Acceleration of neutrals in a nanosecond laser produced nickel plasma

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Time of flight dynamics of slow neutrals, fast neutrals, and ions from a nanosecond laser produced nickel (Ni) plasma are investigated. Species arrival times confirm the hypothesis that fast neutrals are formed by the recombination of fast ions with free electrons. Both neutrals and ions are found to accelerate for a short interval immediately after ablation, which is attributed to internal Coulomb forces which create electrostatic potentials resulting in the charged particle acceleration. This process is further enhanced by laser-plasma energy coupling. Emission from neutrals could be measured for longer axial distances in the plume compared to that of ions confirming that the ions recombine to form neutrals as they move away from the target surface.

INTRODUCTION

Plasmas produced by pulsed lasers are transient in nature. The plasma plume evolves quickly with time after target ablation, and the plume characteristics vary dynamically during expansion. Fundamental studies to understand the dynamics of plume expansion have been of interest to researchers owing to the wide range of applications including pulsed laser deposition,1 higher harmonic generation,2,3 production of X-rays4 and EUV,5 and nanoparticle and nanocluster generation.6 Studies such as Optical Emission Spectroscopy (OES), Time of flight (TOF) spectroscopy, Thomson Scattering, Interferometry, Plasma imaging, Langmuir probing, Shadowgraphy, etc., can be used to investigate the properties of laser produced plasmas (LPP). Parameters relevant to the plasma generation and dynamics, such as temperature, number density, plasma frequency, refractive index, etc., can be evaluated using the above mentioned techniques.7–9 In order to realize a given application, the plasma characteristics may be optimized by tuning the laser energy,10 wavelength,11 pulse duration,12 and nature and pressure of the ambient gas.13 Target ablation occurs in a few picoseconds after laser irradiation if the pulse intensity equals or exceeds the ablation threshold.7 The solid target transforms to the gaseous phase forming a high temperature, high pressure, partially ionized gas cloud, during the first few nanoseconds of ablation.14 This is followed by a rapid expansion leading to the evaporation of neutral atoms, electrons, and ions.15 The velocity of the plasma front will depend on the incident laser energy and the atomic mass number of the target.15,16 During the early stages of plume expansion (i.e., in the first few nanoseconds), the inner region of the plasma will be opaque to laser radiation due to its high electron density,17,18 but it will become transparent when the density falls below a critical value. Therefore, if the laser pulse is several nanoseconds

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an Nd:YAG laser, to produce the plasma. The laser is set to run at 10 Hz for energy stability, but the experiment is performed in the single-shot mode by means of an electronically synchronized fast mechanical shutter positioned in the beam path, which allows only a single pulse to pass through when the shutter is opened. In order to avoid pitting of the target surface, after each laser pulse, the target is moved about 500 μm using a stepper motor driven XY translator, so that the next pulse will irradiate a fresh spot on the target surface. Optical emission spectra in the visible region were recorded using a CCD (Synapsec, Horiba Jobin Yvon) attached to the front exit of a high resolution (~0.06 nm) monochromator (iHR 320, Horiba Jobin Yvon), and spectral lines were identified by comparison with the standard NIST database.27 The optical TOF (OTOF) spectra of neutrals and ions were recorded by a fast photomultiplier tube (PMT) (R943-02, Hamamatsu) positioned at the side exit of the monochromator. The PMT signal was recorded and digitized using a fast oscilloscope (DPO 7354, Tektronix). By keeping the same laser spot size (~300 μm) and background pressure (5 Torr, which is the optimum pressure for maximum OTOF signal), measurements were taken for different laser energies to understand the influence of laser fluence on the generation of fast atomic species. After fixing the fluence at approximately 10 J/cm² (by focusing fast atomic species. After fixing the fluence at approximately 10 J/cm² (by focusing

\[ I_{\text{min}} = \frac{\rho L_s k^4}{\Delta T^2}, \]  

where \( \rho \) is the density of the material, \( L_s \) is the latent heat of vaporization, \( k \) is the thermal diffusivity, and \( \Delta T \) is the laser pulse width. \( I_{\text{min}} \) is calculated to be 3.89 × 10⁸ W/cm² for ns excitation, with the corresponding laser fluence being 2.72 J/cm² in the present case. Arrival times of ions and neutrals are measured for various laser energies and at different pressures.

