Size distribution and mass fraction of microclusters in laser-irradiated plasmas

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Laser interactions with a mixture of a gaseous plasma and microclusters depend strongly on the cluster-size distribution, which is usually difficult to measure directly. We present a new method for recovering the cluster-size distribution and cluster mass fraction from measurements of refractive index of the medium. The refractive index is determined from power absorption and phase shift of the probe measured for various delays between the two pulses. The primary cause of absorption is plasma resonances in clusters with peak density above the critical density, which makes the approach especially suitable for determining the tail of the cluster-size distribution. We demonstrate the feasibility of the method by analyzing the data from recent pump-probe experiments at the University of Texas. We have determined that the distribution of clusters depends on the initial cluster radii for these experiments is well approximated by a lognormal distribution shifted with respect to zero radius.

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1. Introduction

Laser interactions with a mixture of a gaseous plasma and microclusters exhibit a variety of interesting phenomena such as fusion neutron production [1,2] and high-harmonic generation [3–5]. The plasma-cluster medium combines the advantages of gas targets (extended interaction length, possibility of phase-matching, capability of operating at high repetition rate) and solid targets (high saturation intensity, control via pre-expansion), which can be beneficial for making hot plasmas with energetic ions [6,2], for generating pulsed X-rays [7] and optical harmonics [8], and for designing plasma waveguides [9] and table-top laser-driven accelerators [10]. Microclusters are generated in laboratory experiments by a supersonic gas jet expanding into a vacuum. Gas condensation produces large solid density clusters of atoms (more than 10^{6} atoms per cluster) if the gas jet backing pressure exceeds several atmospheres. The clusters are converted into dense nanoplasmas when irradiated by a high-intensity laser pulse. It is usually difficult to measure the initial cluster-size distribution in the jet, which introduces significant uncertainties in quantitative interpretation of laser-cluster experiments.

The knowledge of the initial cluster-size distribution is especially critical for applications that involve large clusters from the distribution tail. One such example is production of fusion neutrons by expanding microclusters. There are several techniques for determining various integral characteristics of the cluster distribution, but a diagnostic method for determining the distribution itself is missing. The presently used tools are Rayleigh scattering [11,12], electron impact ionization mass spectrometry [13], and use of the Hagena’s empirical scaling law [14].

In this paper, we demonstrate how the cluster-size distribution and cluster mass fraction can be inferred from the time-dependent refractive index of the plasma-cluster medium. We find the refractive index from measurements of power absorption and phase shift in a pump–probe experiment with a variable delay between the two pulses. The role of the pump pulse is to ionize microclusters and heat the electrons inside them. The resulting hot electron pressure drives cluster expansion. A delayed low-intensity probe pulse, with a duration much shorter than the cluster expansion time, follows the pump pulse.

The absorption of probe pulse energy in a single cluster is determined by the evolving cluster density profile at the location of the plasma resonance. As the cluster expands, its peak density decreases and eventually becomes subcritical. Once the plasma resonance disappears, the energy absorption drops significantly. The corresponding expansion time is shorter for larger clusters than for larger ones, which means that mainly larger clusters contribute to the total power absorption at longer delays. Therefore, the power absorption measured as a function of the time delay carries the information about the cluster-size distribution.
The power absorption is determined by the imaginary part of the refractive index, whereas the phase shift is determined by the real part. The dominant absorption mechanism is the resonant absorption in those clusters whose peak density is above the critical density. This feature allows us to recover the cluster-size distribution directly from the measured absorption without any knowledge about the ambient plasma. On the other hand, the plasma contribution to the real part of the refractive index is generally comparable to the contribution from clusters. We thus use the phase shift measurements for the probe beam to find the fraction of clusters in the gas target.

In what follows, we first describe the experiments for measuring the absorbed power and the phase shift (Sec. 2). We then present the basic concept of finding the cluster-size distribution and the fraction of atoms in clusters from the measurements (Sec. 3). The concept is illustrated in Sec. 4 by applying the simplified model described in Sec. 3 to analyze the experimental data. Finally, in Sec. 5 we summarize the results, discuss the limitations of the simplified model, and outline the steps for enhancing the model.

