Heliyon 8 (2022) e10621

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

Investigation of ion dynamics of laser ablated single and colliding carbon plasmas using Faraday cup

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ARTICLE INFO

Keywords:
Faraday cup
Laser-produced plasma
Colliding plasma
Carbon plasma

ABSTRACT

We report a comparative study of a single plasma and a colliding laser produced plasma, investigated using a Faraday cup. An enhancement in ion emission and stagnation is observed in colliding plasma plume compared to single plasma plume. We observed that fast ion generation in laser ablated plasma can be achieved at large laser intensity on to the target. As laser intensity increases ionic yield increases for both colliding and single plume and at a fixed laser intensity ionic yield decreases with increase in ambient pressure. The double peak structure is observed in the ion signal at large fluence where the peaks correspond to fast and slow species. A Faraday cup composed of nine collectors is used to measure the spatial/angular distribution of ion of expanding plasma plume. Ionic yield is found to be larger in the colliding plasma plume than the single plasma plume at all spatial/angular positions.

1. Introduction

Laser produced plasmas have been the topic of significant interest for a long time because of its applications in the field of nanoparticle synthesis, thin film deposition, harmonic generation, as a source of neutral and ions beams, etc. [1, 2, 3, 4, 5]. Ions extracted from the laser plasma have important applications in medical physics, lithography, ion injectors and in thin film deposition [1, 2, 3, 4, 5, 6, 7]. There have been reports of using carbon ion beam therapy, as heavy carbon ion deposits more energy to tumour tissue and is more effective in destroying the tumour tissue compared to protons [7]. Recently the scientific community have shown their interest towards the interaction of two plasma plumes known as colliding plumes [3, 8, 9, 10, 11, 12, 13, 14, 15] Colliding plasma plume is of particular interest as compared to single plume as it has found its potential applications in droplet free thin film deposition, getting an ionic beam (since stagnation evolution is dominantly single species), inertial confinement fusion, and approximations to astrophysical plasma [8, 14, 15, 16]. Various target configurations have been used for the study of colliding plasma plumes e.g. orthogonal, flat target geometry [17, 18]. In each configuration the colliding plumes are produced by focussing two laser beams at two different points on the target surface. M. Favre et al. have used orthogonal geometry to study the dynamics of colliding carbon plasma plume [8]. In their work they have demonstrated the effect of background pressure in the post collision dynamics [8]. Rumsby et al. were first to investigate the colliding carbon plasma in flat target geometry [19]. They concluded that observed luminosity in the interaction region depends on the collisional process [19]. Whenever collisional ionization dominates over recombination process it leads to enhancement of the observed luminosity [19]. We have used the flat target geometry for the study of colliding plumes. Flat target geometry has some advantage over others because in this geometry the seed plume expands collinearly normal to the target surface and lateral (slower) component of velocity of these plume causes the inter penetration and longitudinal component of velocity gives net physical movement to colliding species. In this geometry we can optimize the collisional process and luminosity of colliding region by just varying the inter plume distance [3]. If we keep two plumes very close, then denser region of two plume will interpenetrate and causes rapid collisional ionization which leads to an enhancement in the observed luminosity and deceleration of ions leading to high degree of stagnation [10, 19]. However at large inter plume distance low dense region of seed plumes will penetrate and mainly electron -electron or ion-electron interaction will dominate [19, 20, 21]. Whether the stagnation or interpenetration will result is determined by the so called collisionality parameter (ζ) [Eq.(1)] introduced by Rambo et al. [10, 21, 22].

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https://doi.org/10.1016/j.heliyon.2022.e10621
Received 26 July 2022; Received in revised form 3 August 2022; Accepted 8 September 2022

$$\zeta = \frac{P_d}{\lambda_{ii}} \quad (1)$$

here P_d is inter-plume distance and λ_{ii} the ion-ion mean free path.

The ion-ion mean free path [Eq.(2)] is given by [10, 21, 22].

$$\lambda_{ii} = \frac{m_i^2 \upsilon_{12}^4}{4\pi e^4 Z^4 n_i \ln \Lambda_{12}} \quad (2)$$

where m_i υ_{12} Z n_i, $\ln \Lambda_{12}$ are mass of ion, plume collision velocity, ion state, ion density and coulomb logarithm. Thus in the collinear colliding plumes, the properties/dynamics of colliding region can be effectively tuned by changing the inter-plume distance and laser fluence [10, 21].

