

NOTES ON TWO-PHOTON DOUBLE IONIZATION OF THE HELIUM ATOM

P. Lambropoulos
IESL-FORTH and Univ. of Crete
Heraklion 71110, Crete, Greece

In view of the extensive discussions on the problem of two-photon double ionization, during the workshop on “Theoretical Challenges in Attosecond Laser Science” , May 12-24, 2008, as well as the rather rapidly increasing rate of new papers on the subject, it is perhaps useful to attempt a summary of the issues, as well as a guide to the relevant literature.

The discussion that follows has been divided in two parts:

Part (A) is based on the realistic parameters of the sources, available presently and in the foreseeable future, for the experimental investigation of the process. Specifically, it takes into consideration the fact that the intensities and pulse durations are such that LOPT (Lowest non-vanishing Order Perturbation Theory), in terms of the relevant transition amplitudes and cross sections, is the most appropriate theoretical framework for the elucidation of the underlying physics. A rather extensive recent discussion of the conditions for the validity of LOPT in this context, as well as a useful background on multiphoton processes accumulated over the last 35 years or so, can be found in [1].

Part (B) refers to aspects of the process under pulses of extremely short duration and higher than presently envisioned intensities. Such pulses may possibly become available in the future, but, in any case, theoretical investigations in that range of parameters may be valuable in elucidating the finer details of the processes involved.

PART A

1). The idea of 2-photon double ionization of Helium, as a route towards the exploration of direct double ionization (DDI), which is nonsequential, (NS) but fundamentally different from its counterpart under long

wavelength (which relies on recollision), was introduced and analysed semiquantitatively in [2], almost 10 years ago. The importance of the judicious choice of photon frequency, in order to spectroscopically separate the direct from the sequential, was one of the main points in [2], where the structure of the photoelectron (angle-integrated) energy spectra was illustrated and discussed (see Figs. 3 and 4 of ref.2).

2). The question of an optimum photon energy for the experimental investigation of the direct process has been explored quantitatively in [3], where it was shown that photon energy of around 45 eV is indeed the optimum, because then the yield of the sequential is at a relative minimum.

3). Given the status of the radiation sources in the photon energy range needed for the process (from about 40 to 80 eV), it is understood that for the time being and the foreseeable future, the intensities and pulse durations are such that the processes under consideration are describable in terms of cross sections obtained through LOPT of the appropriate order. The fact that the relevant experiments involve pulsed sources, simply means that, for the quantitative description of experiments, the cross sections will enter a set of rate equations, governing the evolution of the atomic and ionic species during the pulse, in which a realistic description of the temporal and spatial structure of the pulse should be employed (see, for example, ref. 4). In special cases, not envisioned to be of relevance in the present context (at least for the time being), a more complete description should be cast in terms of the density matrix, which reduces to the rate equations when the coherences involved in the off-diagonal matrix elements are negligible.

4). For photon energies below about 54 eV, the sequential represents a three-photon process (1 photon to ionize the neutral and two more photons to ionize the ion in its ground $1s$ state) and, as already mentioned in (1) above, it consists of two peaks spectroscopically separated from the direct. A quantitative calculation in that regime (including realistic temporal and spatial pulse shapes) has been given in [4]. Above that photon energy, the sequential is an overall two-photon process, exhibiting again two peaks, which however overlap energetically with the continuous spectrum of the direct (see Fig. 2 of ref.[2], as well as a most recent version in Fig(1) of ref.[5]).

