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Two-photon dissociative ionization of H_2 by ultrashort xuv laser pulses

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Synopsis Ab initio calculations of H_2 photoionization induced by low-intensity ultrashort xuv laser pulses are reported in the region of two-photon absorption. Our computational approach is based on the solution of the time-dependent Schrödinger equation including all electronic and vibrational degrees of freedom. We find that, as one approaches the one-photon ionization threshold, dissociative ionization largely dominates over non dissociative ionization. We show that this behavior is mostly induced by the presence of molecular autoionizing states in this photon energy region.

Recent advances in laser technology allows for the production and manipulation of femto (fs) and even attosecond (as) laser pulses by means of high-order harmonic generation (HHG) within a regime that ranges from vacuum ultraviolet (vuv) to soft x-ray radiation and tunable intensities from 10^9 (perturbative) up to 10^{14} W/cm^2 (non-linear effects). The dynamics of simple many-electron molecules like H_2 subject to laser pulses is still far from being fully understood in many aspects due to the computational difficulty in accounting for i) all degrees of freedom, ii) the electron correlation and iii) the ensuing multichannel scattering problem. Fully correlated calculations on H_2 photoionization studies have also been attempted in the past although using the frozen nuclei approximation (FNA), thus avoiding the crucial role that nuclear motion plays.

The importance of including the nuclear motion in the multiphoton ionization in H_2 has been pointed out recently [1]. The latter authors show that the inclusion of nuclear motion leads to a substantial increase of resonance enhanced multiphoton ionization (REMPI); they also suggested to control the ratio of dissociative ionization (DI: H_2 $+2\hbar\omega \rightarrow H+H^+ + e^-$) to nondissociative ionization (NDI: $\rightarrow H_2^+ + e^-$) by varying the pulse duration in the region of two-photon absorption. In fact, it was found that for laser pulses (polarized along the molecular axis) longer than 10 fs and $\hbar\omega > 0.45$ a.u., the DI process becomes the dominating one. We show here that the reason why DI dominates is entirely due to the opening at $\hbar\omega \sim 0.45$ a.u. of autoionization from the $\mathcal{Q}_1$ doubly excited states of H_2, an effect that is rigorously incorporated in our theory [2]. Figure I shows that for pulses of 10 fs and I=10^{12} W/cm^2, DI dominates in the region of photon energies $0.45 < \hbar\omega < 0.55$. At these photon energies, the lowest Q_1 $^1\Sigma_g^+$ doubly excited state is populated.

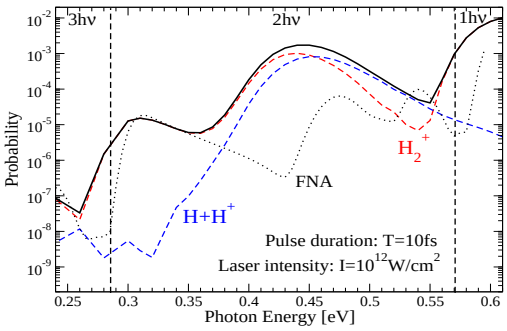

Fig. 1. Total ionization probabilities vs. photon energy. Solid line: total ionization probability (black), Dashed lines: Dissociative ionization (blue) and non dissociative ionization (red), Dotted line: frozen nuclei approximation (black). Vertical lines indicate the position of the N-photon ionization thresholds within the FNA approximation for an infinite pulse duration.

References

[1] Palacios A, Bachau H and Martín F 2006 *Phys. Rev. Lett.* **96** 143001; Palacios A, Bachau H and Martín F 2007 *Phys. Rev. A* **75** 013408

[2] Sanz-Vicario J L, Bachau H and Martín F 2006 *Phys. Rev. A* **73** 033410

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