Part IV

Hydrogen in Strong Fields and Chaos

Multiphoton Transition to the Continuum of Atomic Hydrogen

N.K. Rahman

Dipartimento di Scienze Chimiche, Università di Trieste,
Piazzale Europa, 1, I-34100 Trieste, Italy

The current experimental and theoretical status of multiphoton transition to the hydrogenic continuum, its prospects in the near future and the relation of these studies vis-a-vis standard QED are described.

1. Introduction

Non-linear processes involving transitions to the continuum brought about by high intensity pulsed lasers are the subjects of numerous investigations in the area of atomic, molecular and optical physics. New processes are being experimentally uncovered at a rapid rate, and the field promises to be of continued vigour due to the remarkable progress that we are witnessing in the development of high power lasers. Almost all of these studies have been done with non-hydrogenic atoms (or molecules). These processes are treated theoretically with necessarily approximate inputs and at some stage of the calculations, wave functions, matrix elements, summations over the entire spectra, what you will, that are utilized give us results that can hardly be considered as exact. Often the agreement between theory and experiment is either fortuitous or non-convincing for many reasons and more often, the agreement between theory and expriment is simply lacking.

The mathematical structure of the quantum mechanical amplitudes of non-linear processes involving continuum for system is rather complicated and for non-hydrogenic systems can be the theoreticians nightmare. To cite an example, a large number of experimental results have been reported in the last few years with the rare gases, but no reliable calculations for these experiments exist. Essentially the detailed confrontation between theory and experiments have been given up in this area and very often one is satisfied with the order of magnitude estimates.

Hydrogen atom is in the pleasant exception to this rule: the theoreticians are expected to produce accurate estimates of these processes. The experiments then serve to test these benchmark calculations and the satisfactory conclusion of this dialogue serves as the foundation of the atomic physics of non-linear processes involving continuum just as the photoionization process of the hydrogen atom [1] serve as the cornerstone of all photoionization processes. In what follows, I shall explain what three recent experiments have measured in this area and how theoretical calculations have

The Hydrogen Atom Editors: G.F. Bassani · M. Inguscio · T.W. Hänsch

helped to explain quantitatively these measurements of three highly non-linear processes involving multiphoton absorption in the continuum of the hydrogen atom.

Further on, I shall discuss a few other such processes which have not yet been experimentally measured for the hydrogen atom. These could be the next set of experiments with the hydrogen atom and intense lasers. The future of this class of processes vis-a-vis the next generation of high power lasers useful for basic atomic physics experiments, and the non-linear processes viewed from the perspective of QED theorists are the final comments in this paper.

2. REMPI, ATI and REATI

There is a large number of processes that one can envisage being induced by intense lasers with the hydrogen atom. I have selected here three of them, the reasons being that in the last three years, three experiments [2,3,4] have been performed to study them. These processes may be called with the acronyms REMPI (Resonance enhanced multiphoton ionization), ATI (Above threshold ionization) and REATI (Resonance enhanced above threshold ionization) [Fig. 1]. All of these three processes are non-linear, i.e. the probabilities are not linear functions of the intensity of the lasers.

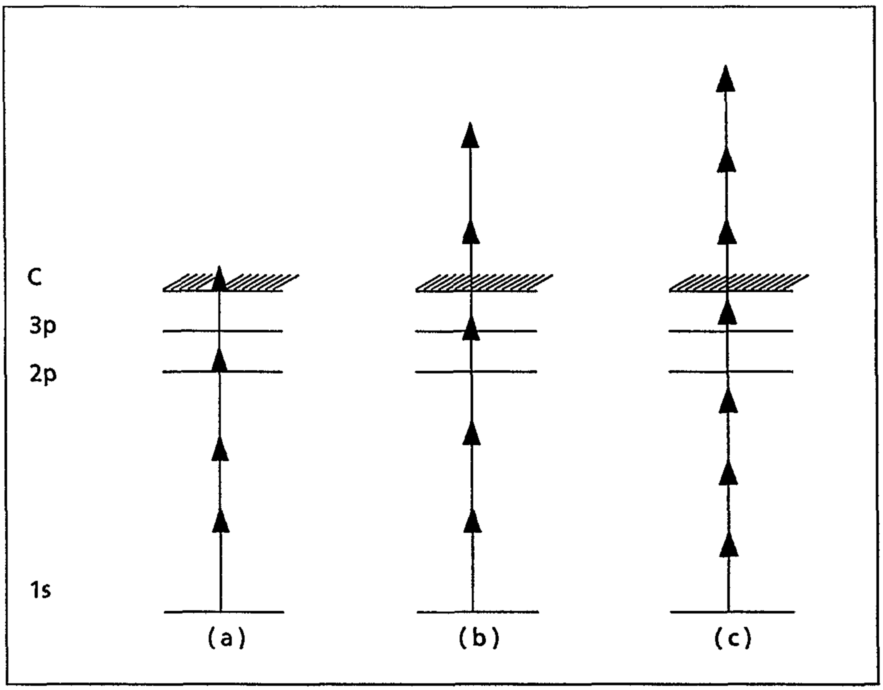

Fig.1. Examples of (a) REMPI, (b) REATI and (c) ATI for the hydrogen atom. Each arrow represents a photon absorption.

