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Impurities good and bad: Doped cluster nanoplasmas in intense laser fields and characterization of impurity level

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Abstract. Doping of cluster-based targets can bring out considerable modifications in the evolution of the nanoplasma formed from clusters in intense laser fields. The consequence could be either an increase or, a decrease (depending upon the properties and proportion of the dopant) in the emission of the resulting charge particles or photons from nanoplasma. As we can control the percentage of CS_2 in the doped Ar-CS_2 cluster, we can have argon-doped CS_2 cluster (when argon constitutes about 10–40%) and CS_2-doped argon cluster (when fraction of CS_2 is 10–40%). In the experimental studies of electron spectra and X-ray emission from pristine Ar_n ($n \leq 25,000$) and doped Ar-CS_2 clusters at laser intensities of about 10^{15} W cm^{-2}, it is observed that there is more than an order of magnitude enhancement in those emissions in doped Ar-CS_2 clusters than in the former case. Conversely, a significant reduction in those emissions was found in the latter case. Such observations signify the importance of characterization of these targets. In this direction, we demonstrate a simple method for the characterization of doping level based on the Rayleigh scattering measurements.

Keywords. Doped clusters; nanoplasma; intense laser fields; X-ray emission.

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1. Introduction

Ready availability of intense, ultrashort laser sources has driven widespread interest in studying the response of various forms of matter to intense ultrashort light. Investigations have led to the discovery of several fundamental phenomena and many avenues for technological applications have opened up. Gas phase clusters are uniquely placed between the two density regimes. The large local density in a cluster gives rise to solid-like behaviour but, at the same time, sizes of clusters are small enough to ensure that atomic and molecular effects do not lose their relevance. This combination of high local density and low bulk density leads to various spectacular phenomena [1]. Clusters absorb nearly 100% of the incident laser energy [2]. This absorbed energy (owing to the absence of any solid-like energy dissipation mechanisms) is converted into keV electrons [3], Mev ions [4] and photons of upto a few keV energies [5,6]. The extent of acceleration in deuterium clusters is shown to lead to nuclear fusion [7] which indicates the potential of the nanoplasma to be used as a tabletop neutron source. Efforts were made to control and optimize the brightness and yields of these emissions using parameters like laser polarization, pulse width, wavelength and chirp [1]. As is evident, all the control parameters are pertaining to laser only. We propose the possibility of controlling cluster ionization dynamics not by modifying the laser parameters but rather by engineering the constituents of the cluster target itself.

In earlier reports, similar possibilities have been demonstrated while studying the X-ray emission from nanocluster plasmas that are formed by interactions of water-doped argon clusters with intense femtosecond-duration laser pulses. It was shown in [8] that when very large argon clusters (up to 40,000 atoms) are doped with water molecules, the changes in the ionization dynamics and the enhanced propensity for ionization of molecular entities give rise to significant changes in the nanoplasma behaviour. The experimental findings in water-doped clusters can be summarized as follows: (i) more than an order of magnitude enhancement in Ar K-shell X-rays [8], (ii) more than three-fold increase in yields of highly energetic ions [9], (iii) seven-fold enhancement in the highly energetic electron yields [10] and (iv) two-fold increase in the integrated laser energy absorption [11]. The qualitative change in the dynamics of the evolution of cluster nanoplasma upon doping was shown numerically in [10]. The time-dependent electron density (generated by low ionization energy constituents of the cluster) rises in the early stages of the laser pulse and consequently, changes the cluster explosion dynamics resulting in the coupling of more laser energy into the cluster nanoplasma.

