Ultrafast Dynamics in Explosions of Atomic Clusters
Irradiated by Ultra-High Intensity Laser Pulses

by

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Approved by
Supervising Committee:

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To Marion
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Andreas Florian Henig, M.A.
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Supervisor: Todd Ditmire

The focus of this experimental work is to gain deeper insight into the explosion dynamics of nanoscale atomic clusters irradiated by ultra-high intensity femtosecond laser pulses. Energy distributions of ions accelerated in the expansion of small low Z as well as large high Z clusters are studied.

A spatial anisotropy peaked along the laser polarization direction and superior energies for an increased pulse duration are found for hydrogen clusters, suggesting the activation of cluster electron multiple-pass vacuum heating that was recently predicted in literature. Through irradiation of large argon clusters with a timed sequence of two ultrashort infrared pulses a strong enhancement for an optimum delay is observed, indicating the presence of a resonance in collisional heating.
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Chapter 1

Introduction

In past years, high-intensity (> $10^{15} \text{ W/cm}^2$) short pulse (≤ 1 ps) laser interaction with matter has been a vivid field of research. Typical experiments involved studies of either a low density gas (< $10^{19} \text{ atoms/cm}^3$) or a high density solid target (10$^{23}$ atoms/cm$^3$) exposed to such high laser intensities. A large number of these studies were motivated by the ambition to generate incoherent and coherent x-rays as well as high-energetic particles.

For use in x-ray generation, gas phase targets are attractive since coherent x-ray radiation can be produced via harmonic generation when the laser propagates through the medium. In addition, a plasma channel suitable for x-ray laser gain can be created. However, due to the poor absorption of laser energy from gases their effective use in x-ray generation is limited.

In contrast, solid targets usually absorb a large percentage of the incident laser energy. Particle energies ranging up to $MeV$ in magnitude have been observed as well as copious amounts of x-rays emitted from the hot plasma. Other than gases, solid targets usually require careful alignment and unwanted debris is generated as a consequence of the laser interaction.

A unique combination of both gas and solid phase components can be produced by high-pressure gas jets. Solid density clusters of atoms and molecules are formed by the adiabatic expansion of gas into vacuum. Thus, a very high local density that allows efficient laser absorption and results in the generation of hot plasmas is combined with a low global density and a lack of debris.

In recent years, the observation of keV electrons, $MeV$ ions as well as x-rays in
the keV energy range produced by high intensity laser interactions with clusters has been reported. The ability of clusters to efficiently couple laser energy into fast ions [1] is of particular interest for the development of compact, short-pulse neutron sources ([2], [3]). The nature of the cluster explosion is determined by the fraction of electrons that are extracted from the cluster due to the laser electric field. An ion acceleration through Coulomb repulsion followed by an isotropic explosion is observed from fully stripped clusters. At the opposite limit, when the cluster space charge prevents the vast majority of electrons from escape, the cluster can be pictured as a quasi-neutral nanoscale plasma sphere which undergoes an isotropic hydrodynamic expansion ([10], [39]). In the intermediate case where the cluster is partially ionized neither of these models is a valid description of the explosion dynamics. Two distinct populations are formed by the remaining electrons: a cold electron core within the cluster boundary and a halo of extracted electrons ([15], [16]). Both of them will oscillate along the laser polarization direction, thus breaking the symmetry of the cluster expansion.

This work presents experiments carried out in the hydrodynamic regime as well as in the intermediate case to gain deeper insight into the respective interaction mechanisms and the subsequent cluster explosion.
Chapter 2

Theory of Laser Cluster Interactions

This chapter will give an introduction to the background necessary for a description of the interaction of intense, ultrashort pulses with clusters in the gas phase. Subsequently to a description of the ionization mechanisms relevant processes heating the generated plasma will be presented. In addition, different cluster explosion models and their respective coverage of experimental parameters will be addressed.

2.1 The Ponderomotive Potential

The equation of motion of a free electron driven by an oscillating electromagnetic field is given by

\[ m_e \frac{d\vec{v}}{dt} = -e\vec{E} - e(\vec{v} \times \vec{B}) \]  

(2.1)

with \( \vec{v} \) being the electron velocity, \( e \) its charge and \( m_e \) its mass.

For an oscillation frequency of \( \omega_L \) the electric component of a linear polarized field can be written as \( \vec{E} = \vec{E}_0 \sin(\omega_L t) \) whereas the magnetic component is \( \vec{B} = \vec{B}_0 \cos(\omega_L t) \). For the regime of laser intensities below \( 10^{19} \text{ W/cm}^2 \) the magnetic force is negligible, therefore equation 2.1 solves to

\[ \vec{v} = \frac{e\vec{E}_0}{m_e\omega_L} \cos(\omega_L t) \]  

(2.2)


\[ \Delta E = \hbar \nu \]

| \[ i \] >

\[ I_{p,i} \]

Fig. 2.1. Schematic illustration of multi-photon-ionization. An electron in a bound state \( |i\rangle \) can reach the continuum by absorbing several photons on a short timescale to overcome its ionization potential \( I_{p,i} \).

The cycle averaged kinetic energy of a free electron oscillating in a linear polarized electromagnetic field that is referred to as its ponderomotive potential is thus

\[
U_p = \frac{1}{2} m_e \langle v^2 \rangle = \frac{e^2 E_0^2}{4m_e \omega_L^2} \propto I \lambda^2 \tag{2.3}
\]

It is proportional to the product of the laser intensity \( I \) and the square of its wavelength \( \lambda \).

### 2.2 Ionization mechanisms

To generate free electrons and ions that are subject to acceleration in the field of a strong laser pulse, ionization of the atoms bound to clusters has to happen before. Dependent on the intensity of the laser, different processes gain importance in the theoretical description.

#### 2.2.1 Multi Photon Ionization

Even if the energy of a single photon is below the ionization potential of an atom, an electron can still gain sufficient energy to overcome the potential barrier of the nucleus. In a process called *multi photon ionization* an electron is excited by several photons on a timescale so that it enters a continuum state before decaying back to
the ground state. By expanding the theory of one photon dipole transitions to higher orders an equation for the probability $W_{fi}$ of this mechanism that is schematically depicted in figure 2.1 can be found.

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

$$= 2\pi (2\pi \alpha \omega)^k g^{(k)}(\omega)$$

$$\cdot \sum_g \sum_n \cdots \sum l \frac{\langle f| r | g \rangle \langle g|r|n \rangle \cdots \langle l|r|i \rangle}{[\omega_{gi} - (k - 1)\omega][\omega_{ni} - (k - 2)\omega] \cdots [\omega_{li} - \omega]^k}$$

$$= 2\pi (2\pi \alpha \omega)^k g^{(k)}(\omega)$$

$$\cdot \sum_g \sum_n \cdots \sum l \frac{\langle f| r | g \rangle \langle g|r|n \rangle \cdots \langle l|r|i \rangle}{[\omega_{gi} - (k - 1)\omega][\omega_{ni} - (k - 2)\omega] \cdots [\omega_{li} - \omega]^k}$$

$$= 2\pi (2\pi \alpha \omega)^k g^{(k)}(\omega)$$

Here $k$ is the number of photons that are absorbed, $\alpha$ is the fine structure constant, $\sigma^{(k)}$ stands for the generalized cross section, $g^{(k)}(\omega)$ for the density of states as a function of photon energy $\omega$, $\omega_{ab}$ for the energy between atomic levels $a$ and $b$ and $I$ denotes the laser intensity. The sum is calculated over all possible dipole transitions.

### 2.2.2 Tunnel Ionization

For intensities above $I = 10^{14} W/cm^2$ the laser field is strong enough to cause a significant distortion of the atomic potential. A perturbation treatment is therefore not valid anymore. It is replaced by a quasi-classical approach describing the electron trapped in the atom potential well.

In figure 2.2(b) it is shown how the strong external field of a laser polarized in $x$-direction manipulates the former Coulombic potential so that the previously bound electron is enabled to tunnel through the barrier on one side. This process is referred to as tunnel ionization.

The probability for tunnel ionization in complex atoms has been worked out by Ammosov, Delone and Krainov [6]. The so called ADK-rate $W_{ADK}$ within one optical cycle is given by

$$W_{ADK} = C_{n^*Im}^2 |I_p| e \frac{2(2|I_p|)^{3/2}}{4E_0} \left( \frac{3E_0}{\pi (2|I_p|)^{3/2}} \right)^{1/2} \left( \frac{2E_0(2|I_p|)^{3/2}}{2n^* - |m| - 1} \right)^{2n^* - |m| - 1}$$

5
(a) Bound electron state (marked blue) in absence of an external field

(b) Distortion of the atomic potential by a strong laser field (highlighted red) enabling tunnel ionization

(c) Complete barrier suppression in an even higher field is greatly increasing the ionization rate

Fig. 2.2. Shown is the distortion of an atomic potential subject to a strong laser field polarized in x-direction. Without the presence of the external field the potential is purely coulombic. A strong field is lowering the potential barrier on one side so that the electron is enabled to tunnel through. At even higher fields the barrier gets suppressed below the ionization potential allowing the electron to leave easily.
with \[ C_{n+1} = (2e/n^*)^{n^*} (2\pi n^*)^{-1/2} \] \[ f_{lm} = \frac{(2l + 1)(l + |m|)!}{2|m||m|(l - |m|)!} \] \[ n^* = Z(2|I_p|)^{-1/2} \]

In this equation \( l \) denotes the angular momentum and \( m \) the magnetic quantum number of the electron state. \( I_p \) is the respective ionization potential and \( E_0 \) the laser electric field.

### 2.2.3 Barrier Suppression Ionization

With a further increase in laser intensity, the potential barrier gets eventually suppressed below the ionization potential of the electronic state. Thus, *barrier suppression ionization* (BSI) becomes the dominant mechanism of generating free electrons (see figure 2.2(c)).

The effective potential of an atom with atomic number \( Z \) situated in a linearly (in x-direction) polarized external field \( E_{ext} \) is

\[
V(x) = -\frac{Ze^2}{4\pi\epsilon_0|x|} - eE_{ext}x
\]

By solving \( dV(x)/dx = 0 \) a local maximum of \( V(x) \) can be found, corresponding to a position \( x_{max} \) of

\[
x_{max} = \sqrt{\frac{Ze}{4\pi\epsilon_0 E_{ext}}}
\]

The electric field required to enable barrier suppression ionization can be obtained by equating the effective potential at \( x = x_{max} \) with the ionization potential \( I_p \) yielding

\[
E_{BSI} = \frac{I_p^2\pi\epsilon_0}{Ze^3}
\]
The associated laser intensity $I_{BSI}$ is thus given by

$$I_{BSI} = \frac{c\epsilon_0}{2} |E_{BSI}|^2 = \frac{c\pi^2 \epsilon_0 \mathcal{I}_p^4}{2Z^2 e^6}$$

(2.13)

To give a numeric example, for hydrogen with an ionization potential of $I_{p,H} = 13.6\, eV$ the minimum laser intensity necessary for BSI is $I_{BSI,H} = 1.4 \cdot 10^{14}\, W/cm^2$.

### 2.2.4 The Keldysh Parameter

In order to determine the dominant ionization mechanism prevailing under particular experimental conditions, the Keldysh parameter $\gamma$ is a good criteria. Its definition is

$$\gamma = \sqrt{\frac{I_p}{2U_p}}$$

(2.14)

In the case of $\gamma > 1$ multi photon ionization is the relevant process while tunnel ionization becomes significant for $\gamma \ll 1$.

The experiments conducted with hydrogen were done at peak intensities around $10^{18}\, W/cm^2$ which gives a Keldysh parameter of about $\gamma \approx 10^{-2}$. This strongly indicates the predominance of the tunneling regime over multi photon ionization.

For the expected charge states of argon in the two color experiment, $\gamma$ was also below one.

### 2.2.5 Above Threshold Ionization (ATI)

During an ionization process one speaks of above threshold ionization if an electron is not ionized at the maximum of the laser electric field but at a phase shift $\phi > 0$ with respect to the peak. The increase in electron kinetic energy is then given by

$$K_e = 2U_p \sin^2 \phi$$

(2.15)

For $\phi = 0$ the electron velocity will be zero after the laser puls passed, because the timed average acceleration by the laser field will be zero. A maximum of $K_e = 2U_p$ can be reached if the electron leaves the binding potential well at $\phi = \pi$. 
Considering that the probability of tunnel ionization has its maximum at the peak of the laser electric field the vast majority of electrons will be ionized close to $\phi = 0$. Thus, the gain in their kinetic energy will be small compared to the ponderomotive potential making above threshold ionization an inefficient way of energy transfer into the plasma if tunnel ionization is predominant.

2.2.6 Collisional Ionization

Collisional ionization results from inelastic collisions between electrons and ions. Due to the high density in the cluster (around solid density of about $10^{23}$ atoms/cm$^3$) this mechanism will dominate the production of higher charge states once some electrons are produced by direct optical ionization processes as described above.

The associated collisional ionization rate formula was empirically determined by Lotz [7]. Averaged over a Maxwellian electron distribution the rate per ion is given by

$$W_{\text{coll}} = n_e \frac{a_i q_i}{I_p(kT_e)^{1/2}} \int_{I_p/kT_e}^{\infty} \frac{e^{-x}}{x} dx$$

(2.16)

Here $n_e$ denotes the electron density, $I_p$ is the ionization potential in eV, $a_i$ is an empirical constant equal to $4.5 \cdot 10^{-14}$ eV$^2$ cm$^{-3}$ [7] and $q_i$ gives the number of ion valence electrons.

For the validity of the assumption of a uniform temperature $T_e$ within the cluster as well as an isotropic electron velocity distribution described by a Maxwellian, the electron thermalization time is implied to be small on the time scale of the laser cluster interaction. Thus the small spatial size of the cluster (on the order of a few nanometer in radius) will prevent the buildup of significant thermal gradients.

Considering typical conditions prevailing in a cluster the collisional ionization rate can be quite high. As an example, $W_{\text{coll},kT}$ in a solid density argon cluster for ionization from Ar$^{8+}$ to Ar$^{9+}$ ($I_p = 422$ eV) at a temperature of $T_e = 1$ keV and a density $n_e = 2 \cdot 10^{23}$ cm$^{-3}$ calculates to $0.3$ fs$^{-1}$. In comparison, the tunnel ionization rate for creating Ar$^{9+}$ starts to become significant not until a laser intensity of $> 10^{18}$ is utilized.

Equation 2.16 is solely taking the randomized electron motion into account. Besides that, the electrons will exhibit an oscillation that is driven by the laser electric field.
Calculating an associated collisional ionization rate is difficult for a dense plasma due to the high number of collisions. An estimate can be found by assuming the electron velocity to be purely sinusoidal. This simplification will generally result in an overestimate of the collisional ionization rate since the real maximum electron velocity will be lower than in the peak of a pure sinusoidal oscillation.

With above prerequisite, the collisional ionization rate \( W_{\text{coll,las}} \) per ion due to the electron oscillatory motion is

\[
W_{\text{coll,las}}(t) = n_e \sigma_i e \frac{E}{m_e \omega} |\sin \omega t|  \tag{2.17}
\]

with \( \sigma_i \) being the velocity-dependent cross section for ionization. \( W_{\text{coll,las}}(t) \) is nonzero only if the kinetic energy of the driven electron exceeds the ion ionization potential \( I_p \). In order to calculate a cycle-averaged rate that can be compared to equation 2.6 and 2.16 one thus has to solve the following integral

\[
W_{\text{coll,las}} = \langle W_{\text{coll,las}}(t) \rangle = \frac{n_e}{\pi} \int_{\phi_{\text{min}}}^{\pi} \sigma_i e \frac{E}{m_e \omega} \sin \phi d\phi  \tag{2.18}
\]

where \( \phi_{\text{min}} \) corresponds to the the phase where the electron kinetic energy equals \( K_e \) equals \( I_p \).

