Studies of intense Laser driven Cluster Explosions

by

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To my father
Acknowledgments

The experiments described in this work would not have been possible without the great help of many people. Unfortunately it is only possible here to name a few.

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Recent studies of the explosions of atomic clusters driven by high intensity femtosecond laser pulses have shown that at a sufficiently high intensity and fast laser rise time, the clusters can be stripped of all the electrons. The subsequent explosion is caused by the mutual Coulomb repulsion of the remaining ions. Assuming that the sudden inner cluster ionization approximation (SICI) is applicable, at the temporal onset of the Coulomb explosion all of the relevant electrons are already removed from their host atoms and become unbound electrons. Consequently the ions start from near their initial equilibrium positions in the cluster and expand isotropically. This simple model has been widely applied and has been successful in describing the ion energies observed from the explosions of small clusters (< 1000 atoms) with low charge states (like hydrogen and deuterium). Based on a theory developed by Breizman and Arefiev, if ion and electron motion are not separable, a dependence of the resulting ion energies to the laser polarization should occur. This is caused by a lowered average charge along the laser polarization due to electron motion in the external electric field. This thesis shows experimental results observing this effect.

Furthermore Last and Jortner recently have proposed, based on par-
ticle dynamics simulations that heteronuclear clusters, with a mixture of heavy and light ions under certain conditions will not explode by the simple, equilibrium Coulomb explosion model but that dynamic effects can lead to a boosting of energy of the lighter ejected ions. In this thesis the experimental confirmation of this theoretically predicted ion energy enhancement in methane clusters is presented.
Contents

Acknowledgments v

Abstract vii

Chapter 1 Introduction 1

Chapter 2 Physics of Laser Cluster Interaction 3
  2.1 High Intensity Interactions with Atoms 4
  2.2 Ionization Mechanisms in Clusters 8
  2.3 Models for Cluster Explosions 9
    2.3.1 Coulomb Explosion 10
    2.3.2 Hydrodynamic Explosion 14
    2.3.3 The intermediate Regime 16
  2.4 Heteronuclear Clusters 19
  2.5 Clustering in a Gas Jet 22

Chapter 3 Experimental Apparatus and Techniques 24
  3.1 Introduction to the THOR Laser 25
  3.2 The Gas Jet 28
  3.3 The Vacuum Chamber 30
List of Tables

2.1 Kinematic parameters $\eta$ for methane . . . . . . . . . . . . . . . 20
2.2 Hagen parameter $k$ for different gases . . . . . . . . . . . . . . . 23
# List of Figures

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>Enhanced tunnel ionization in the presence of an external field</td>
<td>6</td>
</tr>
<tr>
<td>2.2</td>
<td>Ion spectra for a cluster Coulomb explosion</td>
<td>13</td>
</tr>
<tr>
<td>2.3</td>
<td>Cluster heating rate</td>
<td>15</td>
</tr>
<tr>
<td>2.4</td>
<td>Electrons confined in a spherical cluster in presence of an external field</td>
<td>18</td>
</tr>
<tr>
<td>2.5</td>
<td>Simulated energy distribution after a heteronuclear cluster</td>
<td>21</td>
</tr>
<tr>
<td>3.1</td>
<td>Schematic drawing of the THOR laser</td>
<td>27</td>
</tr>
<tr>
<td>3.2</td>
<td>Schematic drawing of the gas jet</td>
<td>29</td>
</tr>
<tr>
<td>3.3</td>
<td>The vacuum chamber</td>
<td>31</td>
</tr>
<tr>
<td>3.4</td>
<td>Photon and electron peak in TOF signal</td>
<td>35</td>
</tr>
<tr>
<td>3.5</td>
<td>Typical TOF trace for hydrogen clusters</td>
<td>37</td>
</tr>
<tr>
<td>3.6</td>
<td>Typical TOF trace for argon clusters</td>
<td>38</td>
</tr>
<tr>
<td>3.7</td>
<td>The MCP detector</td>
<td>40</td>
</tr>
<tr>
<td>4.1</td>
<td>Different TOF signal types in the $H_2$ data</td>
<td>46</td>
</tr>
<tr>
<td>4.2</td>
<td>$H_2$ TOF traces for different laser polarizations</td>
<td>48</td>
</tr>
<tr>
<td>4.3</td>
<td>$H_2$ energy spectra for all three measured polarizations</td>
<td>51</td>
</tr>
<tr>
<td>4.4</td>
<td>Averaged TOF trace for $CD_4$</td>
<td>54</td>
</tr>
<tr>
<td>4.5</td>
<td>Comparison of averaging methods</td>
<td>56</td>
</tr>
<tr>
<td>Section</td>
<td>Title</td>
<td>Page</td>
</tr>
<tr>
<td>---------</td>
<td>-----------------------------------------------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>4.6</td>
<td>$CD_4$ energy spectrum to TOF trace from Fig. (4.4)</td>
<td>57</td>
</tr>
<tr>
<td>4.7</td>
<td>Energy spectra of $CH_4$ and $CD_4$ at different laser intensities</td>
<td>59</td>
</tr>
<tr>
<td>5.1</td>
<td>Comparison of $H_2$ cluster simulation results with the analytical solution</td>
<td>66</td>
</tr>
<tr>
<td>5.2</td>
<td>Snapshots of simulated methane cluster Coulomb explosions</td>
<td>68</td>
</tr>
<tr>
<td>5.3</td>
<td>Light ion energy distribution after $(CA_4)_{2109}$ cluster explosion</td>
<td>70</td>
</tr>
<tr>
<td>5.4</td>
<td>Simulated mean and maximum energies after $(C^4+_4A)^+_n$ cluster explosion</td>
<td>71</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

Over the last several years, there has been much activity in studying high intensity, ultrashort laser pulse interactions with atomic clusters. At high intensity ($I > 10^{16} \text{W/cm}^2$) the physics governing the laser cluster interaction is fundamentally different than in previous optical studies on clusters. Many experimental and theoretical studies have shown that when femtosecond short pulses which are comparable to or shorter than the disassembly time of a cluster in the laser field, the entire laser pulse interacts with an inertially confined body of atoms. The most dramatic consequence of this unique interaction has been the observation of high charge states and very fast ions ejected from the irradiated clusters [1, 2, 3]. In fact, ions with many $keV$ of energy are ejected from the exploding cluster, an effect which can be exploited to drive nuclear fusion [4, 5]. The vast majority of studies have centered on the explosions of single species "homo nuclear" clusters such as van der Waals bonded rare gas or hydrogen clusters. It is now well established that for large, high $Z$ clusters subject to intense IR, optical or VUV irradiation and moderate intensity, the space charge forces in the cluster prevent complete escape of electrons photo ionized in the cluster. This
forms a nanoplasma which exhibits a variety of collective electron oscillation effects [2, 6]. However, at higher intensity, particularly in smaller, low Z clusters, the laser can quickly strip the cluster of most of its electrons and the explosion is a pure Coulomb explosion. If the laser rise time is fast enough, the fast ions observed in these explosions are ejected isotropically after the intense laser pulse ionizes and expels the electrons from the cluster. The ions then expand by mutual repulsion in the charged cluster sphere. In this simple picture, the ion kinetic energies are simply related to their initial potential energy at their equilibrium position in the cluster. This model has been very successful in explaining the ion energies observed in a number of experiments with low to mid Z clusters (hydrogen to argon) [7, 8]. However, if the ion expansion starts before all electrons are expelled by the laser, Breizman and Arefiev recently suggested [9] that the remaining electrons start to form a cold electron core inside the cluster. This core adiabatically adjusts to the external field of the laser compensating the electric field in the overlapping volume of electron and ion sphere. The resulting electron core oscillation along the laser polarization lowers the average potential created by the ions thus lowering the energy that the ions can gain due to their mutual Coulomb repulsion. These findings motivated the investigation of hydrogen clusters subject to high intensity laser pulses with different polarizations.

With this developing understanding of laser driven explosions in homonuclear clusters, an interesting question exists as to whether mixed species, heteronuclear clusters will behave in the same manner under intense ultrafast laser irradiation. Recent theoretical results suggest that ex-
plosions of clusters with mixed atomic species may be more complicated and exhibit some quite interesting differences [10]. Initial experiments to study the explosions of heteronuclear clusters have been published, for example explosions of HI clusters driven by 260fs pulses at an intensity of $I > 10^{16} W/cm^2$ were studied by Tisch et al. [13]. However, the dynamics and interplay of the two ion species in such clusters have not been studied in any experiments. The motivation for studying Coulomb explosions of mixed species clusters has been advanced recently by Last and Jortner [10, 11, 12]. They showed, through a series of molecular dynamic simulations, that the energies of lighter deuterons from exploding heteronuclear clusters, like $D_2O$ or $CD_4$ are increased through a dynamical effect in the Coulomb explosion. They observed a clear enhancement of the deuterium energy and peaking of the ion energy of deuterium to higher energy then would be expected with a simple Coulomb explosion picture. In this thesis, an experimental confirmation of this dynamic enhancement of light ion energies from exploding heteronuclear clusters is presented. By comparing ion energy spectra from methane ($CH_4$) and fully deuterated methane ($CD_4$) under otherwise identical conditions, a clear enhancement in the explosion energy of protons compared to deuterons is demonstrated.
Chapter 2

Physics of Laser Cluster Interaction

In this chapter different aspects that are mandatory to understand the purpose and the results of the conducted experiments will be illuminated. Specifically the mechanisms of cluster ionization in an ultra fast high intensity laser field and the subsequent explosion will be explained. For this explosion two different scenarios are conceivable, the electrical one and the thermal one. Both will be explained in detail, as well as an approach to describe the intermediate range. Furthermore the difference between the behavior of homonuclear and heteronuclear clusters subject to an intense laser pulse will be addressed.

2.1 High Intensity Interactions with Atoms

Before the discussion of clusters exposed to high intensity laser fields starts, it is informative to know about the interaction of light with electrons in single atoms. The same physics can then be applied to a cluster ionized by an intense laser prior to the cluster expansion.
There are two different ionization regimes for single atoms due to an intense laser: multiphoton ionization and tunnel ionization. The first regime corresponds to laser intensities that are not strong enough to significantly alter the the field of the bounding nucleus, that means intensities on the order of $I = 10^{14} \text{W/cm}^2$ and below (a more strict definition will be introduced later). In this case, in order for the electron to be able to overcome the atom’s electric field, several photons are necessary, so that the electron gains enough energy in the interaction to be ionized. One can determine the probability for a multiphoton transition up into the continuum by expanding the theory of one-photon dipole transitions to higher orders. This finally gives the following equation introducing the generalized crossection $\sigma^{(k)}$, where $k$ is the number of photons necessary to accomplish the transition and $I$ denotes the laser intensity:

$$W_{fi} = \sigma_{fi}^{(k)} I^k$$

$$\sigma_{fi}^{(k)} = 2\pi (2\pi \alpha \omega)^k g^{(k)}(\omega) \cdot \left| \sum_{g} \sum_{n} \cdots \sum_{l} \cdot \frac{\langle f| r| g \rangle \langle g| r| n \rangle \cdots \langle l| r| i \rangle}{\omega_{gi} - (k-1)\omega} \left[ \omega_{ni} - (k-2)\omega \right] \cdots \left[ \omega_{li} - \omega \right] \right|^2$$

Here $g^{(k)}(\omega)$ is the density of states as a function of photon energy $\omega$, $\omega_{ab}$ gives the energy between two atomic levels $a$ and $b$ and the sum is executed over all possible dipole transitions. That means one has to calculate all transition dipole matrix elements which can be rather unpleasant, especially if one is not dealing with a hydrogen atom.