In order to achieve control over laser plasma-based applications, one must have control over the plasma properties and its dynamics. It necessitates the characterization of laser produced plasma [23, 24, 25, 26]. There are many diagnostic tools that can be used to study the laser plasma viz. interferometry, optical emission spectroscopy (OES), 2D imaging, ion probe etc. [3, 23, 27, 28, 29, 30]. There are several limitations of each diagnostic technique depending upon the experimental conditions. For example, refractive index sensitive optical diagnostics namely interferometry and shadowgraphy are used to have information about the plasma plume at higher ambient pressure only. Similarly OES/2D imaging techniques which gives the information of plasma species in their excited state cannot be used at larger distance from the target surface as plasma emissions become faint there and hence could not be imaged [3, 21]. However, the plasma species have sufficient thermal kinetic energy to travel up to larger distances. To see the dynamics/behaviour of plasma species at larger distance aiming toward its application for ion beam and thin film deposition, electric probe can be used. Electrical probe is commonly used to measure the kinetic energy of ions, most probable velocity, temperature, and ion current density [31]. It is essentially metallic plate/wire at some potential inserted inside the plasma. Langmuir probe and Faraday cup are largely used for diagnosing the laser ablated plasmas for the behaviour of ions. We have used Faraday cup which consists of a collector plate associated with a system of grids at different potential [28]. Typically, the ion signal obtained from Faraday cup shows the shifted-Maxwellian distribution but in this work, we have shown that as one changes the laser fluence on to the target there is significant change in ion dynamics [14, 31]. At very large fluence more energetic ions are formed and ion signal traced by Faraday cup shows deviation from shifted Maxwellian distribution. In expanding plasma plume uniform spatial distribution of ions are required for getting homogeneous thin films. Therefore, to get the spatial distribution of ions we have used an array of Faraday cup composed of nine collectors. In the present work we have comparatively studied the plume dynamics and behaviour of ions/electrons of colliding carbon plasma and single carbon plasma at various laser fluence and ambient pressures using a Faraday cup. This experiment provides the valuable information about the effect of background pressure and laser fluence on the ionic yield.

2. Experimental

A schematic diagram of the experimental setup is shown in Figure 1. A flat graphite target was placed inside the vacuum chamber on a rotating mount. The vacuum chamber was evacuated below the pressure of 10^{-6} mbar and then the chamber was filled with nitrogen gas using a regulator in a controlled manner to obtain the desired pressure conditions. An infrared laser beam of 1064 nm wavelength and 8 ns pulse width from an Nd: YAG laser ((LAB-190-10 series from Spectra Physics) which emits maximum energy up to 1 J per pulse was used for ablating the graphite target. To produce the colliding plasma plume, we have used the wedge prism to divide the incoming laser beam into two equal half energy beam and then focussed it at two different points on the target which produces the two seed plumes. These seed plumes at later stage of evolution collides with each other and form the colliding plasma. The inter plume distance given by $P_d \approx f\gamma(\mu - 1)$ was ~2 mm throughout the experiment where f is focal length(35 cm) and γ is wedge angle ($45'$) and μ is refractive index (1.45) [21,32]. The single plasma plume was produced by just blocking one of the split components produced by wedge prism. This experiment was performed by operating the laser in single shot mode. For fluence scan laser energy per pulse was varied from 50 mJ to 500 mJ using abeam attenuator (Topag). A Faraday cup was inserted inside the vacuum chamber to collect the ions from the expanding plasma. The Faraday cup assembly used in this experiment consists of hollow brass cylinder having a metal collector plates and system of grids. The collectors and grids were separated by nylon spacers. A hollow brass rod was attached on the back of Faraday cup to move it spatially inside the chamber. Optimum biasing voltage was applied on collectors/grids using a power supply (Hamamatsu, C-3350). An oscilloscope was used to record the collector signal of the Faraday cup. The oscilloscope was interfaced with the computer for further analysis of stored data.

