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Doctoral Thesis Report. Dr. Attilio Cafolla, Dublin City University.

Candidate: Mr. Lazaros Varvarezos.

Thesis title: Fast and Ultrafast Multiphoton-Multicolour Ionization and Spectroscopy of Small Quantum Systems.

Report on Thesis:

The research presented in this thesis deals with the interaction of laser produced radiation with small quantum systems. The focus of the thesis is on exploring the application of state-of-the-art light sources to problems that are at the forefront of current atomic and molecular science. Three systems were studied and for each a different light sources and experimental technique was employed. The first project reports on the two photon ionization of krypton (Kr) irradiated by light produces in a Free Electron Laser (FEL); the second describes soft X-ray photoabsorption measurements of the carbon and oxygen K-edges in carbon dioxide and methane gases using radiation from a laser produced plasma; while the third is a comparative study of the fragmentation of methane employing radiation from an ultrafast laser. As such these studies are of strong interest both from a fundamental science point of view and also for researchers in diverse disciplines such as, for example, in environmental science (methane gas is a large contributor to the greenhouse gas effect) and planetary science (methane is major component of the atmosphere of Titan).

The thesis is organised into 6 Chapters, together with a Bibliography, and two short appendices concerning technical aspects of the velocity map imaging (VMI) technique and the optimisation protocol for the laser produced plasma soft x-ray source. The overall organisation of the thesis is very clear and logical with the three experimental chapters (4 - 6) written in a concise and convenient 'stand-alone' format. My only criticism of the thesis is that it lacks a concluding chapter summarising the principal results and more importantly to present a brief discussion of the future directions for each of the projects presented therein. I note that each of the experimental chapters concludes with a brief summary of the findings however, the thesis lacks a concluding chapter summarising the principal results and more importantly a brief discussion of the future directions for each of the projects presented. Finally, it is customary for the candidate to provide a list of publications and presentations arising from their work at the beginning of the thesis, I suggest that this be included.

Chapter 1, ("Introduction") is a very brief historical overview of the interaction of light with matter specifically focussing on the interaction of intense laser light with matter. Several examples of light matter interactions in areas of technological importance are given, including extreme UV lithography for integrated circuit production, soft x-ray laser microscopy and coherent diffraction for imaging of biological samples, light detection and laser ranging for remote sensing. There follows a brief discussion of different types of laser light sources and the output parameters that are relevant to the system under study.

Chapters 2, ("Sources in the NIR region") and 3 ("Sources in the UV and SXR regions") provide the background theory and techniques used to generate the laser radiation used in the experiments described in the subsequent chapters. Chapter 2 goes in detail into the state-of-the-art concerning the generation of laser radiation in the visible and near infrared region of the spectrum. The principles of operation of the two laser sources used in this work, (Ti:sapphire and Nd:YAG), are presented. The Q-switching and mode locking techniques used to generate nano second and femto second laser pulses are described. Chapter 3 concerns a number of laser based XUV and soft X-ray radiation sources. This chapter introduces the principle of operation of the free electron laser (FEL) with emphasis on the microbunching and self-amplified spontaneous emission (SASE) processes. The discussion in this chapter is cast in the framework of the FLASH_FEL facility in Hamburg where the experiments described in Chapter 4 were performed. The second part of this chapter describes the generation of extreme UV and soft X-ray radiation using laser produced plasmas incorporating a double stream gas puff target. Radiation produced by a LPP source was used for the studies presented in chapter 5.

Chapter 4, ("VUV ionisation of Kr"), reports the experimental results of a two-photon double ionisation (TPDI) of Kr irradiated by FEL pulses at 25.2 eV. The background theory of TPDI is described with emphasis on the angular distribution of the emitted photoelectrons as characterised by the beta parameters β_2 and β_4. The angular distributions of the photoelectrons were measured using the velocity map imaging (VMI) technique at beamline B1 at FLASH. This technique permits measurement of the angular distributions at the first and second ionisation steps as a function of the FEL intensity and, through angular integration, the photoelectron spectrum of the emitted electrons. In this experiment it is possible to resolve the spin-orbit components of the angular distributions. The work reported here is ahead of current theory in this field however, the results are in good agreement with reported theoretical calculations for the spin-orbit averaged case. Theoretical calculations are ongoing for the spin-orbit split components of the angular distributions.

Chapter 5, ("Low Temperature Photoionised Plasmas"), reports on soft x-ray absorption (XAS) measurements of the carbon and oxygen K-edges in carbon dioxide (CO_2) and methane (CH_4) gas targets. By varying the distance between the gas target and the radiation source (emission from a laser produced plasma), it was possible to measure the XAS spectrum of both the neutral (non-ionised) molecules and the photoionised molecules. Using this comparative technique the spectral signatures of several ion species were identified in the ionised molecules, (e.g. C^+, $C2^+$, O^+, O^{2+}). To confirm the identity of these ion species calculations were carried out using the Cowan atomic structure codes.

Chapter 6, ("Molecules irradiated by intense laser fields") is a comparative study of the fragmentation of methane induced by ultrafast laser fields. The ultrafast pulses are produced by a Ti:sapphire laser operating at the fundamental wavelength of 800 nm and, through second harmonic generation in a Barium Borate (BBO) crystal, at 400 nm. The ion fragments produced by irradiation ate detected using a time of flight (TOF) spectrometer in conjunction with a VMI spectrometer. The results show that there are significant differences in the fragmentation process between the two wavelengths at low laser intensities (< 1.7×10^{14} W.cm^{-2}). At higher laser intensities (> 4.0×10^{14} W.cm^{-2}) a forward/backward structure is observed in the H^+ and H_2^+ fragments at 800 nm, indicating a Coulomb explosion process is occurring. For the same measurement at 400 nm the forward/backward component is present in the H^+ signal but is absent in the H_2^+ signal which suggests the absence of the Coulomb explosion process indicating that the fragmentation process is different for the two wavelengths employed.

Three appendices complete the manuscript, appendices A and B are technical noted describing the velocity mapping Imaging (VMI) technique and the optimisation of the laser produced plasma source the final appendix is a comprehensive list of references used within the main body of the manuscript and contains relevant literature related to the topic of the thesis.

Overall, the work reported in this thesis is original and well in-line with open issues currently of interest in atomic, molecular and laser physics, in addition to being of interest to the broader scientific community. The candidate has made significant contributions to several hot topics in his area of research. The thesis is very well written and contains very few typographical errors, however there are some minor corrections that could be made to improve the manuscript, in particular attention should be paid to some of the figures where the font size in the annotation is too small to read easily, for example, figures 5.8 and 5.10.

In conclusion, the submitted manuscript demonstrates that the candidate is now prepared to defend his research project. The research presented in this thesis clearly demonstrates the candidate's ability to perform research to a remarkably high standard and I recommend that he be awarded the degree of Ph.D.

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