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## Sequential versus nonsequential two-photon double ionization of the $D_2$ molecule at 38 eV

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**Synopsis** A simple theoretical model is used to interpret recent experimental results for two-photon double ionization (DI) of  $D_2$  at 38 eV. We show that the measured kinetic energy distribution associated with emission of two protons can be interpreted as a sum of two processes: a sequential and an instantaneous absorption of the two incident photons. These processes lead to peaks in different regions of the spectrum.

The motivation of this work is to interpret recent experimental results for two-photon double ionization of  $D_2$  with FEL pulses of 38 eV [1]. We will consider two different processes. In the first process, we assume that ionization is produced by a direct transition from the  $D_2$  ground state to the double ionization continuum. Since the corresponding probability is difficult to evaluate because one has to describe electron correlation in the continuum and at the same time one has to account for the nuclear motion, we have made use of the Franck-Condon approximation. Therefore, for this direct ionization process, the absolute value of the corresponding transition probability will not be determined. In the second process, the two photons are absorbed sequentially so that the first photon absorption leads to  $D_2^+(v)$  in a vibrational state  $v$  associated to the  $1s\sigma_g$  electronic state and the second photon absorption leads to the Coulomb explosion of  $D_2^+(v)$ . Since  $\sim 95\%$  of the one-photon ionization probability leads to  $D_2^+$  in the  $1s\sigma_g$  state, it is reasonable to neglect population of higher electronic states of  $D_2^+$  and the doubly excited states of  $D_2$ . Thus, for a long enough pulse like the one used in the experiment, the total transition probability for this sequential process can be written as a sum of products of two independent probabilities:

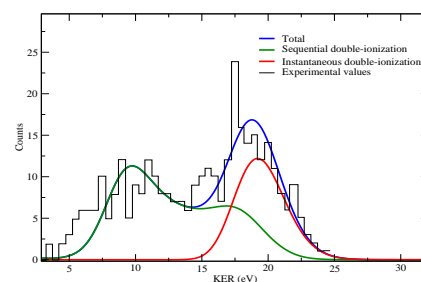
$$P_{seq}^{KER} = \sum_v P_{seq}^{v,KER} = \sum_v P_1^v \times P_2^{v,KER} \quad (1)$$

where

$$P_1^v \propto \left| \int \langle \Psi_{el}^{H_2}(\mathbf{r}, R) | \mathbf{D} | \Psi_{el}^{H_2^+(1s\sigma_g)}(\mathbf{r}, R) \rangle_r \times \chi_{v_i=0}^{H_2}(R) \chi_v^{H_2^+(1s\sigma_g)}(R) dR \right|^2$$

$$P_2^{v,KER} \propto \left| \int \langle \Psi_{el}^{H_2^+(1s\sigma_g)}(\mathbf{r}, R) | \mathbf{D} | \Psi_{el}^{H^++H^+}(\mathbf{r}, R) \rangle_r \times \chi_v^{H_2^+(1s\sigma_g)}(R) \chi_{KER}^{H^++H^+}(R) dR \right|^2$$

The figure shows that the peak at 11eV is entirely due to the sequential ionization process while that at 19 eV is mostly due to the direct ionization process. The KER distribution is very similar to that found in the experiment.



**Fig. 1.** KER spectrum for two-photon double ionization of  $D_2$  for a photon energy of 38eV.

### References

- [1] R. Moshhammer, J. Ullrich and Y. H. Jiang, private communication

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