# Twin peak distribution of electron emission profile and impact ionization of ambient molecules during laser ablation of silver target

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# Twin peak distribution of electron emission profile and impact ionization of ambient molecules during laser ablation of silver target

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Laser-induced plasma generated from a silver target under partial vacuum conditions using the fundamental output of nanosecond duration from a pulsed Nd:yttrium aluminum garnet laser is studied using a Langmuir probe. The time of flight measurements show a clear twin peak distribution in the temporal profile of electron emission. The first peak has almost the same duration as the laser pulse while the second lasts for several microseconds. The prompt electrons are energetic enough ($\approx$60 eV) to ionize the ambient gas molecules or atoms. The use of prompt electron pulses as sources for electron impact excitation is demonstrated by taking nitrogen, carbon dioxide, and argon as ambient gases. © *1998 American Institute of Physics.* [S0003-6951(98)03928-X]

In recent years the study of pulsed laser ablation has become a topic of vital importance both from the fundamental point of view and in the context of applications.[1–9] Even though the ionic, atomic, and molecular temporal profiles have been investigated in depth,[10–12] electron temporal profiles have not been studied in detail. Yet, the multiphoton induced surface and volume photoelectron generation and their angular distributions have been studied during laser interaction with metals.[13,14] Laser heating of the plasma electrons and the role of electrons in plasma shielding during nanosecond and picosecond laser interactions are also of interest in the context of laser matter interactions.[15–18] In this letter we report the observation of a twin peak distribution of the electron pulses occurring during the interaction of infrared radiation from a pulsed Nd:yttrium aluminum garnet (YAG) laser with a silver target. Application of these electron pulses as short duration excitation sources in the collisional ionization of atoms/molecules is demonstrated.

The schematic diagram and details of the experimental setup were already given earlier except for minor modifications required for the present investigations.[19] In brief, the experimental setup consists of a pulsed Nd:YAG laser (Quanta Ray) which emits pulses of 1.06 $\mu$m radiation with 10 ns duration. The laser pulses were tightly focused (estimated spot radius of 50 $\mu$m) on to the surface of a silver target which was kept inside a partially evacuated plasma chamber. The electrons were captured with a positively biased Langmuir probe made of tungsten connected to a digital storage oscilloscope (Tektronix TDS 220) through a 50 $\Omega$ load and the optical emission was monitored using a one meter grating monochromator (SPEX 1704) coupled with a thermoelectrically cooled photomultiplier tube (Thorn EMI). The plasma emission intensities were gated and averaged using a boxcar averager (SRS, SR 250) and fed to a computer for data analysis.

Figure 1 shows the time of flight profile of the electron pulse as recorded by monitoring the Langmuir probe current at a distance 2 cm from the target surface. There are two distinct peaks occurring in the probe current signal. The first one, which is very sharp, almost coincides with the laser pulse with a full width at half maximum approximately equal to the laser pulse width. This indicates that the first peak does not correspond to the thermal electrons found in laser plasmas. These are therefore the photoelectrons which are accelerated through laser absorption by an inverse bremstrahlung process. The second peak appears well after the termination of the laser pulse and with much slower peak velocities of the order of those usually observed for atomic and ionic species coming from the target. Therefore, obviously the second peak corresponds to the electrons accompanying the silver plasma. The electron energy corresponding to the two peaks are evaluated using volt-ampere characteristics of the Langmuir probe. The first peak is found to have an electron energy of approximately 60 eV and the second peak has energy of 2 eV.

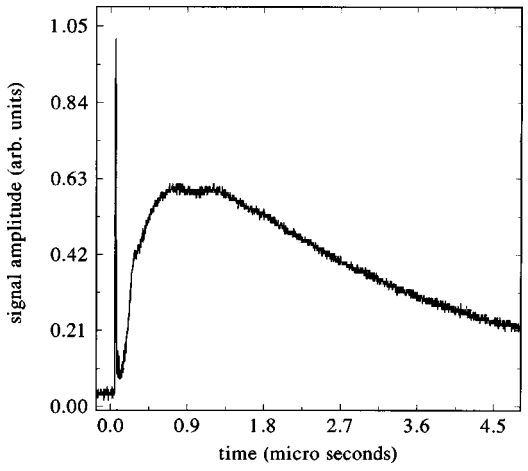


FIG. 1. Temporal profile of the probe signal. The profile shows clear twin peak distribution. The narrow peak appearing early in time represent prompt electrons and the broad peak correspond to the plasma electrons which extends to several microseconds.