2. Experiment

In our experiments clusters are formed by condensation in a room temperature (\(T = 300 \text{K}\)) pulsed supersonic argon jet located inside a vacuum chamber. A Series-9 pulsed solenoid valve from Parker Hannifin (General Valve division), backed by pressure \(P = 41, 350 \text{ mbar} \) (600 psi), opened for 1.5 ms to admit gas into a conical nozzle with orifice diameter \(d = 750 \mu\text{m}\) and half expansion angle \(\gamma = 11^{\circ}\). Hagena’s parameter for these conditions is \(\Gamma^* = 2.20(0.74d/tan\gamma)^{0.85} = 1.2 \times 10^5\), using gas species parameter \(k = 1650\) for argon along with \(P, T\) and \(d\) in the units given above. Measurements of a conical pulsed argon jet similar to ours [11] show that for \(10^5 < \Gamma^* < 10^6\), \(\Gamma^*\) is related to the number \(N_h\) of argon atoms per cluster by \(N_h = 100(\Gamma^*/1000)^{1.8}\), and that \(N_h\) was nearly uniform throughout the jet. We checked this relation independently by collecting 90 Rayleigh scatter of 532 nm pulses from a Q-switched Nd:YAG laser as a function of \(P\) over a range \(200 < P < 600 \text{ psi}\) for which gas dynamic simulations of cluster formation show that cluster mass fraction \(f_c\) remains nearly constant [11,15]. The collected scattered energy \(E_{sc} = n_{c}E_{0}\), where \(n_{c}\) is cluster number density. Noting that \(K \approx N_h^{-1}\) and \(n_{c} = f_{c}N_h/N_g\) and using \(I = P/P\), the above relation yields \(E_{sc} \approx f_{c}E_{0}\), where \(n_{total}P\) is the atomic density averaged over a volume containing many clusters. Our measurements yielded \(E_{sc} = 0.049 \pm 0.05\) in agreement with this scaling law. Assuming spherical clusters with solid Ar atomic number density \(n_{0} = 1.8 \times 10^{27} \text{ cm}^{-3}\), this confirmed relation yields an average cluster radius of about 20 nm.

Figs. 1 and 2 show schematics of the absorption and frequency-domain interferometry (FDI) experiments, respectively. For both experiments 800 nm, 100 fs pulses from a 10 Hz Ti:sapphire laser system were split into two beams. Pump pulses (400 nm, 100 fs) were generated by frequency doubling one beam in a 1 mm thick, type-I phase-matched KDP crystal. The pump pulses, focused with a singlet lens, had a beam diameter \((1/e^2)\) in intensity) of 40 \(\mu\text{m}\). The other 800 nm beam was used as a probe with a delay controlled by a translation stage. We used a Mach–Zehnder interferometer (not shown) to measure the volume-averaged atomic density \(n_{total} = 10^{18} \text{ cm}^{-3}\) with pulses from the Q-switched Nd:YAG laser along the path traversed by the nearly co-propagating pump and probe pulses through the gas jet. In the absorption experiment, the focused pump intensity was \(4.4 \times 10^{15} \text{ W/cm}^2\). An 800 nm pulse was split by a Michelson interferometer into a co-propagating probe pulse and a reference pulse with a fixed temporal separation of 1.6 ps. The probe pulse was focused by a singlet lens to the pumped region of the jet and its diameter was approximately 150 \(\mu\text{m}\). The probe and reference pulses were imaged from the exit of the jet onto the entrance slit of the spectrometer by a focusing lens and a microscopic objective. The two pulses interfere in the spectral domain after dispersion on the grating. A CCD camera recorded the interferogram. The probe pulse was modulated by the plasma-cluster medium and gained additional phase. The phase shift with respect to the reference beam at the center of the pump region for each delay \(\Delta\phi(\tau)\) is extracted from the interferogram by a Fourier analysis program [16]. Each data point is 20 shots average.

