is faster using the 248 nm laser. (*)

Deposition of SnS-enriched Cu₂SnS₃ is faster than deposition of stoichiometric Cu₂SnS₃. The deposition is slightly faster at room temperature than at 150-300 °C.

Morphology

**As deposited**

Films are covered in μm-size droplets from target.

The amount and size of droplets does not change significantly with laser wavelength.

The morphology of the SnS-enriched films (bottom) was quite different with the 355 nm versus the 248 nm laser. However, the stoichiometry was similar as seen on the right (under annealing).

Depositions were done at 25-25 °C.
Substrates were Mo-coated soda lime glass.
Top images: SEM in-lens detector
All other images secondary electron detector

**After annealing with S**

Annealed films of Cu₂SnS₃ by the two lasers had similar composition and appearance.

In contrast, films of SnS-enriched Cu₂SnS₃ by the two lasers were highly distinct. The as-deposited films had a similar atomic composition, but the annealed films differed both in composition and appearance.

All films except the SnS-enriched film deposited at 355 nm contain bubbles; large burst bubbles were seen in the Cu₂SnS₃ film deposited at 355 nm.

Annealed films contain a mix of SnS (brighter in SEM images) and Cu₂SnS₃ (darker).
Composition was measured by Energy Dispersive X-Ray Spectroscopy at 15 kV.

*) similar data on 205 nm deposition previously shown in Ettlinger et al., 2015, App. Surf. Sci., Vol 336, pp. 385-390

This work has been supported by a grant from the Danish Council of Strategic Research