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# Single Shot correlation in VMI- TOF measurements on He nanodroplets at MIR femtosecond laser pulses

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# Abstract

In this thesis a new data acquisition method is explained for correlate single shot VMI-TOF measurements on He droplets ionized by a MIR femtosecond laser. Until now VMI images and TOF data are always treated in a statistical way. With these new method we expect to have some better correlation for single explosion and relate each of the event from an individual way, having specific information that could be lost in the statistical method. The correlated data is acquired by triggering the VMI camera and the TOF oscilloscope with the laser trigger, so both acquisitions begin and end after the same laser pulse. Results for the energies.....here a resumé of the results.....

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# List of abbreviations

|               |   |
|---------------|---|
| <b>ATI</b>    | above threshold ionization                              |
| <b>BSI</b>    | barrier suppression ionization                          |
| <b>CCD</b>    | Charge-coupled Device                                   |
| <b>CPA</b>    | Chirp Pulse Amplification                               |
| <b>CWL</b>    | Central wavelength                                      |
| <b>EM</b>     | Electro Mechanics                                       |
| <b>LASER</b>  | Light Amplification by Stimulated Emission of Radiation |
| <b>LT</b>     | Langmuir-Taylor   |
| <b>MCP</b>    | Micro Channel Plate                                     |
| <b>NIR</b>    | Near Infrared   |
| <b>pBASEX</b> | polar Basis Set Expansion                               |
| <b>PID</b>    | Proportional – Integral – Derivative                    |
| <b>TBR</b>    | Three Body Recombination                                |
| <b>TOF</b>    | Time of flight  |
| <b>VMI</b>    | Velocity Map Imaging                                    |
| <b>VUV</b>    | vacuum ultra violet                                     |
| <b>XUV</b>    | Extreme ultraviolet                                     |

## 0.1 Introduction

Physicists have always wonder to explain and resolve dynamic processes in short scale times, so initial conditions of processes can be describe in a time evolution scale. Describe any system like this requires to acquire data in shorter windows of time, for example a film is only a consecutive sequence of photographs that recreate a large time laps in a smaller time scale pics. For atomic physics, we are talking about a micro-cosmos that varies from microseconds, i.e several bodies dynamics, to attoseconds for atoms, where time scales can go down to  $10^{-9}$  s, requiring to create measurement methods capable to record in shorter time, while the experiment have to be done in a controllable way to ensures its reproductivity, as any scientific method.

The time window of dynamics of a sytem is related to quantum dynamics, in a simple view also to its size. For dynamics happening in a molecule or a many body system interaction, the time window can oscillate between microseconds to femtoseconds, although for millielectronvolt-scale (*meV*) energy spacing of vibrational energy levels implies that molecular vibrations occur on a time scale of tens to hundreds of femtoseconds. The motion of individual electrons in semiconductor nanostructures, molecular orbitals, and the inner shells of atoms occurs on progressively shorter intervals of time ranging from tens of femtoseconds to less than an attosecond. Motion within nuclei is predicted to unfold even faster, typically on a zeptosecond time scale.

To achive this high resolution in space and time physicist have challenged to create systems with a well controlled spatial and temporal gradient. Fortunately nowadays, laser pulses can research up to extreme non-linear optical processes, producing single aisolated pulses of ultra violet(UV) waves as short as 67 *as* [4]. Such fast pulses open up the possibility of time resolved measurements fort short processes like electron dynamics. However, to do this, experimental schemes must be devised that allow these new light sources to be used to perform measurements on the microcosmos. In particular, in the last few years, many studies at atom- and molecule-clusters had been published, From mid-infre red (NIR) interaction to UV or XUV pulses, that not just lead to a broad spectra to study but also to a large range of possible applications such as the generation of energetic electrons and ions in the keV-regime [1], as well as intensive XUV and attosecond pulses [3]. Laser pulses with peak intensitiesof up to  $10^{21}$ W/cm<sup>2</sup> are available nowadays [2] comercially so the difficulty and expensive of the experiments source also are easy.