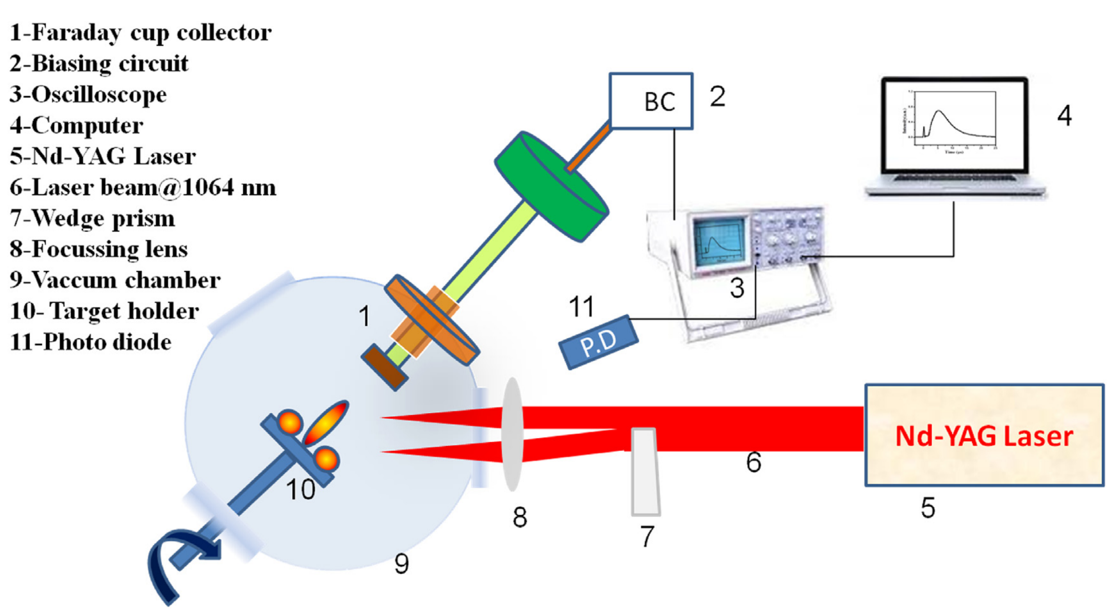

Figure 1. Experimental setup.

3. Result and discussions

When a laser pulse having sufficiently high energy is focused on the target surface this initiates vaporization and eventually to plasma formation which consists of electrons, atoms, molecules, ions, cluster, nanoparticles, and agglomerates [2, 33]. Laser produce plasma species moves away from the target as their thermal energy rapidly converts into kinetic energy [34, 35, 36, 37, 38]. Since plasma comprises of ions, electrons, and neutral species therefore to get the time-of-flight signal of ion flux only, electrons must be prevented to reach on the collector surface. This could be accomplished by maintaining the optimum potentials on the collector/grids. Figure 2a shows the variation of collector signal obtained under the highest laser fluence (70 J/cm^2) in the experiment at various collector bias voltages in vacuum environment of 10^{-5} mbar. A negative collector signal even at zero collector voltage shows that the effective number density of electrons is higher than the ion number density at collector surface. This may be due to the high mobility of electrons and due to significant loss of ion charges due to recombination with ambient and other species. Figure 2b follows the variation of peak of ion signal with respect to collector voltage. This

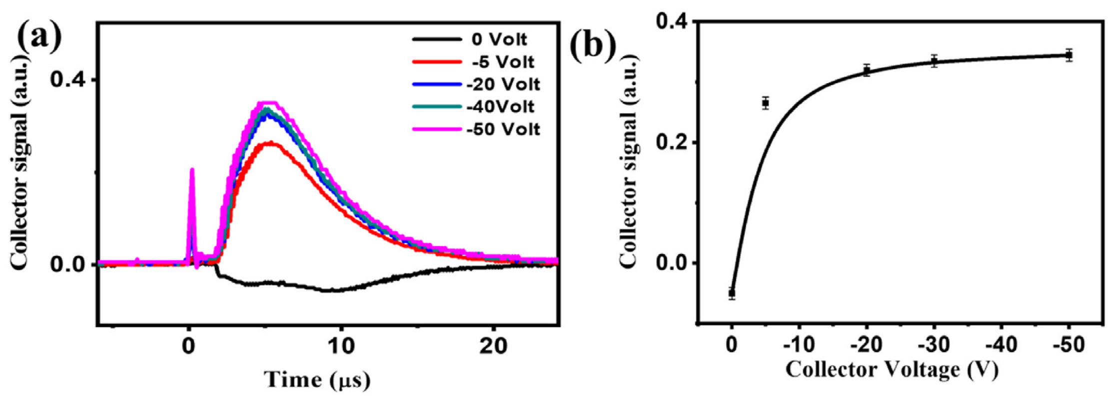

Figure 2. (a) The ion trace of Faraday cup at different collector bias voltage at 10^{-5} mbar ambient pressure. **(b)** Variation of peak collector signal with voltage applied on the collector.