Fortunately, the laser intensities for the experiment described in here are all significantly higher than $I = 10^{16} \text{W/cm}^2$, so that the laser
becomes highly nonperturbative, the described perturbation theory breaks down and tunnelling becomes the dominant ionization mechanism. As shown in Fig. (2.1) the effective potential that the electron experiences is now a superposition of both laser and atomic field.

A reasonable assumption is to consider the introduced external field as time independent since the laser oscillation is much slower than the atomic frequency. The superposition of both external and internal field produces an effective potential of:

\[
V(r) = -\frac{Ze^2}{4\pi\epsilon_0 r} - eEr
\]  

\[
r_{\text{max}} = \sqrt{\frac{Ze}{4\pi\epsilon_0 E}}
\]  

with \( r_{\text{max}} \) indicating the position of the local maximum. Assuming that...
the local maximum of the effective potential \(V_{\text{max}} = V(r_{\text{max}})\) equals the ionization potential \(I_p\), one can make an estimate of what the electric field has to be in order to ionize the atom.

\[
E_{\text{ionize}} = \frac{V_{\text{max}}^2 \pi \epsilon_0}{Ze^3} \tag{2.5}
\]

The according intensity is then given by [22]:

\[
I_{\text{ionize}} = \frac{1}{2} \sqrt{\mu_0 \epsilon_0} E^2_{\text{ionize}} = \frac{1}{2} \frac{V_{\text{max}}^4 \pi^2 \epsilon_0^3}{Z^2 e^6} \tag{2.6}
\]

However, this can only serve as an estimate. In order to calculate correct values, one has to take into account the effective nucleus charge experienced by the electrons.

To determine which ionization regime applies to the conditions used in an experiment, the Keldysh parameter \(\gamma\) was introduced defined as

\[
\gamma = \sqrt{\frac{I_p}{2U_p}} \tag{2.7}
\]

\[
U_p = \frac{e^2 E_l^2}{4m_e \omega^2} \sim I \lambda^2 \tag{2.8}
\]

where \(m_e\) and \(e\) are electron mass and charge and \(U_p, E_l, I\) and \(\omega\) are ponderomotive potential, electric field, intensity and frequency of the laser, respectively.

A value of \(\gamma > 1\) indicates the multiphoton regime and accordingly a value of \(\gamma < 1\) states that tunnelling is the predominant ionization mechanism. For a laser with a wavelength of 1\(\mu m\) and an intensity of 10^{16} W/cm^{2}
the ponderomotive potential for an electron is roughly 1keV. The ionization potential for hydrogen is 13.6eV. Consequently with these parameters the Keldysh parameter becomes $\gamma \approx 0.08$ indicating that at this intensity the tunnel ionization regime is clearly reached.

2.2 Ionization Mechanisms in Clusters

Essentially, there are three ionization mechanisms in clusters [6]. The first mechanism is direct optical ionization of the atoms as was described in section 2.1. This mechanism is most important in the early stages of the laser pulse interaction since it produces the initial electrons that eventually form the plasma. The second mechanism is ionization due to inelastic collisions between ions and electrons. Since the cluster density is usually on the order of solid density ($\rho \approx 10^{23}$ particles/cm$^3$) this ionization process dominates the production of high charge states once some electrons are produced by direct optical ionization. The heating rate can be calculated with the empirically determined collisional ionization rate formula of Lotz [14]. Averaged over a Maxwellian electron distribution, this rate is given by:

$$W_{kT} = n_e \frac{a_i q_i}{I_p \sqrt{kT_e}} \int_{I_p/kt_e}^{\infty} \frac{e^{-x}}{x} dx$$

(2.9)

where $n_e$ is the electron density, $a_i$ is an empirical constant ($a_i = 4.5 \times 10^{-14} (eV)^2/cm^3$), $q_i$ is the number of electrons in the outer atom shell, $I_p$ is the ionization potential in eV and $T_e$ is the initial electron temperature. This rate can be very high, giving rise to the production of high charge states below the according intensity threshold for tunnel ionization [6]. The third ionization mechanism is caused by collisions between ions
and energetic electrons driven by the external laser field. Whereas equation (2.9) only accounts for the electrons whose thermal energy is sufficient for ionization, the velocity the electrons gain by oscillating in the field can also be high enough to contribute to this process. However, Coulomb collisions randomize the sinusoidal oscillation velocity and it is fairly complicated to calculate the ionization rate of electrons driven by the laser inside the cluster. One can try to get an estimate by only considering a pure sinusoidal velocity, however, this treatment will generally overestimate the actual ionization rate since the electron peak velocity is considered to be higher than it is in reality.

### 2.3 Models for Cluster Explosions

There are two conceivable scenarios for the explosion of laser irradiated microclusters: an electrical one (Coulomb explosion) and a thermal one. The electrical scenario assumes that the laser ionizes the cluster completely and the ions are then accelerated by the electric field of their space charge. The thermal scenario emphasizes the electron heating and the according high electron pressure as the mechanism driving the expansion [6]. One main difference is that the thermal scenario preserves quasineutrality in the framework of the cluster size whereas the electrical one does not. Of course there is also an intermediate regime where the laser is sufficiently strong enough to strip off some of the electrons while some are remaining inside the cluster. For the sake of simplicity in the following the clusters are assumed to be spherical and to have a uniform density profile. Furthermore it is assumed that the cluster radius $R$ is much smaller than the laser wavelength $\lambda$. That
way the electric field of the laser can be treated as spatially uniform albeit
time-dependent.

2.3.1 Coulomb Explosion

The easiest model to understand is probably the Coulomb explosion which
implies that the laser quickly pulls out all the electrons of the cluster on a
timescale much faster than the ions are able to move. The stripped electrons
are considered to go to infinity so that they do not contribute to the space
charge forces any more. The cluster then explodes isotropically caused by
the strong Coulomb repulsion between the ions. This model applies to rather
high intensities and smaller clusters, since the laser has to be strong enough
that no electrons remain in the vicinity of the cluster. The intensity required
to drive a pure Coulomb explosion is not easily calculated, it depends on
the dynamics of the driven electron cloud inside the cluster. However, sim-
ulations suggest that a cluster will be fully ionized by the laser field if the
laser ponderomotive potential is equal to or larger than the surface potential
created by the fully stripped cluster. If the laser is not strong enough, it will
only remove a fraction of the electrons until the surface potential reaches a
value on the order of the ponderomotive potential.

The potential created by a hydrogen cluster at the distance $r$ from
the cluster center is given through the Coulomb law. Here $\rho = Z en$ denotes
the charge density with $n$ being the particle density.

$$\Phi = \frac{Q}{4\pi\epsilon_0 r} = \frac{R^3 \rho}{3\epsilon_0 r}$$  \hspace{1cm} (2.10)

Accordingly the surface potential becomes $\Phi(R) = R^2 \rho/3\epsilon_0$. If a solid den-
sity cluster with a charge density of $\rho = e \cdot 10^{23}$ particles/cm$^3$ and a radius
of \( R = 4\, \text{nm} \) is irradiated by a \( 1\, \mu\text{m} \) laser, consequently an intensity of \( I > 2 \times 10^{16} \, \text{W/cm}^2 \) is necessary to drive a pure Coulomb explosion.

If the electrons are removed prior to any ion movement, that is, the laser rise time is short enough, the ion energy spectrum from a single exploding cluster can be calculated easily. Starting with Maxwell’s equation [15]

\[
\varepsilon_0 \oint E \, dA = \frac{4}{3} \pi r^3 \rho
\]

the electric field \( E \) at the radius \( r \) is given by

\[
E = \frac{r \rho}{3 \varepsilon_0}
\]

Thus the electric potential \( \Phi \) inside the cluster can be obtained by integration:

\[
\Phi = - \int E \, dr + C = -\frac{r^2 \rho}{6 \varepsilon_0} + C
\]

Because of the continuity constraint for the potential inside and outside the cluster the integration constant has to be \( C = 3 R^2 \rho/(6 \varepsilon_0) \) and thus the potential energy of an ion with the charge \( Z e \) at the radius \( r \) is given by

\[
E_{\text{pot}}(r) = e \Phi = \frac{(3 R^2 - r^2) Z e \rho}{6 \varepsilon_0}
\]

\[
E_{\text{pot}}(R) = \frac{R^2 Z e \rho}{3 \varepsilon_0}
\]

As one can see, the outermost ions have the highest potential energy in the fully stripped cluster, which is the energy they will gain in the explosion. For inner ions however, Eq. (2.14) cannot be used to obtain the final
kinetic energy. But since the electric field, which is driving the explosion, is zero inside a sphere, the inner ions can just be treated as being the outermost particles of an accordingly smaller cluster. Consequently equation (2.15) can be rewritten to give the final kinetic energy of a particle after a Coulomb explosion as a function of the initial radius $r$ from the cluster center

$$E(r) = \frac{r^2 Ze\rho}{3\epsilon_0} = E_{max} \frac{r^2}{R^2}$$

(2.16)

where $E_{max}$ is the energy given by equation (2.15). An energy spectrum $dn/dE$ can now be derived by weighing the reciprocal value of the radial energy distribution $(dE/dr)^{-1} = r/(2E_{max})$ with the normalized radial density distribution $dn/dr = 4\pi r^2/(4\frac{3}{4}\pi R^3)$:

$$f_{sc}(E) = \frac{dn}{dE} = \frac{dn}{dr} \frac{dr}{dE} = \begin{cases} \frac{3}{2} \sqrt{E(r)/E_{max}} & \text{for } E(r) \leq E_{max} \\ 0 & \text{for } E(r) > E_{max} \end{cases}$$

(2.17)

As one can see, this energy spectrum of a single cluster explosion is actually not a function of the cluster size, only the maximum energy is determined by the initial dimension. In reality, however, the measured ion energy spectrum will be broadened by any distribution in cluster sizes in the target. This distribution is usually considered to be log-normal, which is just a Gaussian distribution, where the variable was exchanged with its logarithm. Hence, in the limit of many particles per cluster the distribution in sizes, denoted by $g(R)$ can be approximated as [16]:

$$g(R) = \beta R \cdot \exp\left(\frac{-(R - R_0)^2}{\beta^2}\right)$$

(2.18)
Figure 2.2: Ion spectra for cluster Coulomb explosions (single cluster and cluster distribution) with an average ion energy of 9 keV. The convolved ion distribution closely approximates a Maxwellian. [Picture adapted from [16]]

Here $\beta$ is a normalization factor and $\delta$ is the variance characterizing the width of the cluster size distribution. Assuming that all clusters are completely ionized, the full ion energy distribution can be obtained by convolving the energy distribution for a single exploding cluster from Eq. (2.17) with the size distribution of Eq. (2.18) [16].

$$f(E) = \int f_{sc}(E)g(R)dR$$

$$= \frac{3\beta}{2} \left( \frac{Ze^2}{3\epsilon_0} \right)^{3/2} E^{1/2} \int_{\infty}^{\infty} \frac{1}{R^2} e^{-\frac{(R-R_0)^2}{\delta^2}} dR$$  (2.19)
Examples for different ion distributions with an average energy of 9keV are shown in Fig. (2.2). For this plot a mean cluster radius of $R_0 = 6nm$ and the variance $\delta = 6nm$ were employed. This comparison shows that the convolved ion distribution signal closely approximates a Maxwellian.