3. Model

3.1. Basic concept

The measured relative absorption \(\alpha(\tau)\) and the phase shift \(\Delta\phi(\tau)\) for the probe pulse can be expressed in terms of the refractive index of the medium \(n(\tau)\),

\[
\alpha(\tau) = 1 - \exp\left(-2 \frac{\omega_m}{c} \text{Im} n(\tau) - 1\right),
\]

\[
\Delta\phi(\tau) = \frac{\omega L}{c} \left(\text{Re} n(\tau) - 1\right),
\]

where \(\omega\) is the frequency of the probe pulse, \(L\) is the distance traveled by the pulse in the medium, and \(c\) is the speed of light. The key assumption here is that \(n(\tau)\) is independent of the location in the jet.1 The refractive index \(n(\tau)\) is close to unity in our experiments and it includes contributions from the background plasma and expanding clusters.

The first term on the right-hand side of Eq. (3) is the contribution from the background plasma whose ion density is \(n_{i}\) and whose ion charge state is \((Z_{\text{tot}})_{i}\). With \(\omega_m = \sqrt{4\pi\varepsilon_0 n_{\text{tot}}e^2/m_{e}}\), note that we will continue using the subscript \(i\) to mark the parameters of the background plasma, which was produced by ionizing the background monomer gas. The second term is the contribution from the ground-state clusters with initial radius \(R_0\). The density of cluster ions in a unit volume that contains clusters is

\[
n_{n} = 4\pi \int_{0}^{\infty} F(R_0) R_0^2 dR_0,
\]

where \(n_{\text{tot}}\) is the initial ion density in the cluster. The cluster mass fraction, which is the ratio of the number of atoms in clusters to the total number of atoms in the jet, is given by \(n_{c} / (n_{c} + n_{m})\).
The measurements of the relative absorption and the phase shift allow us to determine the time-dependent refractive index of the plasma-cluster medium from Eqs. (1) and (2). The goal is to find the cluster-size distribution and cluster mass fraction using the knowledge of the refractive index. Our key premise is that the power absorption is dominated by clusters with plasma resonances. The imaginary part of the plasma contribution to the refractive index can then be neglected, as done in Eq. (3). In our experiments, the peak electron density produced in clusters by the pump pulse significantly exceeds the critical density for the probe pulse. Therefore, there are plasma resonances in clusters that enhance the power absorption, which is the foundation for our premise.

Modeling of pump and probe interactions with the plasma-cluster medium is required to calculate the polarizability $a(s, R_0)$ and the average ion charge state $Z_m$. In order to find $a(s, R_0)$, one has to consider: 1) ionization and electron heating by the pump pulse; 2) cluster expansion following the pump pulse caused by electron pressure; and 3) the response of cluster electrons to the probe pulse. In order to find $Z_m$, one has to consider ionization of the background plasma by the pump pulse. Tunnel ionization is the dominant mechanism in this case, with the collisional ionization being negligible. In what follows, we use the ionization rate given by the Ammosov, Delone, and Krainov (ADK) model [17] to calculate $Z_m$. A model for calculating $a(s, R_0)$ is presented in Sec. 3.2.

Once the polarizability $a(s, R_0)$ is found, Eqs. (1) and (3) yield an integral equation for $L_F(R_0)$. The equation involves only the imaginary part of the polarizability and no knowledge about the background plasma is needed to determine the cluster-size distribution. Instead of directly solving the equation for $L_F(R_0)$, which poses a technical challenge, we employ a simpler approach. We approximate the cluster-size distribution by a function with several free parameters and fit the absorption data to find these free parameters. The quality of the fit depends on how well this function approximates the actual distribution. Nevertheless, we will show that the tail of the distribution determined with the fitting procedure is robust.

