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TIME-RESOLVED SPECTROSCOPY AND IMAGING DIAGNOSTICS OF LASER-PRODUCED PLASMA FROM A CHALCOGENIDE SAMPLE

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Abstract. This paper presents spectroscopic data obtained for the laser ablated plume of a chalcogenide sample during thin films production. The major species including atoms and ions of copper, indium and selenium within the plume have been identified. Based on the results of spectroscopic diagnostics the character of the plasma evolution and characteristics of deposited material were analyzed at various initial conditions of plasma creation.

1. INTRODUCTION

Presently, in a search of new and alternative energy sources, solar cells (SC) have received a great interest as the most suitable choice for creation of clean and effective energy sources. One of the factors constraining a wide application of solar cells is an absence of inexpensive and high efficient materials suitable for industrial introduction. The important role in the decision of these problems will play semiconductor nanomaterials among which triple semiconductor compounds like chalcogenides cause a particular interest, as the most perspective for use in photogalvanic devices. For the multicomponent chalcogenide materials, a preservation of stoichiometry and structure is a challenge; researches of this direction in literature are small in numbers (Acquaviva et al. 2003, Chuchman et al. 2009). But an application of such materials is extraordinary various: solar cells, laser diodes, different detectors and transformers of energy, nonlinear crystalline structures and optical filters. Structures with sizes in a nanometer range can represent a particular interest, due to the expected new properties because of surface and quantum-sized effects. In this connection a control of processes of formation and evolution of laser ablation plume from the chalcogenide samples are extremely important. The physical properties of plasma plume, such as species concentrations and temperature, directly affect the properties of the material being formed.

The distinctive features of laser ablation plasma is a complex spatio-temporal structure and a wide range of varying of plasma parameters during the plasma existence time. In a study of such plasmas one has to combine of a short time scale (ns), strong plasma density gradients and the low density of the active species. Spectroscopic methods give especially valuable information for the initial stages of formation and evolution of laser plasma plume, when the use of other methods is impossible (Burakov et al. 2008).

In the present paper time resolved spectroscopic studies of the laser produced plasma formed near the chalcogenide sample has been performed. Based on the results of spectroscopic diagnostics the character of the evolution of plasma parameters and characteristics of deposited material were analyzed.

2. EXPERIMENTAL

Experiments were made by using a combination of two laser beams generated by pulsed Nd:YAG lasers (LOTIS TII, model LS2134), operating at 1064 nm and 532 nm, each with 45 mJ pulse in a 5-mm beam. The laser beams were focused on the surface of the chalcogenide ($CuInSe_2$) sample placed in the chamber evacuated to a pressure of less than 10^{-4} Torr. The chalcogenide sample was prepared by compressing a mixture of copper, indium and selenium powders taken in the stoichiometric proportion. The sample surface was put in a horizontal plane and the laser beams were focused vertically and at right angles onto the sample surface. The beams were focused by a lens (f_1 = 200 mm) onto the sample placed on the movable holder. The laser repetition rate was 5 Hz.

The emission characteristics of plasmas produced by the 1064 nm laser pulse employing for ablation both singly and after the 532 nm laser pulse were analyzed. The direction of observation was perpendicular to the direction of propagation of the laser beams.

The detection system based on an imaging spectrograph and a time-gated ICCD was used. The detection system allows to characterize one dimensional plume propagation, providing detailed, species resolved information. The optical observation of the plasma emission was executed by imaging the plasma plume onto the entrance slit of the spectrograph such that the expansion direction lies along the orientation of the entrance slit of the spectrograph. The 50-μm entrance slit selected a region of the plasma emission, normal to the sample's surface. At the output of the spectrograph, in the image plane, a time-gated ICCD camera was installed. A delay generator is employed to delay the time gate of the ICCD with respect to the 532 nm laser pulse. So, at its output the spectrograph produced a one-dimensional spatial and spectral image of the expanding plasma, in which the vertical axis corresponds to expansion direction z and the horizontal axis to the wavelength of the emission of plasma species (Fig. 1).

Time evolution of the spectral line emission was separately observed by using another detection system consisting of monochromator equipped by the fast photomultiplier.

