Interaction of ultrashort intense laser pulses with molecular ion beams

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Laser induced dissociation and ionization of H_2^+ have been experimentally studied using intense (10^{13}-10^{15} W/cm^2) short (8-135 fs) laser pulses at 790 and 395 nm. Both fragments from the dissociation of the vibrationally excited H_2^+ beam are measured in coincidence by a 3-dimensional momentum imaging system event by event.

The kinetic energy release distribution following ionization of H_2^+ by 45 fs, 10^{14}-10^{15} W/cm^2 laser pulses, i.e. in the tunneling regime, was measured to be broad and structureless. Its peak shifts toward higher energies as the laser intensity is increased indicating that ionization shifts to smaller internuclear distances. In contrast, a surprising structure is observed near the ionization threshold, which we call above threshold Coulomb explosion [1], defined as resonant multiphoton ionization of H_2^+ during its dissociation along the bond softening (BS), bond hardening (BH) or above threshold dissociation (ATD) pathways. The angular distributions of the two H^+ fragments are strongly peaked along the laser polarization, and the angular distribution is described well by $[\cos^2\theta]^n$, where n is the number of photons predicted for the resonant multiphoton process near the ionization threshold. Our data indicates that n varies as expected with the laser wavelength.

The dissociation results change dramatically with decreasing pulse width, in contrast to the reported trend for longer pulses [2]. At similar peak intensities, determined by using an intensity-difference spectrum method [3], bond-softening is found to be the main feature in 135 fs pulses, while in 45 fs pulses it is a minor process having a low kinetic energy release and a very narrow angular distribution. Above threshold dissociation is dominant in 45 fs pulses whose durations are approaching the vibrational period of the molecule [4]. The measurements with 135 fs pulses at ~10^{14} W/cm^2 reveal a strong correlation between the angular distribution and the kinetic energy released upon dissociation [5]. This correlation is in contrast to the $\cos^2\theta$ distribution of photo-dissociation in the weak-field limit. Bond softening is aligned along the laser polarization much more than weak-field photo-dissociation. In contrast, bond hardening results in wider angular distributions than photo-dissociation.

Preliminary results with few cycle (8 fs) pulses indicate similar trends as discussed above. Attempts to determine the carrier-envelope phase (CEP) of each pulse are underway in order to measure the predicted dependence of the angular distribution of dissociation to H^++D and H+D^+ on the CEP [6].

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