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Electron Rearrangement Dynamics in Dissociating Multiply Charged Iodine Molecules

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We use an XUV-pump–XUV-probe scheme to access electron rearrangement dynamics in dissociating molecular iodine ions. A first pulse of 87 eV, delivered by the free-electron laser FLASH, multiply ionizes and consequently fragments the iodine molecules (I_2). Depending on its delay with respect to the pump pulse, the identical probe pulse induces electron transfer between the dissociating ions, which results in symmetrically charged fragments. In contrast, for large delays electron transfer is blocked and we observe asymmetrically charged ion pairs. By means of a reaction microscope we record the yield of coincident ion pairs from dissociating I_2^{n+} molecular ions as a function of the time delay. This allows to determine the critical internuclear distances and corresponding time scales up to which electrons transfer is possible for various molecular break-up channels. Our results are in very good agreement with predictions of a classical over-the-barrier model demonstrating its validity in an energy regime relevant for FEL, plasma and chemistry applications.

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