5). In all cases, the sequential is a well defined process, involving one-photon ionization of the neutral, leaving (for all practical purposes) the ion in its ground state, followed (depending on the frequency) by one- or two-photon ionization of $\text{He}(+)1s$. In either case, this is a two-rate process, whose proper quantitative description requires the solution of the rate equations (as in ref. [4]). Depending on intensity, pulse duration, etc., a single rate equation may be useful, if nothing else, as a semiquantitative evaluation. This was done in [2], which was actually based on a much earlier related treatment [6]. It is, however, essential that the expression for such a single-rate include the relevant widths of the states involved (see Eq. (4) of ref.[2]). Otherwise, the expression exhibits unphysical singularities because of energy differences in the denominators (as in Eq(4) of ref.[5]), which are due to the incomplete derivation of the expression and not, as argued in [5], to the order of perturbation theory. As for the notion of “virtual sequential” processes [5], it is hardly necessary or even meaningful. It appears to be a term referring to a transition corresponding to the tail of a line which has a certain width, as explained above; and there is nothing virtual about that.

Moreover, certain qualitative arguments concerning the dependence of the sequential on pulse duration can be misleading. For example, according to Eq.(20) of ref.7, the yield of the sequential is proportional to the square of the pulse duration, while that of the direct is proportional to the duration. This, however, is an oversimplification because it fails to take into account the fact that the population of the initial state also changes (is depleted) with time. A more careful analysis, through the solution of the rate equations, shows that the dependence of the yields from the two channels on the pulse duration is much more complicated, depending in fact on the combination of intensity and range of pulse duration.

6). As the photon frequency changes and crosses the threshold at which the two-photon ionization of the ion becomes one-photon ionization, there is no divergence, as has occasionally been implied. Such a change of the order of the process is well known in multiphoton and strong –field physics, as for example, when a channel closes due to the ponderomotive shift. Moreover, given the significant bandwidths of the pulsed sources relevant to this problem, even the threshold behavior of the cross section is of no relevance, as it is overshadowed by the bandwidth; typically more than 1 eV.

7). The direct two-photon double ionization cross section (under the conditions of Part A), conceptually and calculationally, is separate from the sequential processes cross sections, which are well-defined and well-known single ionization cross sections. Although, above ~ 54 eV, direct and sequential overlap spectrally, which means that in a measurement one can not tell where an electron of a particular energy (angle-integrated) has come from, theory can and should provide an answer as to the ratio between the sequential and direct amounts. Photoelectron angular distributions – much more demanding experimentally – can of course provide a further input as to the origin of the electrons. Unlike the direct process, photoelectron energy and angular distributions for the sequential, corresponding to one- or two-photon single-electron ejection, are easily calculated on the basis of methods developed over the last 30 years of multiphoton physics, and studied both theoretically and experimentally in many atoms and molecules.

8). The direct two-photon double ionization cross section has by now been calculated by a number of authors. For a list complete, to the best of my knowledge, up to October 2007 see ref. [1]. For an update (not necessarily complete), references [7-10] should be added, as well as ref.[5], already mentioned above. The reasons for the discrepancies in the values obtained through different approaches are not as yet understood, although they should not be surprising given the complexity of the problem. Cast in terms of LOPT, it requires the calculation of the two-photon transition amplitude from the ground state of the neutral to the double continuum; an unequivocally defined quantity, albeit not easy to calculate. For a discussion of the methods and issues pertaining to this quantity, see the discussion in ref. [1]. Most results on this cross section have, however, been obtained through the solution of the time-dependent Schrodinger equation (TDSE), which provides the double ionization yield at the end of the pulse employed in the calculation. Assuming the intensity employed in the calculation is not too high (say, below 10^{15} W/cm²) and the pulse duration not too short (say, 10 field cycles or more), and with proper care (see [11]), the value of the cross section can be extracted from the ionization yield. It should of course agree with the value obtained through LOPT, especially if the same basis has been used in both. Such comparative calculations within the same basis seem to be a somewhat distant, albeit highly desirable goal.

9). The time-dependent (TD) approaches, relatively more easy to implement, provide the total wavefunction of the two-electron system, at the end of the pulse. Extracting the double ionization signal from that wave-function is not straightforward and the method required does actually depend on the basis employed in the time propagation. A number of issues arise in connection with this final step, and pertain to the “correct” form of the final state employed in the projection. For example, are Coulomb functions for $Z=2$ acceptable or should the final state contain manifestly correlation? And so on. For contrasting views, see refs[7-10]. I will refrain from taking a position on this issue, except for one particular aspect discussed immediately below.

