Observation of non-symmetric side-scattering during high-intensity laser-plasma interactions

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Abstract

Non-symmetric side-scattering has been observed during the interaction between a high-intensity laser pulse and under-dense argon plasma. The angle between the laser’s forward direction and the scattered radiation is found to decrease for increasing electron densities ranging from 0.01 to 0.25\textit{n}\textsubscript{c}, where \textit{n}\textsubscript{c} is the critical density for the laser wavelength. We show that the observed features of the scattering cannot be described by Raman side-scattering but can be explained to be a consequence of the non-uniform density distribution of the plasma with the scattering angle being oriented along the direction of the resulting electron density gradient.

Particle beams, depending on their type and energy, find their applications in a wide variety of fields, such as nuclear fusion, cancer treatment, material structural probing and subatomic physics. Therefore, particle accelerators play a significant role in the generation of these particle pulses with the required parameters. Charged particle pulses are traditionally accelerated by applying an alternating electromagnetic field inside a solid structure, whose size varies depending on the required particle-energy \cite{1}. Relativistic plasmas, generated during the interaction of high-intensity laser pulses with matter, offer an alternative way to realize a potentially more compact accelerator, in which particles can be accelerated by the electric-field distributions inside a plasma structure reaching amplitudes in excess of several TV/m. Plasmas in different regimes of electron-density, namely underdense (\textit{n}\textsubscript{e} \ll \textit{n}\textsubscript{c} = \varepsilon\textsubscript{0}\textit{m}\textsubscript{e}\omega\textsubscript{p}^2/\textit{e}^2, where \textit{n}\textsubscript{e} is the critical density, \varepsilon\textsubscript{0} is the vacuum permittivity, \textit{m}\textsubscript{e} is the electron mass, \omega\textsubscript{p} is the laser’s central frequency and \textit{e} is the electron’s charge) and overdense (\textit{n}\textsubscript{e} \gg \textit{n}\textsubscript{c}) have been widely utilised to study electron \cite{3} and ion acceleration \cite{4} processes, respectively.

During the last decade, the interaction dynamics between an ultrashort laser pulse (\textit{\tau}\textsubscript{\text{p}} < 35 fs) and near-critical plasma (0.1\textit{n}\textsubscript{c} < \textit{n}\textsubscript{e} < \textit{n}\textsubscript{c}) have also been extensively investigated both in experiments and with the help of numerical simulations. These dynamics have been found to be uniquely characterized by phenomena, such as resonant laser absorption \cite{5}, relativistic self-focusing \cite{6}, direct laser acceleration (DLA) \cite{7}, hole boring \cite{8}, soliton formation \cite{9}, vortex formation \cite{10}, laser pulse collapse \cite{11} and gamma ray generation \cite{12}. Of particular interest is DLA, whereby a single electron filament is formed and sustained, within which electrons can be accelerated through a resonant interaction with the laser’s electric field to several hundred MeV. In a near-critical plasma, the energy deposition by the laser pulse in the plasma can be expected to be higher than that of in underdense plasma, as the plasma frequency approaches the laser frequency leading to more resonant energy absorption.

Although several experiments have been carried out in this near-critical regime \cite{11,13,14}, the mechanism leading to the acceleration of electrons still remains elusive. The reason for this could be due to a still imperfect understanding of the interaction dynamics between the near-critical plasma and the driving laser, and also the kind of instabilities that could potentially arise during such an interaction. Such phenomena could be studied in a pump-probe geometry, where a synchronized optical probe pulse backlights the interaction, which is being driven by the (pump) main pulse. Past experiments using foam targets have focused primarily on investigating the dependence of the maximum energy of the accelerated ions on the target’s thickness and density \cite{13,14}. However, because of the nature of foam targets, it is difficult to probe such an interaction \cite{15}. In an another
experiment carried out using a gas target, the phenomenon of laser pulse collapse (strong and almost complete absorption of the laser energy) was observed using optical diagnostics but the experiment could not reveal more details about the dynamics of the laser–plasma interaction \[\text{[11]}\]. Thus, in order to shed more light into the interaction dynamics and in an attempt to explore the DLA mechanism, an experiment was carried out at the JETI-40 laser system at the University of Jena, Germany, in which the phenomenon of side-scattering of electromagnetic radiation has been studied in detail. Such a scattering process has been previously reported \[\text{[16]}\], where it has been attributed to Raman scattering. Here, we show that under our experimental conditions, these observations cannot be explained by Raman side scattering but by another mechanism that we propose here. This mechanism is based on the deflection of the laser light in a non-uniform plasma density distribution.

