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Effects of Inverse Bremsstrahlung in Laser-Induced Plasmas from a Graphite Surface

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Abstract. The kinetic energy of electrons emitted due to laser interaction with a graphite surface was studied with a time-of-flight spectrometer. In addition the yields of carbon atomic and molecular ions were measured as a function of laser pulse energy. Pulse energy thresholds for ion emission are observed to correlate with the observed maximum electron energies. Furthermore, the data suggest that ionic carbon clusters can be dissociated by energetic electrons or photons created in the plasma. We believe that initially photoemitted electrons are accelerated by inverse bremsstrahlung to the energies required for electron impact ionization and dissociation

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In a recent article Phipps et al. [1] developed a theory for hydrodynamic variables of laser induced plasma, which appears to describe macroscopic properties such as pressure and expansion velocity in a convincing way. On the other hand, the microscopic processes which can take place in such plasmas seem to be less well understood. There are, however, indications in the literature that the chemical nature of the products of laser induced desorption is to some extent determined by plasma effects [2, 3].

In this paper we elucidate the mechanisms for electron creation and acceleration in a laser-induced plasma from a graphite surface. Graphite is an appropriate choice for such studies, because here the laser-induced emission of clusters is believed to be a thermal process [4]. Electronic excitations and ionization should therefore be due to processes in the plasma and not in the bulk. We present evidence that inverse bremsstrahlung is the primary mechamism for acceleration of electrons. This is consistent with the theory of Phipps et al. [1]. Subsequent processes involve electron impact ionization/dissociation and emission of UV photons via bremsstrahlung. This latter process may in turn lead to photo-excitation, photo-ionization, and photo-fragmentation.

1. Experimental

The experiments were performed in a vacuum chamber with a base pressure of 10^{-9} mbar. The target was commercial grade polycrystalline graphite. The laser used in these experiments was an injection seeded Nd : YAG laser system with a pulse energy stability of 6–8%. It provided $\sim$ 8 ns pulses at 532 nm and $\sim$ 6 ns pulses at 355 nm. The laser beam was focussed to a diameter of $\sim$ 0.3 mm. The emitted positive ions were measured either with a quadrupole mass spectrometer (QMS) or, as were the electrons, with a time-of-flight (TOF) spectrometer consisting of a 65 cm long mu-metal flight tube and a set of extraction/focusing electrodes at the entrance. The field from these electrodes only acted on the particles for a short distance, and was found to have no noticeable effects on the flight times.

The angle of incidence of the laser beam was $\sim$ 70° C from the surface normal, and the spectrometers were placed with their axes at right angles to the beam direction. The graphite surface clearly changes as a function of radiation exposure. This is manifested as a variation of up to 20% in the pulse energy thresholds for emission of electrons and ions. To minimize exposure, all experiments were done in single-shot mode. The qualitative conclusions drawn from the measurements are not affected by the observed threshold variation.

2. Results and Discussion

Electron, photon, and total ion yields were measured as functions of pulse energy using the TOF-spectrometer. Figure 1 shows the results for 355 nm, s-polarized light.

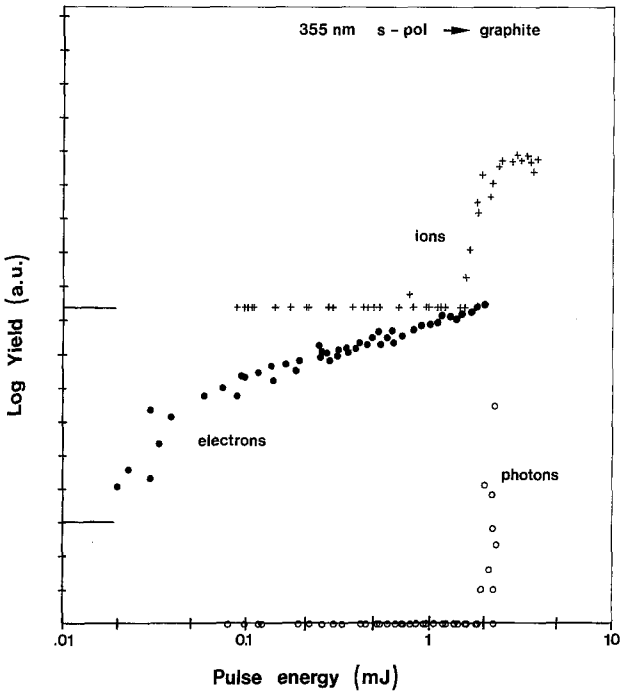

Fig. 1. Electron, photon, and total positive ion yields from a graphite surface vs. laser pulse energy, as measured with the TOF-spectrometer (355 nm, s-polarized light). The electron and ion yields have been displaced vertically for clarity

