

Mechanisms of laser control of desorption through selective surface excitation

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Irradiation of insulators can induce surface decomposition and emission of particles in different charge, electronic, and kinetic energy states. This irradiation is mostly absorbed inside the solid and only a minor part of it is manifested in desorption. The surface of irradiated crystals undergoes dynamic structural changes caused both by the atom desorption and accumulation of radiation defects. It is clear that to achieve control over radiation-stimulated surface processes one must have significant understanding of the reactive mechanism.

In this paper we discuss the mechanisms of laser control of photo-desorption from alkali halides using frequency selected laser pulses and demonstrate that similar concepts can be applied to studying surface reaction and desorption in other materials, such as MgO. We focus primarily on hyperthermal desorption of halogen (and oxygen) atoms and show that the yield, electronic state, and velocity distributions of desorbed atoms can be controlled. We demonstrate that the observed control is due to preferential excitation of surface excitons. This approach takes advantage of energetic differences between surface and bulk exciton states and probes the surface exciton directly. Application of this approach to controlling the yield and state distributions of desorbed species requires detailed knowledge of the atomic structure, spectroscopic properties, and electronic structure of transient surface exciton species. These studies integrate results of detailed electronic structure calculations and ultrasensitive laser pump-probe experiments within a developing surface exciton desorption model. We demonstrate that this model is extendable to other insulators and discuss the necessary conditions for selective excitation of surface excitons and controlling surface processes of a wide variety of materials.