1. Set-up

Laser pulses from the JETI-40 laser system with 800 nm central wavelength and 30 fs (FWHM) pulse duration were focused using an \(/6\) off-axis parabola (as shown in figure 1(a)) into an argon gas-jet whose backing pressure could be varied from 5 to 80 bar. The energy distribution inside the focal spot shown in figure 1(b) was optimized using adaptive optics to reach a focal spot diameter of 6.6 \(\mu\)m in which 34\% of the total energy of 550 mJ (i.e. \(\sim 210 \text{ mJ}\)) was contained, leading to a peak intensity of \(4.4 \times 10^{19} \text{ W cm}^{-2}\). Under these conditions, it was possible to reach an ionization level of up to Ar\(^{16+}\) via over the barrier ionization \[\text{[17]}\], allowing the critical density \((n_c)\) of \(1.72 \times 10^{21} \text{ cm}^{-3}\) to be reached \[\text{[18]}\]. The laser–plasma interaction region was imaged onto a CCD with a magnification of 5 reaching a resolution of 1.3 \(\mu\)m px\(^{-1}\). The interaction region was backlighted using a frequency doubled, 400 nm wavelength probe beam of 30 fs duration, which was synchronized to the main beam and propagating under an angle of 90\(^\circ\) to the main pulse’s propagation direction. Any ions accelerated close to the main pulse’s direction could have been observed using the combination of a scintillator screen and a gateable CCD (G-CCD) covering an observation half-angle of 23\(^\circ\) (0.4 rad), which was followed by an electron beam profiler and a magnetic electron spectrometer, whose entire aperture was covering an opening half-angle of 0.05\(^\circ\) (900 \(\mu\)rad) in the laser’s forward direction capable of detecting electrons with kinetic energies \(>5 \text{ MeV}\). Although the scintillator screen, which has a rise time of 100 ps and decay time of 700 ps, was also sensitive to more energetic particles, the pico-second resolution of the G-CCD could be used to distinguish ions (\(<5 \text{ MeV}\)) from fast electrons and x-rays.

The gas nozzles used in this experiment have been characterized beforehand using a Mach–Zehnder interferometer. Figure 2(a) shows a typical interferogram obtained for a nozzle with 400 \(\mu\)m outlet-diameter. The fringe-shift in the interferogram is due to the phase-shift accumulated by a probe laser pulse when it propagates through the gas density in comparison to the propagation through vacuum. By assuming cylindrical symmetry with respect to the gas jet’s axis of symmetry, this fringe-shift can be directly related to the gas-jet’s density distribution.

2. Measurements

The experiment was conducted for backing pressures ranging from 5 to 80 bar and it must be pointed out that there were neither energetic electrons \(>5 \text{ MeV}\) nor ions \(<5 \text{ MeV}\) to be observed in the laser’s forward direction corresponding to the energy ranges of their respective diagnostics. The absence of energetic electrons also makes
the accelerating ions to kinetic energies $\geq 5$ MeV highly unlikely. Figure 3(a) shows a typical interferogram obtained using the side-view diagnostic showing the laser-plasma interaction region, where the laser pulse coming from the left was focused to the center of the gas-jet at a height of 500 $\mu$m above the nozzle’s surface. It shows filamentary structures originating from the laser axis pointing preferentially in the upward direction. These structures are most likely caused by scattered laser light, which is intense enough to modify the plasma’s refractive index along its propagation path, most likely through ionization. The side-scattering is observed not only to originate from the outer edges of the gas-jet, where the gas density measurements exhibit a complicated density profile [19], but also in the central part of the gas-jet, where the gas density increases in a much smoother fashion.