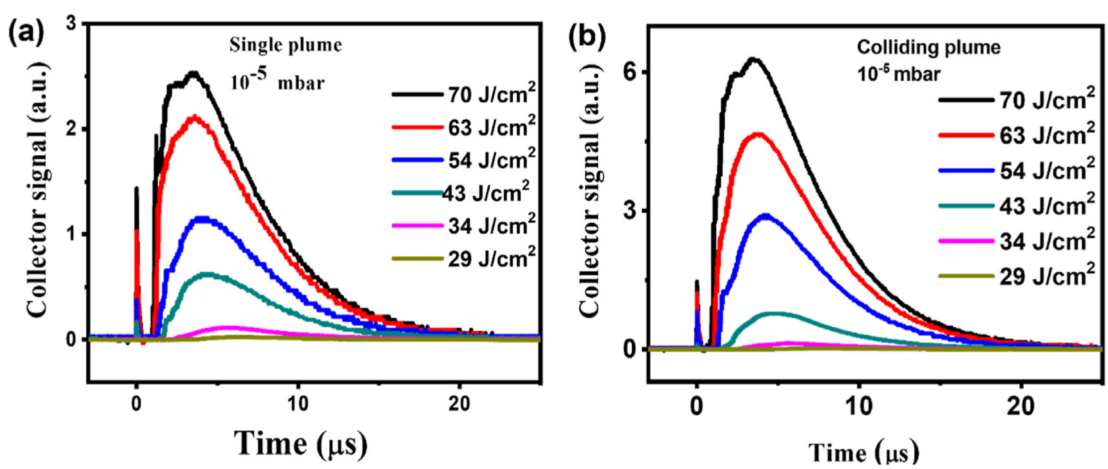

Figure 3. Time of flight trace of ion flux at different laser fluence in vacuum for (a) Single plume (b) Colliding plume.

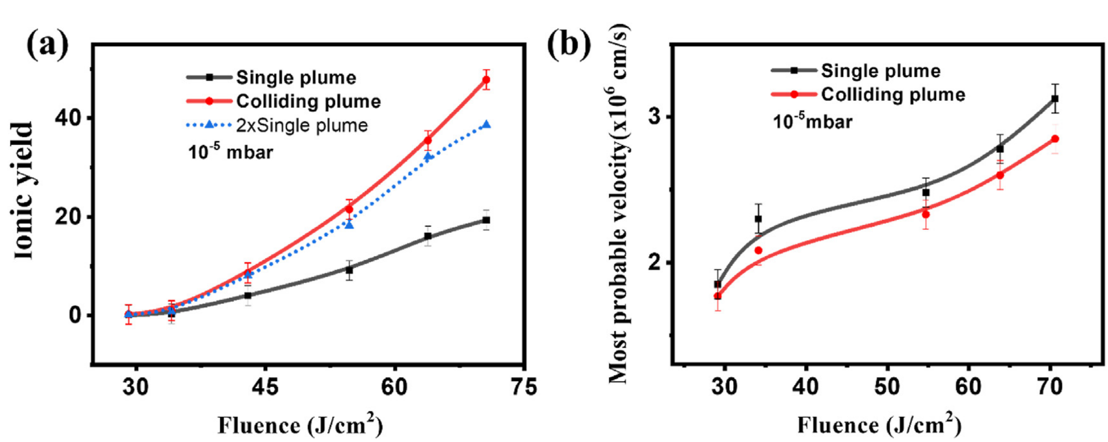

Figure 4. Variation of (a) ionic yield and (b) most probable velocity of single plume and colliding with laser fluence in the vacuum environment of 10^{-5} mbar.