### 2.3.2 Hydrodynamic Explosion

In the case of larger clusters and lower intensities, not all electrons are removed from the cluster but they remain trapped by the electric field of the ions. In this case the cluster can essentially be treated as a nanoplasma. Once a few of the electrons are ionized, the laser deposits energy into the cluster mainly by collisional heating or inverse bremsstrahlung thus most of the energy goes into the free electrons significantly increasing the electron temperature $T_e$. Assuming that the cluster radius is much smaller than the laser wavelength, one can describe the field inside the cluster as the one inside a dielectric sphere surrounded by a constant electric field. This field inside the sphere is then given by [15]

$$E = \frac{3}{|\epsilon + 2|} E_0$$

where $E_0$ is the vacuum field strength and $\epsilon$ is the dielectric constant of the sphere. It can be expressed using the Drude model:

$$\epsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\nu)} \approx 1 - \frac{\omega_p^2}{\omega^2}$$

$$\omega_p = \sqrt{\frac{4\pi^2 n_e}{m_e}}$$
where \( \omega_p, \omega \) and \( \nu \) are the electron-plasma, the laser and the electron-ion collision frequency, respectively.

The heating rate per unit time and volume in the cluster is then given by [6]

\[
\frac{\partial U}{\partial t} = \frac{\omega}{8\pi} \text{Im}[\epsilon] |E|^2
\]

\[
= \frac{9\omega^2\omega_p^2\nu}{8\pi} \frac{1}{9\omega^2(\omega^2 + \nu^2) + \omega_p^2(\omega^2 - 6\omega^2)} |E_0|^2 \quad (2.23)
\]

Note that if \( n_e/n_{\text{crit}} \approx 3 \) (where \( n_{\text{crit}} = m_e\omega^2/(4\pi e^2) \) is the electron density at which the plasma frequency equals the laser frequency) the dielectric constant becomes \( \epsilon \approx -2 \) and the electric field \( E \) inside the cluster
and thus the heating rate go through a maximum. The height and width of this maximum are determined by the collision rate $\nu$. In Fig. (2.3) the heating rate is plotted as a function of $n_e/n_{\text{crit}}$ for three values of the collision frequency $\nu$.

Due to the high pressure associated with the hot electrons, the cluster starts to expand because the heated electrons pull the cold ones and the heavy ions with them. The characteristic speed for this explosion is the plasma sound speed

$$c_s \sim \sqrt{\frac{Z k T_e}{m_i}} \quad (2.24)$$

where $m_i$ denotes the ion mass and $T_e$ is the electron temperature. Another exploding mechanism is again that some electrons leave the cluster completely, thus creating a positive charge buildup on the cluster. As described in detail in section 2.3.1 the mutual Coulomb repulsion between the ions consequently forces them to explode.

### 2.3.3 The intermediate Regime

An interesting case to discuss is when the electron extraction is incomplete. Again, the extracted electrons are considered to go to infinity so that the cluster becomes positively charged on a timescale faster than the ions are able to move. Assuming that the laser frequency is much smaller than the plasma frequency $\omega \ll \omega_p$ one can treat the confined electrons in the plasma adiabatically which means they instantly adjust to the external field. It turns out, for a spherical cluster the equilibrium configuration describing the confined electrons can be found analytically [9]. Neglecting collisions inside the cluster one can assume the electrons stay cold and behave like a
perfectly conducting fluid. To find the equilibrium solution one has to find the boundary of that fluid in the effective field arising from the superposition of the external laser field and the space charge field. The electric field created by an hydrogen like ion background is given by

$$\vec{E}_i = \frac{4\pi n_i e}{3} \vec{r}$$  \hspace{1cm} (2.25)$$

where $n_i$ denotes the ion density. This equation is valid for $r \leq R$ with $R$ again being the cluster radius. Accordingly a uniform electron sphere creates the field

$$\vec{E}_e = -\frac{4\pi n_i e}{3} \vec{r}_e$$  \hspace{1cm} (2.26)$$

where $\vec{r}_e$ is measured from the center of the electron sphere. Of course, the total space charge field in the overlapping volume of the two spheres is then given by superimposing both contributions.

$$\vec{E} = \frac{4\pi n_i e}{3} \vec{r} - \frac{4\pi n_i e}{3} \vec{r}_e = \frac{4\pi n_i e}{3} \vec{d}$$  \hspace{1cm} (2.27)$$

where $\vec{d}$ is the distance between the centers of the two spheres. A schematic drawing of this situation is shown in Fig. (2.4).

Since the electron cloud will adjust to an equilibrium, the electric field due to the displacement of the spheres should just compensate the external laser field $\vec{E}_0$, so that the electric field inside the overlapping volume disappears. Thus the condition for the displacement $\vec{d}$ has to be

$$\vec{d} = -\frac{3}{4\pi n_i e} \vec{E}_0$$  \hspace{1cm} (2.28)$$
Figure 2.4: Electrons confined in a uniform spherical cluster in the presence of an external field. The outer circle marks the ion background, the inner circle marks the boundary of the electron core. At the point where both spheres touch each other a potential leak appears allowing electrons to leave the cluster.

Having found the displacement $\vec{d}$, the radial extension of the electron sphere $R_e$ still needs to be determined although it is clear that it should be somewhere between zero and $R - d$. It turns out that the radius is given by the time history of the laser and its electric field. With no laser the electron sphere radius equals the cluster radius $R_e = R$, if now the field is turned on and increases monotonically, the condition $R_e = R - d$ ensures that at the point where the boundaries of both spheres touch each other a small potential leak exists where electrons can be pulled out. If the laser field passes through its maximum, the field decreases and so does the displacement $\vec{d}$. The electron sphere shifts back towards the cluster center so that the potential leak disappears, no electrons are pulled out any more and the radius of the electron sphere is determined by the previous maximum value of the external electric field $E_{\text{max}}$. 


\[ R_e = R - d_{max} = R - \frac{3}{4\pi n_i e} E_{max} \] (2.29)

Obviously, after the laser passed, electron and ion sphere remain concentric, that is the cluster consists of a neutral core surrounded by a positive ion shell. This shell will then explode on the ion time scale due to mutual repulsion, whereas the electron core will remain at rest.

However, it has to be kept in mind that the described model only applies if the ion background remains stationary and does not expand during the electron core oscillation. The characteristic ion response time can be estimated by calculating the reciprocal ion plasma frequency \( 1/\omega_{pi} \) which leads to an ion motion on a time scale of \( \Delta t_i \approx 5 \text{fs} \). This certainly is much shorter than a laser pulsewidth of \( \Delta t_l \approx 40 \text{fs} \) as used in the experiments described in chapter 4. The expansion on the other hand can lead to another interesting effect depending on the laser polarization. Now the electrons still oscillate in the external field while the ions start to expand due to their Coulomb repulsion. However in the direction of the laser field the average charge acting on the ions is lowered because of the oscillating electron core.

Thus the ions exploding in the direction of the laser polarization gain less energy than the ones perpendicular to it where the effective charge is not altered by the oscillating electrons.

### 2.4 Heteronuclear Clusters

Particle dynamics simulations recently conducted by Last and Jortner [10, 11, 12] suggest that heteronuclear clusters consisting of a light and heavy ion species do not explode by the simple Coulomb explosion model even though
all particles are completely ionized. In fact, the explosion mechanism is more complicated and yields some interesting effects. It turns out that in the case of heteronuclear clusters the relation between the acceleration of the light species $A$ and the heavy one $B$ is determined by the kinematic parameter $\eta$ for each species.

$$\eta_A = \frac{q_A}{m_A} \cdot \frac{m_B}{q_B}$$  \hspace{1cm} (2.30)

Here $q$ and $m$ are the charge and mass of the particles of species $A$ or $B$, respectively. If the charge to mass ratio for both species yields a kinematic parameter of $\eta_A > 1$ the acceleration, the lighter ions experience from their preliminary equilibrium position, is larger than for the heavy ions. Consequently the light ions gain a higher velocity and they start to overtake heavy particles. Since the force on a particle with the distance $r$ from the cluster center is determined by the charge density within the sphere with this radius $r$, by overtaking the heavy ions, the effective charge acting on the light particles increases and so does the acceleration they experience. This stronger repulsion ultimately leads to larger kinetic energies. Since the energy enhancement is caused by overtaking ions, consequently the outer-
most ions do not benefit from this effect since they can not outrun any other particles. Accordingly the maximum ion energy produced in the cluster explosion is still fixed by the potential energy of the cluster surface particles. The average energy however is significantly shifted to higher energies as compared to the energy spectrum of a homonuclear cluster explosion.

The reverse effect happens for \( \eta_A < 1 \). Now the heavy ions attain a higher acceleration leading to an outrunning of the lighter species and an increase of the average kinetic energy of the heavier particles. In the case of \( \eta = 1 \) both species travel with the same speed.

Table (2.1) shows the different kinematic parameters for both atomic
species of methane \((CA_4)\) with \(A\) being either hydrogen \((H)\), deuterium \((D)\) or tritium \((T)\). Furthermore it is assumed that the lighter species is completely ionized and carbon is ionized to either \(C^{4+}\) or \(C^{6+}\).

Fig. (2.5) shows the simulated kinetic energy distribution of the light ions produced in a Coulomb explosion of \((CA_4)_{2171}\) clusters \((R = 31.8\text{Å})\) for \(A = H, D\) and \(T\). The shown results obviously reproduce the predicted energy enhancement. As expected in the methane case the distribution for the lighter ion species is shifted to higher energies if \(\eta > 1\) with hydrogen experiencing the largest dynamic enhancement. The tritium on the other hand shows no enhancement and approaches a Coulomb distribution. The maximum energy for the light ions remains unaffected because it is fixed by the potential energy of the outermost ions that do not experience a dynamic enhancement.

### 2.5 Clustering in a Gas Jet

When high pressured gas expands into vacuum through an orifice the random thermal energy is converted into directed kinetic energy. This adiabatic expansion leads to a substantial decrease in temperature in the frame of the moving gas [17]. As an effect the weak interaction between molecules and atoms at room temperature can become quite attractive, the gas supersaturates and single particles start to condensate into solid density droplets. The attractive behavior is due to van der Waals forces or hydrogen bonding. The strength of this clustering effect is strongly depending on the experimental conditions, such as pressure in the gas reservoir, shape of the nozzle and the gas species used. Hagena and Obert found that the onset of clustering...
and the size of the produced clusters can be described with the empirically obtained scaling parameter $\Gamma$ [18].

This Hagen parameter is given by

$$
\Gamma = k \left( \frac{d}{\tan \alpha} \right)^{0.85} \frac{T_{0}^{2.29} p_{0}}{T_{0}^{2.29}}
$$

where $T_{0}$ denotes the initial gas temperature, $p_{0}$ is the gas backing pressure in mbar, $\alpha$ is the expansion half angle of the nozzle and $k$ is a gas specific parameter. Examples for $k$ can be found in table (2.2).

It was observed that the use of a supersonic nozzle ($\alpha < 45^\circ$) significantly enhances the cluster production. Most studies have shown that cluster production starts at a Hagen parameter of $\Gamma \approx 300$ with the number of particles per cluster roughly increasing as $N \propto \Gamma^{2.0-2.5}$. Since the cluster radius scales with the number of particles as $R \propto N^{1/3}$ consequently the dependence of cluster radius to backing pressure (at constant temperature) or cluster radius to temperature (at constant backing pressure) can be given:

$$
R \propto p_{0}^{0.66-0.83}
$$

$$
R \propto T^{-(1.53-1.91)}
$$

As shown in table (2.2) the clustering is highly dependent on the gas species used. For example for Kr or Ar a backing pressure of $p_{0} \geq 20bar$
with an orifice $d \sim 1\text{mm}$ is sufficient to produce large clusters at room temperature. For $He$ on the other hand a backing pressure of $p_0 \geq 140\text{bar}$ is required for an onset of clustering at all.
Chapter 3

Experimental Apparatus and Techniques

In this section a detailed explanation of the experimental setup will be given including a description of the THOR laser facility and the time of flight chamber. Furthermore comments on preparations necessary to conduct a datarun will be provided as well as basic information on how to analyze the data.