The ionization cross section as derived by Lotz is

\[
\sigma_i = a_i q_i \frac{\ln(K_e/I_p)}{K_e I_p}  \tag{2.19}
\]

From equation 2.15 that is giving the kinetic energy of an electron ionized above threshold from a non-moving atom/ion one can obtain

\[
\sin \phi d\phi = \frac{1}{4U_p \sqrt{1 - K_e/2U_p}} dK_e  \tag{2.20}
\]

This gives a cycle averaged ionization rate of

\[
W_{\text{coll,las}} = n_e \frac{a_i q_i}{\pi I_p} \left( \frac{1}{m_e U_p} \right)^{1/2} \int_{I_p}^{2U_p} \ln(K_e/I_p) 2K_e \frac{1}{\sqrt{1 - K_e/2U_p}} dK_e  \tag{2.21}
\]
Above equation exhibits a maximum in the ionization rate for electron kinetic energies around the ionization potential. The integral can not be solved directly but a reasonable approximation of $W_{\text{coll,las}}$ can be given by expanding the logarithm around small parameters $1 - I_p/K_e$ as follows

$$\ln(K_e/I_p) \approx \frac{3}{2} - 2 \frac{I_p}{K_e} + \frac{I_p^2}{2K_e^2}$$  \hspace{1cm} (2.22)

This approximation is giving good results for electron kinetic energies from around $K_e \approx I_p$ up to $K_e \approx 2I_p$. With equation 2.21 can be integrated, yielding [10]

$$W_{\text{coll,las}} \approx n_e \frac{a_i q_i}{2\pi I_p m_e^{1/2} U_p^{1/2}} \cdot \left\{ \left[ 3 + \frac{I_p}{U_p} + \frac{3}{32} \left( \frac{I_p}{U_p} \right)^2 \right] \cdot \ln \left[ \frac{1 + \sqrt{1 - I_p/2U_p}}{1 - \sqrt{1 - I_p/2U_p}} \right] \right\}$$

$$- n_e \frac{a_i q_i}{2\pi I_p m_e^{1/2} U_p^{1/2}} \cdot \left\{ \frac{7}{2} + \frac{3I_p}{8U_p} \right\} \cdot \left[ 1 - I_p/2U_p \right]$$  \hspace{1cm} (2.23)

$W_{\text{coll,kT}}$ and $W_{\text{coll,las}}$ give comparable rates if plasma temperature $kT_e$ is on the order of the ponderomotive potential $U_p$ inside the cluster.

### 2.3 Plasma Heating

There are several mechanisms to couple energy of the laser field into the plasma generated by aforementioned ionization processes. Due to their lower mass the energy is primarily transferred to electrons. In subsequent collisions or through space charge effects in the case of electrons leaving the cluster, ions get accelerated.

This section will present heating processes that are relevant for absorption of ultra-short, ultra-intense laser light in dense plasmas.

#### 2.3.1 The Critical Density

The propagation of electromagnetic waves in a plasma can be described by following relativistically correct dispersion relation [8]

$$kc = \sqrt{\omega_p^2/\gamma - \omega_0}$$  \hspace{1cm} (2.24)
with $k(\omega_0)$ being the wavenumber (frequency) of the laser light, $\gamma = \sqrt{1 + p_{osc}}$ the relativistic factor, $p_{osc}$ the transverse quiver momentum and

$$\omega_p = \sqrt{4\pi n_e e^2/m_e}$$

(2.25)

denoting the plasma frequency. In the nonrelativistic case which is valid up to intensities of about $10^{18} \text{W/cm}^2$, $\gamma \approx 1$ can be applied. From equation 2.24 follows that $k$ gets zero when $\omega_p = \gamma \omega_0$ which becomes

$$\omega_p = \omega_0$$

(2.26)

at nonrelativistic intensities. This is the condition for the laser to no longer propagate through the plasma but being reflected. The associated electron density can directly be derived from equation 2.25 to

$$n_{cr} = \frac{\omega_0^2 m_e}{4\pi e^2}$$

(2.27)

This density $n_{cr}$ is generally referred to as the critical density.

The area behind the critical surface is slightly penetrated by an exponentially decreasing laser field. The distance over which this happens is determined by the plasma density and is called collisionless skin depth. In the nonrelativistic limit it is given by $c/\omega_p$.

**2.3.2 Collisional Heating**

The transfer of energy from the laser electric field into the plasma is dominated by collisional heating (or inverse bremsstrahlung) in the case of large clusters for which the laser field is strong enough to ionize the cluster atoms but not sufficient to extract a significant number of electrons out of the cluster boundary. Thus, the quasi-neutral cluster can be treated as a small ball of high density plasma [10]. As in section 2.2.6 a uniform temperature and an isotropic electron velocity distribution that is described by a Maxwellian is assumed. To be precise, for the validity of the cluster approximation by a small plasma ball the cluster dimension has to be much
larger than a Debye length \( \lambda_d \)

\[
\lambda_d = \sqrt{\frac{kT_e}{4\pi n_e e^2}} \quad (2.28)
\]

For a plasma electron temperature of \( T_e = 1 \) keV the Debye length is \( \lambda_d \approx 5 \text{Å} \) at solid density which means that large clusters of 50 – 100Å do meet this prerequisite. In addition, the density within the cluster is taken to be uniform.

With above assumptions, the heating rate can be calculated by considering the laser energy deposition rate in a dielectric sphere which is per unit volume given by [9]

\[
\frac{\partial U}{\partial t} = \frac{1}{4\pi} \vec{E} \cdot \frac{\partial \vec{D}}{\partial t} \quad (2.29)
\]

Time averaging this equation over one laser cycle after inserting \( \vec{E} = \frac{1}{2} \hat{x}(E e^{i\omega t} + \text{c.c.}) \) for the laser field inside the cluster and \( \vec{D} = \epsilon \vec{E} \) yields

\[
\frac{\partial U}{\partial t} = \frac{\omega}{8\pi} \text{Im}[\epsilon]|E|^2 \quad (2.30)
\]

where \( \epsilon \) denotes the dielectric constant of the cluster plasma.

Due to the small cluster diameter with respect to the laser wavelength, the field inside the cluster can be determined by examining the field \( E \) inside a uniform dielectric sphere surrounded by a constant field \( E_0 \)

\[
E = \frac{3}{|\epsilon + 2|} E_0 \quad (2.31)
\]

Hence, the heating rate becomes

\[
\frac{\partial U}{\partial t} = \frac{9\omega}{8\pi} \text{Im}[\epsilon]|E_0|^2 \quad (2.32)
\]

The plasma dielectric constant \( \epsilon \) can be obtained using a Drude model, resulting in

\[
\epsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\nu)} \quad (2.33)
\]

Here, \( \nu \) is the electron-ion collision frequency and \( \omega_p \) the plasma frequency as defined in section 2.3.1.
Thus, the collisional heating rate per unit volume in a quasi-neutral cluster is

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

(2.34)

From this equation it can be seen that the laser electric field is shielded for densities \( n_e/n_{cr} >> 3 \), compared to the vacuum electric field \( E_0 \) the cluster exhibits a reduced field \( E \) within its boundary. Therefore, collisional heating becomes inefficient for electron densities above three times the critical density.

However, the heating rate rises with the increase of \( n_e \) until it reaches a global maximum at \( n_e = 3n_{cr} \). At this resonance \( |\epsilon + 2| \) goes through a minimum causing a great enhancement in the electric field \( E \) inside the cluster.

Figure 2.3 shows the heating rate by inverse bremsstrahlung as a function of \( n_e/n_{cr} \) for three different values of the collision frequency \( \nu \) which causes a damping of the resonance.

For a description of the electron-ion frequency the standard Coulomb formulas of Silin [11] can be used in the extreme cases where the electron oscillation velocity
$v_{osc}$ is much smaller than the electron thermal velocity $v_{kT}$ and vice versa.

$$\nu = \frac{4}{9} \left( \frac{2\pi}{3} \right)^{1/2} \frac{Z^2 e^4 n_i}{m_e^{1/2} \langle kT_e \rangle^{3/2}} \ln \Lambda, \quad v_{osc} << v_{kT} \quad (2.35)$$

$$\nu = \frac{16Z^2 e^4 n_i m_e \omega}{E_0^3} \left( \ln \left[ \frac{eE_0}{2m_e \omega v_e} \right] + 1 \right) \ln \Lambda, \quad v_{osc} >> v_{kT} \quad (2.36)$$

In these equations, $\ln \Lambda$ stands for the standard Coulomb logarithm. For the intermediate case at $v_{osc} \approx v_{kT}$, the general equation given in [11] has to be integrated numerically. It has to be noted that at high densities the equations of Silin generally predict collision frequencies that are too large. In a simulation of the heating process this has to be taken into account. However, since the cluster will expand while being heated the density will drop simultaneously and solid density will only be maintained for a short time.

### 2.3.3 Resonance Absorption

An early presentation of resonance absorption can be found in publications by Forslund et al. [12] and Estabrook et al. [13]. The mechanism will be briefly discussed for a laser incident on a plane solid target.

A laser that is hitting the target at an angle $\theta$ with respect to the surface normal can resonantly excite an electron plasma wave at the laser frequency $\omega_0$ if the electric field vector $\vec{E}$ is in the direction of the density gradient $\vec{\nabla} n$ that is present at the vacuum-plasma transition. Electrons are accelerated by the electrostatic field that is associated with this resonant wave.

The fraction of energy transferred from the laser electric field to the electron plasma wave can be derived by including a small amount of damping to be [8]

$$f_{ra} \approx \frac{2.3}{2} \tau e^{-\frac{4\tau^3}{3}} \quad (2.37)$$

with

$$\tau = \left( \frac{\omega_0 L_m}{c} \right)^{1/3} \sin \theta \quad (2.38)$$
Here, $c$ denotes the speed of light and $L_n$ the density scale length that is defined as

$$L_n \equiv n \frac{dx}{dn} \approx C_s \tau_L$$

(2.39)

where $n$ is the plasma density, $dn/dx$ is the associated slope at density $n$, $C_s = \sqrt{k(ZT_e + T_i)/m_i}$ is the ion sound speed, $T_e$ is the electron temperature, $m_i$ is the ion mass, $k$ is the Boltzmann constant and $\tau_L$ refers to the time of the ionizing laser.

Equation 2.37 shows that the resonance absorption rate depends strongly on the density scale length. In the case of ultra-short pulses that are interacting with small, nanoscale clusters, the density gradient at the vacuum-plasma barrier is generally quite steep. Therefore, the density scale length is much too short to let $f_{ra}$ reach its peak. Resonance absorption is thus not a relevant process for energy deposition into cluster plasmas.

### 2.3.4 Stochastic Heating

The mechanism of stochastic heating ([16]) is similar to conventional vacuum heating first pronounced by Brunel ([17],[18]). While aforementioned collisional heating was derived for a quasi-neutral cluster, parts of the electron population will cross the cluster boundary in the case of stochastic heating.

Given a uniform spherical cluster of density $n_0$ and radius $R_0$ in a uniform static field electric field $E_0$ a total charge of $(4/3)\pi R_0^3 n_0 |e|$ will be obtained by removing all electrons, corresponding to an electrical field at the cluster edge of

$$E_{max} = \frac{4}{3} \pi R_0 n_0 |e|$$

(2.40)

The laser electric field will only be able to extract all electrons, if the condition $E_0 > E_{max}$ is fulfilled. In case of $E_0 < E_{max}$ a cold electron core of density $n_0$ will be formed by electrons that remain inside the cluster [15]. This situation is depicted in figure 2.4. The radius of the core is given by $R_e = R_0 - d_0$ where

$$d_0 = 3 \frac{|e| E_0}{m_e \omega_{p0}^2}$$

(2.41)
denotes the core offset with respect to the cluster center with

\[ \omega_{p_0}^2 = 4\pi n_0 e^2 / m_e. \]

The cold core electrons are in equilibrium since the external field inside the electron core is canceled out by the self-consistent field of the cluster.

The number of extracted electrons is found to be

\[ N_{\text{ext}} = \frac{4}{3} \pi n_0 [R_0^3 - (R_0 - d_0)^3] \]

which for a small \((d_0 << R_0)\) core displacement can be approximated to

\[ N_{\text{ext}} \approx 4\pi n_0 R_0^2 d_0 \]  \hspace{1cm} (2.42)

It has to be stressed that above derivation assumes a rise time scale of the laser electric field from zero to \(E_0\) that is much longer than \(1/\omega_{p0}\). This assures that the confined electrons will adjust adiabatically to the electric field.

When moving on from a static external field \(E_0\) to a field that is oscillating with the laser frequency \(\omega\), \(E_0\) will still be effectively static for the electrons since they adjust adiabatically to the laser field. In this picture a key role will be played by the electrons that are extracted from the cluster. Their characteristic energy \(\epsilon_e\) gained...
in the laser field can be approximated by two times the ponderomotive potential

\[ \epsilon_e \approx 2U_p = \frac{m_e}{2} \left( \frac{eE_0}{m_e\omega} \right) \tag{2.43} \]

In comparison, an estimate for the energy required to overcome the binding potential of the cluster sphere is

\[ U \approx e^2 N_{\text{ext}}/R_0 \tag{2.44} \]

The condition for the electrons not to be bound to the cluster \( \epsilon_e >> U \) is equivalent to requirement that the quivering amplitude \( \xi \) of a free electron

\[ \xi = \frac{|e|E_0}{m_e\omega^2} \tag{2.45} \]

exceeds the cluster radius. For \( \xi >> R_0 \) the extracted electrons will never return to the cluster while in the opposite case of \( \xi << R_0 \) they will be bound to it. In both regimes collisional heating by the oscillating cold electron core will take place. However, the associated energy transfer rate is a rapidly decreasing function of electron temperature (see section 2.3.2).

Stochastic heating can provide considerably higher particle energies. Electrons pulled out by the laser are pushed back once the field reverses. The number of these (warm) electrons is roughly given by \( N_{\text{ext}} \) while their energy is \( \epsilon_e \). Due to the absence of an internal cluster electric field internal, the warm electrons propagate freely through the cluster with a velocity of \( \sqrt{\epsilon_e/m_e} \). This process is repeated for the next laser cycles until the warm electrons with density \( n_0d_0/\xi \) fill the entire cluster and extent into the vacuum beyond the opposite cluster boundary which is equal to the time it takes for a warm electron to cross the cluster

\[ \tau \approx \frac{R_0}{\sqrt{\epsilon_e/m_e}} \approx \frac{1}{\omega} \frac{R_0}{\xi} \tag{2.46} \]

From this point on further extraction of warm electrons is suppressed. An estimate of the maximum number of hot electrons can be made to be

\[ N_H \approx 4\pi R_0^2 d_0 n_0 \omega \tau \approx N_0 \frac{\omega^2}{\omega_{\text{pe}}^2} \tag{2.47} \]
Here, $N_0$ refers to the total number of ions within the cluster. Since $\omega << \omega_p$, $N_H$ is much smaller than the number of cold electrons.