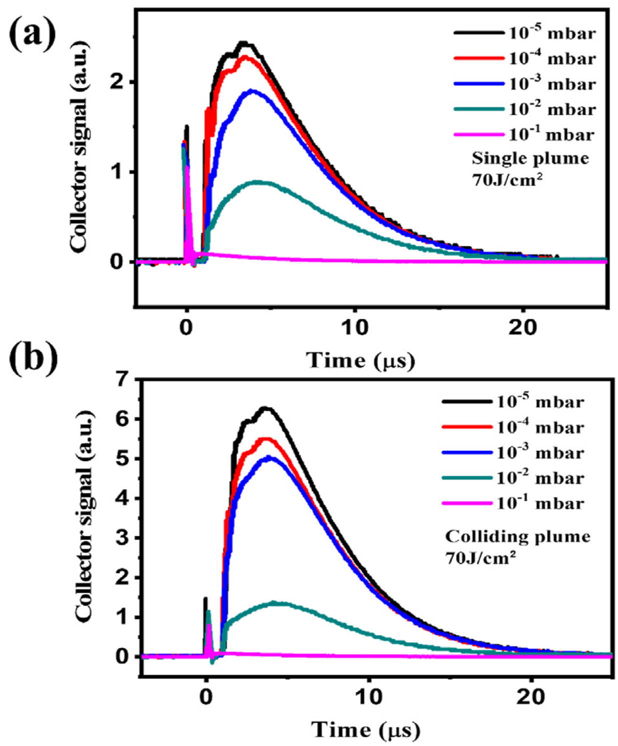

Figure 5. (a) Time of flight trace of ion flux of single plume at different ambient pressure. **(b)** Time of flight trace of ion flux of colliding plume at different pressure.

shows that with increase in collector voltage the ion signal increases positively and become almost constant after a particular negative voltage. This is mainly due to suppression of electrons caused by increasing negative potential field of collector which opposes the electrons to reach on it. Therefore, the rest of the study was done at -50 V which is sufficient to reject the electron flux coming from the plasma. Figures 3a and 3b shows the time of flight (TOF) distribution of ion signal at different laser fluence of single and colliding plume respectively at 10^{-5} mbar ambient pressure. TOF trace shows a prompt photo electron peak followed by the ion signal. The photo electron peak is attributed to emission of high energy UV radiations in plasma which reaches to the collector plate just after interaction of laser pulse with the target. At collector plate these high energy photon beams causes the emission of electrons mainly due to the photo electric effect which results in net increase of positive charges at collector surface which appears as a photo electron peak in ion signal traced by oscilloscope. Both in single plume

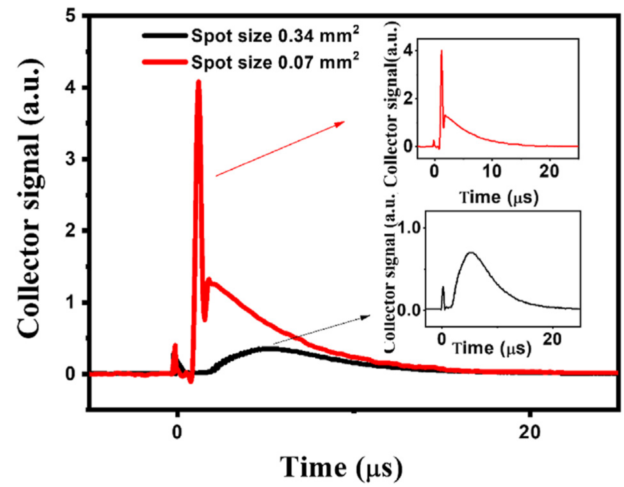

Figure 7. Effect of focussing on time-of-flight trace of ionic yield.

and colliding plume ion signal increases with increase in fluence and peak shifts towards the left side (Figure 3). Observed shift in peak position is attributed to growth in thermal kinetic energy of ions with increase in laser fluence. The two peak structure in time of flight signals of ions at ~70 J/cm^2 is attributed to different expansion velocity of lighter and heavier plasma species [3]. The total number of ions arriving at the collector surface is known as ionic yield. Ionic yield, most probable velocity, and thermal kinetic energy can be calculated with time of flight trace of ions [29]. Ionic yield is calculated by integrating the time-of-flight curve with respect to time. Figures 4a and 4b shows the variation of ionic yield and most probable velocity of both single and colliding plume with increase in fluence. This shows that the ionization and thermal kinetic energy of ions increases with increase in fluence. This is attributed to the physical processes involved in the ablation. In laser ablation process a short laser pulse of high energy is focussed on the target to keep the fluence of the focussed beam greater than ablation threshold of the target material. Ablation threshold of a solid target is given by $F_{th} \approx H_v \rho \sqrt{Dt_p}$, where H_v, ρ , D are heat of evaporation, density, thermal diffusivity of the solid target material respectively, and t_p is laser pulse width [33, 34, 36]. The calculated ablation threshold of graphite is approximately ~30 J/cm^2 where we have used H_v = 38.4 kJ/g, ρ~2.260 g/cm^3, $D \sim 1.57 \times 10^{-3}$ m^2/s and $t_p \sim 8$ *ns* for calculations. This laser beam penetrates the surface layers of the target and rapid heating of the target material takes place resulting in surface evaporation. As we increase the fluence more evaporation takes place. This enhances the collision between ablated species which results in enhancement of ionic signal as shown in Figure 3. We can see from Figure 4a that as compared