Knowing the polarizability and the cluster-size distribution, we can find the phase shift associated with clusters, $(\Delta \phi)_c$, from Eqs. (2) and (3). The total measured phase shift $\Delta \phi$ is a sum of $(\Delta \phi)_c$ and the plasma contribution $(\Delta \phi)_p$. We can thus find $(\Delta \phi)_p$ using $\Delta \phi$ and $(\Delta \phi)_c$ and then, using the calculated value of $Z_m$, we can find $n_mL$ from Eq. (3). On the other hand, Eq. (4) yields $n_cL$ because we can find $L_F(R_0)$ using the procedure described above. The ratio of the quantities $n_cL$ and $n_mL$ yields the cluster mass fraction. We use this procedure in Sec. 3 to analyze the experimental data.

### 3.2. Cluster polarizability

In what follows, we introduce a simplified model for finding the polarizability $a(s, R_0)$ as a function of the time delay $s$ and initial cluster radius $R_0$. The advantage of this model is that it yields a rescaling relation for $a(s, R_0)$, which allows us to evaluate the...
integral in Eq. (3) without calculating $a(\tau, R_0)$ for each cluster-size. We only need to calculate $a$ for a reference cluster and then use the relation to find the polarizability for other initial radii.

The key simplifications of the model are:

1. The ion charge state and electron temperature generated by the pump pulse are assumed to be the same for the entire ensemble of clusters.
2. Cluster electrons are treated as isothermal due to their high heat conductivity.
3. Cluster ions are treated as cold and their expansion during the probe pulse is neglected.
4. The electron collision frequency is assumed to remain small compared to the frequency of the probe pulse.
5. Plasma recombination and additional ionization following the pump pulse are neglected.

We discuss the role of these simplifications in Sec. 5 after we analyze the experimental data.

Taking into account that the expansion time for a typical cluster exceeds the duration of the pump pulse significantly, we neglect the expansion when considering cluster ionization and heating by the pump. The ionization begins with the tunnel ionization of the constituent argon atoms. This mechanism quickly generates seed electrons, after which collisional ionization caused by thermal electrons takes over and dominates until the end of the pulse. The heating of the electron population is determined by the rate of electron-ion Coulomb collisions. The electron distribution is a Maxwellian, because the pulse duration is longer than the time required for electron–electron collisions. We define $Z_0$ and $T_0$ as an average ion charge state and electron temperature in a cluster at the end of the pump pulse. By definition, these quantities are insensitive to the cluster radius. We use the ionization and heating rates given in Ref. [18] to compute $Z_0$ and $T_0$.

Within our simplifying assumptions 1–5, cluster expansion is described by the following set of equations:

$$
\frac{\partial n_i}{\partial t} + \frac{1}{r^2} \frac{\partial}{\partial r} \left( r v_i n_i r^2 \right) = 0, \quad (5)
$$

$$
\frac{\partial v_i}{\partial t} + v_i \frac{\partial n_i}{\partial r} = -Z_0 T_0 \frac{\partial n_i}{\partial r}, \quad (6)
$$

$$
T(t) = T_0 - \frac{2}{3Z_0} \left( \int_0^t \frac{m v^2}{2} n_i r^2 dr \right) \left( \int_0^t n_i r^2 dr \right)^{-1}. \quad (7)
$$

Here $n_i$ and $v_i$ are the ion density and velocity and $T$ is the electron temperature. Eqs (5)–(7) are the continuity, ion momentum balance, and energy conservation equations, respectively. The right-hand side in Eq. (6) corresponds to an ambipolar momentum balance, and energy conservation equations.

Next, we follow the procedure used in Refs. [19,20] to calculate the cluster polarizability for a known electron density profile. We consider interaction of the probe pulse delayed by $\tau$ with a cluster whose initial radius was $R_0$. The pulse is much shorter than the cluster expansion time, which allows us to neglect the cluster density evolution during the pulse and set $n_i = n_i(\tau, R_0)$. Since the laser wavelength is greater than the cluster radius, the laser electric field can be treated as an applied uniform time-dependent electric field $E = E_0 e^{i \omega t - \beta r}$, where $E_0$ is a unit vector. It is convenient to introduce an electrostatic potential $\phi$, such that $\phi = -\frac{1}{4\pi} \int_{B_{cr}} E e^{i \omega t - \beta r} d^3 r$ for the applied field, where $r$ is the radius and $\beta$ is the polar angle in a spherical coordinate system with the origin at the center of the cluster and the polar axis directed along $E_0$.

