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(Laser, Atomic and Molecular Physics)

SINGLE PHOTON DOUBLE IONIZATION OF HELIUM

S.N. Tiwary

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Preface

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SINGLE PHOTON DOUBLE IONIZATION OF HELIUM *

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ABSTRACT

We have calculated the single photon double ionization cross section of helium using the fully correlated wave function for the ground state and partially correlated wave function for the final state. Our present theoretical investigation demonstrates that the inclusion of the full correlation in the initial as well as the final state wave function is indispensable in order to obtain reliable results in the close vicinity of the threshold.

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1 Introduction

Since the discovery of the photoeffect the variation of the photoionization cross section in the vicinity of the threshold has attracted continuing interest of both experimentalists as well as theorists. This is not surprising because the nature of the interaction between photons and atoms or molecules or ions or clusters or solids is most sensitively exhibited in the low energy region, where the excited electrons are still around or slowly leave the system . However, before the advent of the synchrotron radiation as a tunable light source in the VUV and soft X-ray region, most threshold and resonance studies were restricted to the theoretical calculations and crude comparisons far from threshold with the few data points available from discharge lamps or X-ray tubes. Consequently, the first decade of synchrotron radiation work on atoms, molecules, ions, clusters and solids was concentrated on cross section measurements to assess the different theoretical predictions particularly near threshold. For example the review articles by Samson and Starace (1-2) give an excellent overview of the progress made experimentally as well as theoretically up to 1982. In recent years, extensive experimental and theoretical investigations (3-14) have been made in order to understand the threshold effects in atomic and molecular photoionization. Cooper minima, shape resonance and the effect of inter-channel coupling on the cross sectional behaviour of main lines were among prominent issues studied both experimentally and theoretically. Advances in theoretical methods revealed the importance of electron correlation on the cross section and angular distributions of the emitted photoelectrons. The threshold behaviour has proved very helpful in helping to verify the predictive power of the different theoretical approaches beyond the one-particle picture. The avaibility of the synchrotron radiation has accelerated the development in

the measurement of the photoionization cross section. However, today we see that the improvement in theory has led to sophisticated computational methods to meet the challenge of new experimental results, opening a period of fruitful interplay between theory and experiment.

There has been much interest in the study of systems that are dominated by electron correlation. The search for electron correlation in its simplest form has taken place in the world of two-electron processes. A very broad definition of electron correlation would be any effect arising from interaction between electron and electron. Electron correlation seems, however, to play an important role in atomic physics. The concept of the correlation in atomic physics is extended to many areas of research, e.g., molecular physics, condensed matter physics, superconductivity, collision physics. In strongly correlated systems, the independent particle model fails to provide adequate information about the system. One photon double ionization of the helium atom is one of the best model system on which to investigate the effect of electron correlation. Double photoionization consists in the absorption of a single-photon by an atom or molecule followed by the ejection of two electrons. The interaction of a photon with each electron is independent from the others so that double photoionization is a forbidden process unless the electron correlation is taken into account. If two electrons with small kinetic energies leave the residual positive ion, the motion is strongly influenced and controlled by their mutual repulsion due to the Coulomb repulsive interaction ($1/r_{12}$). The interaction leads to the exchange of energy and angular momentum over long distances and therefore implies a correlation between outgoing electrons. The final state consists of an ion and two continuum electrons i.e.

$$h\nu + A \rightarrow A^{++} + e^- + e^-$$

In the case of complex atoms and molecules, the double photoionization process can be divided into two classes (a) the normal Auger process via core ionization and (b) the resonant double Auger process via resonant core excitation (3,8). From theoretical point of view, multi-electron atoms and molecules are extremely difficult because of the core electrons. The subject of double photoionization of noble gas atoms has been of intense growing interest to both experimentalists and theorists because double photoionization (DPI) in noble gases gives basic informations on the electron correlations in atomic targets. Helium, which is the simplest noble gas atom, is more interesting because threre is no complications due to core in the DPI process. A number of experiments (5,6,7,9,15,16,17) and calculations (19,20,21,25-29,30) have been carried out for the DPI of He.

From theoretical point of view, the challenging problem is how to describe the two electrons in the continuum i.e. double-continuum wave function. The subject of various forms of the double-continuum wave functions (DCWF) is of growing interest because it is needed to solve a very broad range of problems, for examples, electron impact ionization, double photoionization. Several asymptotic forms of double-continuum wave functions, which are solution of the Schrodinger equation at large distances for two electron systems in different coordinate systems are available in the literature. In brief, we will describe these wave functions. However, the asympototic wave functions are not adequate at small distances. One needs wave functions which are valid in the entire configuration space i.e. double-continuum wave functions which can be obtained solving the Schrodinger equation for two electron systems without imposing any constraint but unfortunately this is not feasible. For this reason, we will describe several models which are valid in different situations.