3.1 Introduction to the THOR Laser

Before 1985 the main concern to produce high intensity laser pulses was to avoid amplifying the pulses directly in the laser chain without exceeding the intensity damage threshold of the laser optics. The standard solution was to use larger optics and larger amplifying apertures (e.g. up to 50cm for the final amplifying aperture of NOVA at Lawrence Livermore National Laboratory (LLNL)). A new approach was introduced in 1985 by D. Strickland and G. Mourou [19] called the chirped pulse amplification (CPA). They suggested, rather than decreasing the intensity by expanding the beam, to
stretch the pulse in time prior to amplification. Now the intensity would be low enough to use small optics in the amplification stage before eventually the beam gets recompressed to theoretically the same pulsewidth it initially had. Now only for the final stage after the compression large optics are necessary.

To amplify the laser beam a gain material is used. The most common ones are Ti:sapphire and Nd:glass since these materials can amplify a broad bandwidth which is mandatory in order to maintain the pulse shape and keep the pulse duration after compression short. The main difference is the wavelength at which the materials lase (Ti:sapphire - 800 nm, Nd:glass - 1054 nm) and the saturation fluence \( J_{\text{sat}} = \frac{\hbar \omega}{\sigma_G} \) where \( \sigma_G \) is the gain crosssection. The saturation fluence gives the amount of energy one can get out of the crystal per unit area before the gain stops behaving exponentially but starts to become linear (\( J_{\text{sat}} = 0.9 J/cm^2 \) for Ti:sapphire and \( J_{\text{sat}} = 4.4 J/cm^2 \) for Nd:glass). It turns out that the gain in a Ti:sapphire crystal is higher than in Nd:glass although the amount of energy that can be extracted is lower.

The laser used in all the experiments described in this thesis is the Texas High-Intensity Optical Research laser (THOR) (shown in Fig. (3.1)) which is based on the idea of CPA. The initial pulse is created by a mode-locked Millennia Vs J oscillator, a commercially available femtosecond laser. Its pulsewidth is 20 fs centered at a wavelength of 800 nm, the bandwidth is 30 nm full width at half maximum (FWHM). The pulse is send into a stretcher where it is extended to 600 ps lowering the power by a factor of 30,000. The stretcher is a dispersing system which utilizes a diffraction
Figure 3.1: The THOR laser: the oscillator pulse is stretched in time and amplified in three multi-pass stages. After amplification the beam is recompressed and can be send into the experiments. (By courtesy of Prof. T. Ditmire)

grating to introduce different pathlengths for different frequencies. In this case the path for the shorter wavelengths (blue) is longer than for longer wavelengths (red) resulting in a longer pulse in which long wavelengths come first. This beam is now amplified in three multi-pass stages using Ti:Sapphire crystals that are pumped by q-switched Nd:YAG lasers as the gain medium. By changing the timing of the pump beams with respect to the seed pulse one can vary the output energy of the last amplification stage between 6\( mJ \) to 1.3\( J \). After being amplified completely the pulse is sent into the compressor to inverse the effect of the stretcher. That means a second grating is used to introduce a longer path for longer wavelengths.
Theoretically the pulsewidth of the compressed beam can be as short as
the initial oscillator beam. However, loss of bandwidth and dispersion in
the laser chain limit the final pulsewidth to about 40 fs although an optical
fiber was installed to compensate for these effects. Furthermore due to losses
of energy in the compression procedure a final pulse energy of 0.75 J that
can be put on target is achieved.

3.2 The Gas Jet

The clusters in the experiment were created using a gas jet. It is connected to
a high pressure backing line filled with the clustering gas. By opening a valve
the gas freely streams into the vacuum chamber and expands adiabatically.
Consequently the gas temperature drops, the gas supersaturates and the
single particles start to form droplets. The valve used is a General Valve
Corporation series 99 solenoid that can be controlled with an Iota One Pulse
Driver. However, for safety and convenience reasons usually a much more
accurate Stanford timing box was used to control the opening time.

As seen in Fig. (3.2) the nozzle is opened by lifting the poppet with
the magnetic force of the solenoid enabling easy control of the opening time.
Usually durations on the order of 1 ms were used. As was mentioned in sec-
tion 2.5, the efficiency of the clustering process is strongly depending on the
experimental parameters such as gas temperature and nozzle shape. Thus
the valve was surrounded by a close fitting copper jacket that can be con-
ected to a gas supply. By pumping nitrogen gas, which was preliminary
cooled in a separate heat exchanger with liquid nitrogen, through the cryo-
jacket the backing line gas could be cooled down to $T = -173^\circ C$. Varying
the temperature can be done by changing the flow rate through the cooling jacket. The temperature was measured with a thermocouple attached to the nozzle. Furthermore two different nozzles can be used to accommodate to different gas types. For gas that is very likely to cluster (e.g. argon) a sonic nozzle is sufficient, for hydrogen and methane on the other hand a supersonic nozzle with an orifice of $750 \mu m$ and an expansion half angle of $5^\circ$ was used. The supersonic nozzle can easily be attached to the sonic one making it unnecessary to disassemble the gas jet. By varying backing pressure $p_0$ and gas temperature $T$ one can easily control the mean cluster size created in the gas expansion. For hydrogen the nozzle actually had to be cooled in order to produce clusters of significant size.
It is worth mentioning here that the valve performance depends critically on how tightly the valve is fixed to the body. The best diagnostic is the sound the valve makes while tightening it. The optimum position, the so-called "sweet spot", usually narrows down for colder temperatures so that, even though the nozzle works properly at room temperature after assembling it, further testing with a cooled nozzle is necessary. When the optimum position is almost reached, the valve starts to alternate between full and partial opening on successive shots.

3.3 The Vacuum Chamber

The experiments to investigate the physics of laser cluster interactions were conducted inside a vacuum chamber. A schematic drawing of the chamber is shown in Fig. (3.3). It consists of two major parts, the gas jet, where the clusters are created with the solenoid valve (see section (3.2)), and the time of flight chamber (TOF chamber) where the clusters are intercepted by the laser pulse and the energies of the explosion constituents are measured. The chamber parts are divided by a skimmer with an orifice of 750 µm which the clusters have to pass in order to form a collimated beam before being hit by the laser. That way only a small fraction of the beam reaches the TOF chamber. Obviously it is crucial to keep the density of clusters as low as possible since the ideal situation would be to measure the expansion of just one single cluster. In order to further decrease the cluster density in the beam the distance between nozzle and skimmer was increased from initially 33 cm to 73 cm.
Both parts of the vacuum chamber are pumped independently by two turbo pumps. Thus a significant pressure difference between them can be sustained. That is important because although the pressure in the gas jet reaches up to $10^{-2}$ torr after one shot it is mandatory for the experiment to keep the pressure low in the TOF. Otherwise the mean free path for the generated ions becomes too short and collisions between cluster constituents and gas atoms alter the explosion signal. The pressure in both chamber parts is independently measured by two gauges with a pressure range of 1 mtorr.
\( \approx 1.3 \times 10^{-3} \text{mbar} \) up to room pressure \((760 \text{torr} \approx 1013 \text{mbar})\). Additionally, the pressure inside the TOF can be measured by using an ion gauge with a measurable pressure ranging from \(10^{-8} \text{torr}\) up to \(10^{-4} \text{torr}\). When pumped down, the pressure in the chamber usually was on the order of \(p \approx 7 \times 10^{-7} \text{torr}\) \((\approx 9.3 \times 10^{-7} \text{mbar})\), which is sufficiently low to ensure a long enough mean free path. The necessary low pressure in the TOF also limits the rate at which shots can be done. Whereas for methane a pumping time of \(4 - 6 \text{s}\) is usually sufficient to remove the injected gas, it can substantially exceed \(15 \text{s}\) for hydrogen.

After the cluster beam passes the skimmer it is intercepted by the laser beam at the laser focus. The beam is focused using an \(f/4.9\) lens with a focal length of \(205 \text{mm}\) and an aperture of \(42 \text{mm}\). In order to reduce spherical aberration, a refractive graded index lens is used, which means that the refraction index of the lens material is a function of the radius.

The theoretical focal spot size of the lens is given by the beam waist

\[
w_0 = f \lambda
\]

with \(\lambda = 800 \text{nm}\). This assumes a Gaussian beam profile and corresponds to the radius at which the intensity has dropped to \(1/e\) of its maximum value. For the lens used in the experiment, this corresponds to \(3.9 \mu m\). However, serious burnmarks on the lens as well as a not perfectly aligned compressor increase the focal spot size. Measurements using a CCD camera and a resolution target for calibration yield a "cigar shaped" focal spot corresponding to a circular area with a radius of approximately \(15 \mu m\).

After the laser pulse intercepted the cluster beam it leaves the cham-
ber through a window hitting a power meter to determine the pulse energy. Although the loss of energy in the cluster beam is considered to be low because of the low cluster density the measured energy is still an underestimate of the actual on-target energy since reflection and absorption at the window are not taken into account.

To measure the time at which the laser pulse enters the chamber, a photodiode is placed prior to the lens. It triggers an oscilloscope but can also serve as an additional power meter as long as a neutral density (ND) filter is positioned in front of the diode in order to prevent the diode signal from saturating.

After the clusters exploded due to energy deposition by the laser, the energies of the cluster constituents are determined by measuring the time of flight (TOF) in a field free drift tube of 1.14m length. The detector is a micro channel plate (MCP) which basically works as an electron amplifier. When particles with a sufficient energy hit the MCP they can free electrons that are then accelerated in a high voltage gradient (usually $-1.66kV$ were used). These accelerated electrons reach high enough energy so that they free new electrons thus amplifying the initial signal. By connecting an oscilloscope to the MCP the current pulses created by particle impacts can be observed and saved. Together with the photodiode signal the flight time of the particles is obtained. However, not only ions can cause a signal at the MCP. Also photons and high energy electrons that have sufficient kinetic energy to overcome the potential well at the MCP surface cause a signal which comes a few nanosecond after the laser is detected by the photodiode. The MCP has an aperture of 18mm which means it covers about $1/16000$ of the sphere.
centered at the laser focal spot. This corresponds to $7.8 \times 10^{-4} \text{sr}$.

Another way to use the chamber is to do actual mass spectroscopy of the gas inside the TOF without creating any clusters. To do that the laser is also focused into the chamber thus ionizing the gas it contains. By applying a high voltage (usually $U_{\text{acc}} = 10kV$) the created charged particles are accelerated in the voltage gradient. The kinetic energy the ions gain in that field is $E_{\text{kin}} = \frac{1}{2}mv^2 = qU_{\text{acc}}$. Thus the time of flight gives the characteristic charge $q_i/m_i$ where $q_i$ and $m_i$ are charge and mass of the ion, respectively. That also establishes a way to optimize the laser pulsewidth by optimizing the compressor for signal strength of a high charge state from e.g. argon.

### 3.4 Data Analysis

The described setup allows the variation of several parameters such as gas temperature, backing pressure and laser intensity. Also polarization effects can be measured by putting a waveplate in font of the lens thus changing the laser polarization. As mentioned in section 2.3.3 under certain conditions the energies of ejected ions in a cluster explosion can be affected by the electric field of the laser.