Inside the cluster, the electrostatic potential is governed by the equation $\nabla (e \Delta \phi) = 0$, where $e = 1 - \omega_2 \phi(n_i) / (\omega_2 + i \tau)$ is a dielectric constant, $\omega_2 (n_i) = \sqrt{4\pi Z_0 n_i e^2 / m_i}$ is the plasma frequency, and $r$ is the electron collision frequency. The equation is solved numerically for a function $\psi(\tau, r; R_0)$, which is required to converge to $\psi = r$ away from the cluster ($r \to \infty$), with $\phi = -\psi \cos \theta E_0 e^{i \omega t}$. The polarizability of the cluster is related to $\psi(\tau, r; R_0)$ by

$$
a(\tau, R_0) = \frac{\omega_2^{p0}}{\omega_2^2} \int_0^t \left( \frac{\partial }{\partial \tau} + 2 \frac{\partial }{\partial r} \right) \frac{n_i(r, \tau; R_0)}{n_0} r^2 dr. \quad (8)
$$

where $\omega_2^{p0} = \sqrt{4\pi Z_0 n_0 e^2 / m_e}$ and $n_0$ is the initial ion density in the cluster.

Using the rescaling relation for the ion density, $n_i(\tau, r; R_0) = n_i(r_0 a_0 / r_0 a_0, \tau_0 a_0 a_0, a_0 a_0)$, we find from Eq. (8) that the rescaled polarizability is $a(\tau, a_0) = \frac{a_0 a_0}{a_0 a_0} a(n_0 a_0 a_0, a_0 a_0, a_0 a_0, a_0 a_0)$. This relation allows us to evaluate the integral in Eq. (3) without recalculating $a(\tau, R_0)$ for each cluster-size. We calculate $a$ for a reference cluster over a wide range of time delays $\tau$ and then use the rescaling relation to find the polarizability for other initial radii. Even though the collision frequency $\nu$ enters the equation for $\phi$, the dependence of $a$ on the collision frequency is negligible if $\nu$ is sufficiently small to make the resonant layer narrow compared to other radial scales of the cluster.

The imaginary part of the polarizability is enhanced in clusters with a plasma resonance, compared to those without one. Prior to the expansion, the ion density inside every cluster is above the critical density, such that $\omega_2 (n_i) \geq \omega$. The expansion occurs in the form of a rarefaction wave propagating inward and outward from the cluster edge, as illustrated in Fig. 3. Until the wave reaches the center of a given cluster, there is a radial location where the resonant layer is present. Once the rarefaction wave reaches the center, the peak density starts to drop and eventually becomes subcritical everywhere inside the cluster (see Fig. 3). Asymptotically, the polarizability converges to a real value that is independent of the delay,

$$
a(\tau, R_0) \to \frac{\omega_2^{p0} R_0}{\omega_2^2} \frac{3}{4}. \quad (9)
$$

with

$$
n(\tau) = 1 - \frac{1}{2\omega_2^{p0} - \frac{1}{2} \frac{\omega_2^{p0}}{\omega_2} n_i}{n_0}. \quad (10)
$$

In the next section we use the asymptotic expression (10) for the refractive index to find the cluster mass fraction.

4. Data analysis

The measured relative absorption $\kappa(\tau)$ and the phase shift $\Delta \phi(\tau)$ are shown in Figs. (5) and (7). Our first step is to calculate cluster parameters generated by the pump pulse in the absorption experiments. The peak intensity of the pulse was $10^{13}$ W/cm$^2$. We find that the corresponding electron temperature and the average ion charge state at the end of the pump pulse are $T_0 = 200$ eV and $Z_0 = 9$. We take the atomic density of argon in clusters to be equal to $n_0 = 1.8 \times 10^{22}$ cm$^{-3}$. 