At very high incident energies, two ejected electrons are very far away from the residual ion, one can argue that these two escaping electrons do not experince Coulomb force. Under this situation, one can describe two electrons by product of two plane waves. Product of two plane waves gives the threshold law which differs completely from the experimental threshold law. No matter, how far or how fast electrons are, they always experience Coulomb force. Hence this model is the crudest model and is used in the first Born approximation. Since, electrons always experience Coulomb force , so widely used model is product of two Coulomb wave functions. This model has been employed in the calculation of the double photoionization of helium and hydrogen molecule. Since, this model does not contain correlation, hence it gives linear threshold law which disagrees with the experiment. Another possible physical situation may occur in intermediate energy range that one electron is slow and another fast. The slower electron may screen the faster one. In this case, one electron can be described by Coulomb wave and faster one can be described by plane wave. This model gives the threshold law which disagrees with the experiment. For reliable calculations of double photoionization cross sections, one needs correlated double continuum wave functions especially in the vicinity of the threshold where electron correalation plays an extremely important role. Such wave functions are not obtained until now.

The main reasons for choosing the near threshold double photoioniztion of helium are:

(1) to test the validity of the modified Altick wave function which includes monopole and dipole interactions

(2) the DPI has the advantage that this is dominated by electric dipole transition and the resulting final state is pure and well defined. In the case of He, the final state consists of alpha particle and two outgoing electrons and

hence there are no complications due to core electrons and offers the best opportunity to test the validity of double-continuum wave functions.
The single photon double photoionization of helium reaction can be written as:

$$h\nu + He(1s^2) \rightarrow He^{++} + e^- + e^-$$

Recently, Tiwary and his co-worker (25-29) have investigated the effect of correlation on the double photoionization of He employing the fully correlated wave function for the ground state and the Altick wave function (24) (which includes only the monopole interaction) for the final state. Result is encouraging relatively in the high energy region. At low energies, the discrepancy is large. Altick (23) has improved his wave function by including the monopole and dipole terms of the Neumann expansion of the Coulomb interaction $1/r_{12}$. In the present work, we have employed the improved Altick wave function for the final state.

2 Method

The expression for the double photoionization of He can be written as (Tiwary (25)):

$$\sigma^{2+}(E_\nu) = \frac{4\pi^2\alpha a_0^2}{E_\nu}\sum_i \int\int d\epsilon d\epsilon' | < \Psi_f(r_4,r_2)|r_1+r_2|\Psi_i(r_1,r_2) > |^2 \delta(E_\nu - Bi - \epsilon - \epsilon') \tag{1}$$

where α is the fine structure constant and a_o is the first Bohr radius. E_ν is the incident energy, and Ψ_i and Ψ_f are the wave functions of the initial and final states involved in the transition. The form of Ψ_i and Ψ_f are

The configuration-interaction wave function which includes correlation is (Tiwary and his co-worker (53-56))

$$\Psi(LS) = \sum a_i \Phi_i(\alpha_i LS) \tag{2}$$

The form of the Byron-Joachain wave function (30) is

$$\Psi(r_1,r_2) = \frac{1}{4\pi}\sum F_l(r_1,r_2)P_l(cos\theta_{12}) \tag{3}$$

where

$$F_l(r_1,r_2) = \sum A_{mn} f(r_1,r_2) \tag{4}$$

Details are given in (30).
We now turn to the problem of representing the final state Ψ_f of He with two continuum electrons and one alpha particle. We have employed the Altick wave function which takes account of the interaction between two electrons via Coulomb interaction. The general form of the Altick wave function is

$$\Psi \approx \rho_1^{i/k_1} e^{ik_1\rho_1} \rho_2^{i(\xi/k_2+\alpha_L)} e^{ik_2\rho_2} + \rho_1^{i(\xi_2/k_2+\alpha_L)} e^{ik_2\rho_1} \rho_2^{i/k_1} e^{ik_1\rho_2} (1 + Q(\rho_1/\rho_2) \tag{5}$$

