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Thesis To Applied for the Grade of Master of Science in Applied Physics

Albert-Ludwigs-Universität Freiburg

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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. Whit these new method we expect to have some better correlation for single explosion and relate each of the event from a 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 begging and end after the same laser pulse. Results for the energies..............here a resumy of the results..........

Contents

List of abbreviations

ATI above threshold ionization

BSI barrier suppression ionization

CCD Charge-coupled Device

CPA Chirp Pulse Amplification

CWL Central wavelength

EM Electro Mechanics

LASER Light Amplification by Stimulated Emission of Radiation

LT Langmuir-Taylor

MCP Micro Channel Plate

NIR Near Infrared

pBASEX polar Basis Set Expansion

PID Proportional – Integral – Derivative

TBR Three Body Recombination

TOF Time of flight

VMI Velocity Map Imaging

VUV vacuum ultra violet

XUV 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 reproductivility, 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 fentoseconds, 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 higth 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, produccing single aisolated pulses of ultra violet(UV) waves as short as 67 as [21]. 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 [5], as well as intensive XUV and attosecond pulses [14]. Laser pulses with peak intensitiesof up to 10^{21} W/cm^2 are available nowadays [12] commercially so the difficulty and expensive of the experiments source also are easy.

But this is never enough, Lasers is just one huge step in order to control and ignite atomic processes in controlled standard. Other step needed is how to acquire the information we want. For this purpose several techniques are available depending the nature of the process. For this particular work we are interested in two techniques, Velocity map image (VMI) and Time of flight (TOF). Since its invention, this techniques has become two of the most commune and important measurement techniques in high energies 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 $10^{22} W/cm^2$ 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 Mid Infrared (MIR) femtosecond pulses in doped He clusters. The interaction of the dopant with the Laser field result in a energy transfer to the droplet that ignite a 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 [5]. 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. To the analysis of this process we studied the electrons as well as the ion's 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 the Droplet He generation, a short plasma interactions as a basic background of coulomb ionization in order to understand the physical meaning. 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 its detection , going thorough 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 set-up 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 needed to improve this process as well.

1 Theoretical Background

In this chapter we will present all the theoretical background necessary for the development of this project, from the theori and creation of the He dropletrs to the physics behind the plasma and coulomb explosion process to the detection techniques. In order to guuide the reader in an organised way, the chapters are organized ina way that follow the proceses necessaries to the performance of the experiment. this means that all the chapters explained in here occurrence in the ssame order during the experiment.

1.1 Helium Nanodroplets

The combination of cryogenic matrix isolation, discovered in 1954 [20], and the now well defined properties of Helium (He), specially its superfluidity face discovered in 1937 by *Kapitza et. all* [10], have as consequence one of the most powerful and flexible tool in physics, the helium nanodroplets. Helium nanodrops have unique properties that makes it very suitable for the cluster and nanophysics experiments in the last decades. For example, they do not exhibit any optical transitions in the entire infrared, visible and ultraviolet range. They can readily pick up atoms and molecules and form complexes from the species embedded in their interiors, or on their surfaces and act as a ideal matrix for atom, molecules and clusters isolation. [15] [18]. The size of a He cluster can go from of a few thousands up to 10^8 of atoms, and reach temperatures at ultra cold temperature regime (close to $0.37K$ [17]) [4]. 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 espectra and stablishing 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 [13]. Second, because the superfluid fase of the He nanodroplets [6], 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 [18]. The theory of He superfluidity will not be part of this section, this imformation is well documented in other sources, and here we are based on ref. [4] where all theory is well presented to the reader. In the next section we will dedicate a bigger effort on explain the theoretical and technical background of the He nanodroplets creation as well as the physical and technical process to doped it.

1.1.1 He Nano droplets production

At room temperature, helium is a light inert gas. It is odorless, colorless, tasteless, and after hydrogen, the second most abundant element in the universe. [4]. It have a simple 2 atoms structure, exhibing numerous exotic phenomena whose theoretical descriptions are rather complex in many cases, i.e it characteristics of a quantum fluid. From helium exist two stable isotopes ^3He and ^4He. ^4He has two electrons, two protons and two neutrons, no nuclear spin and no total spin, pertaining to the bosonic family, while ^3He with only one neutron has a spin of $I = 1/2$ and belongs to the fermions [1].