This results in several possible experiments, that is scanning one parameter while keeping the other ones constant. The data of the experiments is then obtained with an oscilloscope connected to the MCP and the photodiode, thus recording arrival time of the laser and ion impacts on the detector. The datarun with the according number of shots for a datapoint is then saved on a computer where each file corresponds to one channel on
Figure 3.4: The blue curve shows a photon and an electron peak in the TOF signal. The red curve was taken by applying a 4.6keV retarding potential in front of the MCP to reflect electrons proving that the second peak is indeed caused by electrons. The data were taken before the additional nozzle-skimmer distance was introduced. [adapted from [23]]

The acquired time of flight signal then has to be interpreted in order to extract the physical quantities of interest. The signal that is given by the MCP consists of four different parts, however usually only two are really visible. Shortly after the laser-cluster interaction at $t = 0$, photons ($t \approx 4\text{ns}$) and high energy electrons ($t \approx 30\text{ns}$) are detected by the MCP. The photons are mainly scattered light from the laser, the electrons are produced in the ionization process of the cluster. However, the electron peak is usually not visible since only a few electrons are energetic enough to overcome the

the oscilloscope (usually channel 1: MCP and channel 2: diode signal).
potential barrier of the MCP. Furthermore since the cluster density was reduced by increasing the distance between skimmer and nozzle (see section 3.3) also the number of electrons was significantly reduced. Fig. (3.4) shows experimental data for photon and electron peak before the distance was increased.

The incoming photon peak also offers another method to find out when the laser cluster interaction happened. Since photons travel with the speed of light, they need $3.8\, ns$ to reach the MCP. Knowing this, one has a way to validate the information obtained with the photodiode signal. However, the height or even the appearance of the photon peak was often subject to strong variations making it an unreliable source to determine the arrival time of the laser.

Successive to the laser/electron peak the subsequent peak is caused by the ions created in the cluster explosion hitting the MCP. This signal usually lasts for several $\mu s$ before eventually it fades out and gets superimposed by the signal caused by the expanding background gas plasma. At what time cluster ion and background signal is detected depends on the experimental parameters such as cluster size or the gas that was shot. In the case of hydrogen it usually takes about $10\, \mu s$ for the background plasma to be detected by the MCP, in the case of argon or methane on the other hand that background signal never appeared in the data.

Examples for typical TOF data are shown in Fig. (3.5) and (3.6). Fig. (3.5) shows a shot of hydrogen clusters done at a nozzle temperature of $T_0 = -165^\circ C$ and a backing pressure of $p_0 = 41.4\, bar$, the MCP voltage was, as usual, $V_{MCP} = -1.66\, kV$. Fig. (3.6) shows a TOF spectrum for
Figure 3.5: Typical TOF signal for hydrogen, (a) shows the complete measured interval of 10\(\mu\)s, (b) is a zoom into the plot. The data were taken with the experimental settings \(p_0 = 41.4\text{bar}\) and \(T_0 = -165^{\circ}\text{C}\).

argon done with the experimental settings \(p_0 = 22.4\text{bar}\) and \(T_0 = 23.5^{\circ}\text{C}\). The MCP bias was \(V_{MCP} = -1.6\text{kV}\). The argon data were taken before the distance between skimmer and nozzle was increased, the hydrogen data were taken afterwards. However, this does not alter the fact that the shown plots are representative for the specific gas type and cluster signal. In both cases also a zoom into the data showing a time interval of 300\(\text{ns}\) is displayed. These plots represent original data one laser shot produces. However, to increase readability the displayed plots are the inverted data measured by the oscilloscope. Since the MCP response to impacts is a negative current, so is the oscilloscope signal. Accordingly the pictures have to be converted afterwards and the offset introduced by the oscilloscope has to be removed.

As one can clearly see, the density of peaks in the case of hydrogen is a lot higher than in the case of argon. It is not possible anymore to determine single peaks and distinguish between noise and single ion impacts. On the other hand in the argon data one can clearly discern single peaks
Figure 3.6: Typical TOF signal for argon clusters, (a) shows a time interval of 50$\mu$s, (b) is a zoom into the plot. The data was taken with the experimental settings $p_0 = 22.4\text{bar}$ and $T_0 = 23.5^\circ\text{C}$.

thus making it possible to really count ions hitting the MCP. Because of that obvious difference two ways of averaging the data were established.

The problem is that in any case one single shot does not give reliable information since the experimental parameters are subject to statistical variations. To obtain a more sophisticated picture of a typical TOF trace and energy spectrum it is necessary to average the experimental data over several shots. On that account usually 100 to 200 shots were taken for one datapoint, that is for one setting of parameters. Having obtained the averaged TOF signal one can then start to extract physical quantities and compare these to the theoretical predictions thus validating or falsifying the theory.

In the case of argon the data analysis was pretty straight forward. As was stated before one can easily discern single ion incidents in the MCP signal. To extract that information a peak-count-algorithm was introduced that automatically scans each shot for ion impacts and counts these. It
then creates intervals with a set width of usually $dt = 50\,\text{ns}$ and counts the number of peaks in each interval. By adding the result from each shot, a histogram is achieved showing the probability of ion impacts in the time interval $t$ to $t + dt$. By dividing each "probability" by the number of total counts the result is then normalized for the total measured time interval.

Since the ion mass is known, together with the detection time so is their velocity and energy. Another way to count and characterize peaks is to create energy intervals of a set length and count the number of peaks within these energy ranges. Again by adding the results of each shot an energy histogram is produced that can then also be normalized by dividing the number of counts per interval by the total peak number.

However, in the case of hydrogen or methane the described method of averaging does not work. As was mentioned earlier, no single peaks are discernable any more and it seems ions are detected faster than the MCP is able to respond. However, assuming that the MCP response to a particle impact is characteristic and reproducible, one can obtain an average signal by just adding each shot. On the other hand it is not obvious that this is really allowed. To explain that, the operating principle of an MCP has to be illuminated in more detail.

An MCP consists of two thin plates with each containing an array of thin electron multiplier channels. A schematic drawing of an MCP is shown in Fig. (3.7). In the MCP used in the experiment, the channels have a diameter of $10\mu \text{m}$ with a channel spacing of $12.5\mu \text{m}$. Each plate has a thickness of $0.04 - 0.045\,\text{mm}$. When a voltage $V_{\text{MCP}}$ is applied to the input-side and output-side a potential gradient is built up along the channel.
Figure 3.7: Schematic drawing of the MCP detector. The detector consists of two plates mounted such that the input bias markers point in different directions. That way the correct alignment of the electron amplifier tubes is ensured.

direction. If a particle now strikes an inner channel wall on the input-side with a sufficiently high incident energy a number of secondary electrons are emitted. These electrons are accelerated in the potential gradient and travel along a parabolic path determined by the initial velocity. They then collide with the channel surface thus emitting a second generation of electrons. These secondary electrons are then accelerated in the potential gradient. In this manner the electrons collide repeatedly with the channel wall creating more electrons as they travel towards the output side. The result is a large amplification of the incident signal. To avoid that an ion can pass the MCP undetected the channel direction is not perpendicular to the plate surface.
but with a certain angle. By attaching two plates to each other with one plate rotated by 180° the amplifier tubes are not arranged in a straight line anymore and particles entering a channel necessarily hit the channel wall.

On the other hand, a particle that happens to hit the MCP in between two channels, remains undetected since the emitted electrons are not amplified. Also the MCP response is not necessarily always the same to one particle impact. Two things effect the resulting signal. First of all, the energy of the incident particle determines the number of first generation electrons that are generated by the impact. This of course has a major contribution to the final amplified signal. The second issue is that the strength of the amplified signal is of course depending on where the initial particle impact happened. It is obvious that if the particle hits a channel wall very close to the input-side surface than the amplifying distance is maximized and so is the signal amplification and the peak height on the oscilloscope. However, if the particle travels a certain distance in the amplifier tube before hitting the wall and emitting the first electrons, the distance at which the signal is amplified is accordingly shorter resulting in a lower peak on the oscilloscope even though in both cases the energy might have been the same. Consequently twice the peak height on the oscilloscope does not necessarily mean twice the number of detected particles.

Therefore it might seem that adding the MCP signal directly should not give the same information as counting single peaks, since the MCP signal is not only determined by the particle density but also by the particle energy (kinetic and internal by its charge state) and the way the particles hit the detector.
To find out whether the direct MCP signal still contains valuable information, the MCP current, that is the total integrated area of the signal, as a function of the number of peaks, was scrutinized for hydrogen and argon. As long as peaks were distinguishable and therefore countable the total integrated MCP current was found to increase linearly with the number of registered ion impacts. This observation was made although some of the shots had much more energetic ions than others and therefore should have shown a different total response and despite the fact that surely argon ions were hitting the MCP in several different charge states. By creating a histogram of peak height versus arrival time a slight variation of the MCP response was noticed resulting in higher peaks for faster ions. Eventually this effect was found to be minimized using a bias voltage of $V_{MCP} = -1.66kV$ [25].

These results suggest that the method of adding the original MCP data to obtain an averaged signal may modify slightly but not significantly the results of the initial analysis making it a valid way to analyze the hydrogen and methane data. However, if the data allows to count single ion impacts, this technique of analysis is recommended.
Chapter 4

Experimental Results

In this chapter the experimental results obtained with the TOF chamber are presented. The chapter is divided into two sections with one part devoted to the comparison of Coulombic $CH_4$ and $CD_4$ cluster explosions under identical conditions. The other part will address measurements done with hydrogen clusters to observe a dependence of the ion energies to the laser polarization.

4.1 Polarization Scan of Hydrogen

As was derived in section 2.3.1 to strip a 4nm hydrogen cluster completely of the electrons an intensity of roughly $I \approx 2 \times 10^{16} \text{W/cm}^2$ is necessary. Since all the intensities used in the experiment are significantly higher, one would expect a pure Coulomb explosion of the hydrogen clusters resulting in an isotropic explosion and no dependence of the TOF signal on the laser polarization. In fact, the energy spectrum is expected to look similar to the cluster distribution signal displayed in Fig. (2.2). However, because of the small proton mass, the assumption that the ion background is static during the laser cluster interaction starts to break down very soon. Since
the estimate of the characteristic ion motion time in section 2.3.3 yielded \( \Delta t \sim 10 \text{fs} \) the ions start to move long before the laser pulse has passed. Although all hydrogen atoms are supposed to be ionized, the early laser intensity is not sufficient to extract all electrons and some remain bound in the potential well created by the ion background while the ions start to expand. The resulting electron core motion [9] consequently lowers the average charge in the direction of the electron core oscillation which is in the direction of the laser polarization. Thus the average energy as well as the maximum energy the ions gain along the laser polarization should be lower than perpendicular to it.

However, it turns out that creating a consistent cluster signal over several hours with hydrogen results in some unexpected problems. To create clusters at all, the supersonic nozzle has to be utilized. Furthermore the backing gas has to be cooled with a constant flux of nitrogen gas cooled preliminary in a bucket of liquid nitrogen. One observation was that firing the gas jet while cooling, decreased the nozzle temperature additionally to a value that would not be reached otherwise. Usually that way a temperature of \( T \approx -173^\circ C \) could be reached although during the last run for no apparent reason only \( T \approx -165^\circ C \) was achieved. The major problem however is, that operating the nozzle at these temperatures is more complicated than at room temperature. It always started to leak after some hours of running time making it impossible to continue the experimental run. The leak was caused by the poppet not closing properly again after one shot. Sometimes the leak disappeared after letting the nozzle warm up again to room temperature, but eventually even that did not help anymore making it necessary
to rebuild the gas jet. Although the nozzle was rebuild several times so that it would not leak and work properly even at low temperatures, the leakage always appeared after some hours of operation at low temperatures. Another problem was that one time the poppet got stuck in the open position, so that the backing line gas streamed freely into the chamber. This issue however was solved by changing the springs directing the poppet.