These observations raise certain pertinent questions regarding the observed modifications in the nanoplasma behaviour upon doping of the clusters. These include whether these observed phenomena are water-specific or, any dopant, for that matter, with low ionization energy would exhibit similar behaviour? What would happen if a cluster is doped with a system that has larger ionization energy [12]? To answer such questions more studies with a range of laser and doping parameters are required. The present report is an effort in this direction. We present studies on the carbon disulphide doped clusters. CS_2 has one of the lowest ionization energies and thus is an obvious choice to explore the above-mentioned phenomena. Also, CS_2 being a highly polarizable and vapourizable system, the extent of doping could be enhanced much more than the extent of doping (maximum 8%) obtained using water [11]. These experiments, however, become more involved in that CS_2, being itself a highly condensing material, can produce large clusters of its own. Consequently, when allowed to mix with argon before clustering, the size of CS_2-doped argon cluster will be different from pure argon cluster under otherwise similar experimental conditions (like backing pressure, etc.). Therefore, separate characterizations of CS_2 clusters, Ar clusters and Ar-CS_2 clusters become essential to draw any inferences regarding the effect of doping. In light of this complication, it becomes imperative to form a methodology to characterize doped Ar-CS_2 clusters. This is achieved by using Rayleigh scattering experiments in conjunction with the methods developed by Hagena *et al* [13]. The goal of the present studies is to generalize the role of doping which could, subsequently, result in exploring doping as a tool to control the dynamic evolution of nanoplasma and developing new strategies for laser-based tabletop accelerators.

2. Experimental methods

Our experimental set-up is described in recent publications [8] and only a brief outline is presented here. CS_2 clusters were produced by bubbling He gas through a column of CS_2 liquid, before entering the stagnation chamber and subsequent expansion. The cluster source consists of a general valve (series 99 stainless steel solenoid valve) fitted with copper gasket seal and a Kel-F poppet. To produce clusters of different sizes and densities, different nozzles of 0.5 and 1.0 mm orifices and 45° half expansion angles were used. Intense laser radiation was obtained from a CPA-based Ti:S laser system capable of delivering pulses of upto 55 mJ energy, of 100 fs duration, at 10 Hz repetition rate. The electron emission was measured with a 19 cm field-free time-of-flight (TOF) spectrometer consisting of a retarding potential analyzer (RPA) close to the microchannel plate (MCP) detector. The electron energies were estimated by scanning the voltage on RPA from 0 to -5 kV and integrating the signal near $t = 0$ over time [10]. The X-rays were measured across a thin mylar window, using a 7 mm^2, 300 μm silicon X-ray detector (Amp-Tek, XR-100CR) fitted with 25 μm beryllium filter. For the studies on electron spectra, a 750 μm skimmer was used to deliver the clusters into a differentially pumped chamber maintained at a base pressure of $\sim 10^{-8}$ Torr. For Rayleigh scattering measurements the second harmonic of the 806 nm 100 fs Ti:S laser was used. The Rayleigh signal was found to vary as cubic power of the stagnation pressure of argon gas.

3. Results and discussion

3.1 *Characterization of Ar-CS_2 clusters*

Using argon as a seed gas to generate clusters introduces additional complexity. Unlike helium which barely forms clusters of its own, the condensation propensity (and hence clustering) is very high for argon [13,14]. In this case, mixed Ar-CS_2 clusters could be formed rather than pure CS_2 clusters. Rayleigh scattering measurements alone for the characterization of clusters has severe limitations when both the cluster density and the cluster size are unknown [15]. In this case, therefore, an additional measurement is required to estimate the average cluster size. Also, the standard form of Hagena method cannot be directly used for estimating CS_2-cluster size as these are valid only for the rare gas media [13]. The condensation parameter used is not known for other vapours like CS_2. Therefore, we adopt the approach of corresponding gases to deduce CS_2-cluster size [16,17].

