Benchmark calculations for multiphoton ionization of He at a wavelength of 390 nm

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Introduction

The study of electron dynamics in atoms subjected to intense laser fields has been a focus of experimental and theoretical physics effort over the last 25 years, and will continue to be so for decades to come due to the new experimental facilities coming into play across the world. In addition to the construction of a petawatt laser at the Central Laser Facility of the Rutherford Appleton Laboratory, new developments include VUV and X-ray free-electron lasers at DESY and Stanford, and the emergence of attosecond light sources.

These developments in experimental laser techniques have enabled experimentalists to (continue to) discover new features in strong-field electron dynamics in atoms, such as above-threshold ionization, high-harmonic generation and enhanced double-ionization through dielectronic interactions. However, the most basic atomic process in strong laser fields is single ionization, and recent experiments have demonstrated that we still do not have a complete understanding of this process.

It is of great importance to both theory and experiment to have an excellent understanding of single ionization in a strong laser field. Experimentally, it is very difficult to measure the intensities achieved in the focal spot accurately. Usually, the intensity is measured through single ionization yields. By comparison of the experimental yields with accurate theoretical predictions, one can estimate the intensity of the laser pulse. Accurate theoretical methods and calculations are thus essential to the detailed interpretation of the experimental results.

At present, most experimental results are compared to theoretical calculations of single ionization yields based on single-active-electron models. These single-active-electron models assume that the laser field interacts with only one electron, and that all other electrons do not interact in any way with the laser field. This approximation is appropriate for the alkali atoms, which have one electron outside a closed core. The approximation is found to work well for the noble gases, e.g. He and Ne, since doubly excited states are well separated from the ground state and the low-energy single-ionization continuum. For other atoms, the single active electron approximation is less appropriate. For example, the V ground state configuration is $3d^3 4s^2$, whilst the V^+ ground state configuration is $3d^4$. Single ionization of V thus includes simultaneously the emission of a 4s electron and the transfer of the other 4s electron to the 3d orbital. This single ionization process, recently investigated in experiment[1], cannot be adequately investigated theoretically using single-active-electron techniques.

Despite the limited number of atoms for which single active electron models are appropriate, the Ammosov-Delone-Krainov model[2] (ADK) in particular is commonly used for a wide range of atoms. The ADK model describes single ionization as a tunneling process. However, it is applicable only in a limited intensity region. At low intensities, single ionization must be described as a multiphoton process, rather than a tunneling process, while at high intensities, single ionization must be described as an over-the-barrier ionization process. Despite the fact that the validity of the model has not been assessed for many atoms, and further that it is not known a priori for what intensities it is valid, the ADK model is widely used because it expresses the ionization rate via a simple formula.

Because of the experimental need for accurate single ionization rates, and because these rates can not usually be obtained from single-active electron models, it is important to develop computational techniques, which are able to determine single ionization rates for atomic species that go beyond the single-active-electron approximation. Over the last decade, at Queen's University Belfast we have developed two such techniques: the time-dependent numerical integration (TDNI) of the full Schrödinger equation[3], and the R-matrix-Floquet (RMF) approach for multiphoton processes[4,5]. Although both approaches have been developed at Queen's University Belfast, the two approaches are fully independent: they do not share theory, algorithms, numerical methods or computing code.

In the present report, we compare single ionization rates for He irradiated by 390 nm laser light as a function of intensity. To establish the range of intensities in which the ADK provides a good approximation to the ionization rates, we compare our single ionization rates with those given by the ADK model.

The time-dependent numerical integration of the full Schrödinger equation

The time-dependent numerical integration technique provides a finite-difference solution of the full two-electron Schrödinger equation for the helium atom. It contains no arbitrarily adjustable parameters. To appreciate the accuracy of the approach, the maximum measured errors in the energies of bound and autoionizing states are under 0.01% and 0.1%, respectively, whilst the maximum measured error in the autoionization width is under 1%. Rate calculations are performed by measuring the decay of population within a sphere, centred at the nucleus with a radius of 16 au. The field is ramped on smoothly over 5 field periods. After initial transients have vanished, the population decay approaches steady state in a time-averaged sense.

Full two-electron calculations are performed at 20 intensities between 0.5×10^{14} W/cm^2 and 3.5×10^{14} W/cm^2. To improve the intensity resolution of the calculations, a single-active-electron model is tuned to give quantitative agreement with the full calculations at these intensities. Single-active-electron model calculations are then performed to provide ionization rates at intermediate intensities. These model calculations are also used to estimate the uncertainties in the full calculations due to the spacing of the radial mesh, the box size and the total number of angular momenta retained.

The R-matrix Floquet approach

The R-matrix Floquet calculations combine R-matrix Floquet theory with B-spline basis set techniques. Configuration space is divided into two regions, an inner region, in which the full interaction between the two electrons is included, and an outer region, in which one (outer) electron moves in the field generated by the He^+ ion and the laser field. The boundary is set at 8 au from the nucleus. To link the two regions, we use the so-called R-matrix. By matching the R-matrix with asymptotic solutions, we obtain the energy and ionization rate of a state of the He atom in the laser field.

In the present calculations, we use a Floquet expansion including up to 43 Floquet blocks of which 32 describe absorption and 10 emission. The maximum angular momentum included is L=13. Because of the very large Floquet expansion, we have to restrict our description of He to a minimum: we include only a single target state of He^+, the 1s state. The main restriction in the calculations is thus that at least one electron must be in the 1s state. The effect of this on the single

ionization rate is expected to be small. A comparison of He single ionization rates at a wavelength of 248.6 nm shows that the rates obtained using a 1s-target-state description are about 2% smaller than those obtained using a (1s,2s,2p)-target-state description. These differences are expected to decrease with increasing wavelength, so the 1s-target-state description should be accurate to well within 2% at 390 nm.