On the other hand, if the nozzle was operated at room temperature the leakage problem never appeared, indicating that a change of material properties at low temperatures is responsible for the breakdown. Accordingly trying a different poppet material would be recommended. Although this was not tried before with this particular gas jet, using indium instead of copper gaskets as the sealing material is also suggested. Since indium is softer than copper less force is required to tighten the nozzle to its sweet spot or to unscrew it.

Former experiments done on hydrogen clusters with this chamber exhibited a strong variation in signal types which means that under supposedly the same experimental conditions the TOF signal after one shot ranged from no cluster detection at all to huge saturated signals with no apparent reason for this sudden change. However, a satisfying explanation for the observation could not be found. The same behavior can be seen in data taken for the experiments described in this work as is displayed in Fig. (4.1). All plots show the (inverted) TOF signal observed with an oscilloscope after one shot of hydrogen clusters. Note that even though the signal height is given in arbitrary units, the three traces are on the same scale making them comparable to each other.
Figure 4.1: Different TOF traces for $H_2$ measurements: all TOF signals were generated using the same experimental conditions ($p_0 = 41.4\text{bar}, T_0 = -165^\circ C, I > 1\times10^{17} W/cm^2$). The only difference is the gas jet chamber pressure $p_{\text{gasjet}}$ prior to the shot.

Although every time the experimental parameters such as laser intensity $I$, backing pressure $p_0$ and nozzle temperature $T_0$ were the same the signals look entirely different from each other. However, by observing the pressure buildup in the gas jet a clear dependence of signal type to gas jet pressure right before the shot became apparent. The observation was that this pressure could range from below $10^{-3}\text{torr}$ up to $10^{-1}\text{torr}$ prior to the next shot, which is surprising since the turbo pump should remove the fired gas within seconds bringing the pressure back to below $10^{-3}\text{torr}$ (this is the
The smallest reading of the pressure gauge, the ion gauge can not be used because it shuts down when the nozzle is fired. Even extending the pumping time between two shots up to almost one minute did not change the fact that the pressure sometimes did not go down. This probably can be attributed to the gas jet not closing properly so that gas continued to stream into the chamber. In the case that the pressure fell below $10^{-3}$torr, the subsequent shot usually yielded a ”normal” TOF signal as can be observed similarly for other gas species and is shown in Fig. (4.1a). Note that there is no visible photon or electron peak although the signal looks very good (on this plot the photons are expected to be detected by the MCP detector at $t \approx 100ns$).

In the case that the pressure remained on the order of $10^{-2}$torr the TOF signal exhibited hardly any ion detection at all prior to the expanding background plasma arriving at $t \approx 9\mu s$. This can be explained by taking into account that the clustering is caused by weak forces. Due to the high pressure in the gas jet however the clusters cannot travel freely anymore to the skimmer but undergo collisions with the gas particles which inevitably destroys the clusters. As a result no high energy ions can be observed anymore and the only detectable signal comes from the expanding background plasma created by the incoming laser. This is shown in Fig. (4.1b). The plasma is created because the laser is always intense enough to ionize the atoms in the background gas. Because of the low gas density the plasma temperature can be estimated to be roughly $1/10$ of the ponderomotive potential since heating mechanisms such as collisional heating can be neglected (the temperature is mainly determined by the energy the electrons gain in the ionization process). This leads to a plasma temperature on the order of
Figure 4.2: \( H_2 \) TOF traces for the three measured laser polarizations (0° corresponds to horizontally polarized light so that the electric field points to the MCP detector). The plots are averaged signals over (a) 150 shots, (b) 99 shots and (c) 43 shots.

100eV. The plasma is known to expand with the plasma sound speed \( c_s \) (see Eq. 2.24) which ultimately yields an expansion time on the order of 10\( \mu \text{s} \). Another reason the signal starting at 9\( \mu \text{s} \) is attributed to the expanding plasma is because it always appears and is independent on the timing between incoming laser and firing the nozzle, which means it cannot be caused by laser cluster interactions.

Finally, in the case that the pressure was even above \( 10^{-1} \text{torr} \) at
the time the nozzle was fired, the measured signal intensity was usually extremely high. Since in the previous case the pressure was obviously already high enough to prevent the created clusters from reaching the TOF part of the chamber, at an even higher gas jet pressure it is most unlikely that any clusters can reach the laser focal spot. Thus the signal can not be attributed to high energy ions created in a cluster explosion. The exact cause of the detected signal however still remains to be found. It is possible that the pressure in the TOF part gets too high so that the MCP cannot work properly anymore. Unfortunately an accurate pressure measurement inside the TOF is not possible since the ion gauge always shuts down when the nozzle is fired and the pressure gauge for pressures above $10^{-3}\text{torr}$ shows no visible response. When analyzing the data, this effect made it necessary to manually delete shots with either no cluster ion detection or saturated signals before averaging the data.

Because of these problems only three different waveplate settings could be measured for the hydrogen polarization scan. For the first data point with a laser polarization angle of $\Phi = 0^\circ$ (horizontally polarized) 150 shots were taken, that all showed the expected “normal” TOF signal (see Fig. (4.1a)). The second data point was taken for $\Phi = 40^\circ$ also with 150 shots, but by that time the nozzle obviously started to leak, so that 51 shots showed no ion detection prior to 9µs and can be regarded as useless. Hence to generate the averaged signal only 99 shots were used. For the last data point with $\Phi = 70^\circ$ the pressure variation between each shot eventually got so strong that it did not make sense to make a whole run of 150 shots. Instead every time the gas jet pressure was observed and the TOF
signal was saved (43 shots) whenever the pressure dropped below $10^{-3}\text{torr}$ prior to the incoming laser. The averaged TOF traces for all three polarizations are shown in Fig. (4.2). All measurements were conducted using the experimental settings $p_0 = 41.4\text{bar}$, $T_0 = -165^\circ\text{C}$ and $I > 1 \times 10^{17} \text{W/cm}^2$.

As was mentioned in section 3.4, the averaged TOF trace was obtained by adding every single trace taken for this setting of parameters. Apparently the resulting shapes of the distributions differ significantly for the different measurements which can probably not be regarded as an effect caused by the different laser polarizations since the overall shape should be independent and only a shift to higher or lower energies should be observed. However, taking into account that the number of shots used to calculate the average signal differed by a factor of three, the different shapes are not surprising anymore. Still these plots yield an interesting observation. Despite the noise, the time onset of the cluster signal is apparently earlier for $\Phi = 70^\circ$ than for the other two polarizations.

It is noteworthy, that these TOF traces exhibit a two peak feature as was also observed in prior measurements of hydrogen cluster explosions with this chamber [23]. The first peak is located at roughly $2\mu\text{s}$ after the incoming laser, corresponding to $E \approx 1.4\text{keV}$. The second peak can be found at about $5\mu\text{s}$ and yields an energy of $E \approx 270\text{eV}$. In the previous data the second peak was found to be maximized at vertical laser polarization. The measured setting closest to vertical polarization was $\Phi = 70^\circ$ and it looks as if in this case the second peak actually becomes larger or at least comparable to the first one.
The obtained TOF data was then converted to an energy spectrum using the formula $f(E) = f(t)(dE/dt)^{-1}$. In this case the equation becomes:

$$f(E) = f(t) \sqrt{\frac{ms^2}{8} \cdot E(t)^{-3/2}}$$

(4.1)

where $m$ denotes the particle mass, $s$ is the distance the particle has to travel before reaching the MCP and $E(t) = 1/2m(s/t)^2$ is the kinetic energy of the particles arriving at the time $t$.

The normalized energy spectra for each measured polarization are shown in Fig. (4.3a-c), Fig. (4.3d) shows an overlap of all three distributions. Obviously the different distribution results indicate that the clusters do not
undergo a pure Coulomb explosion. As expected from the TOF traces, the energy spectrum for a polarization angle of $\Phi = 70^\circ$ is clearly shifted to higher ion energies than the other two spectra. The distribution for $\Phi = 0^\circ$ and $\Phi = 40^\circ$ on the other hand look almost the same. The apparent separation of the distributions at high energies strongly indicates an increase of energy perpendicular to the laser polarization.

The shown data can easily be interpreted by using the theory presented in section 2.3.3. The assumption that the ions in the cluster start to expand before the laser has reached an intensity high enough to remove all the electrons, is reasonable since the characteristic time of ion motion is shorter than the laser rise time. Consequently a pure Coulomb explosion is not to be expected and the theory of electrons adjusting to the external laser field becomes convincing. In agreement with the measured energy distributions the theory thus predicts higher energy perpendicular to the laser polarization.

Unfortunately the laser pulsewidth was not measured prior to the experiment, making it impossible to say what either the laser rise time or the peak intensity was. However, it is safe to assume a pulsewidth on the order of $t \approx 50\,fs$ which yields a peak intensity of $I > 1 \times 10^{17}\,W/cm^2$. With that intensity a $7\,nm$ cluster could still be stripped of all electrons. Applying the Hagen scaling law, a mean cluster size of less then $6\,nm$ is to be expected under the conditions used. Hence it is safe to claim that eventually the laser pulse expels all electrons from the clusters. Additionally, the longer pulsewidth makes the overlap of cluster expansion and laser even longer, resulting in further electron oscillations along the laser polarization axis.
4.2 Energy Scan of $CH_4$ vs. $CD_4$

Another campaign of measurements focused on the comparison of exploding $CH_4$ and $CD_4$ clusters under identical experimental conditions to observe the kinetic energy enhancement as predicted by particle dynamics simulations conducted by Last and Jortner [12]. However, there was only enough $CD_4$ gas left to do one energy scan for both $CH_4$ and $CD_4$. To ensure that the experimental conditions such as laser condition and pressure in the TOF chamber were identical, the $CD_4$ run was done directly after acquiring the $CH_4$ data. Fortunately the gas parameter $k$ (Eq. (2.31)) for the Hagena parameter is the same for both methane and deuterated methane ($k = 2360$) [17]. That means by keeping all the other experimental parameters such as backing pressure and nozzle temperature constant the mean cluster size and the cluster size distribution are expected to be identical. Because of the mass difference the only thing that needed to be changed was the timing between incoming laser and firing the nozzle. In the case of $CD_4$ the time difference had to be increased by approximately $0.1\mu s$.

At all the intensities used in the experimental run, the carbon atoms in the cluster are ionized to a charge state of $C^{4+}$. The remaining bound electrons are assumed to be connected so tightly to their host atoms that they do not alter the subsequent explosion of the cluster [12]. Assuming the laser is intense enough to remove all ionized electrons from the cluster boundary, the successive expansion can be regarded as a Coulomb explosion.

A previous characterization [20] of a similar gas jet with an expansion half angle of $\alpha = 9.8^\circ$ and operated with $T_0 = 295K$ and $p_0 = 68.9bar$ found a mean $CH_4$ cluster size of 3nm. Comparing the Hagena parameter from
Figure 4.4: Averaged TOF trace for $CD_4$: for the average 200 shots were used, the marked range corresponds to the time interval in which only deuterium ions are detected.

Eq. (2.31) under these conditions to the one obtained with the parameters used in the experiment described in this thesis, also a mean cluster size of approximately $3nm$ is expected. By applying Eq. (2.10) the laser intensity required to drive a purely Coulombic explosion of the $3nm$ methane clusters can be estimated to be $I \geq 1.4 \times 10^{17} W/cm^2$.