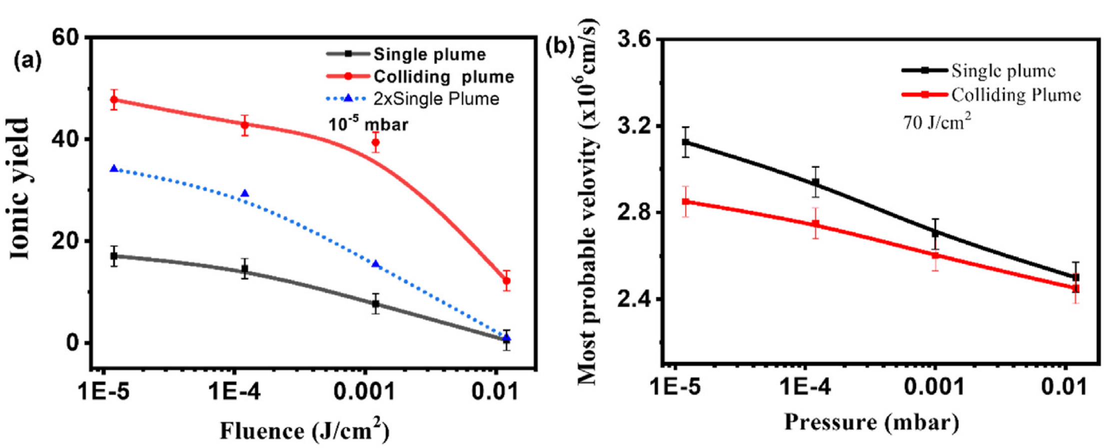

Figure 6. Variation of **(a)** ionic yield and **(b)**most probable velocity of single and plume colliding plume at different pressure.

to single plasma plume ionic yield is 2–3 times larger in case of colliding plasma plume. The ionic yield increases more rapidly with fluence in case of colliding plume as compared to single plume. We can conclude that colliding plume is more efficient ion source at high fluence as compared to single plume. We can further see from Figure 4b that most probable velocity also increases with increase influence for both single and colliding carbon plasma plume. Most probable velocity (v_p) is found to be of the same order (~2–6x10^6 cm/s) but greater in case of single plasma plume compared to colliding plasma plume. This is attributed to retardation of ion species due to the drag forces imposed by the plasma species of seed plume. This is also indicative of stagnation of ions in colliding region. The dynamics of plasma plumes are strongly influenced by the ambient gas pressure. Figure 5a and Figure 5b shows the variation in time of flight distribution of ions of single plasma plume and colliding plasma plume, respectively with pressure. Ion signal reaching at collector decreases with increase in pressure for both single and colliding plasma plume. Variation of ionic yield and most probable velocity of single plasma plume and colliding plasma plume at different pressure is also shown in Figure 6. We can also see from Figure 6a that ionic yield is again 2-3 times larger in colliding plume compared to single plume at all ambient pressure showing the importance of colliding ion source. Most probable velocity of ions in colliding plume is observed lower than that of single plume at each pressure as shown in Figure 6b which is attributed to retardation caused by ion-ion collisions of two seed plumes. We see that expansion of plasma and hence ionic species depends upon the ambient gas and its pressure [11, 39, 40]. Plume expansion in different pressure regime is characterized by the plume length. In vacuum free expansions of plasma species takes place and plume length varies almost linearly with time. Therefore at sufficiently low pressure a maximum ions reach to collector plate in very short time and TOF trace shows the peak at earlier times as compared to other pressure. At moderately high pressure the plasma plume expansion is limited by the ambient pressure [38]. With increase in pressure the ambient gas density increases inside the chamber which leads to increased collision between ions and the ambient species. In such case of highly confined plasma, collisional processes dominate and mean free path decreases. The frequent collision between them results in loss of thermal kinetic energy of ions and hence forward movement of plasma species decreases with increase in pressure which enhances the recombination process. At high nitrogen ambient pressure abundance of slow-moving clusters, atomic/molecular species are found compared to ionic species which leads to decrease in ionic yield [27].