OTOF is an effective nondestructive tool to probe the expansion dynamics of various species in the expanding LPP because the technique essentially depends on the detection of emitted radiation. We measured line emissions at 391.6 nm (3d⁷(2P) 4p → 3d⁶(2D) 4s) and 428.5 nm (3p³d⁹ (2P) 4s → 3p⁶d⁴4s) to characterize the expansion dynamics of neutrals (Ni I) and ions (Ni II), respectively. OTOF dynamics measured for energies ranging from 10 mJ to 80 mJ, at a distance of 2 mm on the plasma plumes longitudinal expansion axis normal to the target surface, for 5 Torr nitrogen ambient pressure, are given in Fig. 1. The vertical axis of Fig. 1 is normalized using the maximum voltage recorded on the oscilloscope for the peak P1 for better clarity. At lower laser energies (10 mJ), only a single peak corresponding to slow neutrals (P2) which represent un-ionized neutral species can be seen at a time delay of about 220 ns. At higher energies (≥20 mJ), the OTOF signal of Ni-I displays an additional peak (P1) which occurs at about 80 ns, indicating fast neutrals. The onset of P1 happens earlier with laser energy but that of P2 is unaltered. Ni II displays a single but broader peak (represented as P3 hereafter) for an irradiation energy ≥20 mJ. Fast neutrals (P1) and ions (P3) are found to arrive almost at the same delay times indicating that fast neutrals are in fact formed by the recombination of fast ions with electrons.

For most practical applications, high quality monoenergetic particle beams of electrons, ions, or neutrals produced from laser produced plasma is needed.29–33 Fast as well as slow ions and neutrals generated along with electrons that leave the initial volume and ions following them to speed up. Therefore, ions present in the inner region

RESULTS AND DISCUSSION

Laser ablation occurs within a few picoseconds after target irradiation, and the ablated species interact with the remaining part of the excitation laser pulse. Material ablation occurs once the intensity of irradiation exceeds the ablation threshold of the material, which is given as28

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(core) of the plume get accelerated to certain distances at the earlier stages of plasma expansion due to the above mentioned space-charge effect. In the present article, an experimental verification on the acceleration of both ions and neutrals occurring to certain distances in an expanding ns laser produced nickel plasma is presented in detail as follows.

Since the nature of the background gas and its pressure will affect LPP expansion dynamics, plasmas can be tuned accordingly for practical applications such as pulsed laser deposition. At lower pressures, the plasma expands adiabatically and the relative scarcity of collisions results in a low number density, leaving more neutrals in the plume. For ns laser pulse irradiation, it is known that the plasma temperature and electron density is enhanced with ambient pressure. The plasma temperature will also increase due to heating via enhanced laser-plasma coupling throughout the plume volume. Indeed the emitted species will interact with...
the relatively long laser pulse and absorb energy via inverse Bremsstrahlung (IB), the strength of which is given by

\[ \alpha_{IB} = 1.37 \times 10^{-35} \lambda^3 N_e^2 T_e^{-4}, \]  

(2)

where \( \alpha_{IB} \), \( \lambda \) (\( \mu \text{m} \)), and \( N_e (\text{cm}^{-3}) \) are the IB co-efficient, irradiation wavelength, and electron number density, respectively. The subsequent rise in electron temperature results in enhanced collisions and further ionization. IB enhancement is possible at higher number densities and the confinement generates hotter plasma.\(^{24} \) At higher pressures, the plasma exhibits a longer history due to plume confinement, but beyond certain optimum pressure plasma temperature and number density will be reduced due to energy loss from collisions with the background gas. This loss mechanism is characterized by the Thermal Leak, given by\(^{23,24} \)

\[ Q_{\text{Leak}} = \frac{2m_e e^2 \sigma_{el} n_B^2}{M_e} \left[ \frac{5kT_e}{\pi m_e} \right]^{1/2}, \]  

(3)

where \( n_B (\text{cm}^{-3}) \) and \( M_B \) (kg) are the density and mass of the background gas, respectively, and \( \sigma_{el} \) is the elastic scattering cross section of the electrons. There exists a certain range of pressures (generally from 10\(^{-3}\) Torr to 10\(^2\) Torr) where a relatively large number of density is observed for the plasma species.\(^{23} \) Accordingly, our measurements were carried out in the pressure range of 5 \( \times 10^{-2}\) Torr–1 \( \times 10^2\) Torr.