Snapshots of a normalized ion density profile \( n_i/n_0 \) obtained by solving Eqs. (5)–(7) are shown in Fig. 3, where \( t = \sqrt{Z_0 t_0/m_i/R_0} \) is the normalized time. The dashed line indicates the normalized ion density corresponding to the critical electron density \( n_{crit} \).

Fig. 3. Snapshots of normalized ion density profile \( n_i(t, \sqrt{Z_0 t_0/m_i/R_0})/n_0 \), where \( t = \sqrt{Z_0 t_0/m_i/R_0} \) is the normalized time. The dashed line indicates the normalized ion density corresponding to the critical electron density \( n_{crit} \).

Snapshots of a normalized ion density profile \( n_i/n_0 \) obtained by solving Eqs. (5)–(7) are shown in Fig. 3, where \( t = \sqrt{Z_0 t_0/m_i/R_0} \) is the normalized time. The snapshots for \( t = 0.55 \) and \( t = 2.21 \) show the inner front of the rarefaction wave propagating towards the center of the cluster. Once the front reaches the center, the peak density begins to drop (\( t = 2.46; 2.61; 3.11 \)). The initial peak electron density in clusters is \( Z_0 n_0 = 1.6 \times 10^2 \) cm\(^{-3} \). The initial peak electron density for the 800 nm probe pulse is \( n_{crit} = 1.7 \times 10^{21} \) cm\(^{-3} \), which is 1.1% of \( Z_0 n_0 \). The peak electron density drops below \( n_{crit} \) at \( t = 6.07 \) and the plasma resonance \( \omega_{pe}(n_i) = \omega \) disappears.

Fig. 4 shows the imaginary part of the normalized polarizability that we find from Eq. (8) for the computed density \( n_i(t, \sqrt{Z_0 t_0/m_i}) \). In order to find \( \alpha(t, R_0) \), we solve for \( \psi \) using the procedure described in Sec. 3 with \( \tau = 10^{-4} \omega_{pe}(n_i) \). The plot of \( \Im[\alpha(t, R_0)] \) has a well pronounced peak \( (\tau = 1.5 R_0) = \sqrt{Z_0 t_0/m_i} \) followed by a cutoff \( (\tau = 5.5 R_0) = \sqrt{Z_0 t_0/m_i} \) that is due to the disappearance of the plasma resonance. The imaginary part of the polarizability has a low residual value for \( \tau > 5.5 R_0 = \sqrt{Z_0 t_0/m_i} \) determined by the value of \( \tau \) used in the calculations. We find that \( \Im[\alpha(t, R_0)] \) peaks at \( \tau \approx 227 \) fs and cuts off at \( \tau \approx 834 \) fs for clusters with \( T_0 = 200 \) eV, \( Z_0 = 9 \), and \( R_0 = 10 \) nm. To find the corresponding dimensional times for a cluster of radius \( R_0 \) one has to multiply the calculated values by the ratio \( R_0/10 \) nm.

The normalized cutoff time is a universal parameter for all clusters, so that the cutoff occurs at longer delays \( \tau \) for larger clusters. Therefore, only large clusters contribute to absorption at long delays and their distribution can then be recovered by fitting the tail of \( \alpha(t) \). We achieve a good fit of the absorption data at \( \tau > 333 \) fs (see left panel in Fig. 5) using a shifted lognormal distribution,

\[
F(R_0) = \frac{c}{4 \pi\sigma |a|} \frac{1}{\sigma \sqrt{2 \pi}} \exp \left( -\frac{[\ln(R_0/a) - \mu]^2}{2\sigma^2} \right).
\]

where \( A = 9.51 \times 10^{-2}, \chi = 0.35, \mu = -2.17, \sigma = 0.93 \) and the radius is normalized to \( a = 20 \) nm. However, the lognormal distribution with these parameters yields an absorption curve that disagrees with the experimental data at short delays (\( \tau < 333 \) fs). The underlying cause is the assumption that the delay \( \tau \) in our calculations is exactly equal to the experimental delay.