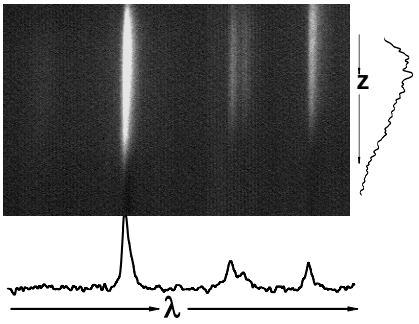

Figure 1: To illustration of the detection of spatial and temporal profiles from the hybrid images of plasma.

3. RESULTS AND DISCUSSION

The typical emission spectrum of the laser-produced plasma of chalcogenide sample is presented in Fig. 2. The most intensive lines in the spectrum correspond to atoms and ions of copper and indium. There are only very weak lines of selenium in the spectrum. As it can be seen from Fig. 2 only ionic line Se II 446,76 nm is unambiguously identified in the visible.

The different stages of the plasma directional expansion from the target surface could be distinguished. In the early stage of expansion a continuous spectrum shows up near the target surface, which can be attributed to bremsstrahlung and photorecombination radiation depending strongly on the density of charged particles in the plume. At the later stage of the plasma evolution individual emission lines could be identified. From temporal profiles of spectral lines obtained at different distances from the target the plasma expansion velocities were estimated. The expansion velocities varied with species, but in each case are typically $\sim 10^4$ ms^{-1} at 10^9 W/cm^2 and decreased with decreasing laser intensity. For example, for copper atoms the velocity is equal 10 and 60 km/s, accordingly for a fast and slow part of a flux

We compared the indicated images and spectra for plasma produced by 1064 nm laser pulse in two different ablation modes: with and without the pre-ablation 532 nm laser pulse (single- and double pulse excitation regimes). The pulse separation in the double pulse mode was fixed at 6.5 μs. The coupling of the second laser beam into the laser-ablated plume caused changes both plume dynamics and intensities of plasma emissions. Specificity of formation and expansion of a laser plume in the double pulse mode allows predicting a possibility of smoothing of a spatial dispersion of a target material during its transportation from the target to the substrate that is important for the optimization of the laser deposition process.

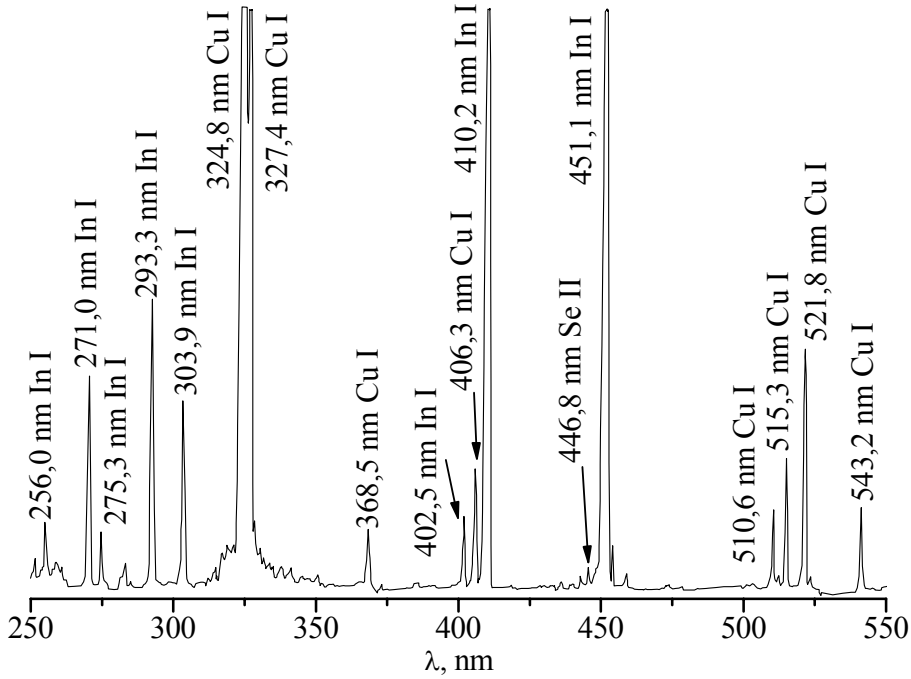

Figure 2: A fragment of the spectrum of laser-produced plasma plume from chalcogenide $CuInSe_2$.

Acknowledgements

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