But Having this is never enough, Lasers is just one huge step in order to control and ignite atomic processe in controlled standard. The other step needes is to

acquire the information we want from this process. For this purpose several techniques are available depending on the nature of the process. For this particular work we are interested in two particular techniques, Velocity map image (VMI) and Time of flight (TOF). Since its invention, these two techniques have become two of the most common and important measurement techniques in high energy physics. But detecting a signal is just one part of the job, the new laser advances like the generation of coherent high-intensity laser pulses with intensities up to  $10^{22} \text{ W/cm}^2$  allow multiphoton ionization that allows to get time resolved measurements. These advances have enabled the development of new research areas, as well as the investigation of ultrafast dynamics in highly excited matter to nanometer size.

In this thesis we focus our efforts on the ionization process by NIR femtosecond pulses in doped clusters from *He*. The interaction of the doped He droplet with the Laser field results in an energy transfer to the droplet that ignites an ionization process, known as a nanoplasma. This resonant interaction of the laser field with a collective oscillation of the electrons in the plasma is driven by the laser field [1]. This process, caused predominantly by electron impact ionization, makes an avalanche-like ionization of the atoms in the cluster, leading to a heating of the plasma and, as a result, to hydrodynamic expansion and Coulomb explosion of it. To analyze this process two techniques were used to study the electrons as well as the ions resulting in the Coulomb explosion. A velocity map imaging and a Time of flight technique are set up in parallel to acquire the data and reconstruct the initial energies and configuration of the plasma in study. In the First chapter we will present a brief introduction to short plasma interactions and a basic background of Coulomb ionization in order to understand the physical meaning of the reaction. In the second chapter a more detailed explanation of the set up used is done, showing from the creation of the He droplets process to the detection process, going through the doping, and ignition process. For the third chapter a detailed explanation on the correlation method for the VMI-TOF measurements is done, and showing the process of the data acquisition and its advantages. In the fourth chapter we present the correlated data and its analysis. Finally the last chapter we present the conclusion of the experiment itself also as the data analysis and future works will be needed to improve this process as well.

I. INTRODUCTION Ion imaging techniques in the field of molecular reaction dynamics photofragment imaging, photoelectron imaging, reaction product imaging have proven to be of high value. 1-3 Especially, the capability of probing the full three-dimensional velocity distribution of scattered particles under study in a single image has contributed to the importance of this method, which has the multiplexing

advantage of detecting particles ions or electrons of all velocities the term velocity refers to the vector quantity whereas speed denotes the scalar! simultaneously. Because the detection of particles often involves multi-photon ionization MPI schemes the ion imaging technique is widely applicable, and is shown to compare well with established one-dimensional 1D time- of-flight TOF 4–6 and Doppler methods.<sup>7,8</sup> As in conventional TOF/MPI techniques, the images can often be obtained in a mass and internal state selective way and, in addition, they can provide information on orientational and alignment effects. <sup>10</sup> In contrast to the conventional time-of-flight method, where kinetic energy release information is contained in the temporal structure in the arrival period of electrons or ions of a specific mass, the ion imaging technique extracts all information kinetic energy and angular distributions from the spatial appearance of the two-dimensional 2D! image. From an image the full three-dimensional 3D information can be reconstructed by means of an Abel inversion or back projection method.<sup>11</sup> This also implies that the kinetic energy resolution attainable is ultimately limited by the quality of spatial mapping by the detection system. In some cases imaging appears less favorable compared to TOF methods in this respect. In order to exploit the imaging method to its full potential one needs to explore methods for improvement of the spatial quality of the 2D image. Since the mapping of 3D distributions of charged particles onto the 2D detector is particularly dependent on the electrode configuration used to form the extracting electric field, this study is concerned with the comparison of conventional grid electrodes versus the application of a simple three-plate electrostatic lens with open electrodes, which can be classified as an “asymmetric immersion lens.” It shall be pointed out how this lens avoids distortions commonly present in imaging with grids, along with having additional appealing features. It turns out that the ion lens can be operated such that particles with the same initial velocity vector are mapped on the same point on the detector, irrespective of their initial distance from the ion lens axis. A more accurate description for the imaging technique using electrostatic lenses is therefore velocity map imaging