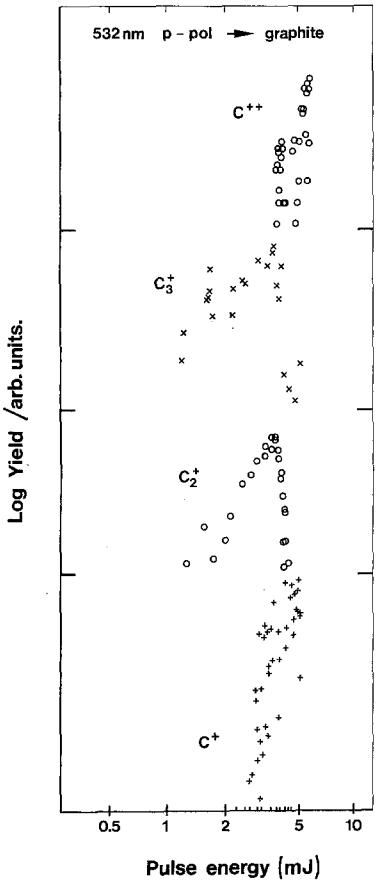

Fig. 2. Positive ion yields vs laser pulse energy as measured with the QMS (532 nm, p-polarized light). The C_2^+, C_3^+, and C^{++} yields have been displaced vertically for clarity

The spectra for 355 nm, p-polarized and 532 nm, s- and p-polarized light have similar shapes. The electron yield first increases very rapidly. Following Lin and George [6] and Strupp et al. [7] we interpret this behavior as due to thermally assisted multiphoton photoemission. As the pulse energy increases the electron yield eventually approaches saturation. We postulate that this is due to the surface becoming partly screened against the incoming light by emitted neutral clusters. Although no positive ions were observed for pulse energies $E_{\text{pulse}} < 1\,\text{mJ}$, we take the saturation of the electron signal as evidence that the graphite begins to evaporate at $E_{\text{pulse}} \cong 0.1\,\text{mJ}$. This is supported by the observation of Fürstenau et al. [5] that the emission of negative carbon clusters sets in at intensities well below the threshold for positive ion emission. Further evidence comes from the fact that the emitted electrons, as discussed in detail below, gain energy from the electromagnetic field; this would not be expected for free electrons in a vacuum, but requires the presence of scattering centers.

As can be seen in Fig. 1, the threshold for ion emission is approximately one order of magnitude higher than the threshold for electron emission. The yields of C^+, C_2^+, C_3^+, and C^{++} vs E_{pulse} measured with the quadrupole mass filter are shown in Fig. 2. At the threshold the molecular ions dominate. C^+ is not observed until E_{pulse} is 2–3 times the threshold energy. We believe that the observed threshold for C^+ emission is determined by the detection limit, and that C^+ is in fact emitted at lower pulse energies. Under conditions similar to ours Berkowitz and Chupka [8] found that the C_2^+ and C_3^+ yields could be significantly larger than the C^+ yield. At approximately 4 mJ/pulse the C_2^+ and C_3^+ yields begin to decrease and eventually disappear, and at almost the same pulse energy the C^{++} yield increases rapidly.

We now present evidence that the variations in the ion yield can be explained as ionization and dissoziation in the plasma induced by electron or photon interactions.

In Fig. 3 we show electron energy spectra for various pulse energies. The spectra have been obtained from the measured TOF spectra using the formula $N(E) = N(t)\frac{dt}{dE}$, where E is the kinetic energy, t the time of flight, and N the yield. At pulse energies close to the threshold for electron emission, the spectra are relatively narrow with widths less than the respective photon energies, as would be expected for a multiphoton photoemission process. As E_{pulse} is increased, a tail emerges on the high-energy side and the maximum kinetic energy increases with E_{pulse}. Kinetic energies in excess of 15 eV were observed. It is interesting to observe that the maximum kinetic energy, $E_{\max}$, apparently varies continuously as a function of E_{pulse}. This would not be expected if the increase in energy were caused by an increase in the order of the multiphoton photoemission process or above threshold ionization. This would appear in the spectra as a stepwise change in electron energy. We therefore interpret the variation in the shape of the electron spectra as being due to inverse bremsstrahlung.