Figure 3(a) also shows the strong intensity modulations caused by the plasma, which renders the interferograms not analysable. Therefore, the corresponding electron-density and its particular distribution along the main-pulse propagation axis needs to be estimated from the measured gas density distribution across the nozzle, the laser focusing geometry, and the intensity thresholds for the different argon ionization states. Here, the Argon atoms are expected to be ionized through the barrier ionization mechanism for which the threshold intensity $I$ can be related to argon’s respective ionization energy ($E_{io}$) and charge state ($Z$) as follows [17]: $I = 4 \times 10^{8}(E_{io}/eV)^{1/Z^2}$. The following ionization energies [18, 20] have been considered for the ionization states $\text{Ar}^{1+} - \text{Ar}^{14+}$: 15.8, 27.6, 40.7, 59.8, 75, 91, 124.3, 143.5, 422.4, 478.7, 539, 618.3, 686.1 and
755.5 eV, respectively leading to threshold intensities between $2.5 \times 10^{14}$ W cm$^{-2}$ (for Ar$^{+1}$) and $6.7 \times 10^{18}$ W cm$^{-2}$ (for Ar$^{+4}$).

Figure 3(b) shows the variation of the scattering angle ($\theta_s$) as a function of the electron-density estimated for the point of origin on the laser-axis. It can be seen that—as the laser pulse propagates towards regions of higher density—the scattering-angle $\theta_s$ decreases. Figure 3(c) shows this scattering angle $\theta_s$ measured for increasing gas pressures. Note that the distances at which the positions of the beginning and end of the scattering process on the laser-axis differ for different backing pressures. At distances over which the side-scattering occurs (300–700 μm in front of the focal plane in vacuum), the laser intensity varies between 1–6 $\times 10^{17}$ W cm$^{-2}$, as estimated on the basis of a focused Gaussian beam propagating in vacuum. This corresponds to $(\gamma) = [1 + a_{0}^2/2]^{1/2} = 1.03–1.13$, which is the time averaged relativistic gamma factor associated with the electrons’ motion in the laser field and $a_0 = (I/W \text{ cm}^{-2})(\lambda_0/\mu \text{m})^2/1.37 \times 10^{18}$ is the normalized vector potential of the main laser-pulse. This means that relativistic self-focusing due to electron mass increase and ponderomotive electron expulsion is just beginning to play a role. Both focussing effects tend to compensate the effect of ionization induced defocusing. For our further analysis, we therefore assume that the resulting electron-density distribution can be estimated by combining the measured gas density distributions from the gas-jet characterization (shown in figure 2(b)) with the degree of ionization of Ar corresponding to the peak laser-intensity along its axis. Although the actual electron density might vary depending on the extent of the interplay among the various focusing and defocusing effects, our estimation nevertheless helps to shed light into the phenomenon of side-scattering, which we have observed for Ar backing-pressures up to 40 bar.

Thus, if the variation of $\theta_s$ is to be plotted against the estimated local electron density, as shown in figure 3(d), it can be seen that $\theta_s$ decreases as the density increases. However, it does not seem to depend on the absolute electron density at the point of origin of the scattering, as different scattering angles were measured for the same absolute electron density ($n_e$), which were achieved for different backing pressures. A possible explanation for this observation will be described in the next section. For increasing backing pressures ($\geq 20$ bar), it could be clearly seen that the scattering process undergoes a progressive transition from the upward direction to the downward direction as the laser pulse propagates towards the nozzle-center as shown in figure 4. For even higher backing-pressures (i.e. $> 40$ bar), side-scattering is strongly suppressed. However, at these higher pressures, we could observe strong filamentation in the forward direction, as shown in figure 4(c). The channels formed by the filamentation process can be distinguished from those caused by the side-scattering process since the former are directed along the propagation direction of the main laser pulse and originate from a relatively large transverse interaction region. Extrapolating the filamentation channels backwards does not lead to the central laser axis, in contrast to the channels (or stripes) caused by the side-scattering process, which indeed seem to originate from the high-intensity region on the laser axis. The dark regions in the images indicate both strong absorption and refraction of the probe-beam by the plasma out of the collection angle of the imaging system. Estimates of the electron density show that the scattering process dominates until $n_e = 0.25n_c$, while filamentation starts to occur at $n_e \sim 0.7n_c$.