For both gas species 200 shots per data point were taken with a time resolution for the TOF trace of $1ns$. The pumping time between each shot was $6s$ which is sufficient to remove the clustering gas from the chamber. In all cases the backing pressure was held constant at $p_0 = (36.7 \pm 0.2) bar$ and the nozzle temperature was $T_0 = (21.4 \pm 0.2)^\circ C$. Also during this data run different signal types as described in section 4.1 were observed.
This probably can also be attributed to the nozzle not closing properly after each shot. However, the variations were not nearly as strong as with hydrogen and the "bad shots" could easily be separated since they yielded no photon peak (in methane this is a much more reliable method to characterize shots than in hydrogen, see Fig (4.1a)). An algorithm was written that automatically removed TOF traces with no photon peak from the averaged data. Consequently the mean TOF trace was usually obtained with less than the saved 200 shots.

In Fig. (4.4) an example for a typical averaged TOF trace is shown. The plot shows CD$_4$ data averaged over 200 shots and obtained using a laser intensity of $I = 2.1 \times 10^{17}$ W/cm$^2$. The way of averaging the data was by adding every single shot to each other and normalize the obtained distribution to unity, so that $\int f(t) dt = 1$ (for readability Fig. (4.4) does not show the normalized plot). Also the peak-count algorithm was applied to average the data, but a closer look to the TOF trace reveals that as well as in the hydrogen data no single peaks are discernable. Accordingly, the counting peak algorithm is not expected to analyze the data accurately. Nevertheless at low energies the averaged signals look almost the same for both analysis methods. But at higher energies the signal obtained by counting peaks becomes increasingly broader whereas the added signal keeps its overall shape. A comparison of both methods is shown in Fig. (4.5) displaying the averaged TOF trace for data obtained with laser intensities of $I_a = 9.6 \times 10^{16}$ W/cm$^2$, $I_b = 1.6 \times 10^{17}$ W/cm$^2$, $I_c = 2.1 \times 10^{17}$ W/cm$^2$ and $I_d = 3.2 \times 10^{17}$ W/cm$^2$. In these plots the blue curve corresponds to added TOF traces, the black curve was obtained by applying the peak-count algorithm.
Since the TOF traces do not directly show whether the kinetic energy enhancement was detected, the averaged data was then again converted into an energy spectrum employing Eq. (4.1). However, for the translation from the time of flight signal to an energy spectrum a certain mass for the ion species has to be taken. According to the gas that was shot either the mass of a proton or of a deuteron was substituted which obviously neglects the carbon detection. Of course, the signal recorded by the MCP is a superposition from both carbon and deuterium or hydrogen ions arriving at the
Figure 4.6: $CD_4$ energy spectrum to TOF trace from Fig. (4.4). The marked range corresponds to the energy interval in which only deuterium ions are detected.

detector and these signals cannot be separated from each other. However, by taking into account that carbon is overtaken by the lighter ion species and thus arrives at a later time, there is a range in the energy distribution for $CH_4$ and $CD_4$ that cannot be caused by the detection of carbon ions. A particle code to simulate the explosion of heteronuclear clusters demonstrated that the maximum energy for Carbon ions is approximately $2.5 \sim 3$ times the maximum energy of the lighter ion species. The simulation will be described in detail in chapter 5.

Since the highest energy particles can safely be assumed to belong to the light ion species, the maximum carbon ion energy and therefore the minimum detection time are determined. Consequently this corresponds to
a characteristic maximum energy in the energy spectrum using the mass of the lighter ion species. Hence every signal above that energy can not be caused by the detection of carbon ions. This characteristic energy is given by $E_{\text{min}} = E_C \cdot m_A / m_C$, where $E_C$ is the maximum carbon energy and $m_A$ and $m_C$ are the light ion mass and the carbon mass respectively. The energy spectrum that corresponds to the TOF trace from Fig. (4.4) is shown in Fig. (4.6). In both figures the energy/time interval is marked in which only deuterium ions are detected.

As shown in section 2.3.1 and 2.3.3 theoretical energy spectra for both homonuclear and heteronuclear Coulomb explosions feature an increase in ion yield with energies up to a sharp cutoff determined by the potential energy of the ions at the surface. This shape of this distribution is not what was observed in the experiments. This difference can be attributed to the distribution of cluster sizes present in the gas expansion. Indeed, as demonstrated in Fig. (2.2) the convolution of a cluster size distribution function with a theoretical Coulomb explosion spectrum results in a spectrum very similar in shape to the ones we have observed.

Fig. (4.7) shows normalized energy spectra at different laser intensities, comparing hydrogen and deuterium data. Clearly the hydrogen energies always exceed the measured deuterium kinetic energies in the energy range presented here. This implies the validity of the theory predicting the kinetic energy enhancement. However an unexpected feature is that there seems to be a significant difference in maximum energy. Furthermore, the separation of maximum energy actually increases with higher laser intensity ranging up to a difference of almost 2.5$keV$ in Fig. (4.7e) with $I = 2.5 \times 10^{17} W/cm^2$. 

58
Figure 4.7: Energy spectra of $\text{CH}_4$ and $\text{CD}_4$ at different laser intensities. The lower curve in each plot always corresponds to $\text{CD}_4$ whereas the higher curve represents the $\text{CH}_4$ data. The black parts of the curve demonstrate the energy range in which only the according lighter ion species $D^+$ or $H^+$ is detected. The hydrogen energies exceed the deuterium energies conspicuously.

Additionally the maximum energy increases with higher laser intensity. As was stated before, in a pure Coulomb explosion, even in the heteronuclear case, the maximum energy is determined by the outermost particles that never experience an energy enhancement. Therefore the maximum energy should be unaffected by the species chosen and should not scale with the laser intensity. The increasing of maximum energy with rising laser intensity also implies a cluster size distribution since at lower intensities larger
clusters can not be fully stripped of the electrons. Accordingly the atoms on
the cluster surface do not experience the repulsion of a fully stripped cluster
and the acceleration as well as the final kinetic energy is lowered.

However, it is not easy to say whether the supposed onset of the
ion signal actually represents the highest energy particles since their flux
might be too weak so that the MCP response would be below the noise
level. Thus the apparent separation of maximum energy might still be the
effect of kinetic energy enhancement.
Chapter 5

The Particle Code

As was described in chapter 4, experiments to observe the predicted kinetic energy enhancement in heteronuclear clusters were conducted (see section 2.4 for the theory and section 4.2 for the experimental results). In order to help interpreting the obtained data a program to simulate methane cluster explosions was created. In the following chapter the basic idea of particle simulations and a detailed description of the code structure is presented as well as the results obtained with the code.

5.1 The Code Structure

In contrast to a homonuclear uniform density cluster undergoing a pure Coulomb explosion a heteronuclear Coulomb explosion can not be solved analytically. However, the motion of every single particle is still determined by Newton’s second law $\vec{F} = m\vec{a}$ giving a set of ordinary differential equations. These equations can always be reduced to the study of sets of first order differential equations. For example the second order equation

$$\frac{d^2y}{dx^2} + f(x)\frac{dy}{dx} = gx$$  \hspace{1cm} (5.1)
can be rewritten to two first order equations

\[
\frac{dy}{dx} = z(x) \\
\frac{dz}{dx} = g(x) - f(x)z(x)
\]

(5.2)

introducing the new variable \(z(x)\). Applying this to a particle in the cluster, the equations in \(x\)-direction become:

\[
\frac{dx}{dt} = v_x \\
\frac{dv_x}{dt} = \frac{F_x}{m}
\]

(5.3)

where \(v_x\) denotes the particle velocity and \(m\) is the particle mass. The force \(F_x\) acting on the particle in \(x\)-direction is determined by the Coulomb repulsion between the ions. The independent variable now becomes the time \(t\). By giving the initial position and charge state of each particle the system behavior in time is determined. The underlying idea of any routine for solving this initial value problem of first-order differential equations is always: derive the function at one given value for the independent variable \(x\) and multiply the result then with a small value \(\Delta x\). This gives an algebraic formula for the change in the function as the variable is "stepped" by the stepsize \(\Delta x\). In the limit of making the stepsize very small a good approximation of the underlying differential equation is achieved. The literal realization of this idea results in Euler’s method, which is however not recommended for practical use since the errors become too big. A more sophisticated approach is the Runge-Kutta method, which propagates a solution over an interval by combining the information from several Euler-style
steps. The stepped functions are then used to match a Taylor series expansion up to some higher orders. However, it has to be kept in mind that errors are inevitably introduced to the results although the correspondence with the correct values can be extremely good.

The particle simulation presented here is based on a fifth-order Runge-Kutta algorithm as described in [24] using the parameters from Cash and Karp to accomplish one step. All particles are treated classically. In this simulation the vertical ionization approximation was utilized, which assumes that on the onset of the cluster expansion \(t = 0\) all of the relevant electrons are removed from the system and do not contribute to the acting space charge forces. This allows a separation of the time scales between the removal of all unbound electrons (outer ionization) and the spatial expansion of the cluster. It was also assumed that the remaining bound electrons are so tightly connected to their host atoms, that they do not alter the subsequent Coulomb explosion of the cluster [12]. Because of these assumptions both bound and unbound electrons are not explicitly treated in the model, which considerably simplifies the simulation since only the positively charged ions remain to be calculated.

Part of the nature of this approach to simulate the explosion is that final energy and particle separation are always overestimated. To calculate the velocity after one step the momentary acceleration has to be taken as valid for the whole subsequent step. In reality, it decreases constantly since the particles move apart from each other and the mutual repulsion weakens. Thus with each step the calculated particle velocity increases too much and so does its energy and the distance from the cluster center. However, by
keeping the step size small enough this effect can be minimized.

A problem that is always connected to sophisticated simulations is the time that it takes to run the calculations. In the case of the cluster explosion modelled, to obtain the force acting on each particle the mutual interaction with every other particle has to be calculated. That means it takes roughly $3n^2$ calculations for one step. A 3nm size methane cluster consists of approximately 10000 particles resulting in approximately $3 \times 10^8$ necessary calculations to increase the time $t$ by one step $\Delta t$. However, one can introduce an algorithm so that the code automatically computes an estimated error after each step and compares it to a reference point. If the estimated error is smaller than the reference, the stepsize is safely increased for the next step. If the error is too big, the calculated step is redone now using a smaller stepsize. In the case of the cluster explosion simulation this results in a constantly increasing stepsize since all particles move away from each other and the repulsion continuously decreases. However, since in this case the behavior of the modelled system here is already known, an easier approach to decrease the simulation running time is to increase the stepsize automatically after a certain step interval. That way also the calculations to estimate the produced error are omitted saving further calculation time.

5.2 Testing the Code

In order to ensure that the code is working properly it was tested for different aspects. The easiest thing to compare is the conservation of energy. Primarily all particles are at rest so that the total energy is the sum over all potential energies. After the last calculated step the sum of obtained kinetic
and residual potential energy is computed and then compared to the initial value. The final energy always was a little too big which is however, as mentioned in section 5.1, unavoidable. The difference between both values was reduced to approximately 1% by reducing the initial stepsize to $1 \times 10^{-17}$ s. This indicates that the simulation is self-consistent, however it still does not say anything about the quality of the results.

An easy way to check the results is to simulate a homonuclear cluster explosion and compare the energy distribution with the theoretically predicted one. However, complete accordance is not to be expected since the code calculates single particles, whereas the analytical solution for a homonuclear cluster uses the charge density $\rho$ to calculate the energy distribution. This results in the possibility of fractional charges acting on each other since the charge density is uniform. By simulating single particles one can define the particle distance as constant, the density though is obviously not constant in every fraction of the cluster. However, in the limit of large clusters the simulation results for inner particles still approach the theoretical solution.