The bosonic state $_4He$ is specially of interest, at temperature T⩽2.8K and under normal pressure has a phase transition from "normal liquid" He-I to super liquid He-II [16], in which the helium can be described by a Bose-Einstein condensation. Even the fermionic ^3He exhibits this phase transition at T⩽ $0.03K$ [8].

The superfluidity of $He - II$, at temperatures close to absolute zero, brings with it some unique features. The essential Properties for this include an almost disappearing viscosity in the superfluid phase, weak interaction, very efficient cooling, and the Transparency for electromagnetic radiation up to wavelengths in vacuum ultraviolets (VUV) Spectral range [4]. 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 [11], it can hardly be polarized and is the least reactive of all the elements.

1.1.1.1 Helium Droplets

The production He droplets had to overcome first one principal problem, its liquefaction. At the end of 19th century many gases were liquefied for the first time by applying pressure at room temperature. However, for He and hydrogen, this method was not successful. In 1922 Kamerlingh Onnes reached temperatures below $1K$ by reducing the vapor pressure above liquid helium to about $2*10^{-5}$ bar with a series of pumps [3]. The Joule–Thomson effect [19] is in this case the responsible for Onnes experiment to reach this low temperatures. The basic idea is that under suitable conditions a gas in expanding performs work against its internal forces. Basically the gas is expanded through a small nozzle thermally isolated from its surroundings. The expansion under theses conditions takes place at constant enthalpy, since the expansion nozzle performs no work. following the next relation:

$$W = H_1 - H_2 = (U_1 + p_1V_1) - (U_2 + p_2V_2) \tag{1.1}$$

where H is the entalpy before and after, $U = \frac{3}{2}Nk_bT$ for ideal gases and $pV = Nk_bT$ [4]. Under Joule–Thomson effect conditions, $W = 0$ so $H_1 = H_2$, this expansion leads to a cooling or a warming and under certain conditions, becomes supersaturated. As a result, condensation takes place and a beam of clusters is formed.

Helium nanodroplets are typically produced by a continuous or pulsed adiabatic Expansion of pre−cooled helium through a small aperture from a reservoir into a vacuum [15]. In this process a droplet jet is formed, and its characteristics (blasting speeds and size distribution) can be changes due the manipulation of the set−up. For example, $\triangle$ pressure between the reservoir and the vacuum chamber (usually in the range of a few to $10MPa$) , the nozzle temperatures(from a few K to $T \leqslant 40K$) or the nozzle size (with pinholes of diameter rounding $5-20\mu m$).

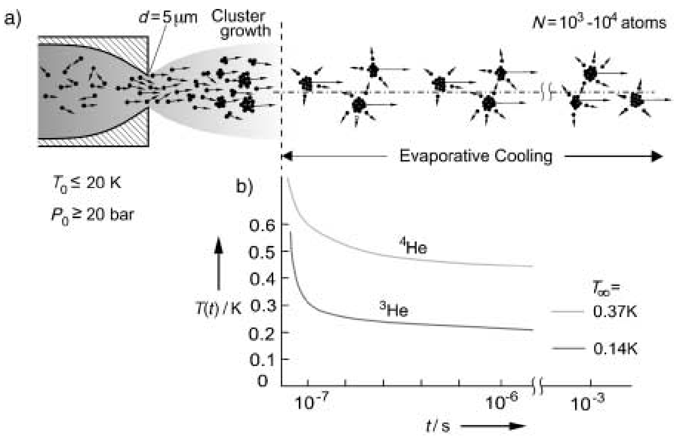

Figure 1.1: a) Schematic representation of the processes leading to the formation and subsequent cooling of helium droplets in a gas expansion. b) Calculated dependence of the droplet temperature on time for ^4He and ^3He droplets after they have left the cluster, taken from [18]

When the Helium expands from the nozzel, its thermal energy is transform in kinetic energy of a supersonic flow field. After the expantion into the vacuum, the gas becomes supersaturated and condensations starts to occurs, creating the beam clusters. This clusters are made of atoms or moleulces held togueter by Wannder wals fores, in this case He-He interaction, that share the same kinetic vector. This means that the two particules travel as close and parallel to each other that a

