Contents lists available at ScienceDirect



Spectrochimica Acta Part B

journal homepage: www.elsevier.com/locate/sab



Review Ion kinetic energy distributions in laser-induced plasma

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ARTICLE INFO

Article history: Received 29 April 2014 Accepted 24 June 2014 Available online 1 July 2014

Keywords: Laser-induced plasma Ion kinetic energy distribution Angular distribution Ion acceleration

ABSTRACT

Studies of ion emissions from laser-induced plasmas (LIPs) provide insights into the hydrodynamic expansion of the plume. Investigations of the kinetic energy distributions (KEDs) of ionized species for various experimental conditions are vital for a fundamental understanding of the formation and expansion dynamics of plasma. This knowledge, in turn, leads to promising improvements in LIP-based technological applications.

This article aims to review some of the dominating mechanisms concerning ion emission kinetics during laser-surface interactions from a basic point of view. The diagnostic methods for ion KEDs are roughly classified. Interesting features of ion KEDs and their angular distributions, as well as the dependence on laser beam properties, ambient surroundings, and target properties, are summarized.

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1. Introduction

Since the introduction of the laser to analytical chemistry in the early 1960s [1–4], laser ablation and ionization have been widely used as flexible and powerful tools for applications such as material

characterization, microanalysis, laser machining, surface treatment, and transient electric field generation [5,6]. For the most important applications, including thin film deposition, ion implantation, and ion accelerators [7–9], the kinetic energy (KE) of the ablated species has significant effects on the deposited film quality, substrate property, or particle beam unity, respectively. Therefore, the basic processes of plume expansion and ion emission and the resulting kinetic energy distributions (KEDs) must be understood to improve many applications.

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The laser beam parameters and target properties affect laser ablation. The physics of the laser ablation process involves laser-target interactions, plasma formation and evolution. Plasma expansion, electron-impact excitation/ionization, recombination, and energy transfer processes result in complicated plasma kinetics [10,11]. Although the plasma should be fully characterized in terms of temperatures, densities, yields, charge state, and so on [12], a special interest for LIP is the KEDs of the beams of ionized species and their spatial distributions. Due to the high detectability of ions, measurements of ion species and their velocity distributions are often conducted because ions can be used as markers to understand plume expansion and laser-target interactions.

Basically, a shifted Maxwell-Boltzmann-Coulomb (MBC) distribution fits well to the overall ion KEDs of a LIP in vacuum [7,12-22]. The shifted energy distributions of ion species prove that the charge separation in the non-equilibrium plasma leads to an accelerating voltage [12,17,19,21–26]. The ablated particles subsequently obtain a certain velocity that preferentially peaks along the direction perpendicular to the target [27]. The anisotropic distribution of the ablated species as well as the unexpected enormous KEs are related to thermal effects (inverse bremsstrahlung absorption, target/plasma heating), supersonic-like adiabatic expansion, and Coulomb effects (ion repulsion, ion-electron attraction, and ion acceleration via charge separation) [21, 27]. Of these effects, the charge separation is the dominant acceleration process for the ultrashort pulse laser regime [28,29]. Typically, an ion KE reaches the order of several keV under a moderate laser power density [30,31], while it extends up to several MeV at an irradiance greater than 10¹⁵ W/cm² [32,33].

The ions emitted from a LIP with a certain charge state, KE, and angular distribution are highly related to the parameters of the employed laser (e.g., laser fluence, laser wavelength, and pulse duration), the ambient surrounding, and the irradiated target material. It is important to evaluate the above factors as well as to determine the underlying mechanisms to make the application viable. In Section 2, a theoretical study of ion KEDs will be presented. Ion KED investigation methods will be briefly classified as time-of-flight (TOF) profile measurements or energy analyzer measurements in Section 3. The dependences of ion KEDs on the laser beam properties, ambient surroundings, and target material will be discussed in Sections 4, 5, and 6, respectively. A conclusion will be drawn in the last section.

2. Theoretical study of ion KEDs

2.1. Mechanism of charge separation and ion acceleration

It is rather instructive to provide basic insight into the physical phenomena involved in the laser-target interactions during irradiation in vacuum. This section mainly focused on the ion acceleration mechanism underlying the dependence on the laser pulse duration.

For irradiation with a longer laser pulse (~nanosecond, ns), the ion ejection during the early stage of the laser pulse preferentially occurs in a direction perpendicular to the target surface and is driven by the pressure gradients [34–36]. The ion velocity distributions become a full-range Maxwell-Boltzmann (MB) distribution with a shifted center-of-mass velocity due to the extensive collisions within the Knudsen layer [10,11,37]. When the laser irradiation terminates, the plume undergoes an unsteady adiabatic expansion and peaks sharply in the forward direction. Typically, experiments have shown that ionic components fit to Anisimov's model, which is based on hydrodynamic motion regardless of electrostatic interaction [38,39]. However, a complete description of the observed ion KEDs is not sufficient because the ion KE is larger than the KE of neutral species and the average energy is substantially higher than the thermal energy [25,40,41]. A series of results have suggested that the ion acceleration process occurs in the LIP [42-47]. In addition, the linear dependence between the charge state and the center-of-mass velocity reveals that both hydrodynamic and electrostatic forces contribute to the expansion velocity of the plasma [48,49]. According to the electrostatic model [41,45,50], electrons that are heated by inverse bremsstrahlung (IB) absorption transfer the absorbed laser energy to ions through ion-electron collisions in the expanding plasma. The scale of the electron-ion thermalization time $(10^{-10} - 10^{-11} \text{ s})$ is much shorter than the laser pulse duration [51]. Thus, ions and electrons reach the same thermal temperatures during the early portion of the laser pulse. Because electrons are the lightest species in the plasma, they obtain the highest thermal velocities and escape from the plasma. The space charge separation between the energetic electrons and the ions that lag behind prevents the electrons from completely escaping the plasma. Because electrons are confined at the leading edge of the plume, a self-electrostatic field is formed, which accounts for the proportionality between the ion peak KE and the charge state. The initially formed ns LIP displays a thermal nature. As the electrons absorb photon energy by an IB process, the plasma reheats at a fixed distance from the target surface, and this reheating sustains the acceleration process [52]. The expression for the final ion KE in the Z charge state is given by the following equation [41]: $E_{kin} = (nZ + 1)kT$. In this equation, the temperatures of the electron and ion are assumed to be equal to $T(T_e \approx T_i \approx T)$; *n* is the average number of times that the ion accelerates in the self-electrostatic field, and k is the Boltzmann constant. The laser pulse duration is much longer than time for the transfer of energy from the electrons to the ions [51]; thus, this equation indicates that the high kinetic energies gained via electrostatic acceleration dominate over those from the relatively low thermal energy [41,53]. The electrons that preferentially eject along the target normal are driven by pressure gradients. The resulting plasma potential is larger in the normal direction and decreases as the angle relative to the target normal increases [27]. The overall processes are illustrated in Fig. 1. A substantial fraction of the final ion KE is gained via the ion acceleration process caused by the charge separation. The difference in the spatial distribution of the acceleration potential leads to the reduction of the average ion KE (Eave) as a function of angle.

Ablation dynamics studies consider the ambipolar electric field (or double layer) effect to be another version of the electrostatic



Fig. 1. Schematic representation of the vital regimes for laser-surface interactions in a vacuum for the ns laser case at an irradiance greater than the ablation threshold for the ion acceleration and the anisotropic distribution.

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