We can now explain the ion yield measurements in the following way: The ionization energies for C_n,

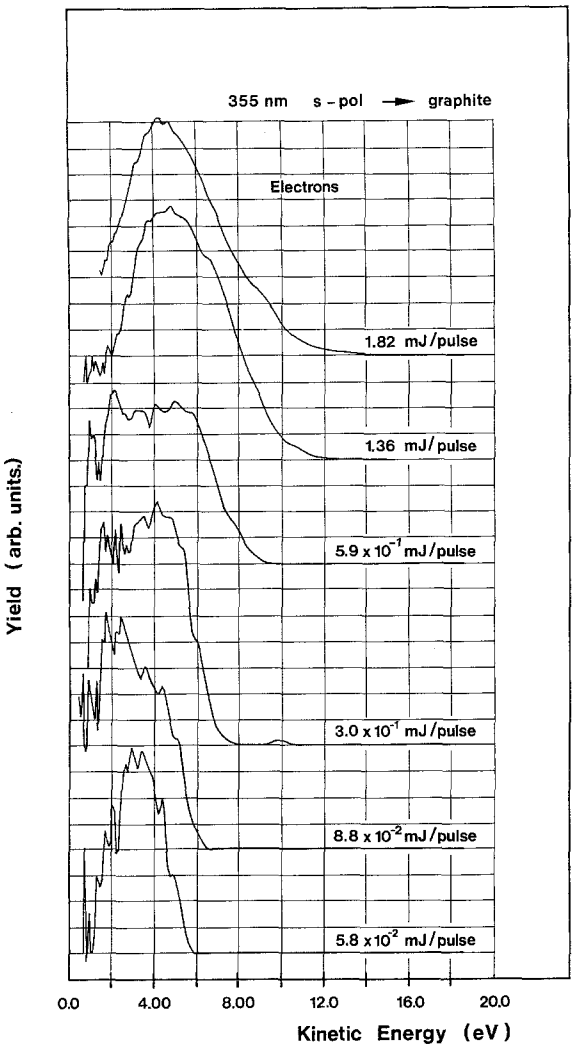

Fig. 3. Electron energy spectra at different laser pulse energies, derived from TOF-spectra

$n = 1, 2, 3$, are all approximately 12 eV [9]. This means that ions can only be formed in the gas phase when $E_{max} > 12$ eV. In Table 1 we list, for different wavelenghts and polarizations, the approximate pulse energies at which the electron kinetic energy has reached 12 eV, together with the thresholds for ion emission. We observe that the onset of ion emission is indeed correlated with the pulse energy where impact ionization becomes energetically possible.

The disappearance of the C_2^+ and C_3^+ ions for $E_{pulse} > 4$ mJ can be readily explained in terms of electron impact dissociation and photofragmentation. The cross-sections for electron impact dissociation are unknown, but we note that van Zyl and Dunn [10] observe thresholds for electron impact dissociation of O_2^+ and N_2^+ which are significantly higher than the adiabatic dissociation energies. An additional contribution to the observed behaviour is that, as the electrons become very fast, they can emit bremsstrahlung with enough energy to cause photofragmentation. Geusic et al. [11] have shown that photofragmentation of ionic carbon clusters is possible for photon energies in the range expected in our plasma (up to ~ 30 eV). However, exact thresholds and cross-sections are unknown. A signal due to C^{++} appears at $E_{pulse} \cong 3.5$ mJ. We believe that the C^{++} ions are created by electron impact ionization of the C^+ ions. This would require ~ 24 eV [9].

Table 1. The pulse energy, E_{12}, at which the electron energy has reached 12 eV, and the threshold pulse energy E_{ion} for emission of ions for different wavelengths and polarizations

		E_{12} [mJ/pulse]	E_{ion} [mJ/pulse]
355 nm	s	1.4	1.6
	p	1.4	1.2
532 nm	s	3.4	3.2
	p	1.5	1.2

We rule out the possibility that the observed ionization and dissociation processes are due to multiphoton excitations and ionization caused by the incoming laser beam. Firstly, the ionization of C_n; $n = 1, 2, 3$ would require at least 6 photons at 532 nm. A multiphoton process of order 6, however, is highly unlikely at the intensities used here ($< 10^9$ W/cm^2). Similarly, multiphoton ionization of C^+ to form C^{++} would be at least an 11-photon process which is even more improbable. And, finally, multiphoton-induced dissociation of C_2^+ and C_3^+ would, according to the work of Geusic et al. [11], be expected to require at most 4 photons at 532 nm. This, however, cannot explain the rather abrupt decrease in the yields of C_2^+ and C_3^+.

3. Conclusion

We have shown that electrons in a laser-induced plasma can be accelerated to energies in excess of 15 eV. The energy distribution of the electrons, which depends on the laser pulse energy, was found to have a strong influence on the chemical state of the emitted species.

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