The above equation can also be written as:

$$\Psi \approx \rho_1^{i/k_1} e^{ik_1\rho_1} \rho_2^{i(\xi/k_2+\alpha_L)} e^{ik_2\rho_2} f_{l_1 l_2 L}(y) + \rho_1^{i(\xi_2/k_2+\alpha_L)} e^{ik_2\rho_1} \rho_2^{i/k_1} e^{ik_1\rho_2 y - \xi_1'} (\frac{1-\xi_1/\xi_2 y}{1-\xi_1/\xi_2})^{\epsilon_1' - \epsilon_2'} \tag{6}$$

$$\rho_i = zr_i, \quad i = 1,2 \tag{7}$$

$$\epsilon = 1 - z^{-1} \tag{8}$$

$$Q = a_0 + \sum a_n (x_1/x_2)^n + \sum b_m (x_2/x_1)^m \tag{9}$$

Finally, Q has been expressed in terms of the hypergeometric functions (see Altick (24)).

Modified Altick wave function (23) which includes the monopole as well as the dipole can be written as follow:

$$\Psi \approx \rho_1^{i/k_1} e^{ik_1\rho_1} \rho_2^{i(\xi/k_2+\alpha_L)} e^{ik_2\rho_2} f_{l_1 l_2 L}(y) + \rho_1^{i(\xi_2/k_2+\alpha_L)} e^{ik_2\rho_1} \rho_2^{i/k_1} e^{ik_1\rho_2} {}_1F_1(\xi_1' - \xi_1; 1; ik_1\rho_2(y-1)) \left(\frac{1-\xi_1/\xi_2 y}{1-\xi_1/\xi_2}\right)^{\xi_1'-\xi_2'} g_{l_1 l_2 L}(y) \tag{10}$$

Details are given by Altick (23-24).

3 Results and discussion

Figure 1 displays our present results of DPICS of He along with the other available experimental as well as theoretical results. For the first time, Byron and Joachain (19) performed theoretical calculation for the DPI of He using uncorrelated wave functions for both the ground state as well as the final state. They found that the probability is too poor for the DPI process. They also calculated the DPI of He using the correlated wave function for the ground state and the uncorrelated wave function for the final state. They described the final state as a product of two unscreened Coulomb wave functions with effective charge two for both electrons. Results of DPI of He with uncorrelated wave functions for both the initial and final states

involved in the transition are extremely small which indicates that the probability of DPI process is very poor without correlation. Results with almost fully correlated wave function for the ground state and uncorrelated wave function for the two continuum electrons are in excellent agreement with the first experiment of Carlson (16). The excellent agreement suggests that the correlation is important in the initial state not in the final state. The most recent experiment of Holland et al (15) disagrees considerably through out the energy range of consideration with the experimental data of Carlson and theoretical prediction of Byron and Joachain. This experimental result suggests that the correlation, probably, is important equally in the both initial and final states involved in the transition matrix element. Results of Holland et al (15) are in accord with the experimental points of Schmidt et al (11) at low energies and tend to lie lower than the curve of Wight and van der Wiel (17). Brown (18) has reevaluated the DPI cross section of He using a Hylleraas-type wave function without decomposition into partial waves and Coulomb function for the final state. His results favour the oldest experimental data and the theoretical results of Byron and Joachain (19). Amusia et al (20) have also investigated this problem in the limit of high, non-relativistic photon energies. Their method leads to a greatly overestimated cross section in the energy range of recent measurements. Yurev (21) and Varanavshikh and Labzovskii (22) have performed the calculations for the DPI cross section of He in the threshold energy region using perturbation and variational methods respectively. Their results are in qualitative agreement with each other, but are limited to the low energy range (not shown in Figure 1). Tiwary (25) has performed calculations for the DPI cross section of He using the position and momentum dipole matrix elements. Tiwary has employed almost fully correlated wave function for the ground state and partially correlated wave function of Altick (24) for the fi-

nal state to evaluate matrix elements. The values obtained from the length formulation tend to lie higher than those obtained using the velocity formulation. Both differ considerably from the first experimental observation and first theoretical prediction and tend to lie close to the recent reliable experimental curve (see Tiwary (25)) of Holland et al (15) especially in the high energy range. There is a considerable discrepancy between theoretical results of Tiwary and experimental data of Holland et al in the vicinity of the threshold. This indicates clearly that (1) inclusion of correlation is extremely important, (2) partial correlation is not adequate to obtain reliable results in the close vicinity of the threshold i.e. two outgoing electrons are very slow and (3) the effect of correlation decreases with the increase of the incident photon energy. It seems to be plausible because when escaping electrons are slow, they have enough time to develope correlation. Carter and Kelly (30) have performed calculations for the DPI cross section of He using the many -body perturbation theory (MBPT) incorporating full correlation in the both initial and final states. Their results are in good agreement in the entire energy range except in the neighbourhood of the double ionization threshold with the most recent and reliable experimental data of Holland et al. (15).