Cluster formation is regarded as a homogeneous gas reaction consisting of bimolecular collisional processes between atoms and cluster, following a unimolecular decay of first order. From the principle of corresponding gases (eqs (12) and (4) in ref. [17]), the modified Hagena parameter can be written as

$$\Gamma^* = (n_0 r_{\mathrm{ch}}^3) \times \left(\frac{d_{\mathrm{eq}}}{r_{\mathrm{ch}}}\right)^q \times \left(\frac{T_0}{T_{\mathrm{ch}}}\right)^{0.25q-1.5}. \tag{1}$$

In eq. (1), the terms used are defined as $r_{\mathrm{ch}} = (m/\rho)^{1/3}$, and $T_{\mathrm{ch}} = \nabla h_{\mathrm{s}}^0/k$, where m represents the mass, ρ the solid density, ∇h_{s}^0 the enthalpy of sublimation of the cluster media and k, the Boltzmann constant while d_{eq} represents the nozzle diameter. n_0 is the number density of the species which is proportional to the stagnation pressure P_0 and T_0 represents the temperature of the stagnation chamber. The term q summarily represents the specific heat ratios and other gas-specific properties relevant in the condensation process ($q = 0.85$ for argon). The experiments are done at room temperature. The vapour pressure of CS_2 at room temperature (298 K) is 0.464 bar. This gives the value of the parameter Γ^* to be ~5900, giving out CS_2 cluster size of $\langle N_{\mathrm{CS}_2}\rangle \approx 2110$ at 1 bar of backing pressure of He or Ar [13,18]. Under similar experimental conditions $\langle N_{\mathrm{Ar}}\rangle \approx 50$. As a result, we have now the size of CS_2-cluster at 1 bar backing pressure. To estimate the sizes at different pressures, we need to know the partial pressure of CS_2-vapour in the experimental chamber at each backing pressure. We perform the Rayleigh scattering measurements to achieve this objective.

We begin with two separate experiments of Rayleigh scattering measurements. The scatterers are argon clusters and CS_2 clusters respectively. We keep all the experimental parameters, except for the backing pressure and (hence) the cluster size, the same. Under these conditions, for a given number of cluster constituents, i.e. $\langle N_{\mathrm{Ar}}\rangle = \langle N_{\mathrm{CS}_2}\rangle$, Rayleigh signal of CS_2 will increase by a factor of

$$S_{\mathrm{RS}} = \frac{f(\mu_{\mathrm{CS}_2})}{f(\mu_{\mathrm{Ar}})} \times \left(\frac{r_{\mathrm{CS}_2}}{r_{\mathrm{Ar}}}\right)^6 \times \left(\frac{P_{\mathrm{CS}_2}}{P_{\mathrm{Ar}}}\right), \tag{2}$$

where $f(\mu) = (\mu^2-1)^2/(\mu^2+2)^2$, μ is the refractive index of the medium and r is the average size of the scatterers [19]. Let us take η as the number density given by ρ/m ($\eta_{\mathrm{CS}_2} \approx 9.9\ \mathrm{nm}^{-3}$ and $\eta_{\mathrm{Ar}} \approx 25.8\ \mathrm{nm}^{-3}$). Then, further consideration of the condition $\langle N_{\mathrm{Ar}}\rangle = \langle N_{\mathrm{CS}_2}\rangle$ gives us, $\eta_{\mathrm{Ar}}/\eta_{\mathrm{CS}_2} = r_{\mathrm{CS}_2}^3/r_{\mathrm{Ar}}^3$. Now, substituting these along with the known values of $f(\mu_{\mathrm{CS}_2}) = 0.1455$ and $f(\mu_{\mathrm{Ar}}) = 0.03285$ [20–22], we estimate from eq. (2) that $S_{\mathrm{RS}} \approx 29.9 \times P_{\mathrm{CS}_2}/P_{\mathrm{Ar}}$. Again, using the condition $\langle N_{\mathrm{Ar}}\rangle = \langle N_{\mathrm{CS}_2}\rangle$ in Hagena relation [13], we have $P_{\mathrm{CS}_2}/P_{\mathrm{Ar}} = (\Gamma^*_{\mathrm{CS}_2}/\Gamma^*_{\mathrm{Ar}})^{1/2.35}$, which under the experimental parameters used for the current studies reduces to $P_{\mathrm{CS}_2}/P_{\mathrm{Ar}} = 0.34$. With the available experimental data at 1 bar (corresponding to $\langle N_{\mathrm{CS}_2}\rangle \approx 2110$, P_{Ar} is estimated to be 5.76 bar for the same size), the Rayleigh scattering measurement at this value gives $S_{\mathrm{RS}} \approx 6.15$. By now, we have all the numbers required. To get the partial pressure of CS_2 vapour the steps involved are as follows: (i) divide the CS_2 Rayleigh scattering data by 6.15, (ii) calculate the corresponding argon stagnation pressure using the cubic fitting of the argon Rayleigh scattering data [8] and get the corresponding pressure (for argon) which would produce argon clusters of the same size as those of CS_2 clusters, (iii) estimate the equivalent argon cluster size using Hagena parameter [8] which gives us the elusive CS_2 cluster size, and (iv) using eq. (1) along with the pressure dependence of Γ^*, get the partial pressure of CS_2 vapour. Now before elaborating the final step, we note that the partial pressure of CS_2-vapour, when CS_2 is backed by argon gas is expected to be approximately the same as that in CS_2 backed by helium. Therefore,