While the warm electron energy is $\epsilon_e$ for $t < \tau$ these electrons heat up from $t > \tau$ on, gaining an energy of $\epsilon_e$ every time they leave the cluster after passing through. This leads to an electron energy of

$$\epsilon_{el} \approx m_e \frac{t^2}{R_0^2} \left( \frac{eE_0}{m_e\omega} \right)^4$$

(2.48)

The process of stochastic heating becomes uneffective as soon as the electrons reach a velocity so high that they do not loose their phase anymore while transiting the cluster, i.e. $R_0/\sqrt{\epsilon_e/m_e} \geq 1/\omega$ is no longer valid. Hence, the maximum energy that can be transferred to an electron via stochastic heating is

$$\epsilon_{el,max} \approx \frac{1}{2} m_e R_0^2 \omega^2$$

(2.49)

This energy by far exceeds the quiver energy of a free electron subject to laser field.

### 2.4 Dynamics of the Cluster Explosion

The respective cluster explosion scenario obviously depends on the experimental parameters, such as laser pulse duration, intensity in the focus and the radius of the clusters. In the following some cases of interest will be addressed.

#### 2.4.1 Small Clusters - Pure Coulomb Explosion

For $R_0 < d_0$, i.e. the cold core displacement exceeds the cluster radius and all electrons will be extracted from the cluster, leaving a sphere composed of bare ions behind. If this occurs on a sufficiently short time, the ions can well be assumed to remain at their initial position during the process.

Subsequently, the ions will be accelerated by Coulomb repulsion forces due to the electric field of their own space charge. This scenario is often referred to as Coulomb explosion. An individual ion will thus gain an energy $\epsilon_i$ of [16]

$$\epsilon_i = \frac{1}{3} m_i \omega_{pi}^2 r_0^2$$

(2.50)
where $r_0$ denotes the ion’s initial position in the cluster and $\omega_{pi} = \sqrt{4\pi n_0 e^2/m_i}$.

An approximate of the time scale attributed to a cluster Coulomb explosion (i.e. a doubling in radius) can be given by

$$\tau_i \approx 1/\omega_{pi} \quad (2.51)$$

### 2.4.2 Medium Clusters

In the case of $d_0 < R_0 < \xi$ electrons will only be partially removed from the cluster. Again, the ions are assumed to be immobile during that process, resulting in a neutral core concentrically surrounded by a positive ion shell. These shell ions will be radially accelerated by Coulomb forces, reaching energies of $[15]

$$\epsilon_i = \frac{m_i \omega_{pi}^2 [r_0^3 - (R_0 - d_0)]}{3r_0} \quad (2.52)$$

The total number of shell ions is given by the number of extracted electrons $N_{ext}$ (see equation 2.42). A time scale associated with the explosion is

$$\tau_i \approx \sqrt{m_i R_0 |e| E_0} \quad (2.53)$$

Hence, the validity of the assumption of non-moving ions can be verified by comparing the actual laser pulse width to $\tau_i$.

### 2.4.3 Large Clusters

In this section large clusters ($d_0 < \xi < R_0$) will be considered that are irradiated by a laser pulse of pulse duration $\tau_{laser}$ so that the condition $\tau \leq \tau_{laser} \leq \tau_{max}$ is fulfilled. Here, $\tau$ is given by equation 2.46 and $\tau_{max}$ is defined as the time were stochastic heating of the electrons stops

$$\tau_{max} \approx \frac{1}{\omega} \left( \frac{e E_0}{m_e \omega^2 R_0} \right)^{-2} \approx \frac{1}{\omega} \frac{R_0^2}{\xi^2} \quad (2.54)$$

After the interaction with such a pulse, the cluster consists of a neutral core surrounded by a thin ion shell and a comparably thick electron halo of approximately equal absolute value of charge. As a result, shell ions get radially accelerated and
gain energy with respect to the hot halo electrons while passing the double layer. Simultaneously, a drop in ion density from $n_0$ to the much smaller hot electron density takes place. As soon as the ion shell thickness equals the electron halo thickness, both ions and hot electrons expand with equal velocity. Due to the adiabatic expansion of the electrons their temperature decreases while the ion kinetic energy rises on the same order as the amount gained during acceleration in the double layer. Once the initial ion shell has expanded, a new one has formed at the edge of the cold electron core so that there is a double layer present at all times until the whole hot electron population has left the core. Thus, equation 2.47 yields the number of fast ions generated in the explosion. Their individual energy is determined by

$$\epsilon_i \approx m_e \tau_{\text{laser}}^2 \left( \frac{eE_0}{m_e \omega} \right)$$

with $\tau_{\text{laser}}$ being the laser pulse duration.

### 2.4.4 Quasi-neutral Clusters - Hydrodynamic Expansion

In the case of a quasi-neutral cluster where virtually none of the electrons has left during the heating process, the expansion will be dominated by the pressure associated with the heated electrons which is simply given by

$$P_e = n_e kT_e$$

The cold heavy ions will be pulled outwards by the hot electrons. For ions of charge $Ze$ the characteristic speed of this expansion is given by the plasma sound speed

$$v_{\text{expand}} \sim \left( \frac{ZkT_e}{m_i} \right)^{1/2}$$
Chapter 3

Experimental Apparatus and Techniques for Data Acquisition and Analysis

In this section, a compressed introduction to the laser system utilized to conduct the experiments described will be given. This will also include a description of the basic concepts associated with high intensity lasers. The target chamber layout including diagnostics and the mechanism of cluster production in high pressure gas jets will be addressed, followed by some general comments on data analysis techniques used.

3.1 The THOR Laser Facility

3.1.1 The Concept of Mode-Locking

To obtain pulsed lasers of ultra high intensity, a technique is needed to store the laser energy in single, ultrashort pulses rather than in cw beams. A breakthrough on this field happened in the late 1960s, when the concept of mode-locking was developed, which will be explained briefly.

First, an inhomogeneous broadened laser well above threshold is assumed. The gain spectrum is implied to be constant over frequency. The electric field inside the cavity
Fig. 3.1. Ideal mode-locked pulses resulting from $N_M = 9$ locked modes and a square gain medium can therefore be written as a sum over all oscillating axial modes

$$E(t) = E_0 \sum_{n=-s}^{s} e^{i[(\omega_0 + n \Delta \omega_{ax})t + \phi_n]}$$

with the frequency spectrum centered at $\omega_0$ and axial mode distance $\Delta \omega_{ax}$. The limits of the sum are given by $s = (N_M - 1)/2$ with $N_M$ being the number of axial modes oscillating above threshold. By introducing a process to lock all modes at a constant and equal value of phase (for simplicity $\phi_n = 0$ is supposed), the total electric field becomes

$$E(t) = E_0 e^{i\omega_0 t} \sum_{n=-s}^{s} (e^{i\Delta \omega_{ax} t})^n$$

$$= E_0 e^{i\omega_0 t} \frac{\sin (N_M \Delta \omega_{ax} t/2)}{\sin (\Delta \omega_{ax} t/2)}$$

The intensity $I(t)$ corresponding to this electric field is shown exemplarily for $N_M = 9$ locked modes in figure 3.1. With an increasing number of locked modes $N_M$, the peak intensity $I_{\text{peak}}$ of the main pulses increases with respect to the average intensity $\bar{I}$ as $I_{\text{peak}} = N_M \bar{I}$ while their FWHM pulse duration $\tau$ decreases as $\tau \approx T_{RT}/N_M$. 

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with $T_{RT}$ being the round-trip time of one pulse in the cavity. Thus, by locking a large number of modes at constant, equal phase, the peak power can be greatly increased until essentially the whole power is stored in the main pulses with a negligible amount in between.

In figure 3.1, mode locked pulses are shown that result from a uniform (square) gain spectrum. Besides the primary pulses, $N_M - 2$ much weaker subsidiary side peaks appear per period. For a Gaussian gain spectrum, mode-locking again yields a train of periodically spaced pulses, but the aforementioned small local maxima are essentially not present. Beyond that, the main pulses now exhibit gaussian shapes in time.

An intuitive time-domain picture of a mode-locked laser corresponds to a single pulse bouncing back and forth inside the cavity. The resulting output is a pulse train with repetition rate $RR$

$$RR = \frac{1}{T_{RT}} = \Delta \nu_{ax}$$  \hspace{1cm} (3.4)

Kerr-Lens Mode-Locking

There are various approaches to achieve mode-locking in a laser cavity. Generally, all of them can be referred to as either active or passive techniques.

An example for active mode-locking would be inner-cavity loss modulation via a fast shutter, i.e. an acousto-optic modulator (AOM). This AOM can be driven in such a way so that except during a small time around the minimum of the introduced loss the cavity is below threshold. Therefore, pulses are forced to form that lie within these loss minima at distances of $T_{RT}$. The pulse duration achieved with this technique mainly depends on two characteristics of the AOM:

- Modulation depth $\Delta_m$
- Modulation frequency $f_m$

A larger modulation depth will result in shorter pulses. This is simply due to the fact, that the width of the "low-loss window" decreases for increasing $\Delta_m$, thus shortening the pulse. For acousto-optic modulators, $\Delta_m$ is generally proportional to the RF power $P_m$ supplied to it

$$\Delta_m \propto P_m$$  \hspace{1cm} (3.5)
and therefore the pulse duration shows a weak dependence on $P_m$

$$\tau \propto P_m^{-1/4} \quad (3.6)$$

A higher modulation frequency $f_m$ will give shorter pulses as well. This can again be explained by the narrowing of the ”low-loss window”, this time due to the higher RF frequency.

However, in practice both modulation depth and frequency are limited, so that the theoretical boundary for the pulse length (which is given by the bandwidth of the gain medium, see section 3.1.2) does not lie within close reach. Therefore, for laser cavities producing pulses of a few fs, different approaches are used. A very popular technique is Kerr-lens mode-locking (KLM), which is referred to as a way of passive mode-locking.

The KLM mechanism is directly related to the optical Kerr effect, which describes a nonlinear change in the refractive index with respect to the electric field. The total refractive index can be written intensity-dependent by means of the equation

$$n = n_0 + n_2 \bar{I} \quad (3.7)$$

where $\bar{I}$ represents the time-averaged intensity of the optical field, given by

$$\bar{I} = \frac{n_0 c}{2\pi} |E(\omega)|^2 \quad (3.8)$$
Since the spatial profile of a Gaussian (or sech\(^2\)) pulse exhibits larger intensities for parts of the beam that are closer to the optical axis, the refractive index will be higher for those parts as well, while the beam is propagating through a nonlinear optical medium. Therefore, a nonlinear optical medium with positive \( n_2 \) will act to a Gaussian pulse just as an intensity-dependent convex lens and focus the beam. This effect is called *self-focusing*.

In figure 3.2 it is schematically depicted how Kerr-lens mode-locking takes advantage of that self-focusing effect. An increase in the intensity of the input beam will result in a decreasing beam diameter at the output. By including an intra cavity aperture into the setup, this intensity dependent beam size modulation results in a subsequent modulation in the propagation losses. While the circulating high intensity pulse mode is able to pass the iris ideally unaffected, the low intensity cw mode gets absorbed to a great extent and is kept below threshold for lasing. As figure 3.2 shows, the iris acts as a filter between both modes of operation. This technique is denoted as *hard aperture mode-locking*.

In contrast to the hard aperture that has to be added to the cavity, there is a second discrimination mechanism that is already present in every setup. Due to the spatial profile of the pump beam exciting the gain medium, the gain profile itself exhibits a spatial modulation as well. This results in the formation of an intrinsic *soft aperture*. Cavity modes that are showing the largest overlap with the excited gain medium are amplified the most on every round trip. Both the hard as well as the soft aperture contribute to a selective attenuation of low intensity modes, which leads to the buildup of short, high intensity pulses circulating in the passively mode-locked cavity.

### 3.1.2 Time-Bandwidth Product

For a square gain spectrum and a number of \( N_M \) modes locked, the pulse width \( \tau \) of the primary pulses is close to (see section 3.1.1)

\[
\tau \approx \frac{T_{RT}}{N_M}
\]

(3.9)
gain medium | HeNe | Ar⁺ | Nd:YAG | ND:glass | semiconductor | Ti³⁺ : Al₂O₃
--- | --- | --- | --- | --- | --- | ---
Δν_{gain} (s⁻¹) | 10⁹ | 3 × 10⁹ | 10¹⁰ | 10¹² | 10¹⁵ | 10¹⁴

**Table 3.1. Examples of the gain bandwidth Δν_{gain} for different materials commonly used**

This can be rewritten with the axial mode spacing \( Δν_{ax} = 1/RT \)

\[
\tau \approx \frac{1}{Δν_{ax} N_M} \tag{3.10}
\]

Assuming that all oscillating modes are locked, the gain bandwidth (or amplification bandwidth) \( Δν_{gain} \), which is defined as FWHM of the gain spectrum, can be approximated by

\[
Δν_{gain} ≈ Δν_{ax} N_M \tag{3.11}
\]

Therefore, the pulse duration of the main pulses becomes

\[
τ \approx \frac{1}{Δν_{gain}} \tag{3.12}
\]

Equation 3.12 denotes the ultimate pulse width that can be obtained from a mode-locked laser with uniform gain spectrum, while

\[
τΔν_{gain} ≈ 1 \tag{3.13}
\]

is referred to as time-bandwidth product. Examples for \( Δν_{gain} \) of prevalent amplification media are given in table 3.1.

Given a gaussian gain spectrum, the pulses obtained from a mode-locked cavity will indicate a gaussian envelope in time as well (see section 3.1.1) having an intensity \( I(t) \) as follows:

\[
I(t) = I_0 \exp \left( -4 \ln 2 \frac{t^2}{\tau^2} \right) \tag{3.14}
\]

The FWHM of one pulse in the time domain is therefore represented by \( τ \).
Fourier transforming the associated electric field

\[ E(t) = E_0 \exp \left( -\frac{2 \ln 2}{\tau^2} t^2 \right) \]  \hspace{1cm} (3.15)

\[ \Rightarrow \tilde{E}(\nu) = \tilde{E}_0 \exp \left( -\frac{\pi^2 \tau^2 \nu^2}{2 \ln 2} \right) \]  \hspace{1cm} (3.16)

results in an intensity spectrum given by

\[ \tilde{I}(\nu) = \tilde{I}_0 \exp \left( -\frac{\pi^2 \tau^2 \nu^2}{2 \ln 2} \right) \]  \hspace{1cm} (3.17)

Thus, the FWHM spectral range or bandwidth \( \Delta \nu \) of the pulse is

\[ \Delta \nu = \frac{2 \ln 2}{\pi \tau} \]  \hspace{1cm} (3.18)

and the time-bandwidth product calculates to

\[ (\tau \Delta \nu)_{\text{gaussian}} = \frac{2 \ln 2}{\pi} \approx 0.4413 \]  \hspace{1cm} (3.19)

Similarly, the time-bandwidth product of a \( \text{sec}^2 \) pulse can be ascertained to be

\[ (\tau \Delta \nu)_{\text{sec}^2} = \frac{4 \times \text{arccosh}^2 \sqrt{2}}{\pi^2} \approx 0.3148 \]  \hspace{1cm} (3.20)

For a given spectral width \( \Delta \nu \) of a laser pulse, a lower limit of the temporal width \( \tau \) can be obtained from its respective time-bandwidth product. A pulse at that minimum duration level is often referred to as transform-limited.

