Nanostructured surfaces

Laser-driven surface modification of As₂S₃ nanolayers: *in-situ* synchrotron radiation photoemission, surface-enhanced Raman and DFT study

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Laser and thermal assisted transformations of ultrathin (~10 nm) As_2S_3 chalcogenide nanolayers were investigated *in-situ* using surface sensitive synchrotron radiation photoelectron spectroscopy. Surface transformations and bond rearrangements were observed both in thermally annealed and laser irradiated samples in comparisons with as-deposited nanolayers. The type of changes were found to be dependent on laser photon energy used for irradiation . In contrast with thermal annealing, the irradiation of As_2S_3 chalcogenide nanolayers by laser operating at 403 nm leads to increase the concentration of As at the surface (compositional changes). Simultaneously, the changes of relative contribution of the As and S atoms occupied different chemical states in whole As 3d and S 2p signal were observed. Also, the photo-chemical transformation is found to be reversible in several "laser irradiation – thermal annealing" cycles.

To access the total spectral behavior and to elucidate the structural origin of the chalcogenide surface transformations, the As 3d and S 2p core level spectra were fitted using different As- and S-centered structural units (*s.u.*) and their binding energy positions were calculated using *ab initio* DFT method [1]. Surface-enhanced Raman spectroscopy have been also employed for *ex-situ* characterization and study of molecular level details of the laser induced structural transformations occurred at the surface of As₂S₃ nanolayers.

1. Kondrat O., Holomb R., Popovich N., Mitsa V., Veres M., Csik A., Tsud N., Matolín V., Prince K.C. Local surface structure and structural properties of As–Se nanolayers studied by synchrotron radiation photoelectron spectroscopy and DFT calculations // J. Non-Cryst. Sol. - 2015. -410. - pp. 180–185.