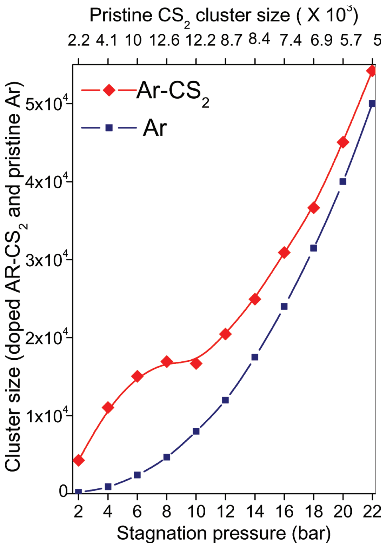

Figure 1. Estimate of the argon, CS$_2$ and mixed Ar-CS$_2$ cluster sizes. The solid square represents the argon cluster size while the data depicted by solid diamond represent mixed Ar-CS$_2$ clusters. The sizes of computed pure CS$_2$ clusters are enumerated on the top axis. The line joining the data points is just to guide the eye.

to estimate the size of Ar-CS$_2$ cluster, as a first approximation, we first account for CS$_2$ clusters and the Ar clusters that would be formed at a given backing pressure and then we attribute the excess in the Ar-CS$_2$ Rayleigh scattering to the mixed Ar-CS$_2$ clusters.

Figure 1 shows the estimated cluster size of pure argon and mixed Ar-CS$_2$ cluster as a function of stagnation pressure. The number of constituents in both Ar-CS$_2$ and argon clusters are shown on the same scale. The sizes of the pure CS$_2$ clusters are shown in the top axis. The point to be noted is that the size of the CS$_2$ cluster initially increases upto 8 bar of backing pressure and then starts decreasing afterwards. Below 8 bar, kinetic flow of CS$_2$ bubbled through helium enables formation of larger clusters. At higher pressures, however, the condensation of CS$_2$ in the stagnation chamber would dominate and there would be decrease in the CS$_2$ throughout about the nozzle giving rise to the formation of smaller clusters. The fraction of CS$_2$ in Ar-CS$_2$ clusters is found to vary from more than 90% at 1 bar to 40–50% at intermediate pressure of 6–8 bar down to less than 20% beyond 16 bar. Further, to estimate the errors associated with the computed size, we compute the Rayleigh scattering signal that would be measured from the mixed Ar-CS$_2$ clusters. The parameters were kept the same as that were used in the experiments. Both the computed and experimentally measured data were compared. A maximum discrepancy of 27% was found in the measured and computed Rayleigh data. This error when projected on to the computed size of the clusters, turns out to be around 13%.