10). In some recent papers [7,8,10], the direct cross section (as a function of photon energy) has been found to begin rising steeply at around 54 eV; i.e. the threshold of the transition of the sequential from three- to two-photon process. I would argue that I see no reason for the value of the direct to exhibit such a rise. The two-photon direct double ionization cross section is just another two-photon cross section, and as such it should exhibit a smooth behavior as a function of photon frequency; unless the first photon hits an intermediate resonance with a real atomic state, which is not the case. In fact, even for a photon frequency (around 56 eV) on resonance with one of the doubly excited autoionizing states, there will be no discernible resonance peak (see, [12]). The only reason I can see for the rise reported in the above references is the unintended inclusion in the result of the contribution from the sequential, which at that photon energy begins overlapping spectrally with the direct. It is therefore important to ascertain that my argument above is incorrect (a distinct possibility) and that the sequential has been excluded. In that case, however, the responsibility for an explanation for the reason of the rise of the direct rests with its authors. The justifications, written or otherwise, offered so far are too vague to be of any value. Actually, the direct two-photon double ionization cross section, physically and computationally, is not that much different from the corresponding one-electron ATI (two photons from the ground state of the neutral). And that cross section is not expected to, and does not, exhibit a similar rise. Put otherwise, a two-photon cross section, within LOPT is ...well, a two-photon cross section; i.e. the same sort of animal, irrespectively of whether it involves one or two electrons.

PART B

1) If the pulse duration is extremely short, certain of the premises in Part A above are not valid. Obviously, there are no sharp demarcation lines between long, short and extremely short. Be that as it may, let us consider some limiting cases. If the pulse becomes one or even two cycles, not to mention shorter, the process may still be perturbative, but the notion of the cross section is no longer valid. If the intensity becomes too large (a good criterion being the ponderomotive energy being much larger than the photon energy, which here means more than 10^{19} W/cm²), even for a pulse duration of 10 cycles or more, the process becomes non-perturbative. In either case, the solution of the TDSE is necessary, while the extraction of a cross section from the ionization yield is not meaningful. If extracted, nevertheless, it would depend on peak intensity and pulse duration, and as such of doubtful usefulness. Comparisons between results from different calculations, in that intensity regime, are therefore meaningful if the same (in all aspects) pulse has been employed, and if it is the ionization (single or double) yield that is being compared. And, a propos, the ratio of singly to doubly ionized ions is an extremely useful quantity for comparison with experimental data (see e.g. [4]).

2) The case of “short” pulse and not too large intensity merits somewhat closer examination, as it can shed some light on the processes involved. What is short in this context? An extreme limit would be a duration such that the helium ion does not have the time to relax to its ground state. Clearly, in that case the energy peaks of the sequential – whatever the photon frequency might be in the region between 40 and 80 eV – would broaden so much that they would no longer be distinguishable, even conceptually, from the direct. The easiest way to get a calibration for that limit is to look at the energy distance between the sequential peaks and convert that to time by taking the inverse. Thus, for example, for photon energy of 45 eV, the distance between those peaks is about 15 eV. [4], while for 55 eV, it is about 30 eV [3]. Roughly speaking, the relevant times are less than about 400 attoseconds. Therefore, given that the pulse durations available or envisioned (for the time being), in that frequency range, are at best about 10 fs or more, the features of the sequential would be as described in part A above. For a more detailed analysis of the “extremely” short time behavior, relating it also to a

measure of a correlation time, I would refer the reader to ref.[13].

3). Finally, two-photon double ionization is not limited to Helium. For a case in much lower photon energy, and consequently intensity, range, see ref.14 pertaining to Mg. That analysis serves also as an illustration of the complexities appearing in systems with more than two electrons, where the conditions for the spectral separation between direct and sequential processes become much more difficult to achieve.

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