## chapter Theoretical Background

In this chapter we will present all the theoretical background necessary for the development of this project, from the theory and creation of the He droplets to the physics behind the plasma and coulomb explosion process to the detection techniques. In order to guide the reader in an organized way, the chapters are organized in a way that follow the processes necessary to the performance of the experiment. This means that all the chapters explained in here occur in the same order during the experiment.

## 0.2 Helium Nanodroplets

The combination of cryogenic matrix isolation, discovered in 1954 [?], and the now well defined properties of Helium (*He*), specially its superfluidity first discovered in 1937 by *Kapitza et. al.* [?], have as a consequence one of the most powerful and flexible tools in chemistry [?]. Helium droplets can grow from a cluster of a few thousand atoms up to  $10^8$  of atoms, and can go to ultra cold temperatures (close to 0.37 K [?]) [?]. Two main advantages of this cooling properties arise. First, dopants in the He nanodroplet are set to their absolute vibronic ground states, avoiding all other possible spectra and stabilizing the cluster in a specific state. More important, the fast cooling helps to the formation of isomers that are difficult or impossible to generate with other methods [?]. Second, because the superfluid phase of the He nanodroplets [?], the bond between dopants and He is weak. Therefore, in contrast to spectroscopy in other matrices with higher temperatures, the optical transitions of many dopants are barely influenced by the He matrix [?].

A helium atom has two electrons. This means that the chemical properties of this element, everything important is said, because everything else opens up. Directly from this fact: The electron configuration of the ground state is  $1s^2$ . So it has a completed shell and is therefore a noble gas. Since with helium just the first shell it is the noble gas with the most strongly bound electrons. Thus, helium is also the smallest of all atoms. The first excitation energy of helium is 19.8 eV, the first ionization energy is 24.6 eV, for the ionization of the second electron one needs additionally 54.4 eV [NIS13]. Helium has therefore in the complete visible spectrum no transitions from the ground state. Through the noble gas configuration, helium has a spherically symmetrical electron distribution. It can hardly be polarized and is the least reactive of all the elements. Neutral Helium can only form chemical bonds via Van-der-Waals forces, covalent bonds do not exist. Therefore, helium forms only very weakly bound complexes as helium dimers. Helium dimers are bound with



only 0.0001 meV. For complexes from several helium atoms the binding energy per atom slowly increases until it reaches the value of bulk helium<sup>1</sup> for more than 10 000 atoms, wherein

Understanding the properties of matter starting from the interplay of atoms and molecules has been achieved to a great deal by the study of small or model systems containing only a few atoms. A detailed view on geometric as well as electronic properties has been brought forward largely through the application of spectroscopic tools. These tools are continuously being improved, in particular with the aid of the availability of sophisticated new laser systems, setting new milestones in terms of repetition rate, power and the time and frequency structure of ultrashort pulses. On the other hand, the path from small model systems to complex functional structures is arduous to climb. In recent years, it has become clear that one approach towards the understanding of complex structures of atoms and molecules is to start with well-defined structures and well-defined distributions of populated states. This calls for spectroscopic studies at very low temperatures and now a new field emerged dealing with cold molecules or ultra-cold chemistry. In this regard, spectroscopic experiments involving helium droplet beams (HElium NanoDroplet Isolation (HENDI)) proved, since their introduction in 1992 [ 1 ], to be a versatile method that provides temperatures below 1 K and offers the possibility of studying well-defined and complex structures of atoms and molecules. Moreover, the quantum nature and in particular the superfluid properties of the droplets allow one to investigate these quantum properties in a size-limited aggregate on the nanometre scale.

## List of Figures

## List of Tables

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# Danksagung

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## Erklärung

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