Fig. (5.1a) shows the simulation results for the ion energies after homonuclear Coulomb cluster explosion as a function of the square initial equilibrium distance from the cluster center. The results were obtained by simulating a $(H_2)_{7153}$ cluster with an initial density of $1.2 \times 10^{22}$ atoms/cm$^3$. Each data point corresponds to one particle. Additionally the theoretical solution $E_{\text{final}} \sim r^2$ for the same conditions is plotted as a straight line. As one can see, for the ions with an initial position close to the cluster center, the simulation results are in good agreement with the theoretical prediction.
Figure 5.1: The comparison of the simulation results for a homonuclear Coulomb explosion of \((H_2)_{7153}\) with the analytical solution shows that the model accurately describes the explosion.

Only at higher energies, corresponding to particles located on the outside of the cluster, the difference becomes apparent.

Another thing to check is whether every particle involved in the explosion approaches a finite velocity for \(t \to \infty\). A \(v(t)\) plot that was generated in the same simulation of a \((H_2)_{7153}\) cluster explosion is shown in Fig. (5.1b). Clearly the predicted behavior is observed. This characteristic development of the velocity in time looks almost the same for each particle although, of course, the final velocity differs.

### 5.3 Simulation Results

The particle simulation was used to model the Coulomb explosion of methane clusters \((CA_4)_n\) where \(n\) corresponds to the number of molecules per cluster and \(A\) denotes the light ion species. In order to compare the simulation results with the experimental data most simulations were done for \(CH_4\) and \(CD_4\) but also some calculations for tritium containing methane.
were conducted. To reproduce the experimental parameters the light ion species had a charge state of $A^+$ whereas the carbon was set to a charge state of $C^{4+}$. The clusters were modelled as consisting of uniformly distributed molecules using an underlying cubic pattern. A $C - C$ spacing of 4.4Å and a $C - A$ distance of 1.09Å was used corresponding to a density of $\rho = 1.2 \times 10^{22} \text{molecules/cm}^3$ which is a little lower than the density of liquid methane ($\rho_{\text{liq}} = 1.6 \times 10^{22} \text{molecules/cm}^3$). The light ions were placed in a tetrahedral assembly around the carbon ion imitating the molecule structure.

Simulations on differently sized clusters $(CA_4)_n$ were performed with $n$ equals 257, 515, 925, 1419 and 2109 molecules. The corresponding radii are 1.8, 2.2, 2.6, 3.1 and 3.5nm respectively. The cluster explosion due to mutual repulsion was calculated on a near picosecond time scale ($0 - 4 \times 10^{-13}s$) starting with $1 \times 10^{-17}s$ timesteps. This small stepsize is necessary to ensure enough accuracy even at the very beginning of the cluster expansion when the local forces are at their maximum. Using short timesteps also ensures that the distance the particles travel in that time interval is much shorter than their mean free path which prevents sudden jumps in the acting forces.

A very satisfying result is shown in Fig. (5.2) where the position of every particle for a simulated $(CA_4)_{123}$ cluster explosion after $t = 10^{-13}s$ is shown. Fig. (5.2a) shows the case of $CT_4$ with $\eta = 1$, Fig. (5.2b) displays $CD_4$ with $\eta = 1.5$ and Fig. (5.2c) finally shows the same explosion for $CH_4$ with $\eta = 3$. 

67
Figure 5.2: Snapshots of the simulated expansion of methane clusters at $t = 10^{-13}\text{s}$. (a) shows a $(CT_4)_{123}$ expansion with $\eta_T = 1$, (b) displays a $(CD_4)_{123}$ with $\eta_D = 1.5$ and (c) illustrates an expanding $(CH_4)_{123}$ cluster with $\eta_H = 3$. Orange corresponds to the light ion species whereas blue denotes carbon ions. The supposedly same size of the expanded clusters is caused by different spatial scales of the plots.
The effect of kinetic energy enhancement as predicted by the theory (see section 2.4) is clearly visible for both \(CH_4\) and \(CD_4\) because obviously the lighter ion species, denoted by the orange dots, have overrun the heavier ions, denoted by the blue dots. Since the kinetic parameter \(\eta\) is larger for hydrogen than for deuterium the dynamic enhancement is expected to be stronger for hydrogen. Obviously the simulations yield the same result since the separation of hydrogen to carbon is clearly bigger than the one of deuterium to carbon after the same time interval. In the case of \(CT_4\) however, where \(\eta = 1\) both ion species are expected to travel with the same speed and no kinetic energy enhancement should occur since the lighter ions do not overtake the heavier ones. Again the simulations yield the same result.

Simulations addressing heteronuclear methane cluster explosions were also conducted by Last and Jortner \[12\], however they used a much more sophisticated approach including the description of electrons and the initial laser cluster interaction. The other main differences are that they used a higher cluster density of \(\rho_{\text{liq}} = 1.6 \times 10^{22}\text{molecules/cm}^3\) and a stepsize of \(10^{-18}\text{s}\) which results in smaller radii for the same number of molecules. Despite these differences, their findings still represent a good opportunity to check if the "simple-man" approach yields useable results.

A similar plot to Fig. (2.5) showing ion energy spectra is displayed in Fig. (5.3b). This time, however, the results were obtained with the simple particle code. It demonstrates the energy spectrum for hydrogen and

\[1\text{The reason for the different used densities is that in [12] the assumed C-C distance in a methane cluster was explicitly said to be 4.4\text{Å} which results in the density used for the code described in this thesis. The density used by Last and Jortner for the simulations however corresponds to a C-C spacing of \(\approx 4.0\text{Å}\).} \]
deuterium ions after a simulated ($CA_4)_{2109}$ cluster explosion. Also in these results the typical features of kinetic energy enhancement attributed to the isotope effect can be observed such as increased average energy for both ion species. Again for hydrogen this effect is stronger than for deuterium. Clearly the correspondence of the spectra obtained with both particle simulations is impressive. For comparison Fig. (5.3a) shows the theoretical energy spectrum for a homonuclear cluster explosion as given by Eq. (2.17) (in contrast to Fig. (2.2) the spectrum is plotted on a linear scale).

In Fig. (5.4) the mean and maximum energies calculated with the particle code are displayed for all ion species as a function of molecules per cluster. For comparison the results for a ($CA_4)_{2171}$ explosion found by Last and Jortner in [12] are also given. The energies calculated with the simpler code tend to be smaller than the ones published by Last and Jortner. However the difference is quite small considering the quality of the different approaches. Important is, that one can clearly see the separation of mean
energy between $H^+$ and $D^+$ in the recent results, whereas the maximum energy shows practically no dependence on the molecule species. On the other hand both maximum and mean energy for $C^{4+}$ differ considerably and are ranging up to tens of keV. Both the maximum and the mean energies of the $C^{4+}$ are affected by the kinematic effect as all the ions are overtaken by some fraction of the light ions which originate at smaller radii. Likewise, the $C^{4+}$ from the $CH_4$ which has $\eta = 0.33$ suffers more than the $C^{4+}$ from the $CD_4$ which has $\eta = 0.67$.

Comparing the energy distribution obtained with the simulation results with the one found experimentally, one has to admit that they do not look alike. This difference can be attributed to the distribution of cluster sizes present in the gas expansion. Indeed, the convolution of a cluster size distribution function with a theoretical Coulomb explosion distribution results in a spectrum very similar in shape to the ones we have observed (see section 2.3.1).
Chapter 6
Conclusion and future Work

In this work studies of the explosion of nanometer sized clusters subject to intense ultra short laser pulses were presented. The theory crucial to understand the laser cluster interaction was discussed in chapter 2. The ionization mechanisms, which initially lead to the cluster expansion, were described in detail as well as the different possible explosion types. However, the Coulomb explosion is assumed to be the prevailing explosion mechanism in the experiments described in this thesis. That means the laser was expected to be strong enough to remove all electrons from the cluster resulting in an ion expansion due to mutual Coulomb repulsion. Additionally a model describing the behavior of heteronuclear clusters and the unique resulting effect of kinetic energy enhancement was presented in section 2.4.

To study a model that predicted the dependence of energies the ions gain in the cluster explosion to the laser polarization, the expansion of hydrogen clusters were scrutinized. The results, presented in section 4.1, show a clear energy enhancement in the direction along the laser polarization, although more data points are necessary to support this result.
A validation of the theory predicting kinetic energy enhancement of the lighter ion species in the Coulomb explosion of heteronuclear clusters is given in section 4.2. The measurements of the energy spectra of $CH_4$ clusters compared to $CD_4$ clusters created under identical conditions clearly yield a higher average energy for hydrogen than for deuterium. However, in stark contrast to the theory the maximum energy for both light ion species $H^+$ and $D^+$ differed significantly from each other even though the theory predicts a maximum kinetic energy independent of the ion species. Consequently for $CH_4$ and $CD_4$ it should only depend on the cluster size which is supposed to be the same because of the experimental settings.

Additionally to help interpreting the data a particle code was written and used to simulate methane cluster explosions under the experimental conditions. The code was presented in chapter 5. Its results are in surprisingly good agreement with simulations conducted by Last and Jortner even though the simulation described in here does not take into account any electrons or laser cluster interactions.

In the following some suggestions for future experiments and experimental improvements are provided.

As should be clear, the laser intensity is crucial to the experiments described in here since it determines the mechanism under which the cluster explodes. In fact, describing an intermediate regime of neither pure Coulombic nor hydrodynamic explosion is not simple at all although a possible approach was presented in section 2.3.3. Because of the lens used at the moment, the peak intensity is limited to $I < 3.2 \times 10^{17} W/cm^2$ thus significantly limiting the parameters under which clusters undergo a Coulomb
explosion. The easiest approach is to introduce a lens with a bigger aperture so that the entire beam can be focused and the full beam energy contributes to the peak intensity. Furthermore in order to have a reproducible cluster signal, the gas jet has to be fixed so that it does not change its characteristics when cooled. As mentioned in section 4.1 trying a different poppet and sealing material might help.

When the gas jet is working properly, an interesting experiment would be to continue the started polarization scan of hydrogen clusters to gain more data points and confirm the found results. This time, however, with a stable cluster signal over the shooting period and a known pulsewidth to compare the laser rise time with the characteristic time of ion motion. Also the unexpectedly found difference in maximum energies for hydrogen and deuterium in the energy scan of methane clusters warrants further research.

Another experimental idea that unfortunately could never be realized is to do a pump probe experiment with clusters to study the time evolution of the ion expansion. The idea is to use an initial pulse to start the cluster explosion in the hydrodynamic limit and then use the second harmonic of this pulse to further heat the plasma a short time after the explosion onset [21]. If the timing is chosen correctly the second laser pulse can hit the cluster such that the electron density in the cluster has dropped to three times the critical density \( n_{\text{crit}} \) which leads to an optimum heating rate as described in section 2.3.2. Thus with two pulses less energy is necessary to create high energy ions than is required if just one pulse was used.
Bibliography


Vita

Matthias Hohenberger was born in Berlin-Lichtenberg, Germany on March 23rd in 1980 as the son of Claus and Christa Hohenberger. After finishing his military service in spring 2000, he went to college at the Bayerische Julius Maximilians Universität Würzburg, Germany. There he took the intermediate examination for the diploma program in the summer of 2002. In the fall of 2003, after one year of graduate studies in Würzburg, he became a graduate student at the University of Texas at Austin. Since then he has been working for Prof. Todd Ditmire in the physics department.

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