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Assessment of dissertation
Fast and Ultrafast Multiphoton-Multicolour Ionization and Spectroscopy of Small Quantum Systems

Submitted by
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Introduction
The dissertation entitled "*Fast and Ultrafast Multiphoton-Multicolour Ionization and Spectroscopy of Small Quantum Systems*" is written by M. Sc. Lazaros Varvarezos, and describes experimental work carried out under the guidance of Prof. John Costello from Dublin City University and Dr. hab. inz Andrzej Bartnik from Military University of Technology.

The dissertation comprises 112 pages of scientific text and is structured in six chapters followed by two appendices and the bibliography with 201 references. Additional 13 pages in the beginning contain the declaration of authorship, an abstract, lists of figures and tables as well as the content. A final page is dedicated to the outcome from the work: M. Sc. Lazaros Varvarezos is first author on two papers sent to J.Phys.B. where one is accepted and the other under review. In addition he is coauthor on two further papers, where one is published in New J. Phys. and the other is under review at J.Phys.B.. Furthermore he contributed to eight conferences with his work.

Assessment

The Einstein one photon ionization process of valence shell electrons is well understood and at the foundations of quantum mechanics. Nowadays lasers achieve high intensities so that new ionization regimes can be explored going from multiphoton ionization to field ionization. Free electron lasers deliver high intensities in the VUV spectral region, allowing for multiphoton ionization processes in this spectral region as well. Laser produced plasma sources deliver photons in the soft x-ray regime and are therefore suited as lab based instruments to explore inner shell ionization processes of molecules. This dissertation focuses on these aspects experimentally with the help of model systems like atoms or small molecules. To that end, after an introduction, the light sources are described in chapter 2 and chapter 3 and the mentioned prototype experiments are reported in chapters 4 to 5. In the following each chapter will be shortly commented on.

Chapter 2 is a useful compilation of textbook knowledge paired with illustrative pictures from laser manuals of the existing Ti:Sa amplifier system at DCU and will help newcomers to the lab to understand the physics behind the technology. In the context of the thesis this is the high intensity near infrared (NIR) light source. In the corresponding experiments at European XFEL, the Ti:Sa laser installed there was used.

Chapter 3 now describes sources in the vacuum ultraviolet (VUV) and soft x-ray (SXR) region. Here the physics behind free electron lasers with an emphasis on FLASH is illustrated and only very shortly the physics and technology of the laser produced plasma (LPP) sources is addressed. The latter technology is then addressed more complete in Chapter 5.

In Chapter 4 two photon ionization experiments at FLASH on Kr at different intensities (below an order of magnitude) at a wavelength of 25.2 eV are described. Making use of a velocity map imaging spectrometer (VMI) the photoelectron angular distributions are recorded and analysed. Whereas the introduction, the report on state of the art, the description of the experiment and the data analysis are very well done, the physics of the observed trends in angular distributions is only merely touched and referred to (ongoing) calculations. The presented data sets will serve as a benchmark for the theory development.

SXR absorption experiments on CO_2 and CH_4 molecules around carbon and oxygen K-edges are reported in Chapter 5. Here the LPP source is employed and described in more detail. The $1/r^2$ scaling of this point source is used to disentangle contributions from neutral absorption and ionized targets. This is the chapter that is also covered in the published J.Phys.B. paper mentioned above.

Results on femtosecond laser ionization and fragmentation at 800 nm and 400 nm central wavelength on the CH_4 prototype using the Laser at the European XFEL are reported in Chapter 6. Intensities are varied in a Keldysh range around one and different fragmentation patterns where observed when comparing the 800 nm excitation to the 400 nm excitation. In the discussion different ponderomotive energies of the recolliding electron are taken into account. The set of data sheds additional light on the difficult regime between multi photon ionization and field ionization.

Recommendation

In conclusion regarding the new contributions to science and the concise written form of the PhD together with the very illustrating and helpful figures this PhD thesis fulfils with all respects the requirements for the award of the PhD degree.

(Thomas Baumert)