A very distinct behaviour in ion dynamics was observed when the laser was focussed to the spot size of 0.07 mm^2 compared to the 0.34 mm^2 as shown in Figure 7. This distinct behaviour is attributed to the change influence with slight variation in focussing conditions. The laser pulse derived from Nd: YAG laser have the Gaussian beam profile. When we focus the laser pulse on the target material the beam spot size of focused beam at the target surface depends upon the lens to target distance. As we focus the laser pulse using lens the beam radius decreases along the propagation direction and at some point, its radius becomes minimum and after that again beam radius increases. Therefore, the laser fluence will be maximum when target is placed at beam waist, and it decreases on changing the target position on either side of the beam waist. The depth of focus is the range over which the intensity of focussed beam remains same. In our case the calculated depth of focus is approximately 1 cm. Therefore, any change larger than 1 cm in distance from lens to target cause a significant change in spot size of the beam on to the target surface and hence the fluence changes. Therefore, one can change the fluence not only by varying the pulse energy but by changing the focussing conditions. We see that when the laser was tightly focussed, the time-of-flight distribution of ions exhibits the formation of a fast ion peak followed by slow (thermal) ion peak. The fast ion peak completely disappeared in loose focussing. The formation of double peak structure at high laser fluence is associated with the generation of fast ions [41]. This

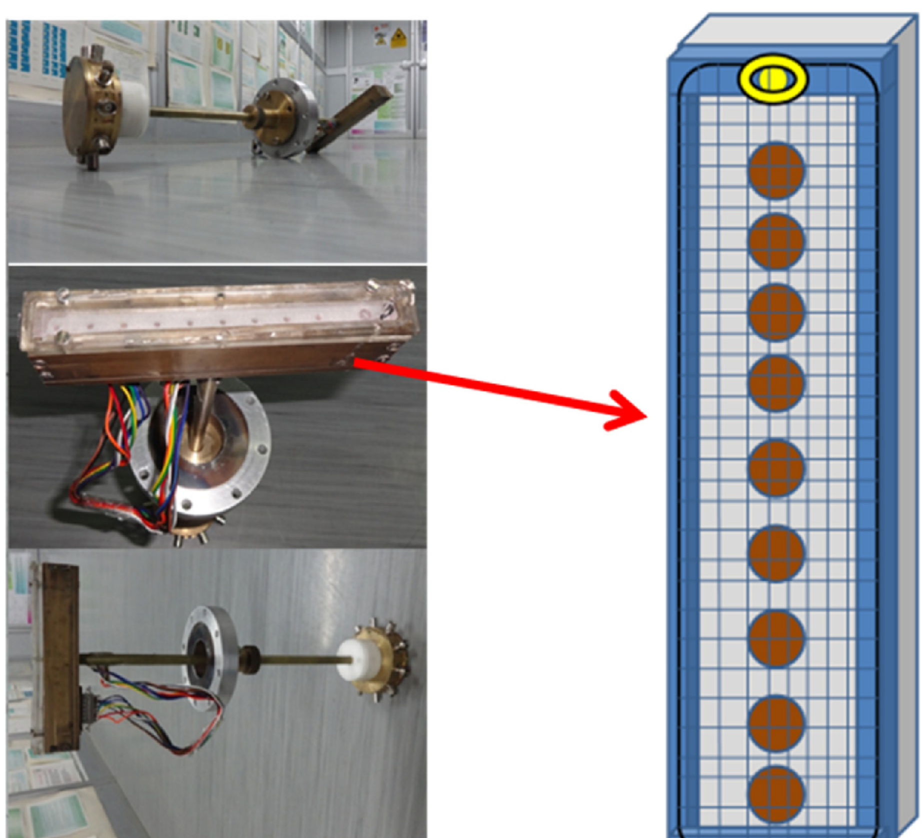

Figure 8. Photo of composite Faraday cups used for characterization of spatial/angular distribution of ions.