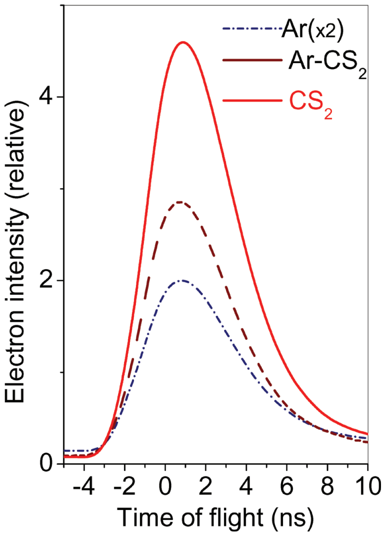

Figure 2. Time-of-flight pertaining to electron signal of Ar clusters (dash-dotted line) and doped Ar-CS_2 clusters (dashed line). The electron intensity in pure CS_2 cluster (solid line) is also shown. The argon data are multiplied by 2 to make representation on the same scale. The data are averaged over 1000 laser shots. The laser pulse with central wavelength 800 nm, width 100 fs and intensity $\sim 10^{15}$ W cm^{-2} was used.

3.2 *Enhancement in yields of high-energy electrons*

A look at the electron time-of-flight (see figure 2) shows an enhancement in electron yields in doped Ar-CS_2 clusters. The data are taken at 8 bar backing pressure and averaged over 1000 laser shots. The estimated size of the doped Ar-CS_2 clusters at 8 bar pressure is about 17,000 whereas that of pure Ar clusters are about 4700 each. It is important to note that while there is huge enhancement in the electron yields in both pure CS_2 and Ar-CS_2 clusters compared to pure the argon clusters (the electron signal of argon cluster is multiplied by 2), there is a significant drop in the electron yields in doped Ar-CS_2 cluster compared to pure CS_2. This is very crucial and will be discussed later.

Figure 3 shows the ratio of electron yields between Ar-CS_2 clusters and Ar clusters as a function of backing pressure at various electron energies (while integrating the electron signals, the background photons have been duly taken care of). The order of magnitude increase in electron yields of energies within the intermediate range of 600 eV to about 2 keV is observed. The enhancement is observed over the whole range of parameters used in these experiments. These observations prove that the presence of low ionization energy materials, in the heteronuclear clusters, changes the dynamics of laser–cluster interactions, i.e., the high-energy electrons and the charge states of ions [9,10]. As per the observations reported in refs [8–10], it is established that water-doped cluster is not unique in the sense that doping with any species having low ionization energy significantly enhances the electron yields.

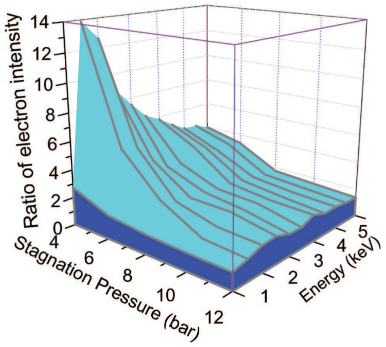

Figure 3. A 3D representation of the enhancement of electron yields in Ar-CS_2 clusters compared to pure Ar clusters as a function of backing pressure at various electron energies. The x-axis represents the stagnation pressure in bar. The y-axis shows electron energies in keV. The z-axis enumerates the ratio of electron yields (Ar-CS_2 clusters divided by pure Ar clusters).