We have effectively neglected the finite duration of the pump pulse by interpreting \( \tau \) in the calculated polarizability as the delay between the two pulses. By definition, the experimental delay is the time interval between the peak intensities in the pump and the probe. In our model, the cluster expansion starts at \( t = 0 \) with \( T = T_0 \) and \( Z = Z_0 \). The electron temperature and the average charge state become equal to \( T_0 \) and \( Z_0 \) only at the end of the pump pulse. Therefore, the calculated delay is shorter than the measured delay by roughly half of the duration of the pump pulse. In order to account for that, we shift the time in the calculated polarizability by adding 70 fs.

The adjustment of the calculated time delay allows us to achieve a good fit of the absorption data for the entire range of measured delays (see right panel in Fig. 5). We again use a shifted lognormal given by Eq. (11) for the cluster-size distribution. The fitting procedure yields \( A = 1.82 \times 10^{-1}, \chi = 0.27, \mu = -2.33, \sigma = 0.93 \). Fig. 6 shows the cluster-size distributions determined with and without the adjustment. The main difference between the two is at smaller radii (\( R_0 < 20 \) nm), whereas the tail of the distribution is insensitive to the adjustment. The tail is robust because the characteristic expansion time for the corresponding clusters is significantly longer than the duration of the pump pulse.

Our next step is to find the cluster mass fraction using the determined cluster-size distribution function. For technical reasons, the peak amplitude of the probe pulse in the phase shift experiment was \( 4.4 \times 10^{15} \) W/cm\(^3\) and not \( 10^{15} \) W/cm\(^3\), as in the power absorption experiment (see Sec. 2). The corresponding electron temperature and the average ion charge state at the end of the pump pulse are \( T_0 = 370 \) eV and \( Z_0 = 14 \). We find that the ion charge state in the background plasma is \( Z_{crit} = 4.6 \). The measured phase shift \( \Delta \phi \) is shown in Fig. 7 as a function of the delay between the two-pulse. It contains two distinct contributions: a contribution from clusters and a contribution from the background plasma. We can distinguish them because of the difference in their characteristic time scales. The plasma contribution reaches its asymptotic value on a time scale comparable to the duration of the pump pulse. The time evolution of the cluster contribution is determined by the characteristic cluster expansion time, which is considerably longer than the pump pulse duration. It then follows from Fig. 7 that the asymptotic contributions from the plasma is \( \Delta \phi_{plasma} = -1.4 \). Our current measurements alone are not sufficiently conclusive to determine the asymptotic value of the cluster contribution \( \Delta \phi_{cluster} \), because the phase shift was measured only for delays less than 1400 fs.

Fig. 4. Time dependence of the imaginary part of the normalized polarizability, \( \Im[\alpha(t, R_0)]/|\alpha| \), for a single cluster, where \( t = \sqrt{Z_0 t_0/m_i/R_0} \) is the normalized time.
In order to find \((\Delta \phi)_c\), we calculate the polarizability for \(T_0 = 370\) eV and \(Z_0 = 14\) and then use it in Eq. (3) together with the determined cluster-size distribution (solid curve in Fig. 6) to find the cluster phase shift. The asymptotic value is \((\Delta \phi)_k \approx -0.83\). It follows from Eqs. (2) and (10) that \((\Delta \phi)_k/(\Delta \phi)_p = Z_0 n_c/Z_m n_m\), where \((\Delta \phi)_k\) and \((\Delta \phi)_p\) are the asymptotic values of the cluster and plasma contributions. We thus find that the mass fraction of clusters in our experiments is 17%, with

\[
\frac{n_c}{n_m} = \frac{(Z_m/\Delta \phi)_c}{Z_0 (\Delta \phi)_p} = 0.20. \tag{12}
\]

5. Discussion

Current absorption and phase shift measurements have been taken with two different pump pulses that have different peak intensities. The mass fraction result would be more reliable if both measurements were taken simultaneously. It is in our future plans to make that improvement, which would automatically increase the range of time delays for the phase shift measurements. Extending the range would also allow us to measure directly the asymptotic behavior of the cluster contribution to the total phase shift.