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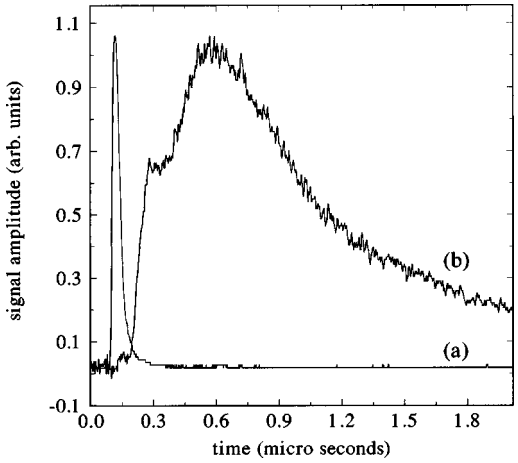


FIG. 2. Temporal profiles of (a) the emission from $N_2^+$ at 427.8 nm and (b) that of silver atomic emission at 546.5 nm.

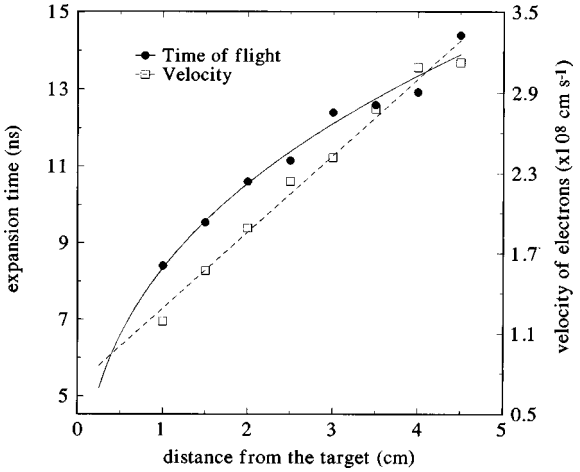


FIG. 3. Variation of the time of flight as well as velocity of electrons with distance from the target.

In metals, the laser interaction occurs within the skin depth. The incoming laser radiation is absorbed by the conduction electrons and the absorbed energy is transferred to the lattice through electron–phonon interactions.[20,21] The electrons are heated well above the lattice temperature within a few picoseconds[22,23] and the surface electron temperature follows almost the same temporal profile as the heating laser pulse whereas the heat is transferred to the lattice with a delay time resulting in bond breaking and generation of the plasma.[23] A fraction of the laser heated electrons may escape from the interaction region without losing energy to the lattice. These fast electrons on collision with the atoms/molecules in the ambient atmosphere ionize them and this is revealed by the characteristic emission from such ionized species.

Figure 2 summarizes the results of measurements done with nitrogen at low pressure (0.2 mbar) in the plasma chamber. It gives a comparison of the temporal emission profiles from the atomic silver and ionized molecular nitrogen in the plasma from a segment situated at a distance 0.5 cm away from the target surface. Figure 2(a) shows the emission temporal profile at the wavelength 427.8 nm due to the $B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ vibrational transition in $N_2^+$ molecule. Figure 2(b) gives the emission profile due to silver at 546.5 nm and this is broad and indicates a larger time of flight. The ambient gas molecules can be ionized by the direct absorption of the ultraviolet (UV) radiation from the plasma core, through collisional excitation by energetic electrons or through the multiphoton absorption of the laser light. Figure 3 shows that the time of flight for these electrons increases with distance from the target even though the variation is not strictly linear. The time of flight ($t$) is related to the distance from the target ($R$) as $R \propto t^{0.34}$ shown by the solid curve in Fig. 3. This eliminates the possibility of ambient ionization and electron generation through the absorption of the UV radiation originating from the focal spot since with the present length scales such an absorption should be instantaneous. Also the UV radiation from the plasma core does not have sharp temporal profiles and in that case the prompt electron pulse would have become much broader than the laser pulse width. The velocity of prompt electrons are of the order of $10^8$ cm $s^{-1}$ and suffers a linear increase with distance from the target as shown by the dotted line in Fig. 3. This is because the thermal energy is converted into kinetic energy as it expands.[3] It is also seen that the intensity of $N_2^+$ emission is directly proportional to the laser power density. In the case of direct multiphoton absorption and excitation, the emission intensity should be proportional to the $n^{\text{th}}$ power of the laser power density where $n$ is an integer. Therefore, multiphoton absorption is not a likely process in the ionization of ambient gas. This fact is supported by the increase of electron density with distance from the target to be described later in this letter. The linearity of $N_2^+$ emission with laser power density

