Imaging the femtosecond time scale correlated electron-nuclear dynamics in surface photodesorption

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We employ time-resolved two-photon photoemission to characterize electronic structure and photoinduced dynamics of chemisorbed alkali atoms on noble metal surfaces. Photoinduced charge transfer excitation of the lowest energy sigma resonance of Cs on Cu or Ag surfaces turns on repulsive forces between atom and surface initiating nuclear wave packet motion on a dissociative potential energy surface. Energy, momentum, and time resolved measurements of photoemission from desorbing atoms provide information on the nuclear wave packet motion and the concomitant changes in the surface electronic structure. In particular, we use time-dependent momentum imaging of photoemission from the excited state to explore the correlation between the electron and nuclear motions.