### 3.1.3 Chirped Pulse Amplification

Up to the mid 1980s, the ambition to build lasers with focused intensities in excess of \( 10^{15} \text{W/cm}^2 \) did not seem feasible. In order to obtain those intensities in the focus, one has to keep the unfocused intensities in the amplification stages under the damage threshold of the gain material. At that time, the common concept to deal with this problem was to increase the beam diameter prior to amplification, thus reducing the pulse intensity. This evolution led to laser systems incorporating optics with huge apertures such as SHIVA and NOVA, both located at Lawrence
Livermore National Lab (LLNL). With an aperture of 50 cm in the final amplification stage, NOVA was capable of producing around 100 kJ of infrared light at 1064 nm wavelength in a 2-4 ns pulse. Due to the enormous costs of such systems, implementation was restricted to National Lab facilities.

In 1985, this quandary was all about to change with the presentation of a new, revolutionary concept by D. Strickland and G. Mourou [4]. Rather than decreasing the intensity of the pulses by spatial stretching, they proposed to introduce a stretch in the time domain. This approach, referred to as Chirped Pulse Amplification (CPA) takes advantage of the pulse bandwidth and is schematically displayed in figure 3.3.

By including a dispersive element such as a pair of gratings into the laser chain prior to the first amplification stage, the pulse instantaneous frequency becomes linearly time-varying, the pulse is chirped. This reduction in intensity allows the use of small optics for all amplification stages. In order to regain a short pulse duration, the chirp of the fully amplified pulse can be almost completely compensated by a second dispersive optic. Due to the fact that a small beam diameter is sufficient for the whole laser with the exception of the final compressor, tabletop terra watt laser systems became possible.

3.1.4 Thor Laser Layout

The Texas High Intensity Optical Research facility accommodates such a modern 20 TW tabletop CPA laser device. Its structure is depicted in figure 3.4. The seed pulse is generated in a Femtolaser Femtosource Scientific s20 oscillator. A
thin highly doped Ti:sapphire crystal serves as gain medium in the folded compact cavity, amplifying on a broad wavelength range centered at around 800 nm (gain bandwidth $\Delta \nu_{\text{gain}} \approx 10^{14} \text{s}^{-1}$, see table 3.1.2). A 5W diode-pumped frequency-doubled Nd:YVO$_4$ Millennia Vs J laser pumps the Ti:sapphire crystal at 532 nm. Group Delay Dispersion (GDD) is controlled by chirped multi-layer dielectric mirrors. The cavity is passively Kerr-lens mode-locked, emitting 1nJ 20fs pulses at a repetition rate of 75 MHz. Subsequently, this frequency is reduced to 10 Hz by a Pockel’s cell before the pulses arrive at the stretcher.

The stretcher consists of a pair of gratings build in such a way so that different wavelength travel different path lengths, hence causing a chirp in the pulse, without introducing a spatial divergence. In the THOR setup, higher frequency parts have to cover a longer distance which is why the stretcher output yields a pulse with high wavelengths at the beginning, followed by gradually decreasing wavelength components. The FWHM pulse width at this stage is approximately 600 ps.

Now the first amplification stage, a 20 pass regenerative amplifier (regen) is en-
tered. Its Ti:sapph crystal is pumped at 532 nm by a q-switched frequency-doubled Quantel Big Sky Nd:YAG laser. At the point, where the gain starts to saturate, the amplified pulse is switched out by a high speed Pockel's cell. The so generated 3.5 mJ pulses get further amplified to an energy of 20 mJ, after the beam size is doubled to 4 mm. As is the regen, the Ti:sapph gain medium of this 4-pass stage is pumped by the same Nd:YAG laser. After exiting, the beam is spatially filtered and enlarged to about 15 mm in diameter. The following 5-pass Ti:sapph stage amplifies the pulses to their final energy. The 20 mm crystal is pumped from both sides by two q-switched frequency-doubled Spectra-Physics Pro Series Nd:YAG lasers. Via changing the timing of those pump lasers, the 5-pass output energy can be continuously varied from 8 mJ up to 1.2 J.

At this point, the beam can travel several different passes, depending on the experiment which should be conducted with it. One possibility is to use the uncompressed 600 ps pulse. In the more likely case that shorter pulse durations are needed, two separate grating compressors are embedded in the system. The smaller one, a two-grating-compressor, is referred to as "Air Pulse Compressor" (APC) since it is not placed in a vacuum chamber. Prior to entering this compressor, the beam is expanded to one inch in diameter by a vacuum telescope. Pulse energies up to 100 mJ at a FWHM of 42 fs can be obtained. Through changing the grating distance an output of longer pulses at higher energies is feasible. While used as a probe pulse compressor, sending a fraction of the total pulse energy in, the Air Pulse Compressor allows an independent choice of the probe pulse duration with respect to the main pulse. Furthermore, with only one mirror on a magnetic base, it can be utilized to switch quickly between two separate experiments, one using the Air Pulse Compressor, the other one the bigger second compressor. Implemented in a chamber that allows vacuum levels of $10^{-5}$ Torr, this main compressor is denoted as "Vacuum Pulse Compressor" (VPC). Before accessing that stage, the beam is spatially filtered and expanded to a diameter of 70 mm in order to protect the gold coated 40 cm grating from damage. The dispersion in the VPC eliminates the chirp introduced by the stretcher almost completely, compressing the pulses down to a 1.4 times transformation limited duration of 35 fs with a spectral width of $\Delta \nu = 12.6$ THz. Given a sech$^2$ envelope, the minimum temporal width of the pulse would thus be $\tau = 25$ fs. The difference of the obtained 35 fs to the original 20 fs oscillator output is mainly caused by two separate effects. Firstly,
gain narrowing in the Ti:sapph amplification media reduces the pulse bandwidth. Secondly, the beam propagates through various materials like glass windows, amplification crystals and air that add dispersion effects of different orders. In order to compensate for the second unintentional manipulation of pulse properties, a custom made fused silica fiber is inserted into the system after the output of the stretcher. It partially corrects aforementioned dispersion effects up to the 5th order. The final output of the THOR Vacuum Pulse Compressor is a beam of 7 cm in diameter with pulses of up to 700mJ compressed down to 35 fs at 10Hz, yielding a maximum power of 20 TW on target.

3.2 The Gas Jet

The cluster targets that were used in the experiments were generated by a gas jet. A schematic picture of such a device is depicted in figure 3.5. The valve is a General Valve Corporation Series 99 solenoid. It is connected to a high pressure backing line that supplies the respective clustering gas. An Iota One Pulse Driver that is externally triggered by a Stanford Research Systems timing box controls the opening and closing of the valve. When a pulse from the driver comes in, the poppet is lifted via the magnetic force of the solenoid. The gas that is filling the backing line is now allowed to freely stream into the vacuum chamber and expand adiabatically. Therefore, rapid cooling occurs which causes the gas to supersaturate and results in the formation of nanosize droplets composed of single atoms or molecules. As soon as the signal out of the pulse driver stops, a return spring puts the poppet back to its origin, thus closing the valve again.

For the experiments described in this thesis, opening times between 0.9 ms and 2 ms were used. Usually one wants to keep the opening times as short as possible in order to allow a high repetition rate of shots. Over a certain iteration limit, the speed of the turbo pump will be insufficient to prevent the pressure under the gas jet from gradually increasing.

The size of the clusters is strongly dependent on parameters such as nozzle shape, gas temperature and backing pressure. For all experiments conducted, a supersonic nozzle, i.e. one with an expansion half angle below 45°, was used which increases the size of the generated clusters. The orifice of this nozzle has a diameter of 750 µm and an opening angle of α = 5°. Besides that, a sonic nozzle (α > 45°) is also
available for the jet and can be easily mounted instead.

While most common gases cluster at room temperature, hydrogen for example has to be cooled prior to its entrance into the chamber. For this purpose, a close fitting copper jacket surrounds the valve. It contains a coiled copper line that can be connected to a gas supply. Temperatures down to $T = -173^\circ$ are achieved via pumping gaseous nitrogen through the jacket, that has been cooled preliminary in a separate liquid nitrogen heat exchanger. By changing the nitrogen flow rate, the temperature can be continuously varied. A thermocouple that is attached to the jet measures the respective actual temperature.

The most convenient way to control the cluster size is by changing the pressure in the backing line. Values ranging up to 1000 psi were used for the different experiments. Considering the gas jet assembly it turns out that the performance of the device depends critically on the tightness of the valve - body connection that is sealed with a copper gasket. One can determine the optimum torque by listening to the sound of the running jet while tightening it. An alternation between good and bad
shots can be observed when the position is almost right. Finding the best spot was especially an issue when the nozzle had to be cooled, i.e. for shots on hydrogen clusters. In that case, the range for a good connection is much smaller compared to room temperature. Therefore, the valve has to be tested while cooled to ensure a reliable performance.

3.3 The Cluster TOF Vacuum Chamber

The Cluster Time Of Flight (TOF) Vacuum Chamber (CTVC) (see figure 3.6) is generally composed of two parts that can be treated separately. One part is the area under the gas jet, where the cluster production by adiabatic cooling of gas streaming through the pulsed valve takes place. The second part is the TOF section, where the clusters are intercepted by the laser focus and kinetic energies of expelled ions are measured. A skimmer with an orifice of 750 µm separates those two tracts. Clusters that are produced at the gas jet have to propagate through the skimmer, hence only a small fraction reaches the interaction region forming a small collimated cluster beam.

The chamber is differentially pumped by two turbomolecular pumps, one mounted above the skimmer and the other one below. This is essential, since the pressure in the upper part jumps up to about $10^{-2}$ Torr every time the gas jet fires. At these pressures, the mean free path of the ions in the TOF tube would not be sufficiently long enough anymore, collisions would thus have an influence on the measured energy distributions of cluster constituents.

To keep track of the pressure, one gauge with an operating range from 1 mTorr to room pressure (760 Torr) is installed in each part. In the TOF section an ion gauge complements the setup. It is capable of a working range of $10^{-4}$ down to $10^{-8}$ Torr. While conducting experiments, the pressure in the TOF tube was usually on the order of high $10^{-7}$ Torr which is sufficiently low to neglect the possibility of collisions.

As mentioned in section 3.2, the shot repetition rate is dependent on the opening time of the gas jet. Besides that, one also has to account for the molecular weight of the clustering gas. Heavy molecules are pumped much more efficiently than light molecules. While the runs with argon clusters were done at an iteration of one shot every two seconds, the down time for the hydrogen experiment was twelve seconds,
Fig. 3.6. Picture of the Cluster TOF Vacuum Chamber. Two turbopumps maintain a differential pressure in the two sections. This allows to keep the pressure in the field free TOF tube low enough so that collisions between the cluster explosion constituents can be neglected. After the clusters are being hit by the laser in the focus, high energy ions are detected by a MCP. Their energy distribution can be determined from their respective time of flight. (Drawing by M. Hohenberger, [23])

even though a lower backing pressure was used.

The beam is focused onto the clusters by a f/1.6 LightPath GRADIUM graded index lens with a diameter of 80 mm and a focal length of 128.5 mm. Exhibiting a refractive index that is a function of radius, spherical aberration is reduced and smaller focal spot sizes are obtained. An anti-reflection coating keeps the reflectivity below 1 % in the broad wavelength range between 400nm and 800nm. The lens is mounted directly on a flange, thus additional losses from an entrance window are avoided.

The moment when the laser hits the clusters is determined by a photodiode that is
placed in front of the chamber measuring the backreflection from the lens. After the clusters are intercepted by the laser, they explode and their constituents are expelled in all directions. Some of them are detected by a Micro Channel Plate (MCP) that is located at the end of a field free drift tube of 1.14 m in length. The MCP consists basically of an array of electron multiplier channels, capable of measuring single ion impacts (a detailed description can be found in section 3.4). The active area of the MCP used has a diameter of 18 mm, corresponding to $1.96 \times 10^{-4}$ sr of a sphere centered at the laser focus. An oscilloscope that is connected to the MCP is recording the detected events. By simultaneously observing the photodiode signal which sets the temporal zero, the flight time of the particles can be obtained.

Besides experiments related to clusters, the chamber can also be used to do mass spectroscopy of the gas inside the TOF. Therefore, the laser beam is used to ionize the gas filling the chamber. The created charged particles are accelerated by an external field generated via a high voltage gradient on two grids placed symmetrically on each side of the focus ($U_{acc} \approx 10 kV$). Hence, the kinetic energy gained by the ions of mass $m$ and charge $q$ in that field can be described by

$$E_{kin} = \frac{1}{2}mv^2 = \frac{1}{2}qU_{acc}$$  \hspace{1cm} (3.21)

By measuring the time of flight of the ions, their velocity and thus their characteristic charge $q/m$ can be determined.

### 3.4 The Micro Channel Plate

A MCP is closely related to an electron multiplier. The one that was used for all conducted experiments is composed of two plates, each consisting of an array of miniature electron multiplier channels. A schematic drawing of the configuration of such a device is depicted in figure 3.7. The channels are parallel orientated, exhibiting a small angle with respect to the surface normal. In the case of the particular MCP utilized, the channels have a diameter of 10$\mu$m with a channel spacing of 12.5$\mu$m. Each plate has a diameter of 18mm at a thickness of 0.04 - 0.0045mm. The two plates are rotated against each other by 180°, yielding a combined v-shape of the tunnels.
In order to build up a potential gradient along the plate surface normal, a high negative voltage $V_{MCP}$ is applied to the input side of the front plate with respect to ground. A voltage divider gives equal potential differences $V_{plate}$ on each plate. The space between the two plates remains field free.

In case of the actual MCP model, the circuit looks as shown figure 3.7, with values of $R_1 = 2.5M\Omega$ and $R_2 = 500k\Omega$. The maximum voltage allowed is $V_{MCP} = -2.2kV$, corresponding to $V_{plate} = -1kV$ on each plate.

Due to the angle of the channels with respect to the plate surface normal, an energetic particle or photon that is entering the small channel orifice is likely to hit the channel wall. Subsequently to the impact secondary electrons will be emitted from the channel surface if the energy of the incident particle or photon is sufficient. These electrons are accelerated by the potential gradient, thus being capable of generating another generation of secondary electrons when they are striking the inner wall again. Hence, a cascade of secondary electrons is produced that leads to...
a huge magnification in signal strength. Once the electrons have reached the back side of the front plate, they fly through the field free region between the plates, hit a channel on the entrance of the second plate and start a second cascade which leads to a further amplification of the signal. At the output, a narrow signal peak with a width of a few nanoseconds can be observed. The fact that the plates are rotated against each other by 180° guarantees that any particle that enters a channel will not be able to propagate through both plates without hitting the inner channel wall at all.

Nevertheless, a MCP will not detect all particles/photons arriving, even if their energy is sufficient for the generation of secondary electrons. In case the impact does not happen within a channel but somewhere on the front surface of the first plate, the particle will not be recognized.

Furthermore, the height of the signal peaks is not necessarily linear dependent on the number of particles that are detected simultaneously. One reason for this is, that incident particles of higher energy will be able to extract a higher number of secondary electrons on the first impact, therefore yielding a higher output signal. In addition, the position where the particle initially hits the channel wall will have an effect. In case this impact happens very close to the input-side plate surface, the number of secondary electrons generated will be maximized, resulting in a maximized signal height for a single particle at a certain energy. If however the particle travels some distance before striking the wall first, less amplification of the signal will take place.

3.5 2nd Order Autocorrelation

In the 1960’s it was realized that the only way to measure the shortest pulses in time was by utilizing themselves as a probe. At that time the concept of the 2nd order autocorrelator was developed.