Results

Figure 1 shows single ionization rates for the ground state of He subjected to 390 nm laser light as a function of intensity between 0.5×10^{14} W/cm^2 and 2.5×10^{14} W/cm^2, obtained using the RMF and TDNI approaches. At these intensities, single ionization of He at these intensities requires absorption of at least nine photons. The rates obtained by the two entirely different approaches are in superb agreement with each other, generally well within 10%.

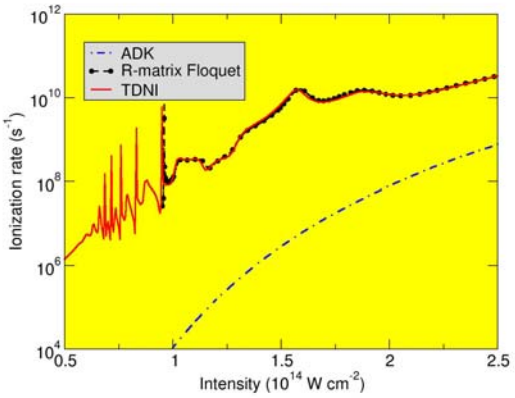

Figure 1. Single ionization rates for He subjected to 390nm laser light as a function of intensity. R-matrix Floquet results (black dotted line with circles) and TDNI results (solid red line) are compared with ADK results (dot-dashed blue line).

Particularly impressive is the agreement between the two approaches for the resonance structure. The resonance structure is no longer the field-free atomic structure; it is strongly perturbed by the laser field. Analysis of the resonances shows that, for intensities above 0.3×10^{14} W/cm^2, the 1s3d state actually lies below the 1s3s state. The good agreement between the two calculations thus demonstrates that the non-perturbative effects of the laser field and the electron-electron interactions are fully accounted for.

Figure 1 also compares our single ionization rates with those obtained by the ADK approach. Although the ADK approach is not strictly valid in this region, the approach remains widely used within the low-intensity regime. It can be seen that the ADK approach severely underestimates the photoionization rates at low intensities. The differences increase with decreasing intensity, from a factor 45 at an intensity of 2.5×10^{14} W/cm^2, to a factor 10000 at an intensity of 1.0×10^{14} W/cm^2.

The intensity range over which a detailed comparison of TDNI and RMF calculations can be carried out is rather limited. The RMF calculations require the total single ionization rate to be larger than about 10^7 s^{-1} as an iterative root-finding procedure is used to obtain the energy and width of the initial state. To achieve convergence, the difference between the initial guess for the energy and the actual energy must be comparable to the width. This becomes quite problematic at low intensities. At high intensities, the R-matrix Floquet calculations encounter another difficulty. The laser field is described using the length gauge in the inner region and the velocity gauge in the outer region. At the boundary, a frame transformation is required. The numerically accurate calculation of this frame transformation gets more difficult when the intensity is increased, as larger and larger Floquet expansions are required.

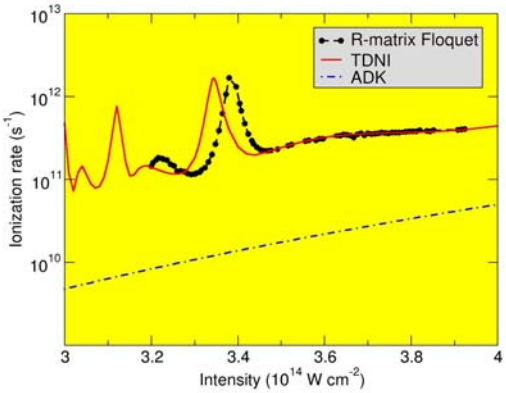

Figure 2. Single ionization rates for He subjected to 390 nm laser light as a function of intensity. R-matrix Floquet results (black dotted line with circles) and TDNI results (solid red line) are compared with ADK results (dot-dashed blue line).

The maximum intensity, for which reliable R-matrix Floquet calculations can be performed reliably using the present expansion, is about 4×10^{14} W/cm^2. The TDNI approach, on the other hand, has been applied for intensities up to 30×10^{14} W/cm^2. In Figure 2, we compare single ionization rates for He irradiated by 390 nm laser light for intensities between 3.2×10^{14} W/cm^2 and 4.0×10^{14} W/cm^2. Apart from the resonance positions, discussed below, we observe excellent agreement between the RMF and the TDNI results. However, the R-matrix Floquet results show errors due to the finite Floquet expansion. Both numerical approaches obtain an ionization rate about a factor 10 larger than the rate predicted by the ADK model.

The main difference between the rates obtained by the RMF and TDNI approaches is the position of the resonance at an intensity of 3.3×10^{14} W/cm^2. The two approaches obtain slightly different intensity shifts of the He ground state, resulting in a difference in resonance intensity of about 1%. The same shift is also noticeable in Figure 1, but the lower intensities in Figure 1 mean that the difference in resonance intensity is smaller.

Conclusions

We have obtained rates for single ionization of He subjected to laser light with a wavelength of 390 nm using the R-matrix Floquet approach and the time-dependent numerical integration of the Schrödinger equation. The two sets of rates are generally in superb agreement with differences typically less than 10%. We thus consider these rates as benchmark single ionization rates for He at 390 nm. For the present range of intensities, the ADK model underestimates the actual rates by at least one order of magnitude.

References

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