3.3 *Consequence of doping: The other side*

For a particular stagnation pressure, the sizes of pure CS_2 clusters, Ar clusters, and Ar-CS_2 clusters are all different. For example, at 2 bar of stagnation pressure $\langle N_{\mathrm{Ar\text{-}CS_2}}\rangle \approx 4300$ (with fraction of CS_2 being 95%) while the same size of pure argon cluster is obtained at 7.7 bar, given the experimental parameters used in this study. Similarly, at 4 bar, $\langle N_{\mathrm{Ar\text{-}CS_2}}\rangle \approx 11{,}000$ (with fraction of CS_2 being 90%) while the same size of pure argon cluster is obtained at 11.5 bar. Figure 3 shows electron yields as a function of stagnation pressure, not sizes. To establish the effect of doping in a particular phenomenon, it is essential to compare the X-ray emission or high-energy electron emission from the clusters of the same size. Only under these circumstances, we can answer the question raised above. In addition, we could also throw light on the question as to what happens when the cluster is doped with a material having higher ionization energy (IP) than the base material constituting the cluster. It could also help in explaining the experimental observations as depicted in figure 2 where a significant drop in the electron yields in doped Ar-CS_2 cluster is observed compared to pure CS_2.

To address these issues, the integrated yields of electron and X-ray emission are sorted according to the respective cluster sizes of both pure and doped clusters. A tabulated compilation is presented in table 1 for X-ray emission (energy range 2.6–3 keV) sorted out for different sizes of Ar-CS_2 and Ar and compared with pure Ar clusters.

First column of table 1 shows the cluster size. The second and third columns represent the stagnation pressures of pure argon and doped Ar-CS_2, corresponding to the respective cluster sizes presented in column one. The proportion of CS_2 in each doped Ar-CS_2 cluster is shown in the bracket in column three. The fourth column gives the gain for doped Ar-CS_2 cluster with respect to pure argon cluster of the same size. The table shows that for a given cluster size the gain in the yields of these photons is more than an order of magnitude at the cluster size of 4300.

Table 1. Gain in X-rays of energy range 2.6–3 keV in doped clusters.

Cluster size	Stagnation pressure (Ar)	Stagnation pressure (Ar-CS_2)	Gain factor
4300	7.7	2 (95)	15
11000	11.5	4 (90)	5.8
15000	13.1	6 (84)	4.3
17000	13.8	8 (72)	3.6
21000	15.1	10 (52)	2.8

The huge enhancement in the doped environment shows potential of such targets to be used as tabletop X-ray sources.

A close look at the third column in table 1 would show that the fraction of argon in doped Ar-CS_2 cluster is 5% and 10% for cluster sizes of 4300 and 11,000 respectively. Therefore, these clusters can be treated as CS_2 clusters doped with argon. In this case, the dopant (argon) has ionization energy more than the base material. In other words, we can address the question of how different the nanoplasma is if the cluster is doped with a material having higher ionization energy. The X-ray emission measurements in Ar-CS_2 cluster target compared to that in pure CS_2 clusters show a reduction in the X-ray emission upon doping with argon. Recalling figure 2, where a significant drop in the electron yields in doped Ar-CS_2 cluster is observed compared to pure CS_2, we computed and compared electron emission yields as a function of cluster sizes. In Ar-CS_2 clusters of size 4300 (5% argon) the total flux of electron dropped by about 51% when compared to pure CS_2 clusters of the same size whereas in another case, when the doped Ar-CS_2 cluster size was kept 11,000 (10% argon), the reduction in the electron flux increased to 55%. So we do see a suppression in electron emission in a mixed cluster with a dopant of higher ionization energy.

4. Conclusion

We aimed to generalize the doping phenomena in the cluster-based targets for laser-produced nanoplasmas. It is experimentally observed that when a pristine cluster is doped with a material of low IP, the dynamics of the nanoplasma (formed upon interaction with intense laser fields) is modified in such a way that electron and X-ray yields are drastically enhanced. These results show that the phenomenon observed with argon–water clusters [8–10] are much more general and are applicable for any dopant. In contrast, when a CS_2 cluster is doped with a dopant (argon) that has higher ionization energy (5–10% of Ar) the electron yield is reduced by about 50%. A general method for characterizing such clusters is also presented. The reported results could help in developing doping as a tool to control the dynamic evolution of nanoplasma and thus, eventually, developing laser-based tabletop accelerators.

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