The analysis of the data presented here is based on the concept that the measured absorption is primarily attributed to the resonant absorption in clusters. We have made a number of assumptions to simplify the analysis without losing the key feature — the resonant response by cluster electrons. The strength of the model is the rescaling relation for polarizability that allows one to compute the power absorbed by a cluster ensemble by calculating the polarizability for just one cluster-size. As pointed out in Sec. 4, the current model effectively neglects the finite duration of the pump pulse. It is necessary to account for the finite duration of both pump and probe pulses in order to enable the model to recover the size distribution of small clusters whose characteristic expansion time is comparable to the pulse duration. Our current model is most appropriate for those applications that are primarily concerned with the tail of the cluster-size distribution.

The rate of electron collisions is fixed in our model and it is taken to be sufficiently low, such that the resonant absorption is independent of the rate. This assumption is justified for Coulomb electron-ion collisions at the resonance for the initial value of the electron temperature \(T_0\) generated by the pump pulse in the absorption experiments. However, the electron cooling caused by the expansion increases the rate of electron-ion collisions. As the electron temperature drops, the three-body recombination eventually begins to counteract the cooling. Therefore, we need to investigate the effect of the electron cooling and recombination in order to calculate the electron collision rate self-consistently.

Besides affecting the energy balance, the recombination also changes the ion charge state in clusters. Our current interpretation of the phase shift measurements is based on the assumption that

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**Fig. 5.** Measured and calculated relative absorption \(\kappa(t)\) as a function of the delay \(t\) between the pump and the probe. Open circles with error bars show the experimental data (the same in both panels). The solid curve in the left panel is the absorption calculated for a lognormal distribution (11) with \(A = 9.51 \times 10^{-2}, \chi = 0.35, \mu = 2.17, \sigma = 0.93\), and \(a = 20\) nm. The solid curve in the right panel is the absorption calculated for a lognormal distribution (11) and shifted by 70 fs, with \(A = 1.82 \times 10^{-1}, \chi = 0.27, \mu = 2.33, \sigma = 0.93\), and \(a = 20\) nm.

**Fig. 6.** Cluster-size distributions determined by fitting the experimental absorption data. The dashed curve yields the absorption shown in left panel of Fig. 5. The solid curve yields the absorption shown in right panel of Fig. 5 after being shifted by 70 fs.

**Fig. 7.** Measured phase shift as a function of the delay between the pump and the probe. The dashed line indicates the asymptotic plasma contribution.
the ion charge state remains equal to $Z_0$ during the expansion. Since
the average ion charge state decreases due to the recombination,
the absolute value of the cluster contribution to the total phase shift
would asymptotically decrease instead of staying flat. There are
indications of this behavior in the experimental data plotted in
Fig. 7. Including the recombination into our analysis of cluster
expansion would involve a kinetic treatment of ions. Cluster ions
with the same charge state at the same location would have
different velocities depending on the time history of their recom-
bination. A preliminary analysis of a single cluster expansion within
a hydrodynamic model that includes the recombination and uses
a self-consistently calculated electron-ion collision rate indicates
that the imaginary part of the cluster polarizability remains peaked
when the plasma resonance is present inside the cluster [21].

6. Summary

We have presented time-resolved measurements of power
absorption and phase shift for a plasma-cluster medium obtained
in a two-pulse pump-probe experiment. We have also presented
a new concept for recovering the cluster-size distribution and
cluster mass fraction using these measurements. The measure-
ments have been analyzed using a simplified model to determine
the cluster-size distribution and cluster mass fraction. We have
found that the cluster-size distribution is well approximated by the
lognormal distribution function (11) with $A = 1.82 \times 10^{-1}$, $\mu = 0.27$,
$\sigma = 0.93$. The cluster mass fraction corresponding to
this distribution has been estimated to be 14%.

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