Resonance in scattering and absorption from large noble gas clusters

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Abstract: Light scattering in large noble gas clusters irradiated by intense laser pulses was studied and compared to absorption measurements. The scattering signal shows the presence of a peak, when the pulse width was varied, similar to one previously reported in absorption measurements. The peak of the scattering, however, occurs at a longer pulse width than for absorption. This result disagrees with a simple simulation and may be due to propagation or non-linear effects not included in the model.

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OCIS codes: (290.0290) Scattering; (320.7120) Ultrafast Phenomena

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In recent years there has been great interest in the interaction of intense laser light with large noble gas clusters[1, 2, 3]. The high local density within a cluster, coupled with the low average density of the overall gas, creates the conditions for energetic interactions. It has been shown that these targets can absorb nearly all of the incident laser radiation[4], producing XUV emission along with high energy electrons[5] and...
ions[6]. This is in stark contrast to irradiation of a pure monatomic gas, where low temperature plasmas and much smaller yields of XUV radiation are produced.

More recently work has been conducted to study the temporal dynamics of the cluster interaction [7, 8]. Looking at the absorption of laser energy, as well as XUV emissions, these studies have given good evidence for a plasma model of cluster expansion[9]. For a large cluster with a high Z species, space charge forces will confine a majority of the ionized electrons to the cluster sphere. A small plasma, much smaller than the wavelength of the laser, will be formed. Collective effects will give rise to an absorption resonance which will lead to an energetic interaction. This resonance occurs when the electron density equals 3 times the critical density. At this point the laser frequency and the natural frequency of the spherical plasma are equal; energy is efficiently transferred from the field into charge separation within the cluster. The energy stored in the dipole will be transferred to the random motion of the clusters via collisions in the dense plasma. By using both pump probe style experiments and a variable pulse width, it was seen that by letting the cluster expand to this resonance condition the interaction can be made more energetic, increasing absorption.

As the laser interacts with the cluster, light will be scattered as well as absorbed. While the amount of scattered light is small, the signal can be used to diagnose the interaction. One would expect the resonance which leads to increased absorption to also lead to increased scattering. We have examined this, performing simultaneous absorption and scattering measurements. We find that there is a similar effect in the scattering as seen in the absorption. However the enhancement seems to occur at a different pulse width than the absorption, indicating that a simple model does not contain a complete description of the interaction.

The laser used for these experiments was a Ti:Sapphire CPA laser system operating at a wavelength of 810 nm, capable of producing about 50 mJ in a 50 fs pulse (the experiments described here only used up to 10 mJ). The pulse width of the laser could be adjusted by detuning the compressor. The experimental setup is basically the same as described previously [7]. Briefly, the laser was sent into a 60 cm spherical target chamber and focused using a f/3 off axis parabola. Approximately 70% of the energy was contained in a 9 µm gaussian spot, with the remaining energy in low intensity wings. The maximum focal intensity in vacuum was \(2.4 \times 10^{17} \text{W/cm}^2\) for a 50 fs pulse. The clusters were produced by a Mach 8 supersonic Laval nozzle backed with up to 14.8 bar xenon or 42.4 bar argon. The cluster radii ranged from 85Å to 205Å for xenon and 110Å to 165Å in argon. The average atomic density 1 mm below the nozzle, where the laser was focused was up to \(6.9 \times 10^{17}\) atoms/cm\(^3\) for xenon and up to \(2 \times 10^{18}\) atoms/cm\(^3\) for argon. The transmitted light was collected with an f/2.3 lens and measured with a pyroelectric energy meter. Another f/2.3 lens was used to collect the 90° scattered light, that is the light scattered perpendicular to the direction of propagation. The signal was detected with a silicon energy probe. An 810 nm bandpass filter, with a 10 nm FWHM bandwidth, was placed in front of the energy head to filter both room light and most plasma light. A \(\lambda/2\) waveplate before the chamber allowed us to rotate the polarization. All measurements are 50 shot averages.

We first examined the radiation pattern to ensure that the plasma dynamics were not interfering with the scattered signal. By rotating the waveplate we can map out the scattered intensity with respect to the laser polarization. The characteristic of Mie scattering, in the Rayleigh limit, is a dipole radiation pattern. The radiation pattern for 42.4 bar argon is shown in Fig. 1. The \(\cos^2(\theta)\) dependence is exactly what we expect. To be sure plasma light was not interfering with the measurement, we put a filter to block the scattered laser light in front of the detector. We simply used a dielectric mirror which reflected the band from about 770-870 nm. The radiation detected was isotropic.
Fig. 1. Emission pattern for scattered laser light. The target was argon at 600 psi. This fit is a \( \cos^2(\theta) \). The intensity was \( 1.3 \times 10^{16} \) W/cm\(^2\). The pulse width was 800 fs.

as would be expected from plasma light. This indicated that the signal measured in Fig. 1 is from scattering and not plasma emission.

The scattering from both argon and xenon was studied (they produced similar results). We measured the scatter perpendicular to the polarization of the incident field, collecting 0.35% of the total solid angle. The total amount of energy scattered was small, the maximum detected signal was 16 nJ for 14.8 bar xenon (this was well above the detection limit of about 30 pJ). Figure 2 shows the scattered signal (bottom) from the cluster target, along with the absorbed energy (top), for several backing pressures of xenon as a function of pulse width. Both the scattering and absorption show similar behavior. There is a rapid rise to a peak and then a gradual decrease in signal. Both the magnitude of the signal and the pulse width for peak signal are larger for higher backing pressure, which corresponds to larger clusters.