Altick has improved his asymptotic double continuum wave function including the first and second terms of the Neumann expansion of the repulsive Coulomb interaction between electrons ($1/r_{12}$). We have employed the improved Altick wave function for the final state and obtained the encouraging results. Our present result is in better agreement with the most recent experiment of Lablanquie (5) compare to our earlier work(25) but the discrepancy remains in the close vicinity of the threshold. This suggests that the fully correlated wave function is needed in order to obtain accurate results in the close vicinity of the threshold.

4. Conclusions

Great experimental and theoretical advancement has been made in the case of the double photoionization of He but there is discrepancy in the close vicinity of the threshold where the correlation plays an extremely important role. To obtain accurate double-continuum wave function in the vicinity of the threshold is still challenging problem for theorists. Kossmann et al (7) have measured the slope for the DPI of He but there is no theory to evaluate directly the slope. Dynamic screening i.e. energy dependent screening, is crucial especially when two electrons are escaping the positive ion. However, there is no method to include the dynamic screening in the Coulomb wave function, Altick wave function, distorted wave function or similar other wave functions. Unscreened Coulomb wave function gives linear threshold law which disagrees with the experiment. This suggests to develop a new method to incorporate dynamic screening. Angular distribution and energy sharing of two escaping electrons have not been studied but these can offer the best opportunity to test the validity of a theoretical model as the exponent does in the threshold law:

$$\sigma = \sigma_0 \epsilon^{\beta}$$

Extensive theoretical and experimental investigations of the threshold laws for various escape processes, for example, double photoionization, electron impact ionization, in atoms, molecules and ions have been of growing interest because these laws provide answers of many fundamental questions in physics. A number of theoretical (33-46) and experimental (3-7, 11-14) studies of these laws have been made. Our present theoretical investigation demonstrates that the inclusion of the first and second terms of the Neumann series in the Schrodinger hamiltonian and resulting wave function

yield better results than the first wave function of Altick which includes only the first term of the Neumann series. However, the the situation in the close vicinity of the threshold is unstisfactory. This reflects that the electron correlation plays an extremely important role in the DPI process and it is indispensable to incorporate full correlation in both the initial as well as the final states in order to obtain an excellent agreement with reliable experimental observations in He.

There are numerous difficulties in obtaining the accurate double-continuum wave functions. However, we would like to make some constructive and fruitful suggestions for obtaining accurate double-continuum wave functions which may be the future directions:

(1) Very recently, Burke et al (47, 49) have developed a new R-matrix method which may be very useful for the double photoionization and electron impact single ionization of atoms and molecules

(2) the standard R-matrix of Berrington et al (48) may be combined with the Altick asymptotic wave function

(3) the standard R-matrix method may also be combined with the hyperspherical wave function

(4) solving the Schrodinger equation including the higher terms of the Neumann series as Altick did for the first and second which will be a very systematic improvment and will provide to study the DPI process in regular and systematic way

(5) developing some new techniques which provide to include the dynamic screening in the Coulomb, Altick and distorted wave functions

(6) developing sophisticated numerical procedure to describe two continuum electrons.

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References

1. J. A. R. Samson, in Corpuscles and Radiation in Matter, (Edited by W. Mehlhorn), Handbuch der Physik Vol. 32 p. 23, Springer, Berlin (1982).
2. A. F. Starace, in Corpuscles and Radiation in Matter, (Edited by W. Mehlhorn), Handbuch der Physik Vol. 31 p. 1, Springer, Berlin (1982).
3. I. Nenner, P. Morrin and P. Lablanquie, Comments At. Molec. Phys. 22, 51 (1988).
4. P. Lablanquie, J. H. D. Eland, I. Nenner, P. Morrin, J. Delwiche and M. J. Hubin-Franskin, Phys. Rev. Lett. 58, 992 (1987).
5. P. Lablanquie, K. Ito, P. Morrin, I. Nenner and J. D. Eland, Z. Phys. D 16, 77 (1990)
6. P. Lablanquie, Ph. D. Thesis, LURE, Orsay, France, 1989.
7. K. Kossmann, V. Schmidt and T. Anderson, Phys. Rev. Lett. 60, 1266 (1988).
8. B. Crasemann, Comments At. Molec. Phys. 22, 163 (1989).
9. V. Schmidt, Appl. Opt. 19, 4080 (1980).
10. A. F. Starace, Appl. Opt. 19, 4051 (1980).
11. V. Schmidt, N. Sander, H. Kuntzmuller, P. Dhez, F. Wulleumier and E. Kallne