REMPI is the bound-free, i.e. ionization, process in which N (N>1) photons are needed to ionize the atom and the frequency of the photons is such that M(M<N) photons can cause a bound-bound transition from the initial state of the atom. This latter condition causes an enhancement of the probability of the usual multiphoton ionization with no frequency condition.

ATI is the ionization by the absorption of N (N>1) photons by the atoms while in order to ionize the atom, one needs M (M<N) photons. Thus the electrons that emerge in this processes acquire energy due to the absoption of additional photons in the continuum. The process involves an admixture bound-continuum and continuum-continuum transitions. The electron spectra is the tell-tale sign of such processes showing how many photons have been absorbed beyond the minimum number M.

REATI is the above threshold ionization in which P (P<M,N) photons can cause a bound-bound transition in the atom, M photons are needed to ionize and N photons are number of photons absorbed for the overall detected signal. In contrast to (P,M,N) which specifies the REATI, the duo (M,N) that specifies ATI and REMPI can be in principle any set of integers with N>l. For the three experiments that have been done, they are (P = 3, M = 4, N = 5) for REATI, (M = 6, N = 9) for ATI and (M = 3, N = 4) for REMPI.

3. Current Status

The theory of these processes for a non-relativistic hydrogen atom involves utilizing perturbation theory of the N-th order as the basic starting point. The amplitude is given by:

$$T_{fi}^{(N)} = <i \left| V^- G_o V^- G_o \ldots G_o V^- \right| f>$$

where G_0 is the Coulomb Green's function and V^- is the photon absorption part of the interaction hamiltonian.

The above expression contains for 4 photon processes (for example provoked by radiation at 355 nm) 3-fold infinite summation (over all the discrete states of H°) and integration over the entire Coulomb continuum. This can be done for the hydrogen atom with a variety of methods, only one of which will be relevant for our purpose and we shall discuss that later. The ATI process starts with utilizing the same amplitude as in Eq.(1) with N = 5 (at the same frequency 355 nm). The remarkable feature of the amplitude now is that while previously the basic radial part of the amplitudes are real, the corresponding ones are now complex. This happens due to the fact that in ATI the last intermediate integration has a pole in the denominator, producing thereby a complex amplitude. This fact recurs for all the higher order ATI processes. For example, at 532 nm, we need to consider N = 7, 8 and 9 for ATI [5] . The integration through the pole occurs one, two and three times respectively. The resulting amplitudes are then utilized to write down the so-called differential cross-section, which gives us the angular distribution of the ejected electrons. These angular distributions of the ejected electrons once measured, give us an excellent check on the validity of the calculation.

How are these calculations done? The work requiring the N-fold infinite summations and integrations is a non-trivial task. A large amount of computational effort has been devoted to their calculation. The methods range from brutal computation, to special representation of the Coulomb Green's function (the notheworthy one being that in terms of the Strumian functions) and the implicit method utilizing a bypass by solving coupled differential equations. This latter method has turned out to be the most effective for the calculations regarding the experimental data in [3]. The method has been utilized to calculate ATI at 355 nm (N,M = 5,4 and 532 nm (N,M = 9,6) and have reproduced rather strikingly all the experimental angular distributions [5]. One may conclude that the lowest order perturbation theory is satisfactory for ATI under the given experimental condition of the intensity of the laser and the photon frequencies.

The REMPI experiment involves coupling 1S and 2P or 3P states by a 3 photon bound-bound transition and then bound-continuum transitions to produce ionization. The process is both non-linear as well as non-perturbative, the latter being manifested by the non-observance of the I^N power law. The calculation essentially uses a formalism that ensures the strong coupling of the two relevant states while the rest are treated perturbatively. The shifts and these states as well as the width of the resonant state appear naturally. The hydrogenic energy formula $E_n = 1/(2n^2)$, also creates the curiosity that the exact resonance with one state is also the threshold of ionization, i.e. the electron should be ejected with zero velocity. This produces certain asymmetry to the probability of ionization as a function of frequency. Taking the experimental pulse structure as well as coherence properties of the laser into consideration, the calculation has been completed to the extent that the results can be directly confronted to the experiment. The agreement is indeed very satisfactory.

The third experiment which concerns REATI is of interest because it measures the branching ratio between the electrons issued with the minimum energy and the ones that had acquired the energy of one photon, under the condition of REMPI. A minor modification to the theory of REMPI allows one to calculate REATI and therefore the branching ratio. The agreement between the experiment and the theory is within 8%.

4. Other Processes

There is a large variety of non-linear processes for hydrogen that have not been studied experimentally, but have been theoretically investigated. It would not be possible for me to discuss them all here, but let me briefly mention a few.

Probabilities of multiphoton ionization have been calculated for the hydrogen atom for N quite large [8,9,10], up to even N = 16. These are rather formidable calculations and no experiments of these have still been made. (There exists a remarkably early experiment [11] with hydrogen atom). The theoretical values of these probabilities arise from large order of the perturbation series. It is noteworthy that some of these processes have been calculated utilizing independent methods (Sturmian function representation of the Green's function and coupled hierarchical set of differential equations) and the results agree with one other.