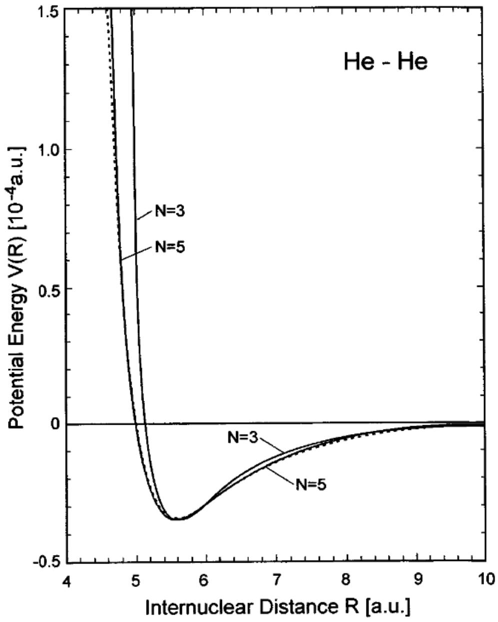

Figure 1.2: Waan der Wals potential for He-He interaction

bonding is possible, see fig 1.2. From the reference frame of the cluster, each of its molecules are close to cero movments, in He this enhace the conditions to be liquid and in consecience superfluidity is achive [7].

There is no mathematical approach of the physics behind this cooling expansion but usually, Raleigh scattering measurements in combination with an empirical scaling law [7] are used to estimate the mean cluster size giving a certain degree of control over the cluster size distribution by adjusting the nozzle width and the source pressure. The droplet size distribution during supersonic expansion in the follows a log-normal distribution of the form [9].

$$p(N) = \frac{1}{\sqrt{2\pi}N\sigma} \exp\left[-\frac{(ln(N/N_0)^2}{2\sigma^2}\right] \tag{1.2}$$

Where N is the number of atom in the cluster, σ is the distribution width and N_0 is the most likely numbers of atoms. Following it give a mean value.

$$\bar{N} = \exp\left(\mu + \frac{\sigma^2}{2}\right) \tag{1.3}$$

With a half width maxima of [9]

$$\sigma N_{\frac{1}{2}} = \exp\left(\mu - \sigma^2 + \sigma\sqrt{2ln(2)}\right) - \exp\left(\mu - \sigma^2 - \sigma\sqrt{2ln(2)}\right) \tag{1.4}$$

As show in Figure **??** The conditions in the He (pressure, temperature and nozzle

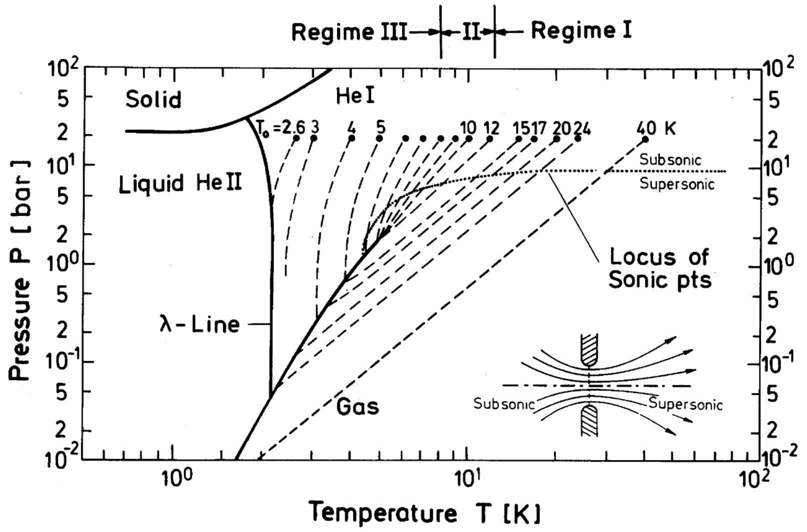

Figure 1.3: Expansion regimes. Pressure-Temperatur phase diagram for ^4He for Nozzle beam expansions starting from a stagnation of 20 bar and a temperatures. As dicusse, quialitatively different behaivors are shown for the regime I - II and II where starting in the gas phase, near the phase trnasition respectivelly. taken from [2].

size) in the free expansion will determine the characteristics of our final He beam. From Here three main regimes can be define.

In regime I or subcritical expansion, begins in the gas phase and leads to droplet formation via condensation. this is the case of most expansion since the pressure are located below the critical pressure P_c. Regime II, also called as critical expansion, is basically and interminable regime that includes all trajectories which are near the critical point, leading to random expansion and difficult control of the beam due the large fluctuations in density. Regime III, the supercritical expansion, starts at low temperatures where the He stops behaving as an ideal gas, expecting flashing or cavitation breaking up the liquid drops jet. [2]

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Danksagung

An dieser Stelle Danke

Erklärung

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