Non-Sequential Two-Photon Double Ionization of He and D_2^+

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One of the most prominent advantages that free-electron lasers (FEL) offer compared to traditional XUV sources is the ultra-high brilliance of the emitted radiation. Due to the high field densities reached, multiphoton processes that could safely be ignored previously in this photon energy region, suddenly become relevant. One example for such a reaction is two-photon double ionization (TPDI) which attracted tremendous theoretical interest during the last few years (see e.g. [1] and references herein). Usually, two reaction mechanisms are distinguished in TPDI. In the sequential case, the photons are absorbed sequentially in time with each photon leading to the emission of one electron, first off the neutral and subsequently from the ionic target. In contrast to that both photons are absorbed simultaneously in the non-sequential mechanism, with both electrons being transferred via a virtual intermediate state directly into the doubly ionized continuum.

Here we report on two benchmark experiments on TPDI of He and D2 performed at the free-electron laser in Hamburg (FLASH). Because the chosen photon energy of 52 eV lies well below the He+ ionization potential of 54.4 eV, the sequential TPDI is energetically not allowed enabling the undisturbed characterization of the non-sequential channel in He. Using a dedicated reaction microscope the momentum distribution of the He^{2+}-ions was measured, which reflects the distribution of the momentum sum of the two emitted electrons. As depicted in Fig. 1 two separated maxima along the polarization direction were observed. The exhibited behaviour clearly differs from that in a previous measurement at a photon energy of 44 eV, where a distribution centred at the origin was obtained [3]. This indicates in agreement with recent theoretical calculations that the energy sharing between the electrons becomes increasingly asymmetric with growing excess energy [2]. While our results qualitatively agree with recent calculations, quantitatively significant deviations exist whose origins will be subject of further investigations.

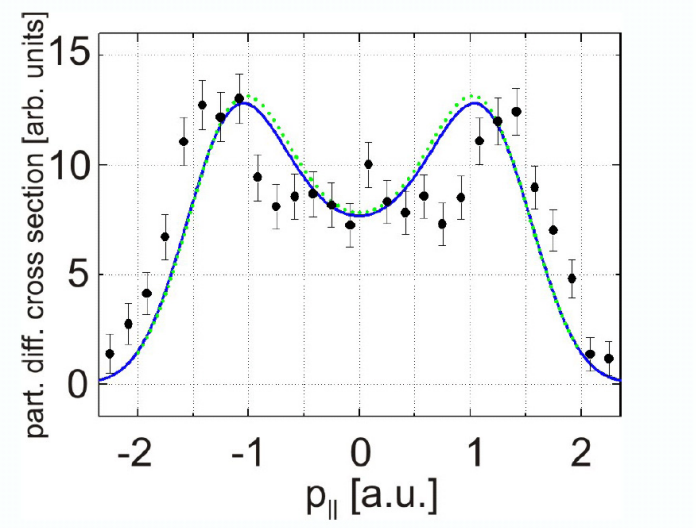

Figure 1: Experimental and theoretical He^{2+} momentum distributions along polarization axis for small momenta in the transverse plane ($|p_{\perp,1/2}| < 0.3$ a.u.). For details see [2].

In the second experiment we investigated the simplest molecular two-electron system H_2/D_2. The left side of Fig. 2 depicts the potential curves of D_2, D_2^+ and D_2^{2+} as a function of the inter-nuclear distance. On the right side of Fig. 2 the distribution of the kinetic energy release (KER), i.e. the sum of the kinetic energies of both coincidently detected D^+-fragments is plotted. The observed two-peak structure can be understood in terms of the underlying dynamics of the different ionization mechanisms. The non-sequential process is occurring only at small inter-nuclear distances since both photons are absorbed simultaneously starting from the ground state of the neutral D_2 molecule. For the sequential channel the situation is more elaborated. After ionization a superposition of states is formed and a nuclear wave packet oscillates along the inter-nuclear axis with a period on the order of the FLASH pulse length (~ 30 fs). The peak at small KERs in the obtained distribution in Fig. 2 can therefore be interpreted as sequential ionization events which occurred close to the outer turning point while the peak at large KERs originates from sequential ionization at the inner turning point in addition to non-sequential ionization. Hence, the KER spectrum encodes the time in between the absorption of the two photons through the internal nuclear dynamics and enables a distinction between sequential and non-sequential mechanism. By comparing our experimental KER-distribution with recent calculations we were able to quantify the relative contribution of the two double ionization channels [4].

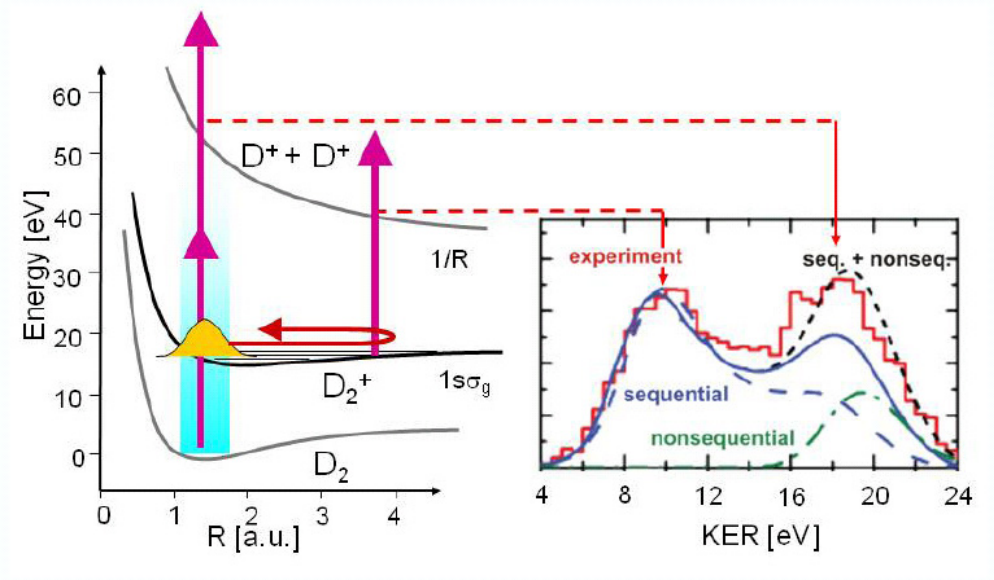

Figure 2: Left side: potential energy surfaces of D_2. Right side: experimental and theoretical KER spectrum for the $D^+ + D^+$ channel [4].

References

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