The principle setup of this technique is shown in figure 3.8. The input pulse is split into two separate pulses by a 50 percent beam splitter. One of them propagates through a delay stage which is introducing a variable delay \( \tau \) with respect to the other pulse. Afterwards, the pulses are overlapped with an angle on a nonlinear Second Harmonic Generation (SHG) crystal, e.g. made up of Potassium Dihydrogen Phosphate (KDP). The optical axis of this uniaxial birefringent crystal is orientated
in such a way so that the two incoming ordinary beams generate a frequency doubled extraordinary beam propagating along the bisecting line of the primary beams. This nonlinear process is referred to as noncollinear type I phase-matching. The electric field $E_{SHG}$ of this second harmonic pulse is proportional to the product of the electric field of the two incident pulses

$$E_{SHG}(t, \tau) \propto E(t)E(t-\tau) \quad (3.22)$$

where $\tau$ denotes their temporal delay. The intensity $I_{SHG}$ is thus given by

$$I_{SHG}(t, \tau) \propto I(t)I(t-\tau) \quad (3.23)$$

The CCD camera is way too slow to measure this signal, it will therefore integrate over it, yielding

$$AC^{(2)}(\tau) = \int_{-\infty}^{\infty} I(t)I(t-\tau)dt \quad (3.24)$$

Only parts of the input energy on the SHG crystal are actually converted to frequency doubled light. The remaining fundamental pulses are spatially separated.
Fig. 3.9. Shown is an exemplary 2nd order autocorrelation image. The x-axis is directly related to the delay $\tau$ between the two pulses incident on the SHG crystal. In the analysis each line is fitted separately by assuming a gaussian (or sech$^2$) pulse envelope shape and thus a mean FWHM of the convoluted original pulse is obtained.

from the second harmonic pulse and can therefore be blocked by an iris. That way the autocorrelation is nearly background free.

A typical 2nd order autocorrelation image is shown in figure 3.9. In order to calibrate this image, the delay stage is moved which causes the incoming pulses to be coincident in time on a different position on the crystal. Obviously, this also causes the autocorrelation signal to move on the CCD chip. For the calibration that spatial displacement is simply equated to the change in delay. In the further analysis, each line is fitted by a gaussian (or convoluted sech$^2$) and thus a mean value of the FWHM of $AC^{(2)}(\tau)$ can be obtained. The FWHM of the intensity $I(t)$ of the input pulse is gained by multiplying with the gaussian (or sech$^2$) deconvolution factor that calculate to

\[
df_{\text{gaussian}} = \frac{1}{\sqrt{2}} \approx 0.7071 \quad (3.25)
\]

\[
df_{\text{sech}^2} \approx 0.6482 \quad (3.26)
\]

It is important to notice that the 2nd order autocorrelation is not capable of measuring $I(t)$ due to the ambiguity of the convoluted signal $AC^{(2)}(\tau)$. Thus, an envelope shape (such as gaussian or sech$^2$) must be assumed to retrieve the FWHM from the recorded image. More sophisticated techniques like Frequency Resolved Optical Gating (FROG) do not require such an assumption.
3.6 Characterization of the Focal Spot

Besides the knowledge of the pulse duration, detailed information on the focal spot characteristics is necessary to interpret experimental results reliably. In theory, the radius \( w(z) \) of a gaussian beam with waist radius \( w_0 \) propagating in z-direction evolves according to

\[
w(z) = w_0 \sqrt{1 + \left( \frac{z}{z_R} \right)}
\]  

(3.27)

where \( z_R \) denotes the Raleigh Range that is given by

\[
z_R = \frac{\pi w_0^2}{\lambda}
\]  

(3.28)

for a beam of wavelength \( \lambda \). The Raleigh Range stands for the distance that the beam has to travel from its waist before the beam diameter increases by a factor of \( \sqrt{2} \).

Inserting equation 3.28 into 3.27 yields

\[
w(z) = w_0 \sqrt{1 + \frac{z^2 \lambda^2}{\pi^2 w_0^4}}
\]  

(3.29)

In order to calculate the beam radius at the focus, \( w(z) \) is equated to the beam radius at the focusing optic and \( z \) is equated to the focal length.

For a wavelength of \( \lambda = 800\text{nm} \), a focal length of the chamber lens of \( z = 128.5\text{mm} \) and a beam radius at the lens of \( w(z) = 30\text{mm} \) the focal spot radius calculates to

\[
(w_0)_{\text{theo}} = 1.09\mu\text{m}
\]  

(3.30)

This theoretical spot size is often referred to as *diffraction limited*, giving the minimum waist radius for a perfectly gaussian beam under these conditions.

The actual focal spot radius has to be measured however. To do this, the beam was attenuated by a zero degree mirror and several Neutral Density (ND) filters. Subsequently, the chamber was opened on the rear and a rail mounted on a xyz-stage was inserted. A Newport Corporation 20x microscope objective attached to the rail imaged the focal spot onto a CCD camera.
Fig. 3.10. Image of the focal spot measured with a 20x microscope objective. The beam diameter at the focal lens is 60mm, the diameter of the focus was determined to be 13.6µm. This corresponds to about six times the diffraction limit.

For aforementioned conditions, the focus is depicted in figure 3.10. This image was calibrated using a Newport Corporation USAF resolution test target to determine the length and width of a pixel. The area of the focal spot was obtained by calculating the number of pixels above a threshold of $1/e^2$ of the highest pixel value. Hence a focal spot area of $A = 145\mu m^2$ is received, corresponding to a radius of a circular focal spot of about six times the diffraction limit

$$w_0^{exp} = 6.8\mu m$$
$$\approx 6 \times w_0^{theo}$$

(3.31)

The peak intensity of a gaussian focus is given by

$$I_{peak} = \frac{2E}{\tau A}$$

(3.32)

For an energy of $E=35$ mJ and a pulse duration of $\tau = 38$ fs as used in the hydrogen shortpulse experiment (see chapter 5) it calculates to

$$(I_{peak})_{hydrogen, short} = 1.27 \times 10^{18} \frac{W}{cm^2}$$

(3.33)
Fig. 3.11. The plot is showing a closeup view of the early stages of a MCP TOF trace. Both the photon and the electron peak are revealed in the burnt orange curve, while the black trace that was taken with a 4.6 keV retarding potential in front of the MCP does not include the second peak. Therefore, the latter can indeed be attributed to electrons (adapted from [24]).

3.7 Data Acquisition and Analysis

For data collection, both the MCP and the photodiode were connected to an oscilloscope. The photodiode that is looking at the backreflection from the chamber lens, thus setting the time were the laser hits the clusters, is used as triggering signal. A PC saves both the MCP and the photodiode trace for every shot taken. Typically a MCP cluster signal is composed of four distinguishable parts. Photons, that are mainly generated through scattering are detected almost instantly (at \( t \approx 7\text{ns} \)) with the laser-cluster interaction. Shortly after this a small peak is noticeable which is due to electrons that were produced in the ionization process of the cluster. Since electrons have to overcome the potential barrier of the MCP front side in order to get detected, this peak is barely visible compared to the following ion signal. A closeup view of the early stages of a MCP trace is shown in figure 3.11, revealing both the photon and the electron peak. Subsequently to the electron peak, ions arrive at the MCP. Events from these can
be observed over a range of several microseconds. The exact width of the ion peak depends strongly on experimental parameters such as backing pressure, intensity in the focus and the type of clustered gas investigated.

In the last phase of the signal trace, the low energy ion tail gets superimposed by spikes resulting from expanding background plasma. This effect is usually noticeable on large observation periods in extent of 10µs.

For one data point, i.e. one TOF trace at identical experimental conditions, 150 to 300 shots were taken to get good statistics. By looking for the rising edge in the photodiode signal the origin of the time axis was determined. Each shot was reduced from its constant offset and inverted for better readability so that the MCP current yields positive values.

In order to extract an energy spectrum out of such a series of raw TOF traces, one has to take a closer look at the signal characteristics. Since all TOF experiments have been done using argon or hydrogen as the clustering material, attributes of these two gases are compared in the following.

### 3.7.1 Argon

A typical raw argon cluster signal is shown in figure 3.12(a). This measurement was taken at room temperature ($T_0 = 293\, K$) at a gas jet backing pressure of $p_0 = 34.5\, \text{bar}$. The MCP voltage has been set to the maximum of $V_{MCP} = -2.2\, kV$.

![Fig. 3.12. Argon cluster trace taken at room temperature at a jet backing pressure of $p_0 = 34.5\, \text{bar}$. The closeup view reveals that single peaks are clearly discernable allowing a peak count algorithm technique in the data analysis.](image-url)
corresponding to $V_{\text{plate}} = -1 \text{kV}$ on each plate.

A zoom into the plot (see figure 3.12(b)) reveals that single spikes are clearly discernable. Each of these peaks can be related to the current resulting from one ion hitting an inner wall of a MCP channel and creating a cascade of secondary electrons. Thus, a peak counting algorithm was utilized in the analysis to obtain one histogram containing all events detected by the MCP within the respective number of shots for one data point. The width of one column in that histogram was set to $\delta t = 50 \text{ns}$. Subsequently, this histogram was normalized to gain the ion distribution function $f(t)$ in time. Converting $f(t)$ into an energy distribution function $f(E)$ is done by evaluating

$$f(E) = f(t) \left| \frac{dE}{dt} \right|^{-1}$$

(3.34)

With the simple equation $E = 1/2m(s/t)^2$ for the kinetic energy this transforms to

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

(3.35)

where $m$ stands for the ion mass, $s$ denotes the propagation distance from the laser focus to the MCP and $t$ represents the actual time of flight.

### 3.7.2 Hydrogen

In comparison to argon, a characteristic TOF spectrum for exploding hydrogen clusters as shown in figure 3.13(a) and 3.13(b) exhibits a different structure. This shot was taken at a temperature of $T_0 = 81 \text{K}$ and a backing pressure of $p_0 = 17.2 \text{bar}$ with $V_{\text{MCP}} = -1.66 \text{kV}$ and $V_{\text{plate}} = -755 \text{V}$. While in case of argon single spikes were unambiguously specifiable, the hydrogen cluster trace features a much higher density of peaks that are mostly superimposing each other. Obviously, the aforementioned peak count algorithm is therefore not applicable to investigate the hydrogen data and a different approach had to be found. Instead of adding one count for each peak to the respective column in the histogram, the analysis was done by integrating directly over the right timed offset reduced inverted normalized TOF traces to convert them to energy spectra by means of equation 3.35.

As mentioned in section 3.4, there are some objections about doing this since the
Fig. 3.13. Hydrogen cluster trace taken at a temperature of $T_0 = 81$ K and a jet backing pressure of $p_0 = 34.5$ bar. The closeup view reveals that single peaks are not unambiguously specifiable. Thus, a peak count algorithm technique can not be utilized in the data analysis.

area under a peak might not be proportional to the number of impacts happening simultaneously. However, the total area under an argon TOF trace was found to increase linearly with the number of peaks counted [23]. This suggests that integrating the hydrogen TOF signal to extract energy spectra is a valid technique. To minimize the effect that faster ions are creating slightly higher peaks, the MCP voltage was set to $V_{MCP} = -1.66 \text{kV}$. 
In order to interpret experimental results reliably, a knowledge of the cluster size is crucial since all theories depend strongly on this parameter. This chapter will provide a description of two different techniques to obtain a mean cluster size for the respective conditions used in an actual experiment. A complete all optical characterization of the gas jet has been done for argon, xenon and hydrogen. The results of these measurements will be presented in the following.

4.1 The Hagena Parameter

The strength of the clustering process depends heavily on the experimental conditions, such as gas jet backing pressure, temperature, nozzle shape and gas species used. An empirical formula describing the onset of clustering and the size of the clusters generated was found by Hagena and Obert [19]. This scaling parameter $\Gamma$, that is referred to as Hagena parameter is given by

$$
\Gamma = k \frac{(d/\tan \alpha)^{0.85} p_0}{T_0^{2.29}}
$$

(4.1)

where $d$ is the orifice diameter in $mm$, $\alpha$ the expansion half angle of the nozzle, $p_0$ the jet backing pressure in $mbar$, $T_0$ the initial gas temperature in $K$ and $k$ a material constant that is related to bond formation. Table 4.1 gives some examples
Cluster production can be significantly enhanced by utilizing a supersonic nozzle (i.e. $\alpha < 45$). Several experiments reveal that the onset of cluster formation occurs at a Hagena parameter of roughly $\Gamma \approx 100$. The number of particles per cluster increases as

$$N_c \propto \Gamma^{2.0-2.5}$$  \hspace{1cm} (4.2)

where the exponent depends on the specific jet parameters. Due to the fact that the cluster radius $R$ scales with the number of particles $N_c$ as $R \propto N_c^{1/3}$, one can directly derive the dependence of $R$ to temperature (at constant backing pressure) and vice versa

$$R \propto T_0^{-(1.53-1.91)}$$  \hspace{1cm} (4.3)

$$R \propto p_0^{0.66-0.83}$$  \hspace{1cm} (4.4)

Thus, the number of particles per cluster $N_c$ and the cluster radius $R$ can be determined via a Rayleigh scattering technique. The scattering signal $S_{RS}$ is hereby proportional to the product of monomer density $n_0$ and $N_c$. Knowing that the monomer density before clustering is exactly proportional to the backing pressure $p_0$, one gets

$$S_{RS} \propto p_0 N_c$$  \hspace{1cm} (4.5)

With equations 4.1 and 4.2 the dependence of the Rayleigh scatter signal $S_{RS}$ on the backing pressure $p_0$ is obtained [5]

$$S_{RS} \propto p_0^{3-3.5}$$  \hspace{1cm} (4.6)

<table>
<thead>
<tr>
<th>Gas</th>
<th>$k$</th>
<th>$H_2$</th>
<th>$D_2$</th>
<th>$N_2$</th>
<th>$O_2$</th>
<th>$CO_2$</th>
<th>$CH_4$</th>
<th>$He$</th>
<th>$Ne$</th>
<th>$Ar$</th>
<th>$Kr$</th>
<th>$Xe$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k$</td>
<td>184</td>
<td>181</td>
<td>528</td>
<td>1400</td>
<td>3660</td>
<td>2360</td>
<td>3.85</td>
<td>185</td>
<td>1650</td>
<td>2890</td>
<td>5500</td>
<td></td>
</tr>
</tbody>
</table>
Hence, by measuring the Rayleigh scattered intensity with respect to $p_0$, one will receive the exact exponent in above equations to calculate the cluster radius. However, it turns out to be difficult to unambiguously determine the onset of clustering from the recorded data. Together with the quite rough number of $\Gamma \approx 100$ attributed to the onset of clustering, this method of measuring the cluster size exhibits some drawbacks.

4.2 Combined Rayleigh Scattering and Interferometry Technique

In order to avoid aforementioned objections related to the unambiguous determination of the starting point of cluster formation a different experimental technique has to be utilized. Approaches that have been carried out include Rayleigh scattering with mass spectroscopy, extreme ultraviolet Rayleigh scattering and absorption, high-energy electron diffraction, fragmentation-corrected time-of-flight mass spectroscopy and atom scattering. The downside of most of these methods is that the experimental setup required is challenging and may not be easily compatible with the main cluster experiment conducted. In addition, it has to be noted that some techniques involve a fragmentation of the cluster due to ionization processes. In the subsequent analysis, this fact has to be carefully considered resulting in some complications one might like to avoid.

A possible all-optical method that combines Rayleigh Scattering and Interferometry [20] will be presented in the following, including jet characterization data taken for Argon, Xenon and Hydrogen. This technique allows both the determination of the cluster radius vs. pressure and the cluster number density vs. pressure.