3. Analysis

In an attempt to find the origin of this side-scattering process, two possible mechanisms, namely Raman scattering, and scattering off a periodic plasma density variation (an equivalent to a grating) were first considered.

Raman scattering results from the decay of a light-wave into a plasma-wave and another light-wave, where its phase-matching condition is given by $k_0 = k_p + k_s$, where $k_0$, $k_p$, and $k_s$ are the wave-vectors of the incoming light-wave, plasma-wave and scattered light-wave, respectively. Furthermore, energy must also be conserved for the process as described by $\omega_0 = \omega_p + \omega_s$, where $\omega_0$, $\omega_p$, and $\omega_s$ are the frequencies of the incoming light-wave, plasma-wave and scattered light-wave, respectively. Side-scattering could be understood as the result of the wave-vectors not being aligned collinearly, [21] as shown in figure 5(a).

As the laser propagates inside the plasma, it locally excites electron density oscillations along its path due to the ponderomotive force of the laser pulse [22]. This co-moving periodic density perturbation is also called a plasma-wave. If such a plasma-wave is to be viewed as a grating structure, as shown in figure 5(b), for a longitudinal spatial extent of the laser-pulse exceeding the plasma wavelength $\lambda_p$, side-scattering can be expected to occur at (1st order) diffraction angles $\theta_{sc}$ depending on the local plasma wavelength $\lambda_p$. The laser pulse, which in our experiments has a FWHM longitudinal extent of 9 μm, exceeds the plasma wavelength $\lambda_p$ for densities $n_e > 0.025n_c$, where $\lambda_p = 2\pi\eta_e/\omega_p$ with $\omega_p$ being the plasma frequency, $c$ being the speed of light in vacuum and the plasma’s refractive index $\eta = (1 - n_e/(\gamma))^1/2$.

Both Raman scattering and plasma grating mechanisms are, however, expected to result in symmetric scattering around the interaction region, meaning both upward and downward scattering should have been seen.
Figure 4. Interferogram obtained for (a) 20 bar (b) 40 bar backing pressures showing the progressive transition of the scattering-angle from upward to downward directions, and for (c) 60 bar backing pressure showing the filamentation process in the laser’s forward direction, where the side-scattering is strongly suppressed.

Figure 5. (a) Phase-matching condition for Raman side-scattering, (b) plasma-wave acting as a grating structure to diffract the incoming laser beam, and (c) comparison of the measured scattering angle with Raman scattering and plasma grating mechanisms.
This is in stark contrast to the scattering pattern that was observed. Furthermore, figure 5(c) shows the resulting scattering angle of both phenomena with respect to the electron density \(n_e\). Both Raman scattering (\(\theta_{Ra}\)) and 1st order plasma grating diffraction (\(\theta_{gr}\)) angles can respectively be related to \(n_e\) in the following way [21]

\[
\sin \theta_{Ra} = \left( \frac{4}{\gamma_e} \frac{n_e}{n_c} \right)^{1/4} \quad \cos \theta_{gr} = 1 - \left( \frac{1}{\gamma_e} \frac{n_e}{n_c} \right)^{1/2}.
\]

Figure 5(c) shows that both mechanisms exhibit an increase in scattering angle as \(n_e\) increases, which is also in clear contrast to the experimental observations. Higher-order Raman processes [23], which could be relevant at these relativistic intensities, would lead to a similar behaviour, but at larger angles. Therefore, we can conclude that neither of these two mechanisms can be the source of the scattering process.