The basic shape of these curves is understood as being caused by the resonance between the spherical target and the laser frequency. At 50 fs, the pulse length is small compared to the hydrodynamic expansion time. An overdense plasma is formed from each cluster, but by the time it expands to the resonance condition, the laser pulse is already past. As the pulse gets longer the resonance begins to occur during the laser pulse, enhancing the absorption and scattering. Eventually, the signal begins to fall off as the resonance occurs early in the pulse and most of the laser energy interacts with a bulk plasma of relatively low density. Simulation done previously on the absorption data (shown in Fig. 2, top) agreed well with this picture of the interaction.

When we compare the scattering and absorption data we see that the scattering signal peaks at a longer pulse width than the absorption. The code that was developed to calculate the absorption\[7\] from the target was modified to calculate scattering also. This code considers a uniform density cluster plasma, taking into account tunnel and collisional ionization, ATI and inverse bremsstrahlung heating, along with the hydrodynamic and Coulomb explosion aspects of the expansion. Shown in Fig. 2 (bottom), it is seen that the scattering model results peak at a similar time as the absorption, disagreeing with the data.
To try to understand this discrepancy we examined the basic equations used in the model. Figure 1 showed a dipole nature, indicating small object scattering. In this limit the scattering and absorption cross sections are

$$\sigma_{sca} = \frac{8\pi}{3} k^4 r_6 \left| \frac{\epsilon - 1}{\epsilon + 2} \right|^2,$$

$$\sigma_{abs} = 4\pi k r_3 \text{Im} \left\{ \frac{\epsilon - 1}{\epsilon + 2} \right\}. \quad (1)$$

These are similar in form and, to first order, we would expect to see similar behavior.

While absorption generally dominates for very small particles [10], it is well known that as the radius increases the stronger scaling with the radius of the scattering cross section compared to the absorption cross section will lead to scattering becoming greater than absorption (Fig. 3). With a longer pulse, the resonance will be reached early in the pulse so a cluster will have a larger radius during more of the pulse than it would for a short pulse. However, this scaling only holds for a constant index of refraction. As a cluster expands, the density drops and hence the index of refraction drops. We can write the dielectric function of the plasma as

$$\epsilon = 1 - \frac{n_0}{n_{crit}} \left( \frac{r_0}{r} \right)^3 \left( 1 + \frac{i\nu}{\omega} \right)^{-1} \quad (2)$$

where $n_0$ is the initial electron density, $n_{crit}$ is the critical density, $\omega$ is the laser frequency, $\nu$ is collisional frequency, and $r_0$ is the initial cluster radius. This is plotted in Fig. 4 for an expanding plasma sphere. The weak field limit of the formulas of Silin [11] were used to calculate the electron-ion collisional frequency (assuming a 300 eV plasma ionized to $Z=12$). Both real and imaginary parts start off very large and then approach their vacuum values. Using a Mie code [12], we can calculate the cross-sections for a radially expanding cluster using this dielectric constant. Seen in Fig. 5, both cross-sections...
Fig. 3. Calculated scattering (dashed) and absorption (solid) cross sections from Mie theory. The index of refraction was constant for all radii.

Fig. 4. Calculated dielectric constant for an expanding plasma sphere. Plot shows both real (dashed) and imaginary (solid) components.
reach their peaks at the same time. The scattered signal does fall off slower than absorption once the peak is reached. However the cluster would be rapidly expanding at this point and hence the contribution to the time integrated signal will be small.

We also calculated the heating and scattering of an individual cluster using the full code. Figure 6 shows the calculated scattered signal (as measured in the experiment) along with the total heating as a function of time. This calculation shows what we expect from Fig. 5. Both of these show a rapid increase at the same time. This resonant behavior, dominating both absorption and scattering, occurs at the same time.

From all the above, we see that the current modeling does not agree with the data. We also investigated reabsorption of the the scattered light in the plasma, but this seems to be a small effect and does not cause a shift on the scale observed. It is possible that non-linear effects of the laser propagating through the plasma are the cause of the difference. The ions in the plasma may also have an effect. Equation 2 only accounts for the free electrons in the plasma. The ions still have a large number of bound electrons around the nucleus. These are tightly held and one would not expect this to be a large effect. However, it is possible that electron recombination into outer orbitals are creating effective scatterers.

In conclusion there is clearly an optimal pulse width to maximize the scattered radiation. While this is related to the resonance seen in the absorption, the fact that the peak occurs at a different pulse width indicates that the scattering dynamics are more complicated than indicated by the simple model. Calculations show that both absorption and scattering should behave in the same way, and the resonance should affect the absorption and scattering at the same time. It is not clear if the discrepancy is due to scattered light before the resonance, during the resonance, or after the resonance. Further experiments measuring the scattered light in a pump-probe configuration should help determine this and resolve the disagreement between theory and experiment.

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Fig. 6. Calculated scattering (dashed) and heating (solid) for a 100 Å xenon cluster. The pulse width was 450 fs and the intensity was $4.1 \times 10^{15}$. by Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.