The energy $\Delta E_{\text{lens}}$ Rayleigh scattered by the cluster beam into a collecting lens for a laser that is intersecting with the clusters on a distance from $x$ to $x + \Delta x$ can be written as [20]

$$\Delta E_{\text{lens}}(x) \approx E_{\text{in}}\bar{\sigma}_{\text{lens}}(x)n_c(x)\Delta x$$

(4.7)

Here, $E_{\text{in}}$ denotes the laser energy incident on the scattering volume, $\bar{\sigma}_{\text{lens}}$ stands for the cluster size distribution-averaged cross section for Rayleigh scattering into the collecting lens and $n_c$ is the average number density of clusters.
The detected energy $\Delta E_{lens}$ is generated by several independent individual scattering processes with randomly distributed clusters over the scattering volume, therefore representing an incoherent sum. The cross section for $90^\circ$ scattering and $kR \ll 1$ is given by

$$\sigma_{lens} = \int_{lens} \frac{d\sigma}{d\Omega} d\Omega = \pi k^4 |\gamma|^2 (\alpha^2 - \alpha^4/4) \quad (4.8)$$

with

$$\gamma = R^3 \frac{\epsilon - 1}{\epsilon + 2} \quad (4.9)$$

where $k$ is the wave number, $R$ is the cluster radius, $\gamma$ is the cluster polarizability under the assumption of a spherical shape and $\alpha = \tan^{-1}(r_0/h) \approx r_0/h$ is the scattering collection half-angle of the lens with radius $r_0$ placed at a distance $h$ to the scattering volume.

Combining equations 4.7 and 4.8 yields

$$\overline{R^n n_c} = \frac{1}{\pi k^4} \left| \frac{\epsilon + 2}{\epsilon - 1} \right|^2 \frac{\Delta E_{lens}}{\Delta x} \frac{1}{E_{in} \Delta x} (\alpha^2 - \alpha^4/4) \quad (4.10)$$

where $\epsilon$ refers to the dielectric function associated with the cluster bulk material and the bar indicates an average over the cluster size distribution. It has to be noted, that a possible contribution to the signal from monomers in the cluster beam has not been taken into account. This is a reasonable assumption since the scattered intensity is negligible.

A second independent equation incorporating $R$ and $n_c$ that is needed in order to determine the cluster radius and number density is obtained via interferometry. The fringe shift caused by the clustered gas is employed to measure the real radial refractive index shift

$$\Delta n_r(x) = n_r(x) - 1 = 2\pi n_c \gamma_r + \delta n_m \quad (4.11)$$

where $\gamma_r = \text{Re}(\gamma)$ and $\delta n_m = 2\pi n_m \gamma_m$ denotes an additional index contribution by monomers of density $n_m$ and polarizability $\gamma_m$. For simplicity, $\delta n_m = 0$ shall be
assumed, resulting in following equation

\[
\bar{R^3} n_c = \frac{\Delta n_r(x)}{2\pi} \left( \frac{\epsilon + 2}{\epsilon - 1} \right) \tag{4.12}
\]

An effective radius \( R_{\text{eff}} \equiv \left[ \bar{R^6}(x)/\bar{R^3}(x) \right]^{1/3} \) as well as an average number density \( n_c \) of the cluster ensemble can be gained by combining equations 4.10 and 4.12. Figure 4.1 and 4.2 show the principal experimental setup for 90° Rayleigh scattering. The cluster jet utilized was identical to the one described in section 3.2 that was used for the experiments presented in the following two chapters. Both the Rayleigh scatter and the interferometry measurements were conducted in a different chamber than pictured in section 3.3, allowing easier beam access to the gas jet at a short
Fig. 4.2. Top view of the setup used for the Rayleigh scattering and interferometry measurements. In the case of Rayleigh scattering, a lens was used to focus the beam interacting with the clusters while the other one was blocked. At an angle of $90^\circ$ and a distance of $h = 260 \text{ mm}$ from the focus the energy scattered into a half angle of $\alpha = 0.097$ was collected and imaged onto a CCD-camera. On the top of the exemplary interferometry image one can identify the nozzle silhouette.

distance below the nozzle. Thus, the Rayleigh scattered intensity as well as the interferometrical determined phase shift was increased. For all measurements the solenoid was operated at $1 \text{ Hz}$ with a valve opening time of $3 \text{ ms}$.

4.2.1 Rayleigh Scattering Measurements

One of the frequency doubled Nd:YAG pump lasers of the 5-pass was synchronized with the gas jet opening time and focused into the cluster beam at a distance of $3 \text{ mm}$ below the nozzle by a lens of diameter $d = 50.8 \text{ mm}$ and focal length $f = 250 \text{ mm}$. 
Pulses yielding energies of 0.84 mJ were stored in the 532 nm beam that was expanded in size to a diameter of approximately 25 mm prior to focussing. The Rayleigh scattered energy was collected at 90° and a distance of $h = 260$ mm to the focus by a lens of diameter $2r_0 = 50.8$ mm. Hence, the associated scattering half angle was $\alpha = \tan^{-1}(r_0/h) = 0.097$. At the imaging plane of the collecting lens, a CCD-camera connected to a PC recorded the data.

A typical sample image is shown in the inset of figure 4.2. For each backing pressure 5 Rayleigh scattering images were taken and averaged after each was reduced by a background shot that was recorded with the identical setup but without firing the gas jet. Figure 4.3 shows line outs through the center of the averaged scatter signal for argon, xenon and hydrogen with respect to backing pressure. The respective collected Rayleigh scattered energy was obtained by integrating over all pixel values of the averaged background reduced images and is depicted on the right hand side of figure 4.3. Each plot includes a least-squares fit giving a scattering yield dependence on backing pressure $p_0$ for the respective gas of

- Argon: $S_{RS} \propto p_0^{2.2} (N_c \propto p_0^{1.2})$
- Xenon: $S_{RS} \propto p_0^{3.4} (N_c \propto p_0^{2.4})$
- Hydrogen: $S_{RS} \propto p_0^{3.0} (N_c \propto p_0^{2.0})$

While both the xenon and hydrogen relations agree well with the theory of Hagena presented in section 4.1 the exponent is significantly lower than the predicted range of $N_c \propto p_0^{-2.5}$ in the case of argon. Similar results were found in earlier experiments [20]. This indicates that the Hagena scaling law overestimates the size of argon clusters for the gas jet characterized.

A composite two-dimensional (2D) Rayleigh scattering profile taken at a distance of 3 mm below the nozzle exit orifice is shown in figure 4.4. It was obtained by scanning the cluster jet that was mounted on a xyz-stage transversely with respect to the probe beam. This image verifies the cylindrical symmetry of the cluster beam that will be a prerequisite for the Abel inversion in the interferometry analysis described below. Also remarkable is the very sharp boundary with vacuum.

To determine the ratio of incoming energy $E_{in}$ to the collected scattered energy $\Delta E_{lens}$ (see equation 4.10) a 45° mirror was placed below the nozzle to redirect the
Fig. 4.3. Shown on the left hand side are line outs through the center of the Rayleigh scattering images with respect to gas jet backing pressure. The data was taken at a distance of 3 mm to the nozzle exit orifice. For each gas the respective collected Rayleigh scattered energy was obtained by integrating over all pixel values of the averaged background reduced images. The results are plotted on the right hand side, including a least-squares fit revealing the dependence of the Rayleigh scattering signal $S_{RS}$ on backing pressure $p_0$. 

(a) Argon at room temperature (293 K) and a valve opening time of 3 ms

(b) Xenon at room temperature (293 K) and a valve opening time of 3 ms

(c) Hydrogen at 83 K and at a valve opening time of 0.9 ms
laser pulse onto the CCD-camera through the chamber window and the collecting lens with no clusters present. The beam was attenuated by several ND filters that were calibrated at 532 nm in advance.

**Time variation of the cluster production**

In order to examine the build up and decrease of cluster formation, the CCD-camera was replaced by a photodiode at the imaging plane of the collecting lens and the time delay of the incident laser pulse with respect to the nozzle opening time was varied. It was found that cluster production eventually reaches steady state in the case of room temperature operation. This situation is depicted in figure 4.5(a) for argon at a backing pressure of 700 psi and a nozzle opening time of 3 ms.

In contrast, at lower temperatures the Rayleigh scattering signal exhibits a sharp peak before it slowly falls off (figure 4.5(b)). This behavior can be attributed to ohmic heating of the solenoid which is limiting the clustering process significantly in the case of gases that require cryogenic operation such as hydrogen. Thus, an optimum delay between the opening of the nozzle and the laser pulse timing exists for pre-cooled gases that has to be taken care of in actual experiments.
4.2.2 Interferometry

As discussed in earlier in this section a second independent measurement is needed to determine the cluster radius since the Rayleigh scattering yield also depends on the cluster density. An interferometry technique can be employed to obtain the refractive index shift due to the clustered gas. The setup utilized is a Mach-Zehnder interferometer that is schematically depicted in figure 4.2. The lens used to focus the beam on the clusters for Rayleigh scattering was removed. Due to the phase shift $\Delta \Phi(y)$ the probe acquires while propagating through the cluster beam a shift in the horizontally orientated fringes is noticeable.

In order to obtain the refractive index shift, fringe patterns were recorded with a CCD-camera, both in the case of gas being present and not. These raw interferograms were Fourier transformed and the resulting frequency spectrum was reduced by everything but the fringe frequency. Shifting the signal to the origin in frequency space and subsequently employing a Fourier back transformation results in a two-dimensional phase map that exhibits discontinuities (i.e. jumps of $\Delta \phi$ by $2\pi$) at the position of the original fringes. By subtracting the $z$-coordinates of the unshifted discontinuity from the position of the shifted discontinuity for each fringe, dividing the respective distance by the fringe spacing and multiplying it by $2\pi$, the phase shift $\Delta \phi(y, z)$ in two dimensions can be extracted.

A sample of $\Delta \phi(y, z)$ is shown in figure 4.6.
The radial refractive index shift $\Delta n_r(x)$ was obtained from $\Delta \Phi(x)$ through the process of Abel inversion which is valid due to the cylindrical symmetry of the cluster beam.

### 4.2.3 Resulting Cluster Radii and Densities

In addition to the $\Delta E_{\text{lens}}(x)/E_{\text{in}}$ and $\Delta n_r(x)$ gained from above mentioned experiments, values of the dielectric constant for solid argon, xenon and hydrogen at $532 \text{ nm}$ are needed. The numbers used are $\epsilon = 1.5053$ for argon ([21]), $\epsilon = 2.1966$ for xenon ([21]) and $\epsilon = 7.29$ in the case of hydrogen ([22]).

With above specified parameters and the described Rayleigh scattering and interferometry measurements an effective radius $R_{\text{eff}} \equiv \left[\frac{R^6(x)}{R^4(x)}\right]^{1/3}$ as well as an average number density $n_c$ of the cluster ensemble can be gained by combining equations 4.10 and 4.12. The results for argon, xenon and hydrogen are presented in figure 4.7.

It has to be noted that the cluster size distribution was not taken into account in this experiment. However, for a nonpathological cluster size distribution (e.g. Gaussian
Fig. 4.7. Shown on the left hand side is the effective cluster radius $R_{\text{eff}}$ with respect to gas jet backing pressure. On the right hand side the obtained cluster density at a distance of 3 mm below the nozzle exit orifice is depicted.

In the case of argon and xenon $R_{\text{eff}}$ shows a nearly linear increase with pressure while the density remains approximately constant (argon) or decreases slightly (xenon). In contrast, hydrogen exhibits a different behavior. The cluster density peaks around 400 psi before falling off to a low density tail at high pressures. At the same time the radius increases faster than linear.
or log normal) the effective radius $R_{\text{eff}}$ will be a good representation of the mean cluster radius associated with a certain distribution \[20\].

Besides the cluster population there will always be a non-clustered fraction of the gas volume present. In general, this will not affect the amount of Rayleigh scattered energy collected but it will contribute to the interferometric phase shift. Since the monomer density is not taken into account in the analysis, the measured cluster radius $R_{\text{eff}}$ will give an underestimate of the real radius. The difference in radius is given by the factor $(1 - \delta_m)^{-1/3}$, where $\delta_m = n_m / (n_m + n_c \bar{A})$ is the relative concentration of monomers with $\bar{A}$ being the mean atom number. Due to the factor’s cube root dependence the effect of the uncertainty in $\delta_m$ on $R_{\text{eff}}$ is greatly mitigated.
Chapter 5

Studies of Anisotropy in the Explosion of Hydrogen Clusters

In previous experiments, anisotropy in the energy distribution of particles accelerated in the explosion of nanoscale clusters has been observed. These measurements were done by irradiating Ar and N\textsubscript{2} clusters ([25], [26], and [27]) whose expansion is assumed to be dominated by Coulomb forces and large Xe clusters that are expected to behave hydrodynamically ([28], [29] and [30]). An interpretation was found through microscopic particle in cell (MPIC) simulations that were conducted for the explosion of large ($N_c > 10000$) clusters of multi-electron atoms [31]. In this picture, the electric field inside the cluster is enhanced by charge separation due to the laser field pulling the electron cloud out of the cluster on one side thus depleting the opposite pole. Consequently, higher charge states are generated which attain superior kinetic energies in the cluster explosion. The ion spectrum will hence be shifted towards higher energies when measured in the laser field direction.

Anisotropic effects in explosions of laser irradiated H\textsubscript{2} clusters have not been observed yet. In previous experiments, Sakabe et al. [32] irradiated small H\textsubscript{2} clusters of $R_{c} \approx 2.5 \text{ nm}$ at $6 \cdot 10^{16} \text{ W/cm}^2$ and observed proton spectra in good agreement with the model of a fully stripped cluster undergoing pure Coulomb explosion. Measurements on large H\textsubscript{2} clusters at the same intensity were done by Mendham et al. [33] leading to the conclusion that hydrodynamic pressure has been the predominant expansion mechanism in that situation. Anisotropy of $\approx 15\%$ was found in deuteron energies from a dense D\textsubscript{2} cluster medium that was irradiated with a peak intensity.
of $2 \cdot 10^{20} \text{W/cm}^2$ [35]. This effect could be partly attributed to the aspect ratio of the laser focus that leads to asymmetric expansion on the macroscopic scale of a plasma filament which forms at high density [16]. However, simulations conducted when the cluster vertical ionization (CVI) approximation is violated (i.e., in case that ion motion during the presence of the laser pulse has to be taken into account) are consistent with this degree of anisotropy [36].

5.1 Experimental Setup

All measurements were conducted on the TOF vacuum chamber depicted in section 3.3. The cluster source used is described in section 3.2. A $\lambda/2$ waveplate of 50.8 mm in diameter mounted in front of the chamber GRADIUM lens was employed to rotate the linear polarization direction of the incoming laser beam centered at 800 nm in wavelength. By recording the reflection from the chamber lens using a photodiode, the time when the laser pulse hits the cluster was determined. Energies of fast ions accelerated through the cluster explosion were detected by a MCP (see section 3.4) measuring the time of flight in a field free drift tube of 1.14 m in length.

In the experiment, the gas jet was operated at a backing pressure of 250 psi and cooled down to $T_0 = 100 \text{K}$ by a nitrogen flow through the copper cooling jacket surrounding the jet thus allowing the production of hydrogen clusters. The nitrogen gas was precooled while pumped through a coiled copper line submerged in a dewar filled with liquid nitrogen. An estimate of the hydrogen cluster size can be obtained from the Hagena scaling law $\Gamma = k \left( \frac{d}{\tan \alpha} \right)^{0.85} p_0^{0.29}$, where $d = 750 \mu\text{m}$ is the jet orifice diameter, $\alpha = 5^\circ$ is the nozzle expansion half angle and $k = 184$ for $H_2$ bonding. This gives a cluster radius of $R \approx 2 \text{nm}$.