Phys. Rev. A 13, 1748 (1976).
12. S. D. Price and J. H. D. Eland, J. Phys. B: 23, 2269 (1990) .
13. G. C. King, M. Zubek, P. M. Rutter, F. H. Read, A. A. McDowell, J. B. West, and D. M. P. Holland, J. Phys. B: 21, L403 (1988).
14. F. H. Read, J. Phys. B: 17, 3965 (1984).
15. D. M. P. Holland, K. Codling, J. B. West and G. V. Marr, J. Phys. B: 12, 2465 (1979
16. T. A. Carlson, Phys. Rev. 156, 142 (1967).
17. G. R. Wight and M. J. van der Wiel, J. Phys. B: 9, 1319 (1976).
18. R. L. Brown, Phys. Rev. A 1, 586 (1970).
19. F. W. Byron and C. J. Joachain, Phys. Rev. 164, 1 (1967).
20. M. Ya Amusia, E. G. Drukarev, V. G. Gorshkov and M. P. Kazachkov J. Phys. B: 8, 1248 (1975).
21. M. S. Yurev, Opt. Spectros. 38, 4 (1975).
22. S. M. Varnavshikh and L. N. Labzovskii, Opt. Spectros. 47, 24 (1979).
23. P. L. Altick, Phys. Rev. A 25, 128 (1982).
24. P. L. Altick, Phys. Rev. A 21, 1381 (1980).
25. S. N. Tiwary, J. Phys. B: 15, L323 (1982).
26. S. N. Tiwary, ICPEAC, Berlin, West Germany (1983).
27. S. N. Tiwary and G. Dujardin, ICPEAC, New York, USA (1989).
28. S. N. Tiwary, unpublished.
29. S. N. Tiwary, LAMP, ICTP, Trieste, Italy (1990).
30. S. L. Carter and H. P. Kelly, Phys. Rev. A 24, 170 (1981).
31. H. P. Kelly, Phys. Scr. T17, 109 (1987).
32. U. Fano, Rep. Prog. Phys. 46, 97 (1983).
33. G. H. Wannier, Phys. Rev. 90, 817 (1953).
34. A. R. P. Rau, Phys. Rev. A 4, 207 (1971).
35. A. R. P. Rau, J. Opt. Soc. Am. B 4, 784 (1987).
36. R. Peterkop, J. Phys. B: 4, 513 (1971).

37. R. Peterkop, Phys. Lett. A 62, 81 (1977).
38. M. R. H. Rudge and M. J. Seaton, Proc. Roy. Soc. 283, 262 (1965).
39. D. S. F. Crothers, J. Phys. B: 19, 463 (1986).
40. C. H. Green and A. R. P. Rau, J. Phys. B: 16, 99 (1983).
41. H. Klar, J. Phys. B: 14, 3255 (1981).
42. H. Klar and W. Schlect, J. Phys. B: 9, 1699 (1976).
43. J. M. Feagin, J. Phys. B: 17, 2433 (1984).
44. A. Temkin, Phys. Rev. Lett. 49, 365 (1982).
45. A. D. Stauffer, Phys. Lett. A 91, 114 (1982).
46. S. Spence, Phys. Rev. A 11, 1539 (1975).
47. P. G. Burke, C. J. Noble and P. Scott, Proc. Roy. Soc. 410, 289 (1987).
48. K. A. Berrington, P. G. Burke, M. Le Dourneuf, W. D. Robb, K. T. Taylor, and Vo Ky Lan, Comput. Phys. Commun. 14, 367 (1978).
49. T. T. Scholz, H. R. J. Walters, P. G. Burke and M. P. Scott, Mon. Not. R. Astr. Soc. 242, 692 (1990).
50. P. Selles, J. Mazeau and A. Huetz, J. Phys. B: 20, 5183 (1987).
51. T. N. Chang and R. T. Poe, Phys. Rev. A 12, 1432 (1975).
52. J. B. Donahue, P. A. M. Gram, M. V. Hynes, R. W. Hamm, C. A. Fost, H. C. Bryant K. B. Butterfield, D. A. Clark and W. W. Smith, Phys. Rev. Lett. 48, 1538 (1982).
53. S. N. Tiwary, Chem. Phys. Lett. 96, 333 (1983).
54. S. N. Tiwary, Astrophysical Journal 272, 781 (1983).
55. S. N. Tiwary, A. E. Kingston and A. Hibbert, J. Phys. B: 16, 2457 (1983).
56. S. N. Tiwary, A. P. Singh, D. D. Singh and R. J. Sharma, Can. J. Phys. 66, 405 (1988

