Time-resolved investigation of laser-induced diffusion by SHG microscopy

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We report the development of an experimental technique to investigate the fluence-dependent dynamics of laser-induced surface processes like desorption or diffusion. In our previous work we have shown that diffusion of adsorbates on vicinal metal surfaces induced by femtosecond laser pulses can be sensitively monitored by optical second harmonic generation (SHG) [1]. The strong non-linear dependence of the diffusion rate on laser fluence made it possible to apply a two-pulse correlation (2PC) technique to study the dynamics of the energy transfer between laser-excited substrate electrons to the adsorbate degrees of freedom in the time-domain on a femtosecond time scale [2]. A non-linear dependence on laser fluence is characteristic of all kinds of laser-induced surface processes. It results in a strong variation of the rate across the laser beam profile, which requires either a correction or a spatially selected detection. For this purpose we have extended our experimental setup by a SHG microscope which makes it possible to observe laser-induced diffusion or desorption for a range of laser fluences simultaneously. We illustrate its operation on the basis of a time-domain study on the diffusion of atomic oxygen on vicinal Pt(111). We observe an increase of the width of the 2PC as a function of laser fluence, a phenomenon that should occur for all substrate mediated laser-induced surface reactions [3]. The results are discussed on the basis of different variants of the electronic friction model.

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