In the experiment, two configurations were used to irradiate the clusters, one at a pulse energy of 36 $mJ$ and full width half maximum (FWHM) duration 40 $f s$, the other at a pulse energy of 71 $mJ$ and duration 250 $f s$. The respective pulse durations were measured using the 2nd order autocorrelation setup described in section 3.5. A focal spot radius of 6.8 $\mu\text{m}$ was determined for the focussing lens (see section 3.6), corresponding to a peak intensity of $\approx 1 \cdot 10^{18} \text{W/cm}^2$. 

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5.2 Measured TOF Traces and Associated Energy Spectra

Figure 5.1 and 5.2 show the time of flight traces obtained for the short pulse and the long pulse run. Each curve represents an average over 150 shots that were offset reduced, inverted and shifted to the correct origin in time which was recorded for each shot by aforementioned photodiode looking at the reflection from the focusing lens. Shots for polarization angles from 0° to 90° were done in steps of 10°, where 0° refers to horizontally polarized light (i.e. in the direction of the MCP) and 90° is attributed to vertically polarized light (i.e. perpendicular to the direction of observation). The curves for different polarization angles were hereby recorded in an arbitrary pattern, thus eliminating the possibility of observing a trend in the data that is just due to a continuous drift in experimental conditions.

Both in the short pulse and the long pulse case a continuous increase of the mean and minimum ion time of flight is clearly noticeable while going from 0° to 90°, referring to a steady decrease in mean and maximum ion energies. For comparison, the traces of 0° and 90° polarization are combined to one plot in figure 5.3. Besides the clear anisotropy in each case, irradiation with the long pulse causes the ion signal to shift to shorter times of flight associated with higher kinetic energies. This effect becomes obvious in the measured ion energy spectra depicted in figure 5.4. Both mean and maximum ion energies are enhanced for a polarization angle of 0°. With the 40 fs pulse an average energy of $\epsilon_{av} \approx 1\, keV$ and a maximum energy of $\epsilon_{max} \approx 9.5\, keV$ is observed at 0° while at 90° $\epsilon_{av} \approx 0.6\, keV$ and $\epsilon_{max} \approx 8\, keV$. In the case of 250 fs pulse duration $\epsilon_{av} \approx 2\, keV$ and $\epsilon_{max} \approx 13\, keV$ at 0° and $\epsilon_{av} \approx 1.6\, keV$ and $\epsilon_{max} \approx 11.5\, keV$ at 90°.

Immediately striking is the large increase in mean as well as maximum energy resulting from the use of the longer 250 fs pulse. The mean energies as a function of polarization angle are shown for both pulse durations in figure 5.5. In each case the energies are enhanced with decreasing angle of polarization, reaching a maximum at 0°. The observed degree of anisotropy $[\epsilon_{max}(0°) - \epsilon_{max}(90°)]/\epsilon_{max}(0°)$ was $\approx 40\%$ for the 40 fs pulse and $\approx 25\%$ for the pulse duration being 250 fs. Since the detection plane is perpendicular to the propagation direction of the laser, the aspect ratio of the laser focus has no bearing on the anisotropy measurement. Even the use of the 40 fs pulse exhibits anisotropy, thus indicating that the associated
Fig. 5.1. TOF spectra obtained by irradiating hydrogen clusters with a short, 40 fs laser pulse center at a wavelength of 800 nm. The linear polarization of the laser was rotated from 0° to 90° where 0° refers to a detection of ions in the polarization direction. Each curve represents an average over 150 shots that were offset reduced, inverted and shifted to the correct origin in time which was recorded for each shot by a photodiode looking at the reflection from the focusing lens. A continuous increase of the mean and minimum ion time of flight is clearly noticeable while going from 0° to 90°, referring to a steady decrease in mean and maximum ion energies.
Fig. 5.2. TOF spectra obtained by irradiating hydrogen clusters with a long, 250 fs laser pulse center at a wavelength of 800 nm. The linear polarization of the laser was rotated from $0^\circ$ to $90^\circ$ where $0^\circ$ refers to a detection of ions in the polarization direction. Each curve represents an average over 150 shots that were offset reduced, inverted and shifted to the correct origin in time which was recorded for each shot by a photodiode looking at the reflection from the focusing lens. A continuous increase of the mean and minimum ion time of flight is clearly noticeable while going from $0^\circ$ to $90^\circ$, referring to a steady decrease in mean and maximum ion energies.
Fig. 5.3. Comparison of TOF spectra obtained at 0° and 90° polarization for a 40 fs pulse and a 250 fs pulse, respectively. Besides the clear anisotropy in each case, irradiation with the long pulse causes the ion signal to shift to shorter times of flight associated with higher kinetic energies.

explosions do not strictly adhere to the fully stripped CVI cluster model which predicts symmetric expansion. In addition, earlier simulations [36] of small, 3.4 nm D$_2$ clusters irradiated with pulses of FWHM from 5 – 100 fs showed a steady decrease in ion energy while increasing the pulse duration since expansion reduces the charge density driving the explosion. This is in direct contradiction to the strong increase in energy measured for the longer pulse. The observed energy enhancement can be attributed to the presence of larger clusters in the cluster size distribution which cannot be described with a purely Coulombic scenario since the extracted electrons begin to influence the expansion.

5.3 Models of the Heating and Expansion Mechanisms

Depending on the cluster radius $R$, the laser cluster interaction can be classified into three regimes. In the following, the formalism of [16] will be briefly reiterated for a uniform spherical cluster with ion density $n_0$ in a laser field of strength $E_0$ and frequency $\omega$ with the approximation that the ions are static. A bare ion cluster sphere creates an electric field that increases from zero at the center to $E_{\text{max}} = n_0|e|R/3\epsilon_0$ ($e$ is the electron charge, $\epsilon_0$ the permittivity of free space) at the cluster edge and then decreases radially outside the cluster.
If the laser field exceeds $E_{\text{max}}$ all the electrons can be extracted from the cluster. In the case of $E_0 < E_{\text{max}}$, some electrons will remain inside the cluster forming a spherical cold electron core of radius $R - d$ and density $n_0$ which will be offset from the center of the cluster in opposite direction of the laser field by an amount of $d = -3\varepsilon_0 E / n_0 |e|$ ($E$ is the electric field strength within the cluster boundary). The core responds adiabatically to the laser field, oscillating with the laser frequency $\omega$ and remaining in equilibrium.

With increasing intensity the laser extracts electrons from the cluster until a maximum core displacement of $d_0$ is reached. Given that the cluster radius $R$ is much smaller compared to the excursion length of an electron in the laser field, $\xi = |e|E_0/m_e\omega^2$ ($m_e$ is the electron mass), the extracted electrons escape from the cluster vicinity leaving a neutral core that is surrounded by a positive ion shell of thickness $d_0$.

In both of these regimes Coulomb forces are solely driving the ion acceleration and
the final energies reached are directly related to the ion initial position in the cluster:

$$\epsilon(r) = \frac{e^2 n_0 [r^3 - (R - d_0)^3]}{3e_0 r}$$

(5.1)

for \((R - d_0) < r < R\). The cluster will be fully stripped if \(d_0 = R\) and above equation will simply give the CVI energy distribution.

A third regime describes clusters of radius \(R > \xi\). In that case the electrons exhibit a two-component distribution: the cold core of electrons confined within the cluster and a cloud of electrons which have been extracted from the cluster by the laser field but cannot be totally removed from the cluster vicinity. These electrons can gain energy via a multiple-pass vacuum heating process as they sweep through the shielded core of the cluster ([37], [15], [16], [38]). An optimum of that heating mechanism should be reached if the condition \(R \approx \xi\) is fulfilled. In the case of large clusters \(R \gg \xi\) the electrons can dephase due to their long cluster transit time compared to one laser period. The electrons undergo stochastic heating (see section 2.3.4) and are able to reach energies greatly exceeding the ponderomotive energy \(U_p\) of the laser field [16].

It is obvious that anisotropy is inherent in the explosions of partially evacuated clusters. The force acting on the ions during the laser pulse is not symmetric since
the internal electron core oscillates along the laser field direction. When a cloud of extracted electrons remains bound to the cluster, further anisotropy arises due to the enhanced electron pressure along the laser field axis. A confirmation of this picture was found by PIC simulations [16] for clusters with $R \gg \xi$ and in the regime $R \approx \xi$ the ion yield is expected to show a similar angular dependence. Anisotropy that reduces with an increasing fraction of electron extraction was also shown in molecular dynamic simulations of $D_2$ cluster explosions [36].

5.4 Interpretation of the Obtained Energy Spectra

For a CVI ion energy distribution and a single cluster size

$$f(\epsilon)d\epsilon \propto \sqrt{\epsilon}d\epsilon \text{ for } \epsilon \leq \epsilon_M = n_H e^2 R^2 / 3 \epsilon_0$$

(5.2)

where $n_H$ is the initial ion density ($\approx 5 \cdot 10^{22}$ atoms/cm$^3$) and the average energy is $\frac{3}{8} \epsilon_M$. The maximum energy gainable from a cluster of radius $R$ is given by the CVI limit. The scaling with cluster radius indicates that large clusters are necessary to generate high energy ions. The cluster radius associated with the longer pulse that is generating proton energies up to $\approx 13$ keV is thus at least 7 nm. Together with the expected cluster size of $\langle R \rangle \approx 2$ nm this estimate can be used to construct a log-normal distribution of cluster sizes of the form

$$f(N) \propto e^{-\frac{\ln^2(N/N_0)}{2w^2}}$$

(5.3)

where $N_0$ is the model cluster size (number of atoms per cluster) and $w$ is proportional to the FWHM of the distribution. The energy distribution function for this ensemble of clusters is $F(\epsilon)d\epsilon \propto g(\epsilon)\sqrt{\epsilon}d\epsilon$ [35], where $g(\epsilon) = \int_{N_\epsilon}^{\infty} n_C(N)dN$ is the density of clusters larger than the size $N_\epsilon$ for which $\epsilon_M = \epsilon$. Figure 5.6 shows the energy spectrum generated with $w = 0.88$ that is including all clusters with $f(N) > 10^{-7}$. Spectra for narrower distributions of $w = 0.25$ and $w = 0.5$ and identical mean cluster radius $\langle R \rangle$ are also illustrated. The data exhibits the less sharp energy cut-off associated with a broader size distribution which is consistent with the results of Sakabe et al. [32]. Over much of the energy range, the shape of the CVI spectrum is reasonably close to that of the 40 fs data set. The offset in
absolute yield may be due to normalization, since not all energetic ions from every cluster in the distribution are detected by the MCP. The energy cut-off in the data at \( \approx 9.5 \text{ keV} \) indicates that the short pulse cannot drive CVI explosions of clusters exhibiting radii in excess of \( R_{\text{cutoff}} \approx 6 \text{ nm} \).

This behavior can be explained by considering the magnitudes of \( d_0 \) and \( \xi \) compared to \( R \). A range of intensities is contributing to the measured signal but the ions in the high energy part of the spectrum (\( > 1 \text{ keV} \)) probably originate from clusters in the high intensity regions of the focus. For the given peak intensity of \( \approx 10^{18} \text{ W/cm}^2 \) the cold electron core offset is \( d_0 \approx 10 \text{ nm} \). Therefore, all the clusters are expected to be fully stripped thus exhibiting a much weaker anisotropy than observed. This indicates that the main contributing intensity has to be somewhat lower than this. Assuming an intensity of \( 10^{17} \text{ W/cm}^2 \) one obtains \( d_0 \approx 3.2 \text{ nm} \) and \( \xi \approx 27.5 \text{ nm} \). Incidentally this results in \( R_{\text{cutoff}} \approx \frac{1}{\xi} \) which was chosen by Breizman et al. in [15] as the maximum size for which extracted electrons do not have to be accounted for in the laser cluster interaction. However, both this validity condition and the choice of intensity to be \( 10^{17} \text{ W/cm}^2 \) are rather arbitrary.

For clusters of \( R < 3.2 \text{ nm} \), the electrons are completely extracted and escape the cluster vicinity since \( R \ll \xi \). In the case of \( 3.2 \text{ nm} < R < 6 \text{ nm} \), the clusters are
only partially stripped but electrons once extracted do still not return which is why
the explosion is still driven by Coulomb forces between the ions. Since only a small
fraction of electrons are retained within the cluster ($\approx 10\%$ for $R = 6\,nm$, calculated
by assuming this percentage to be equal to the ratio $(R - d_0)^3/R^3$ of the volume
of the cold electron core to the volume of the cluster) the final ion energies remain
close to the CVI limit. The high energy ions which are accelerated from the outer
layers of the cluster should not be greatly affected by the small core of unextracted
cold electrons. Simulations of $D_2$ clusters show that the ion energy spectrum can
remain very similar to the CVI distribution even when the CVI approximation is
violated to a certain degree [36]. Nevertheless, an asymmetry is introduced in the
expansion by the motion of the electron core along the laser polarization direction
([36],[16]).

The situation changes for clusters of $R > 6\,nm$ which is when the radius becomes
comparable to the excursion length $\xi$. Now the dynamics of the interaction are cru-
cially altered by extracted the electrons that remain in a cloud around the cluster.
At this point the electrons can pick up energy through the aforementioned vacuum
heating process ([37], [15], [16], [38]). The more times the electrons transit the
cluster, the more energy they gain. An asymmetry in the ion expansion will arise
from the higher electron pressure along the laser field direction. In order to generate
energetic ions in this regime, either the electrons must leave the cluster leaving bare
ions subject to Coulomb repulsion, or the electrons must transfer their energy to
the ions in an ambipolar expansion.

In the short pulse spectrum the absence of ions more energetic than $9.5\,keV$ indi-
cates that neither of these scenarios occurs and no effective heating takes place for
the large clusters. In contrast, energies up to $13\,keV$ are generated by the longer
pulse, signifying that electron heating becomes more efficient with the increased
number of laser cycles.

Once the electron energy exceeds $U_p$, their excursion becomes much larger than $R$,
therefore allowing some electrons to completely escape the cluster. Even though the
condition $\xi > 5R$ for a removal of extracted electrons from the cluster [15] may not
hold for the initial excursion, it can become valid through vacuum heating of the
electrons. In short, the longer pulse is capable of energizing and removing parts of
the electron cloud thus initiating charge-driven explosions of clusters with $R \approx \xi$,
whereas in case of the $40\,fs$ pulse the cloud remains bound and relatively cold so
energy is not effectively transferred to the ions. It has to be noted that ion motion during the presence of the laser has to be taken into account for the longer pulse interaction since it takes place on the same timescale as the electron extraction. Due to the associated reduction in density the cluster potential is lowered and ion energies are expected to decrease. However, the measured shift of the spectrum to higher energies shows that this effect is overcompensated by the superior vacuum heating driven by the longer pulse. To finally resolve this issue, further simulation of the impact of ion motion on the process at the specific experimental conditions used would be required. Additionally, in case very large clusters of $\approx 10 \text{nm}$ are produced, they could experience a resonant plasma response to the laser field ([10],[39]) which would result in a rapid increase in the electron temperature at the point where the plasma frequency drops into resonance. For hydrogen clusters of radius $10 \text{nm}$ irradiated by a 90 $fs$ pulse at $6 \cdot 10^{16} \text{ W/cm}^2$ Mendham et al. [33] concluded that the hydrodynamic pressure is predominant in the cluster response. At the conditions used in the experiment presented in this thesis a very low abundance of such large clusters is expected; for the distribution shown in figure 5.6 it comes out to be as low as $f(10 \text{nm}) < 10^{-10}$. Hence, it is a valid assumption not to expect any significant contribution to the obtained signal.
Chapter 6

Time-Resolved Studies of the Expansion of Large Argon Clusters

In contrast to small hydrogen clusters that are either fully stripped or at least reduced by the vast majority of electrons via irradiation with a high-intensity short pulse laser, large noble gas clusters can maintain quasi-neutrality during the interaction process. Obviously, such clusters thus exhibit completely different heating and expansion mechanisms than presented in the last chapter. A pump-probe experiment was designed to study the expansion dynamics of large argon clusters. The mean energy of ions generated in the cluster explosion was found to be greatly enhanced at an optimum delay between two infrared ultrashort pulses irradiating the cluster in agreement with theoretical predictions.

