Laser-coupled R-matrix calculations for high-harmonic spectroscopy

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Abstract: We develop R-matrix calculations for systems embedded in a quasi static field. We report the field effects on the Rydberg series of Helium, and on the tunnel ionization rates of CO_2.
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1. Introduction

The three-step model provides a simple physical picture for explaining a range of processes initiated by a strong laser field [1]. In this model an electron is first removed from an atom or molecule (i), oscillates in the continuum (ii), and comes back to re-collide with the parent ion (iii). Effects such as high harmonic generation, double ionization and electron diffraction originate from electron–ion recombination or elastic/inelastic scattering in the presence of the strong laser field. It is commonly assumed that the strong laser field does not affect electron-ion recombination and scattering [2]. However, the situation may change in the presence of structured continua, as has been recently shown theoretically [3] and demonstrated experimentally [4]. We aim at developing an *ab initio* approach to investigate electron-ion collisions in the presence of the strong laser field. Our approach is based on the *ab initio* R-matrix method and should allow for the treatment of complex molecules. The idea is to extend the R-matrix method to consider scattering and recombination in a strong static field. Moreover, by analyzing the electron-cation scattering for energies below ground ionic state threshold, we can extract the tunnel ionization rates of the studied system, what is of key importance in high harmonic spectroscopy, as state of the art theory struggles to give the correct ionization rates even for some small molecules.

To benchmark our developments we study the field effects in both a weak and strong static field regime. In the weak field regime, we investigate the field induced dynamics on the Rydberg series of He converging to the first excited ionic state. We show that the static electric field strongly couple the asymptotic channels, resulting in a redistribution of the resonances width and position, in good agreement with theoretical predictions and previous complex scaling calculations[5]. In the strong field limit, we benchmark the field effect by analyzing the alignment dependent tunnel ionization rates of CO_2. We observed a peak ionization rate varying from 40^o to 55^o with an increasing laser field intensity, which is in good agreement with previous experiments and simulations[6].

2. The R-matrix method

The R-matrix method consists of partitioning the space into a complicated inner region containing all the electron-electron interactions ($r < a$), and a greatly simplified outer region ($r > a$) far enough from the atom or molecule, such that only the target long range potential and the laser interacts with the scattered electron. For *ab initio* simulations, we use the UKRmol suite developed by Tennyson *et al*. [7]. The suite is registered as a project on CCP-Forge, which provides an on-line repository for the codes.

In order to adapt the UKRmol suite to the investigation of strong field physics, Harvey *et al*., developed the module CDENPROP which calculates the dipole transitions between continuum-continuum and continuum-bound states [8]. The module STCFLD includes the field effects in the UKRmol package, by adding to the Hamiltonian the field induced continuum-continuum couplings in between the field free eigenvectors of the system. For simplicity, the Hamiltonian is diagonalized in the field free basis.

3. Results

<u>*Weak field regime:*</u> The atomic orbitals of the first nine states of He^+ were generated by a linear combination of Gaussian basis set (cc-PVTZ+even tempered scheme). We build continuum orbitals up to l=5 and spanning energies from 0 to 3 Hartrees using a set of optimized GTO to represent the Coulomb functions [9]. With this model, we could characterize the Rydberg series up to a distance of 0.57 eV from the first excited ionic threshold (40.5eV). When the static field was turned on in the z-direction, the resonances split as the parity (π) and the total angular momentum (L) are no longer good quantum number for the system, and must be replaced by the projection of L in the z-axis (M). Another interesting effect observed is the redistribution of resonance peaks and widths, that arises due to the channel coupling induced by the field. The interaction among resonances can be accessed by the total

resonance strength (S), which simply measures the area of the cross section in a given energy range. This analysis shows that the $(2,5a)^1D^e$ and $(2,5a)^1P^o$ resonances shown in Figure 1 are strongly coupled to each other, as S in the vicinity of these resonances remains almost constant for a wide range of field strength. The photoionization cross section also shows a very interesting feature, as the $(2,5a)^1D^e$ resonance, which is a forbidden in the field free case, rapidly increases with stronger fields. The same happens to the S-type resonances found at 39.86eV. Once again, the emergence of these peaks is explained by the couplings induced by the static field.

Strong field regime: The molecular orbitals of CO_2^+ were generated by a state averaged CASSCF procedure using MOLPRO [10]. Eight states were included in the averaging. Continuum orbitals up to the f-orbitals were obtained by the same procedure described for He. We show that the angle dependent peak ionization rate is shifted towards larger angles as the static field increases. The general behavior of our calculations agrees well with previously reported calculations [6].The dynamical shift of the peak ionization maps the influence of the molecular orbitals involved in the process.

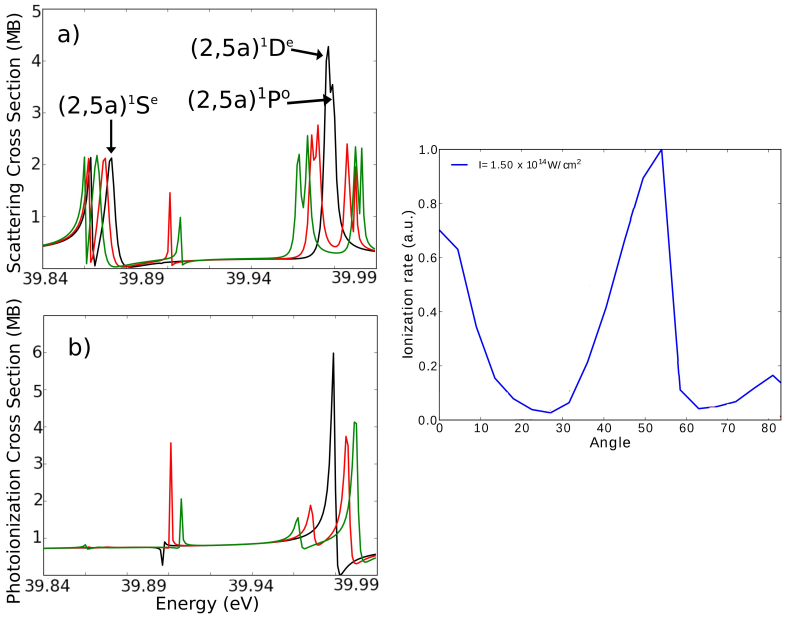

Figure 1: In the left we show the e-He^+ scattering (a) and He photoionization (b) cross section in the vicinity of the first excitation threshold for the Ag+B1u irreducible representation. Black, red and green curves represent electric fields of 0.0, 3.0, and 5.0 a.u. respectively. The energy axis of figure (b) was shifted by the ionization potential of Helium (24.56 eV), so we could compare its resonances with (a). The right panel shows the alignment dependent ionization rates of CO2 for different laser field intensities.

4. References

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