Photoionization of H_2 using the molecular R-matrix with time approach

J Benda[1], Z Mašín[2,*], G S J Armstrong[3], D D A Clarke [3], A C Brown[3†], J D Gorfinkiel[1‡] and H van der Hart[3]

[1]School of Physical Sciences, The Open University, Walton Hall, MK7 6AA Milton Keynes, UK
[2]Institute of Theoretical Physics, Faculty of Mathematics and Physics, Charles University, V Holešovičkách 2, 180 00 Prague 8, Czech Republic
[3]Centre for Theoretical Atomic, Molecular and Optical Physics, School of Mathematics and Physics, Queen's University Belfast, Belfast BT7 1NN, United Kingdom

Synopsis We present results of the first calculations using the variational *ab initio* molecular R-matrix with time approach. We have calculated two and four-photon ionization cross sections for H_2 and studied the effects of electron correlation and choice of the Gaussian atomic basis sets. Our results are compared with earlier calculations.

The atomic R-matrix with time dependence (RMT) [1] approach has been applied successfully to the study of the interaction of atoms with ultrashort laser pulses. The approach is based on the R-matrix method that divides space into an inner and an outer region. RMT uses highly accurate wavefunctions in the inner region that describe multielectronic interactions precisely. In the outer region, the ionized electron becomes distinguishable; here the single-electron time-dependent Schrödinger equation is solved, with the electron subject to the long-range potential of the molecule and the laser field.

Recently, we have extended the original atomic RMT software [2] to treat interaction of laser fields with molecules in the fixed-nuclei approximation. The molecular, inner-region data required (transition dipole moments, etc.) are generated using the UKRmol+ suite [3], a highly accurate set of (time-independent) programs initially designed to treat electron and positron scattering from polyatomic molecules.

The RMT approach is very general and allows the study of a range of processes from the perturbative to highly non-linear regimes. Detailed understanding of the molecular RMT models for multiphoton ionization is a prerequisite for its application to processes such as strong-field ionization and high-harmonic generation.

In order to gain this understading and validate the molecular RMT calculations, we have studied two- and four-photon ionization of H_2 for which earlier results are available. Figure 1 shows our simplest results produced using a Gaussian basis set for the bound orbitals and B-splines for the description of the continuum. Our cross sections are in good agreement with those obtained with the diatomic R-matrix Floquet approach [4] but less so with other calculations. We have studied the effect of the modelling of electron correlation for multiphoton ionization and will compare the results of our close-coupling models with similar results obtained using other approaches.

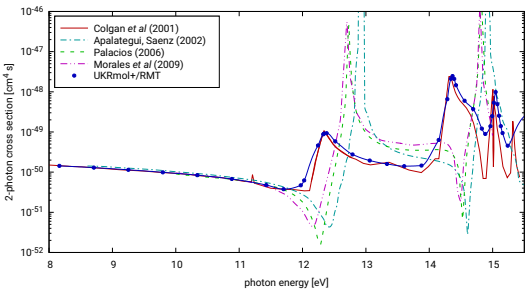

Figure 1. Cross section for two-photon ionization of H_2. Our initial results show good agreement with earlier Floquet calculations.

References

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*E-mail: zdenek.masin@utf.mff.cuni.cz
†E-mail: andrew.brown@qub.ac.uk
‡E-mail: J.Gorfinkiel@open.ac.uk