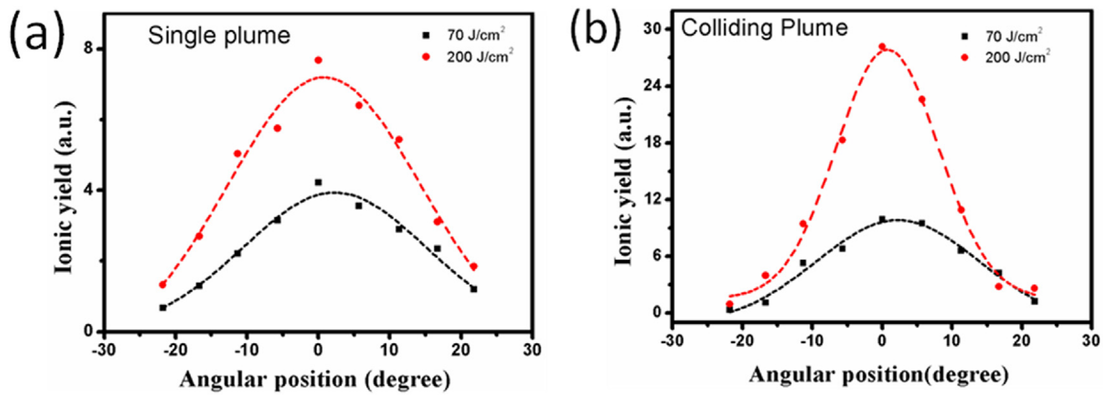

Figure 9. : Distribution of ions making different angles with respect to axis of plasma expansion in **(a)** single plume **(b)** colliding plume in vacuum environment of order of 10^{-5} mbar.

is because when a laser pulse hits the target surface, only a small portion of the pulse is used for vaporization and formation of primary plasma. This primary plasma further interacts with the remaining part of the pulse and due to absorption of photons by inverse Bremsstrahlung (IB) process heating of the plasma takes place [37]. In the IB process, electrons gain energy under the influence of oscillating electric field of laser pulse and at the same time it transfers its energy to other plasma species by making collisional excitation. At low laser fluence larger fraction of the laser pulse is consumed in target heating and only small portion is available for heating of primary plasma. Whereas at larger fluence only small portion of laser pulse is used in target heating and larger portion of the pulse is used for heating through IB process. In IB process electrons gain enough energy to escape from core plasma boundary and creates an electrostatic field [34]. This electrostatic field accelerates the ions of the core plasma which appear as a fast ion peak in time-of-flight trace of Faraday cup [41].

In order to measure the spatial/angular distribution of ions of expanding plasma plume, we have also designed an electric probe (array of Faraday cup) composed of nine collectors (4mm in diameter) fixed on a Teflon slab at 1 cm distance as shown in Figure 8. A hollow brass rod was brazed on the rear end to move it spatially. It was inserted inside the vacuum chamber using a Wilson seal. This array of Faraday cup was mounted vertically inside the vacuum chamber at 10 cm away in front of the target surface. Figure 9a and Figure 9b shows the angular distribution of ions in single plume and colliding plume, respectively at two different laser fluence. Ionic yield is found larger in colliding plume compared to single plume at all angular positions. With increase in fluence from 70 J/cm^2 to 200 J/cm^2 ion yield increases for both single and colliding plume at all angular position. From this figure we can conclude that plasma plume expansion is highly forward peaked in colliding plume with maximum of ion concentration along the axis of plasma expansion making it a more suitable ion source.

4. Conclusion

We have compared here the TOF distributions of ions of single plume and colliding plume at different pressures and laser fluences using a Faraday cup. The ionic yield in colliding plume is found almost 2–3 times larger than the single plume irrespective of the pressure. This shows that colliding plasma is more efficient ion source. Larger time span of TOF signal of colliding plume confirms the stagnation of plasma. A significant decrease in ionic yield takes place with the increase in ambient pressure. These results confirm that colliding plume is a better ion source with large ionic yield compared to single plume at high laser fluence and at all ambient pressure. TheTOF distribution of ions traced by the Faraday cup depends strongly on the laser fluence and focussing conditions. A remarkable change in time-of-flight trace of ions is observed when laser fluence is varied by changing lens to target distance. Fast ion formation is noticed from double peak structure of ion signal at high laser fluence. These study shows that laser ablated plasma at larger fluence can be used as possible source of fast ions for practical applications. The composite Faraday cup gives the spatial distribution of ions. These results confirms that as the fluence increases the larger fraction of ions expand normal to the target surfaces. These results lead to the better understanding of laser ablated plasma in terms of plasma constituents and behaviour of ions/electrons.

Declarations

Author contribution statement

Ravi Pratap Singh: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

D.N.Patel, R.K.Thareja: Contributed reagents, materials, analysis tools or data; Wrote the paper.

Funding statement

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Data availability statement

Data will be made available on request.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

Acknowledgements

The authors would like to thank IIT Kanpur for providing the facilities and workspace.

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