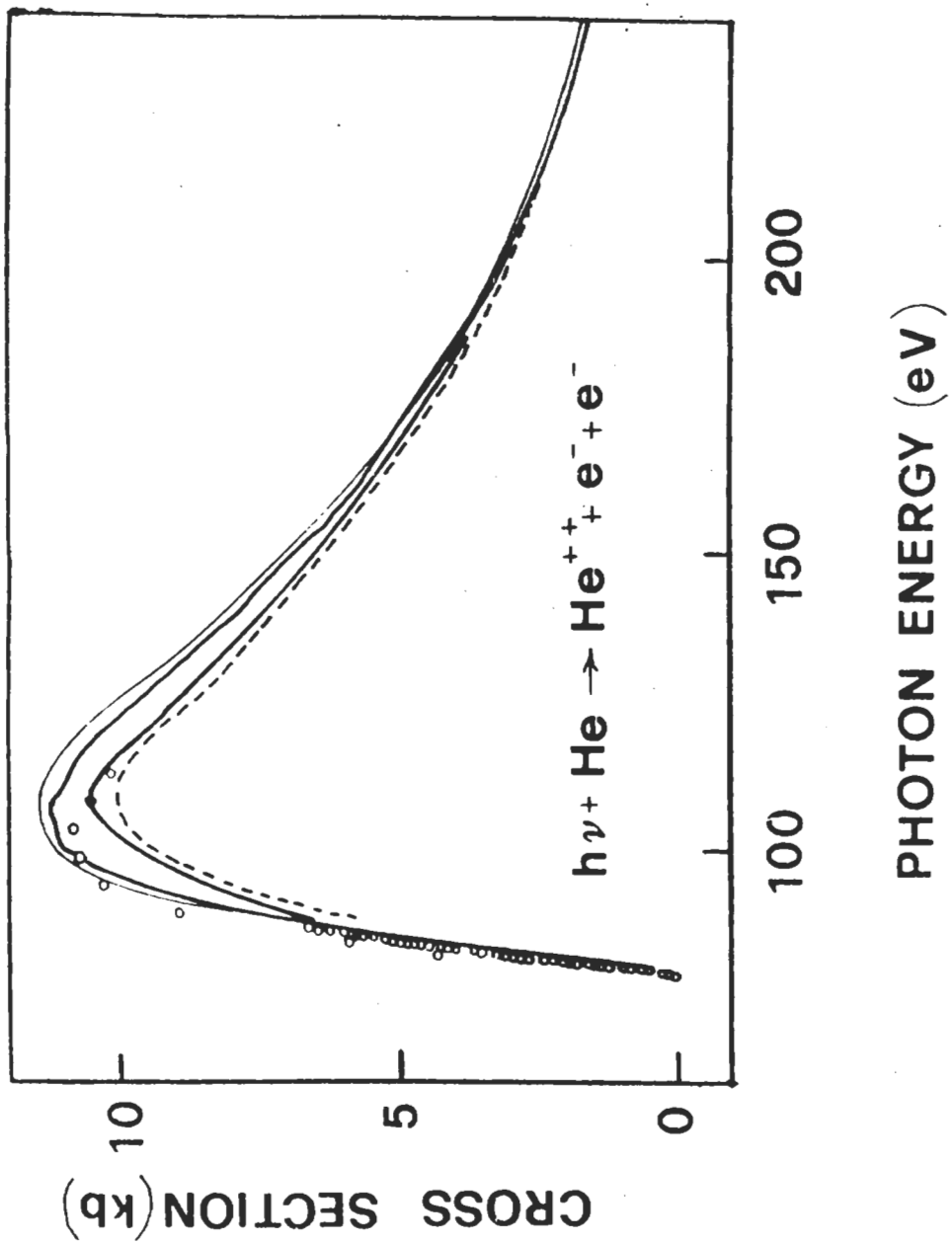

Fig. 1 - Single photon double photoionization cross section in helium; Thickest line: present work; Open circle: Lablanquie et al. (5); Solid thick line: Schmidt et al. (11); Solid thin line: Carter and Kelly (30); Dashed line: Tiwary (25).