Another class of processes involves free-free multiphoton transition. These are the absorption or emission of N photons entirely in the hydrogenic (i.e. Coulombic) continuum. These processes could be multiphoton bremsstrahlung or inverse bremsstrahlung. Exact calculation [12] for 2 photons have been recently reported, which is the natural extension of the classic Sommerfeld calculation for the single photon. Measurement of such processes for hydrogen i.e. electron absorbing N photons in the Coulomb potential have still not been attempted. Another class of processes that have drawn the attention of theorists is in the Raman-like processes involving N Photons from an external source. Laborious calculations and accurate predictions have been made: what are now lacking are the corresponding experiments.

5. Discussion

The agreement between the three recent experiments and the corresponding calculations augers well for the future of the physics of multiphoton processes in which a wide variety of newer processes are to be observed experimentally. To be able to observe them for the hydrogen atom means that one can confront them with exact calculations and the basic understanding of these processes leaves no room of uncertainty.

Hydrogen atom may now prove to be an extremely valuable system for understanding the dynamics of the absorption of photons from extremely intense fields. Intensities of the order of $10^{16\text{-}18}$ Watt/cm^2 with pulse duration in the second region are beginning to be utilized for a newer class of experiments. The interaction term in the hamiltonian $H_{int} = - e/mc\ A \cdot P + e^2A^2/2mc^2$ for these field strengths are on the average comparable or higher than the Coulomb potential. The perturbation theory, at least the lowest order calculation, then become inoperative and one has to take recourse to resummation of the higher order terms of the perturbation series [13,14].

Other techniques such as Floquet theory then may become of much importance. One starts to wander into the uncertain regions of theory and it behooves us to consider above all the hydrogen atom (whose every eigenfunction we know to the T) as our theoretical guinea pig for these calculations. The experiments can then be done to confront these predictions and the strong field problem will then progress with a logical foundation. Technical progress in this direction appears to be really exciting.

Finally it is not unreasonable to place some of these processes and the corresponding experiments in the perspective of quantum electrodynamics. In QED, one nowadays, with large computation effort, calculates the higher order terms which increase the accuracy of a prediction. One thus adds more and more digits to measurable number and test how accurate is QED. The accuracy of the calculation and the corresponding measurement of Lamb shift of the hydrogen atom is truly remarkable. In a broad sense, the multiphoton transition into the continuum of the hydrogen atom is adding another dimension to the picture. The effects are due to the external field and the processes that are being investigated are the physical manifestation of rather high order terms of the perturbation series. Indeed, by means of the external field, one has been able to calculate 16th order term of the perturbation series (no experiment yet) and measure a 9th order term, (the experiment in Ref. 3 and the calculation in Ref.5), essentially exactly. Even without neglecting the fact that the calculations are for the non-relativistic hydrogen atom, the order of these processes certainly is a remarkable testimony of how much progress has been made in the quantum electrodynamics of the external electromagnetic field.

Acknowledgements

Much of the work that I have discussed here, happens to be partly the result of my own involvement along with Dr.Y.Gontier and Dr.M.Trahin of the Institut Recherche Fondamental, Service de Physique Atomique et des Surfaces at Saclay Laboratories. I must thank them for their friendship and cooperation. Thanks are due to Ms. Stefania Grassini for an excellent job of preparing this manuscript with the shortest of notice.

References

1. H.A.Bethe, E.E.Salpeter: Quantum Mechanics of One- and Two- Electron Atoms, (Springer Verlag, Berlin, 1957)
2. D.E.Kelleher, M.Ligare, L.R.Brewer: Phys.Rev. A31, 2747 (1985)
3. D.Feldmann, B.Wolff, M.Wenhöner, K.H.Welge: Z.Phys.D. 6, 293 (1987)
4. M.J.Muller , H.B. van Linden van der Heuvell, M.J. van der Wiel: Phys.Rev.A34, 236 (1986)
5. Y.Gontier, N.K.Rahman, M.Trahin: Europhys.Lett. 5, 595 (1988)
6. E.Karule: J.Phys.B: At.Molec.Phys. 21, 1997 (1988)
7. Y.Gontier, N.K.Rahman, M.Trahin: Phys.Rev. A37, 4694 (1988)
8. Y.Gontier, M.Trahin: Phys.Rev. A4, 1896 (1971)
9. E.Karule: J.Phys.B: At.Molec.Phys. 4, L 67 (1971)
10. A.Maquet: Phys.Rev. A15, 1088 (1977)
11. M.Lu Van, G.Mainfray, C.Manus, I.Tugov: Phys.Rev.A7, 91 (1973)
12. V.Veniard, M.Gavrila, A.Maquet: Phys.Rev. A 35, 448 (1988)
13. Y.Gontier, N.K.Rahman, M.Trahin: Phys.Rev. A14, 2109 (1976)
14. Y.Gontier, N.K.Rahman, M.Trahin: Nuovo Cimento D4, 1 (1984)