6.1 Numerical Modeling and Previous Experiments

Ditmire et al. presented a model that describes the cluster as a spherical nanoplasma [10]. In this picture, collisional heating (i.e. inverse bremsstrahlung) is considered the dominant process for energy transfer from the laser field to free electrons inherent to the cluster. A detailed description of this heating mechanism can be found in section 2.3.2 within the theory chapter, the basic ideas and consequences will be shortly reviewed in the following.
The most striking result is the fact that electron collisional heating shows a resonance when the electron density drops to three times the critical density $n_{cr}$ of the cluster plasma, given by

$$n_{cr} = \frac{\pi c^2 m_e}{e^2 \lambda^2}$$

(6.1)

At this point the electric field in the cluster greatly exceeds the vacuum electric field of the laser causing a rapid increase in electron temperature. Hence, the expansion velocity of the cluster is dramatically boosted when driven at the plasma resonance. The occurrence of two distinct electron populations is also predicted by the model and has been observed experimentally [40]. At $n_e = 3n_{cr}$ hot electrons are generated that create a sharp peak in the electron energy spectrum.

According to the model, an evident approach to enhance the energy of ions generated in explosions of large, high $Z$ clusters is irradiation at the time when inverse bremsstrahlung becomes resonant. A first pulse can be employed to start the cluster expansion, followed by a correctly timed second one that heats the electrons once their density has dropped to three times the critical density hence guaranteeing a very efficient energy transfer.

In previous experiments by Zweiback et al. [41] this has been done using two infrared pulses with a duration of $50\,fs$ centered at $810\,nm$ wavelength. Absorption measurements were conducted on Argon and Xenon clusters with cluster radii in the range of $11\,nm$ to $16.5\,nm$ and $8.5\,nm$ to $20.5\,nm$ respectively. Utilizing a pump-probe setup with a weak $0.54\,mJ$ first and a strong $5.4\,mJ$ second pulse focused to intensities of $1.6 \cdot 10^{16}\,W/cm^2$ and $1.6 \cdot 10^{17}\,W/cm^2$ a distinct peak in the absorption of xenon clusters was observed while scanning the delay between pump and probe. With increasing cluster size the optimum timing was found to increase as well, indicating a lower electron density expansion velocity for bigger clusters in agreement with the nanoplasma model.

Springate et al. found an enhancement in the mean and maximum energies of ions generated in explosions of xenon clusters through irradiation with a correctly timed sequence of two high-intensity laser pulses. In contrast to Zweiback et al. the probe pulse was not at the fundamental frequency as the pump but at the laser’s second harmonic. In addition, longer pulse durations of $260\,fs$ for the first $1.9\,mJ\,(6 \cdot 10^{14}\,W/cm^2)$ pulse at $780\,nm$ wavelength and $185\,fs$ for the second $1.3\,mJ$
(2.5·10^{15} \, W/cm^2) pulse at 390 nm were used. The idea was that due to the large duration of the first pulse the laser electric field would still be present once the cluster electron density drops to three times the critical density. Thus the pump will both start the expansion and resonantly heat the electrons. Since \( n_{cr} \) is dependent on the laser wavelength according to equation 6.1, a second frequency doubled pulse can hit the resonance at a different time, causing a second peak in the electron temperature. The measurements carried out showed reasonable agreement with numerical simulations done using the nanoplasma model.

### 6.2 Setup for a Two-Color Three Pulse Pump-Probe Experiment

In order to take aforementioned previous experiments to the next level, a two-color three pulse pump-probe experiment was designed to gain more insight of the dynamics of large noble gas cluster explosions. In contrast to Springate et al. who used a long pump to start the cluster expansion and heat it resonantly at the laser fundamental, the presented approach incorporates one pump and two probe pulses. The short pump at the fundamental frequency solely initiates the cluster explosion and does not significantly influence the electron heating since the electron density has not enough time to decrease to fulfill the plasma resonance condition. Therefore, the amount of energy deposition is expected to be strongly dependent on the delay of the two short probe pulses, one at the laser fundamental and the other one at its second harmonic. Due to the wavelength dependence of the nanoplasma critical density, the frequency doubled probe should hit the expanding cluster prior to the infrared probe in order to maximize the electron temperature.

The design of the setup is depicted in figure 6.1. A nonlinear crystal of Potassium Dihydrogen Phosphate (KDP) with a diameter of 50.8 mm and a thickness of 1 mm is employed to generate second harmonic light from the incoming femtosecond infrared laser pulse centered at a wavelength of 800 nm. The conversion efficiency achievable in type I phasematching configuration is around 10 – 15%. A dichroic mirror that is highly reflective at 400 nm while transmitting 800 nm separates the frequency doubled beam from the fundamental. The infrared light is subsequently split into two pulses by a 50% beamsplitter. By two 0° mirrors mounted on linear translation stages a continuously variable delay is introduced between those pulses. Afterwards
the infrared beams are sent collinearly onto a second dichroic mirror transmissive at 800 nm wavelength where they are recombined to the same path with the blue arm of the setup.

Two protected silver mirrors are utilized to steer the conjunct output through a 80 mm graded index lens focussing the beam into the low cluster density interaction region. Due to dispersion in the 12.25 mm thick lens the 400 nm light is focussed on a significantly different position in z-direction compared to the infrared beam.

In order to compensate for this effect and overlap the foci in laser propagation di-
rection, a pair of lenses was incorporated into the blue arm to introduce a slight divergence of the 400 nm beam. Obviously the alignment is crucial for the success of this experiment and a perfect spatial overlap of the three foci has to be ensured in every direction of space throughout the duration of one run which usually takes a couple of hours. In order to guarantee that the alignment does not drift over that extended period of time a way had to be found to permanently keep track of the foci spatial position. This is done by a pair of achromatic doublets that image the xy-plane at the focal spot with an approximately 10 times magnification on a CCD camera. The first lens is hereby mounted directly onto the vacuum chamber on a custom-made flange.

To overlap the pulses in time, a drop-in metal mirror is employed to pick off the combined output of the setup after the second dichroic mirror. While blocking one of the infrared arms, this beam is guided onto a Beta-Barium Borate (BBO) crystal generating third harmonic light at 267 nm in case the fundamental and the frequency doubled pulse are coincident in time. A series of interference filters transmissive at 267 nm reduce the output of the BBO crystal by background caused from non-converted 400 nm and 800 nm light. The thus cleaned 3rd harmonic pulse is detected using a photomultiplier tube that is connected to an oscilloscope. By scanning the timing of each infrared pulse with respect to the blue all arms can be brought to zero time delay.

A major issue in this experiment is the wide range of intensities contributing to the cluster time of flight signal, making it difficult to unambiguously interpret the data obtained. In order to restrict the intensity distribution involved in the detected TOF traces, a vertically orientated slit with a width of two times the confocal parameter \( b \approx 0.5 \text{ mm} \) was mounted at the entrance of the time of flight section. With the beam radius \( w(z) \) described by

\[
w(z) = w_0 \sqrt{1 + \left( \frac{z}{z_R} \right)^2}
\]

where \( z_R = b/2 \) is the Rayleigh range and an intensity dependence on radius

\[
I(r) = \frac{2P}{\pi w^2} e^{-\frac{2r^2}{w^2}}
\]

where \( P \) is the total power stored in the beam, the relevant variation of pulse peak
intensity in the z-direction is reduced to a factor of 5 using a 1 mm slit. The slit was carefully aligned by venting the chamber with nitrogen and watching the breakdown at the focus with a CCD camera mounted collinear with the time of flight tube direction (figure 6.2).

6.3 Measured Ion Energy Enhancement from Irradiation of Argon Clusters with a Red-Red Pulse Sequence

As a first run with the setup, argon clusters generated at a backing pressure of 500 psi were irradiated by a sequence of two infrared pulses, while the blue arm was blocked. The associated average cluster radius can be obtained from figure 4.7(a) to be \( \langle R_0 \rangle \approx 9 \text{ nm} \). Both pulses of 800 nm center wavelength incorporated an identical energy of 12.6 mJ and were each focused down to a peak intensity of about \( 1 \cdot 10^{17} \text{ W/cm}^2 \). The pulse duration was measured to be \( \approx 35 \text{ fs} \).

Since the aforementioned slit reduced the number of events detected by the micro channel plate significantly, the distance between nozzle exit orifice and skimmer was reduced by 40 cm from initially 73 cm to 33 cm. Thus, the cluster density below the
skimmer was increased and more ions were generated that were able to reach the MCP. The voltage at the micro channel plate was tuned all the way up to the maximum of $-2165\,\text{V}$, corresponding to $-1000\,\text{V}$ on each plate. For cluster production the gas jet was operated at an opening time of $2\,\text{ms}$ and a repetition rate of $2\,\text{s}$.

Prior to the run, zero time delay between the two infrared pulses was found by overlapping each of them in time with the blue pulse to generate 3rd order signal as described above. Subsequently, one $800\,\text{nm}$ pulse was scanned with respect to the other on a delay range from $-1366\,\text{fs}$ to $+1366\,\text{fs}$ in steps of $130\,\text{fs}$, where $0\,\text{fs}$ refers to the point of coincidence in time. For each delay a number of 300 shots were recorded. The order in which the different delay points were measured followed an arbitrary pattern, thus eliminating the possibility of data misinterpretation due to a drift in experimental conditions. In addition, the alignment was checked every time a new data point was recorded.

The mean ion kinetic energies obtained are depicted in figure 6.3. Each point represents an average over 300 shots. As expected for the use of two identical pulses, the curve exhibits a symmetry to the zero-timing axis indicating constant experimental conditions throughout the whole run. A strong enhancement of ion energies for a
delay of \( \approx 800 \, fs \) is clearly visible. Mean kinetic energies of \( 7.3 \, keV \) are reached at the optimum timing while only \( 3.4 \, keV \) are gained at zero delay, corresponding to an enhancement of more than 100%.

Pronounced results confirm the prediction of a resonance in the cluster collisional heating process as presented by Ditmire et al. [10]. The heating rate is significantly increased once the cluster has expanded to lower electron densities compared to the situation where both pulses hit the cluster at the same time. The optimum delay of \( 800 \, fs \) can be attributed to the plasma resonance at \( n_e = 3 \, n_{cr} \).

Due to a failure of the cluster source, additional runs with a blue and an infrared pulse as well as a three pulse run could not be completed yet.
Chapter 7

Conclusion and Future Projects

In conclusion, ultrafast dynamics of cluster explosions were studied for small low Z as well as large high Z clusters that exhibit completely different heating mechanisms. The cluster source was extensively characterized to provide the basis for a reliable data analysis.

Ion energy distributions from irradiation of H$_2$ clusters of radii $R_0$ approaching the excursion distance $\xi$ of an electron in a laser field were measured. The respective angular distribution of mean ion energies for use of a 40 fs and a 250 fs pulse was found to reach a maximum along the laser field direction. In addition, a strong increase in ion energies is observed with increasing laser pulse duration. Both results can be attributed to multiple-pass vacuum heating mechanisms predicted in this regime in recent theoretical and numerical studies ([37], [15], [16], [38]).

To fully understand these phenomena, additional simulations in the long pulse regime ($>100\, fs$) with $\xi \approx R_0$ are necessary. As an important consequence of the multiple-pass vacuum heating, the acceleration of electrons and protons to energies exceeding $U_p$ could not be definitively measured due to the intensity distribution in the focus. This situation has been improved by adding a slit mounted in front of the time of flight tube to the setup. However, a new design allowing to study the maximum proton energy as a function of laser intensity in the long pulse regime would be desirable in order to provide a sophisticated proof of the theory.

At the opposite end of the target spectrum, heating mechanisms in large high Z noble gas clusters of condensed argon were investigated. A two-color three pulse pump-probe setup was designed and assembled to study the explosion dynamics.
Fig. 7.1. Shown are peaks in the 3rd order signal obtained from overlapping one of the infrared pulses at 800 nm with the frequency doubled blue pulse. Points marked in red refer to the main pulse of peak intensity $I_0$, while black points are associated with local maxima in the 3rd order harmonic generation yield. Prepulses are hereby located on the negative time axis whereas postpulses were attributed to times $>0$.

For irradiation with a timed sequence of two identical short infrared pulses, a strong increase in ion mean energy was found at an optimum delay. This observation indicates the presence of a resonance in the collisional heating rate as predicted by Ditmire et al. [10]. In the very near future, additional runs using an infrared and a blue pulse as well as incorporating all three pulses will provide further knowledge.

Described method of zero-timing the respective pulse paths via 3rd order harmonic generation on a BBO crystal (see section 6.2) can also be used to characterize the pulse regarding the presence of pre- and postpulses.

To do so, one of the infrared pulses was scanned in time by the blue pulse. Since the intensity of the generated second harmonic light is proportional to the square of intensity of the laser pulse incident on the KDP crystal, the contrast ratio of unwanted features to the main peak in the primary pulse will be squared during the process of second harmonic generation. Thus, the blue pulse can be assumed to lack any pre- and postpulses. A scan carried out for delays reaching from $-20\,\text{ps}$ to $20\,\text{ps}$ is shown in figure 7.1. The negative part of the x-axis corresponds hereby to times prior to the arrival of the main pulse centered at 0. For clarity, points being part of the main peak are illustrated in red, while the respective maxima of
pre- and postpulses found in the temporal shape are depicted in black. Prepulses were detected at times of $-5.9\,\text{ps}$, $-11.3\,\text{ps}$ and $-18.7\,\text{ps}$. Postpulses are located at $1.46\,\text{ps}$, $1.7\,\text{ps}$, $2.2\,\text{ps}$, $4.4\,\text{ps}$ and $4.8\,\text{ps}$.

Obviously, such features can have an effect on the dynamics of the cluster expansion. Especially the postpulses are quite high in intensity, reaching beyond $0.01\,I_0$ at $1.46\,\text{ps}$, where $I_0$ denotes the intensity of the main pulse. However, at the time when the first postpulse arrives at the cluster, the electron density in the argon pump-probe experiment will be sufficiently low so that no significant heating will occur. An initiation of the ionization and a slight expansion could be caused by the prepulses of intensities $0.00035\,I_0$ and $0.0007\,I_0$. Thus, prepulses in the beam might alter the timing for resonant collisional heating measured.

In order to improve the temporal pulse shape, a pulse cleaner is currently implemented in the laser system. Using a saturable absorber, this setup will remove prepulses originating from the oscillator.

In addition, a permanent Rayleigh scatter diagnostic for real time characterization of the gas jet is planned to be installed. That way, a drift in experimental conditions resulting from the cluster source can be instantly detected and compensated for.

By adding an electron spectrometer to the cluster TOF chamber, the two-color multi-pulse setup will be used to investigate electron energy distributions generated through cluster explosions, resulting in deeper insight of the dynamics associated with the expansion process.
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