Incongruent and delayed evaporation of multicomponent materials: Manifestations in laser-ablation plumes

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One of the main advantages of pulsed laser ablation (PLA) in nanomaterial synthesis is the ability to produce thin films and nanoparticles with a composition identical to that of the ablated multicomponent target [1-2] However, such favorable conditions do not always occur and, in many cases, laser-produced nanomaterials are non-stoichiometric. This is mainly due to two reasons, (1) selective (incongruent) ablation and (2) component segregation in the ablation plume. To control the material composition during PLA synthesis, the later effect was extensively investigated [3-4] but the role of incongruent target evaporation remains virtually unexplored and often neglected.

Here we review our mass spectrometric and theoretical studies on incongruent evaporation and its manifestations in the laser ablation plume [5-9]. Particular attention is given to the effect of delayed evaporation which occurs under multi-shot PLA conditions for more volatile components due to diffusion from the bulk to compensate for their deficiency on the surface. For targets with a large difference in the component volatilities, this results in the presence of very slow particles in the plume (Fig. 1). We demonstrate that even a small delay in evaporation affects strongly the plume expansion dynamics [7-8]. Furthermore, we show that core-shell PLA-produced nanoparticles can be formed due to delayed evaporation [9].

Figure 1: Time-of-flight distributions of the main plume particles under nanosecond PLA of InP



References: [1] D. Chrisey and G. Hubler, *Pulsed Laser Deposition of Thin Films* (Wiley, New York 1994); [2] S. Barcikowsky, M. Husted, and B. Chichkov, *Polymery* 53 (2008) 657; [3] C.B. Arnold and M.J. Aziz, *Appl. Phys. A* 69 (1999) S23; [4] J. Schou, *Appl. Surf. Sci.* 255 (2009) 5191; [5] A.V. Bulgakov, A.B. Evtushenko, Y.G. Shukhov, I. Ozerov, and W. Marine, *Quantum Electron.* 40 (2010) 1021; [6] O.A. Bulgakova, N.M. Bulgakova, and V.P. Zhukov, *Appl. Phys. A* 101 (2010) 53; [7] A.A. Morozov, S.V. Starinskiy, and A.V. Bulgakov, *J. Phys. D* 54 (2021) 175203; [8] X. Yao, C.W. Schneider N.M. Bulgakova, A.V. Bulgakov, and T. Lippert, *Appl. Phys. A* 129 (2023) 590. [9] S. Tahir, N. Shkodich, B. Eggert, J. Lill, O. Gatsa, et al., *ChemNanoMat* (2024), in press.