Species velocities were measured for various positions along the plume axis progressively away from the target surface, until signals from neutrals and ions were too weak to be detected. Results obtained for fast neutrals and ions are shown in Figs. 3(a) and 3(b), respectively. At the lowest pressure used (0.05 Torr), \( P_1 \) velocity is found to increase from \( \sim 20 \text{ km/s} \) (at 2 mm) to \( \sim 23 \text{ km/s} \) (at 4.5 mm), beyond which the signal disappears. At a higher pressure of 1 Torr, \( P_1 \) accelerates only up to a shorter distance of 3.5 mm (velocity is 25 km/s at 2 mm and 32 km/s at 3.5 mm), beyond which it decelerates. The emission is observed until 7.5 mm. This deceleration occurs due to plume confinement, and therefore, the emission can be observed only for shorter axial distances at the higher pressures of 20 Torr and 100 Torr. From Fig. 3(b), it is seen that at 0.05 Torr the ions accelerate and fly away without being detected beyond 4.5 mm. At 1 Torr, ion velocity increases from 27 km/s to 30 km/s for a movement of 2 mm (from the 2 mm to 4 mm position) along the plume axis. At larger distances (after 4 mm), ions decelerate due to the reduced coulomb pull, though emission can still be detected up to 8.5 mm. At 20 Torr and 100 Torr, the OTOF signal is very weak beyond the 4 mm and 3 mm positions, respectively, which is possibly due to a higher rate of recombination. From Figs. 3(a) and 3(b), it can be seen that the plume length is largest for 1 Torr ambient pressure.

From these results it becomes clear that acceleration of fast neutrals and ions can indeed be observed in expanding nanosecond laser produced plasmas at relatively higher input fluences. Ion acceleration occurs due to the internal electrostatic fields generated by space charges present in the plume, and also due to laser-plasma energy coupling. Ion velocity is reduced at larger distances due to the reduced number density. The experimental observation that fast neutrals are accelerating in a ns LPP for certain optimized conditions of background pressure and laser fluence is novel. The measured acceleration of fast neutrals and ions is consistent when the experiment is repeated for various axial distances and pressures. From the measured arrival times of neutral and ionic species, it is experimentally confirmed that recombination of fast ions with electrons during plasma expansion is the primary mechanism for the generation of fast neutrals in the present LPP. This recombination mechanism was proposed earlier by Amoruso et al.\(^{23} \) in the context of an ultrafast (femtosecond) Ni LPP expanding into a near-vacuum background of 10\(^{-7}\) Torr, but has not been experimentally verified yet.

Fig. 4 shows the velocities of slow neutrals measured at the pressures of 0.05 Torr, 1 Torr, 20 Torr, and 100 Torr, for
a laser pulse energy of 30 mJ. Fast neutrals decelerate quickly so that the slow neutrals catch up with them, and the peaks P1 and P2 eventually merge, in between 4 mm and 6.5 mm positions. In this region, fast and slow neutrals cannot be resolved in time and space. The comparatively large error obtained when the measurement is repeated at 3.5 mm is ascribed to the interactions of fast and slow species in the plume causing relatively large shift in the peak of the OTOF signal.

The above measurements conclusively prove that fast neutrals can indeed be generated in nanosecond laser-produced plasmas. From the arrival times, it is confirmed that fast neutrals are recomposed neutrals produced by the recombination of fast ions with free electrons in the plasma, which is the reason for their initial high velocities. The velocity of fast neutrals and ions increases with the irradiation energy, indicating efficient laser-plasma energy coupling.

CONCLUSION

In summary, a nanosecond laser produced nickel plasma was generated by irradiating a pure solid nickel target with 7 ns laser pulses at 1064 nm. OTOF measurements were performed on the Ni-I (361.9 nm) and Ni-II (428.5 nm) transitions for different ambient pressures, plume axial positions, and laser energies. OTOF peak velocities were measured for various axial positions in the plume. Arrival times of the observed fast neutrals and ions are reduced at higher laser energies, indicating efficient laser-plasma energy coupling. An acceleration of fast neutrals is observed during plume expansion at higher irradiation energies. From the measured species arrival times it is experimentally shown that fast neutrals are generated in the plume by the recombination of fast ions with electrons. This is further confirmed from the observation that fast neutrals decelerate in the later part of plume expansion and co-propagate with the slow neutrals.

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