We believe that the clues to the root of the scattering process lie in the non-uniform nature of the gas-jet’s density profile along the laser’s direction of propagation. The (radial) gas-jet density distributions that could be calculated from the phase-shift obtained from the interferogram (shown in figure 2) indicate the subsonic nature of the gas-jet, where the gas density continuously increases from the outer edges of the gas-jet towards the center resulting in a Gaussian-like distribution. Furthermore, these measurements indicate that there are small-scale local density fluctuations, which are likely either due to the limited amount of phase-shift accumulated by the portion of the laser-beam propagating through the outer regions of the gas-jet and/or the numerical error resulting from the line-by-line Abel inversion process with which the radial phase-shift is obtained from the integrated phase-shift present in the interferograms. Therefore, in order to avoid the influence of these high-frequency local density fluctuations on our overall analysis, the radial density distribution of the gas-jet for 20 bar and lower backing pressures has been modeled by a smooth distribution based on the actual density measurements, as shown in figure 6(a).

A close-up view of the interaction region of laser and gas-jet shown in figure 6(b) reveals the asymmetric nature of the transverse density distribution with respect to the laser axis. The arrows in figure 6(b) indicate the direction of the density gradient, which progressively changes its direction from positive (up) to negative (down) angles relative to the laser’s propagation axis. As the laser pulse enters the gas-jet, the density above the laser-axis is higher than below the laser-axis, then it passes through a region of uniform transverse density distribution after which the trend reverses as shown in figure 6(c). Thus, the resulting asymmetric plasma electron density distribution (shown in figure 6(d)) can be obtained by overlapping the symmetric laser focusing geometry and the asymmetric gas density distribution and calculating the ionization degree of argon. Figure 7(a) shows the scattering-angle measured for 20 bar and its corresponding electron-density gradient \(\nabla n_e\) along the laser-axis, which for positive angles yield a very good agreement. Whereas, at distances close to the nozzle-center, the electron-density estimations might considerably deviate from the experimental values, due to the interplay between the various focusing and defocusing effects that might cause changes in the laser’s intensity and thus in the local electron-density. This could explain the deviation of the direction of \(\nabla n_e\) from the measured values at negative angles. However, an overall agreement between the measured values and the plasma-gradient \(\nabla n_e\) suggests that the

![Image](image.png)
scattering originates from the central interaction region and propagates away along the direction of \( \nabla n_z \). This is also confirmed by measurements carried out for other backing pressures, as shown in figure 7(b).

Meanwhile, it is worth examining the effect of a transverse non-uniformity of the gas density on Raman scattering and plasma grating scattering. In the case of Raman scattering, the transverse non-uniformity might lead to asymmetric (instead of symmetric) Raman scattering, as the density non-uniformity might favor the misalignment of the wave vectors in the upward (or downward) direction compared to the downward (or upward) direction. However, the dependence of the scattering angle on the plasma density (i.e. that the scattering angle becomes smaller for increasing density) is not expected to change for a plasma that is not uniform in the transverse direction. Similarly, in the case of plasma grating scattering, the density non-uniformity is also not expected to influence the dependence of the scattering angle on the plasma density. Like in a grating structure, the diffraction angle primarily depends more on the spacing or the periodicity of the plasma wave than on its exact shape. Therefore, any transverse non-uniformity of the plasma, which might alter the exact (transverse) shape of the plasma wave structure, will not lead to a change in the expected scattering angle. Thus, neither of these mechanisms is able to explain our results even when the transverse non-uniformity of the plasma is considered.