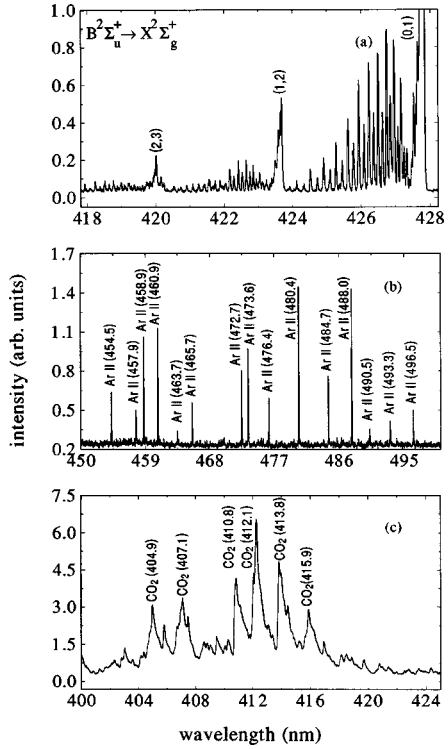


FIG. 4. Time resolved spectra with different ambient atmospheres with a gate delay 0 ns and width 40 ns. (a) High resolution spectrum of the emission $\Delta v = -1$ band from $N_2^+$. Spectrum shows clearly resolved rotational lines with intensity alternations. (b) The spectrum with argon ambient where the spectrum is dominated by the emission from singly ionized argon. (c) The spectrum of carbon dioxide obtained under the same pressure conditions as that for nitrogen ambient.

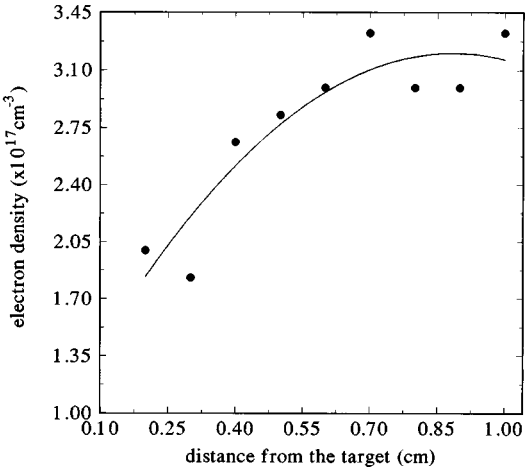


FIG. 5. Variation of prompt electron density as a function of distance from the target at a pressure of 0.27 mbar. The density increases with distance from the target indicating electron generation due to collisions.

actually supports the argument in favor of electron impact ionization of ambient molecules. Collisional ionization requires high energy electrons and the temporal pulse of the $N_2^+$ emission almost coincides with the occurrence of the first electron current pulse at various distances. The decay time of the $N_2^+$ emission profile is found to be considerably larger than that of the width of the exciting electron pulse since various lifetimes of the excited states are involved in the spectral emission process.

Time resolved spectra were recorded with the boxcar gate kept at different delays with respect to the termination of the laser pulse. At short time delays, only the emission spectra of the ambient molecules appears. Figure 4(a) gives a time resolved high resolution spectrum of the $\Delta\nu=-1$ band of $N_2^+$ with band head at 492.8 nm with a time delay of 0 ns and gate width of 40 ns after the laser pulse at a distance of 0.5 cm away from the target. There are clear rotational fine structures appearing in the spectrum with characteristic intensity alternations due to the difference in the statistical weights for even and odd numbered rotational levels of $N_2^+$. Figures. 4(b) and 4(c) show the time resolved spectra recorded with argon and carbon dioxide as ambient gases, respectively. This clearly indicates that electron pulses created during laser beam interaction with metals can effectively be used as short duration collisional excitation sources for atomic and molecular gases.

The spectrum with argon ambient shows various emission lines of first ionized species. Therefore, it can be considered that the laser plasma is preceded by a partially ionized ambient plasma due to prompt electron emission from the metal surface. The electron density as a function of distance from the target is evaluated using the stark broadened emission line width of argon and is given in Fig. 5. Electron densities corresponding to prompt electrons are of the order of $10^{17}$ $cm^{-3}$ and there is an increase in the electron density with distance from the target surface. This indicates that the source of these prompt electrons are not solely from the target surface but further electrons are created in the ambient medium due to cascade ionization. The electron energy decreases due to collisions and therefore there is a saturation of the electron density beyond 0.7 cm.

In conclusion, a twin peak distribution in the electron current pulse is obtained during laser pulse interaction with a silver target. The first peak corresponds to laser heated electrons escaping from the interaction volume before the absorbed energy is transferred to the lattice. The second peak comprising of low energy electrons has a comparatively broader temporal distribution and corresponds to those in the silver plasma. The prompt electron pulses can effectively be used as an excellent short duration excitation source in atomic and molecular spectroscopy. We hereby propose that by using even shorter duration pulsed lasers for irradiating metal surfaces one can get ultrashort electron pulses and this may act as a very effective electron source required for electron-vibration energy relaxation experiments.

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