MAR11-2010-001013

Abstract for an Invited Paper
for the MAR11 Meeting of
the American Physical Society

Probing electron correlations by laser-induced tunnel ionization

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Pairwise electron correlation has been intensely studied by projecting two electrons to the continuum simultaneously via a well controlled perturbation, e.g. a collision with an energetic electron, a fast ion or a single XUV photon. Electron correlation studies using multiphoton ionization remain an exception. One reason may be that recollision aside, studies in rare gas atoms have largely suggested that multiphoton multiple ionization in the tunneling limit proceeds sequentially - each successive ionization stage loosing memory of previous electronic correlations. On the other hand, laser tunnel ionization has been known to access multiple electronic states. Recent evidence, corroborating the notion that tunneling can prepare these correlated multielectron states in a coherent superposition, suggests that sequential multiple ionization may provide insight into dynamical correlations in the parent ion. Here, we demonstrate how dynamics of electron correlation can be investigated using laser-induced tunnel ionization by interrogating valence shell electrons in rare gas atoms with intense laser pulses. We find a strong spatial propensity in the sequential double tunnel ionization regime. For instantaneous emission, we find that the two electrons are preferentially emitted in perpendicular directions. Applying laser scanning tunneling microscopy in a pump-probe scheme we directly observe the periodic charge redistribution in the valence shell of singly charged noble gas atoms that was predicted by Santra and coworkers and recently inferred in an attosecond pump-probe experiment using XUV probe pulses. In contrast to single photon ionization, tunneling is highly directional. Here, we exploit that property of tunnel ionization to remove an electron from a rare gas atom along a specific spatial direction. We then probe the correlation by ionizing a second electron via a laser-induced tunneling gate. Since our tunneling gates are optically controlled, the second gate can be opened at any angle and at any time relative to the first. Hence, not only spatial but also temporal variations of the correlation can be probed. We demonstrate the generality of this concept by extending our measurements to a small molecule (HCl).