Therefore, looking beyond these two scattering mechanisms, the non-symmetric side-scattering can be shown to be a consequence of the deviation in laser pulse propagation direction from the ideal laser-axis (straight line) due to the non-uniformity of the gas density and thus the electron density. The path undertaken by the central ray of laser light, which is schematically shown in figure 8(a) can be calculated by solving the eikonal equation [24] in an asymmetric electron density distribution \((n_e(x, z))\). Figures 8(b) and (c) show the corresponding extent of the light-ray’s deviation from the laser axis and its angle of deviation, respectively, for varying pressures. Thus, the process of side-scattering can be explained in the following way. As \( \theta_{ne} \) continues to decrease, in order for the phase matching condition to be satisfied between the incoming \((\vec{k}_i)\) and refracted light wave \((\vec{k}_r)\), an additional light wave component \((\vec{k}_d)\) can be expected to result with a positive angle as shown in figure 8(d), which can be interpreted as the scattering wave. As \( \theta_{nd} \) starts to increase, figure 8(e) suggests that the scattering can be expected to occur at negative angles. The local minima \( \theta_{nd} \) have been observed for varying backing pressures and could be verified to correspond to the interpolated zero-point of the simulated (and measured) scattering angles. Thus, the side-scattering observed in our experiments can be understood to be the result of momentum conservation as the laser is refracted away from its ideal propagation direction. While it is not necessary to have a scattering light wave to conserve the momentum in plasma, it can be produced as a consequence.

It is important to note that despite the wavenumber of the scattering wave \( k \), being relatively small, the dispersion relation for electromagnetic waves in plasma i.e. \( \omega^2 = k^2c^2 + \omega_p^2 \) indicates that the resulting frequency of the scattered wave \( \omega_s \) is still larger than the plasma frequency \( \omega_p \). Therefore, the scattered light will be able to propagate away from the central laser-plasma interaction region i.e. the plasma is slightly underdense for the scattered light wave.

This analysis can be complemented by the use of the perturbation method. In the expression for the wave-vector \((\vec{k})\) (in its non-relativistic limit) of the laser pulse propagating in a plasma (i.e. \( |\vec{k}| = 2\pi\eta/\lambda_0 = (2\pi/\lambda_0)(1 - n_e/n_c)^{1/2} \)), a perturbation in the electron density \((\delta n_e)\) can be introduced as follows

\[
|\vec{k}| - |\delta\vec{k}| = \frac{2\pi}{\lambda_0} \sqrt{1 - \frac{n_e + \delta n_e}{n_c}} = \frac{2\pi}{\lambda_0} \sqrt{\eta - \frac{\delta n_e}{2n_c\eta}} \tag{2}
\]
where the final expression is obtained using a Taylor series, separating the perturbation terms from the non-perturbation terms and neglecting the higher-order terms, such as $\delta n_{e}^{2}$, $\delta n_{e}^{3}$, etc. Thus, for a non-uniform plasma, the wave-vector component $\delta k$ resulting from the density perturbation $\delta n_{e}$ can be written as $\delta k = (k_{0} \delta l / 2n_{e}) \vec{\nabla} n_{e}$. This also shows that the component $\delta k$ is oriented along the electron-density gradient $\vec{\nabla} n_{e}$, which can be interpreted as the scattered-wave, as was observed in our experiments.

Thus, in addition to the previously known plasma instabilities in under-dense plasma, such as Raman scattering and two plasmon instability, our measurements suggest an additional mechanism by which a fraction of the laser energy is scattered by the plasma up to near-critical densities, which can be termed as plasma-gradient scattering. We have shown that this occurs as a result of the non-uniform plasma density distribution at moderately relativistic intensities. Therefore, the energy loss incurred by the laser pulse through this scattering mechanism can affect the DLA process, where it is desirable that the laser pulse’s energy is effectively utilized for the acceleration process. Moreover, understanding the nature and origin of this side-scattering process, might also prove to be beneficial for applications other than laser driven particle acceleration. For future experiments aiming at exploring the interaction of laser pulses with underdense to near-critical plasma, e.g. to study the mechanisms of DLA or others, it is advisable to choose a density regime above $0.25 n_{c}$, if the plasma density’s transverse uniformity cannot be ensured.

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