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Material transfer in Pulsed Laser Deposition of the solar cell materials Cu_2SnS_3 and Cu_2ZnSnS_4.

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Earth-abundant non-toxic materials Cu_2SnS_3 (CTS) and Cu_2ZnSnS_4 (CZTS) are very appealing for cheap, large-scale production of thin-film solar cells. The cells have recently reached an efficiency of 9.4 % and may have to potential to reach to reach 15%, which will be interesting goal for solar cell manufacture if the price per harvested kWh is sufficiently low. Due to their complex stoichiometry it is very difficult to obtain a single phase material with evaporation or sputtering deposition. Here we discuss the fabrication of thin films of CTS and CZTS made with Pulsed Laser Deposition (248 nm, KrF laser) from sintered targets. Usually, e.g. in PLD of oxide materials, the loss of the most volatile component, i.e. oxygen, can be compensated with the introduction of reactive background gas in the chamber. In our case, however, the least volatile element, copper, is not transferred as expected. The stoichiometry of the deposited films strongly depends on the laser fluence: severe deficit (up to complete lack) of copper in the film is observed when the fluence is below 0.4 J/cm^2, while a copper rich composition is observed at fluence values above 1 J/cm^2. The stoichiometry of the deposited films can be understood in terms of the cohesive energy of the atoms composing the target; a cohesive energy which is very different among copper, zinc, tin and sulfur. A narrow range of fluence values that preserves the target stoichiometry is found. Furthermore, this method allows fabrication of very homogeneous, amorphous thin films of the Zn-Sn-S “alloy”, which is an immiscible ternary system that has no stable solid phase and thus cannot be made with standard film growth techniques.