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STUDIES ON RELAXATION PROCESSES IN LASER-PRODUCED PLASMA

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Abstract. Based on the results of quantitative spectroscopic diagnostics the character of relaxation of parameters (n_e, T_e, n_a) of recombining laser produced plasma has been analyzed.

Laser-produced plasmas in the recombination phase have been used for formation of atomic, molecular and cluster beams, for generation of laser oscillations, as X-ray and vacuum ultraviolet sources etc. (Schriever *et al*, 1998, Ohyanagi *et al*, 1996). For optimization of these applications and refined management of ablation plume characteristics it is of great importance the understanding the character of plasma parameters relaxation.

In the present paper based on diagnostics of plasma composition and its spatial-temporal changes at different irradiation conditions relaxation aspects of laser-produced plasma dynamics have been examined. The main attention has been focused on the temporal evolution of parameters (n_e, T_e, n_a) of recombining laser produced plasma, relaxation dynamics of the ionized and atomic states of Al after action of the excimer XeCl laser radiation (λ=308nm) on the Nd:YAG laser-ablated aluminum plume, the elucidation of self-regulating regime of recombining laser-produced plasma evolution with a frozen ionization state in the late stages.

Details of the experimental apparatus employed in studies of laser-produced plasma composition have been described elsewhere (Burakov *et al*, 1997). Briefly, plasma was produced by focusing of a Nd:YAG laser radiation (1064nm, 10ns, 100mJ, 10^8-10^9 W/cm^2) on the surface of the metallic (Al, Ti) samples in the helium (air) atmosphere at pressures of 0.1-500 Torr. Relaxation dynamics of the ionized and atomic states was studied by the time-resolved emission spectroscopy and laser-induced fluorescence (LIF) techniques. The electron density (n_e) was determined from the emission linewidth measurements according to the Stark broadening theory. The shapes of some selected lines were analyzed at different delay times (with respect to the leading edge of the laser pulse) in order to obtain the temporal evolution of n_e. The electron temperature (T_e) was deduced by the relative intensities of lines from a given state of ionization.

Optical observation of the plasma emission was performed by imaging the section of the plasma plume onto the entrance slit of monochromator equipped by the fast photomultiplier. The emission spectra of plasma were recorded in the UV and visible region (spectral resolution of $\Delta\lambda \geq 3 \bullet 10^{-2}$nm) at different distances from the target surface. The detection of the photomultiplier signals was accomplished by a transient digitizer, connected to a personal computer for data processing, storage and analysis.

For LIF measurements a tunable dye laser radiation at the fundamental wavelength (560 - 630 nm) or its second harmonic was used to probe the ground state atoms and ions with the regular delay times after ablating laser pulse.

The emission spectra, electron temperature and density are found to be influenced by the irradiation conditions and ambient atmosphere. The profiles of the AlI 396.15nm line for different delays and the temporal evolution of electron density deduced from Stark broadening of this line are shown in the Fig.1.

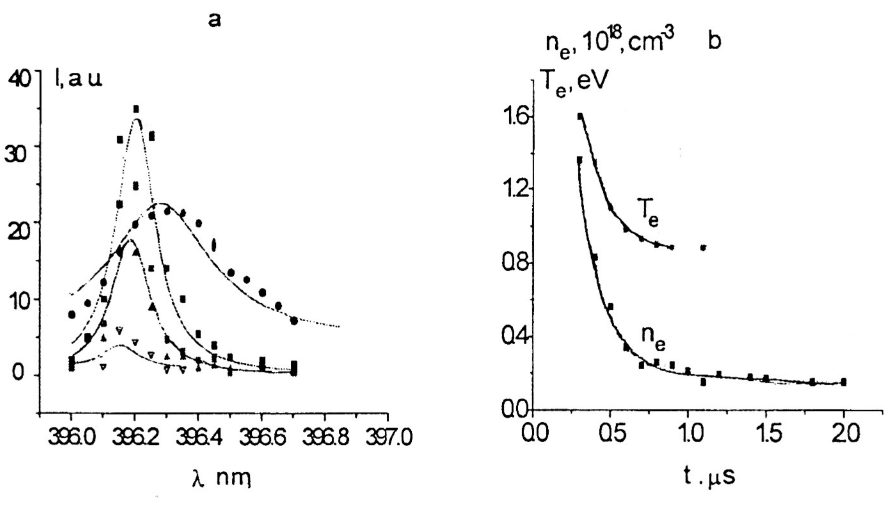

Fig.1. (a) AlI 396.15nm line Stark profiles at delays 0,4(•), 0,7(■), 1,0(Δ), and 2 μs (∇) after the ablating laser pulse and (b) time evolution of electron density (n_e) and temperature (T_e) in aluminum/air plasma at distance of 0.5mm from the target and laser irradiance of 500 MW/cm^2.

The temporal evolution of n_e for each distance can be characterized by an approximately biexponential decrease. The initial more fast decay rate can be attributed to the propagation of the recombining plasma, while the slow decay is preferably due to the recombination processes. The electron temperature rapidly decreases in the time interval where maximal line emission is observed. After this time T_e is not practically changed with time, showing that there is a quasistationary equilibrium between the rates of cooling of electrons and their recombination heating.

The similar behavior of recombination dynamics with a very slow recombination of ions with electrons in the late stage has been observed in recombining titanium/air laser produced plasma. The characteristic recombination time essentially exceeded the gasdynamical expansion one. So, plasma has had time to fly apart without a complete recombination. It is supported by the temporal dependence of relative concentrations of ions and atoms as determined by fluorescence measurements. Such situation in recombination dynamics called "frozen" ionization is caused by decreasing of electron density and delivering the kinetic energy to them in the recombination process and as result there exist a tendency to maintain the plasma temperature.

Relaxation dynamics of the excited atomic states of Al after action of the excimer XeCl laser radiation (λ=308nm) on the Nd:YAG laser-ablated aluminum plume was studied by measuring the time histories of the AlI 396.15nm emission line. The relaxation rates were found to be dependent on distance from the target and XeCl laser pulse delays. The more shorter duration of the AlI 396.15nm signal, its faster decrease at longer delays and the change of the spectral shape (Fig.2) were observed. The results obtained indicated that the resonant photoionization of the Al atoms via $3^2D_{3/2}$ level with radiative recombination (predominantly at short delays), and collisional deexcitation of laser excited atomic level ($3d^2D_{3/2}$) with the excitation transfer to the adjacent $4p^2P^0$ levels resulting in the cascade of emission transitions $4p^2P^0$-$4s^2S$-$3p^2P^0$ were the most probable mechanisms of populating of Al ($4s^2S_{1/2}$) level.

So, time-resolved spectroscopic diagnostics of the recombining laser produced plasma allowed us to make some conclusions concerning character of relaxation of its parameters (n_e, T_e, n_a). The investigations showed, that it possible to distinguish two main stages in the time and space evolution of laser-induced plasma in gas environment. The first stage is associated with a plasma expansion. Plasma evolution in this stage is ruled mainly by processes with charged species (ionization by electron impact, and three-body electron-ion recombination). At the end of this stage plasma involving

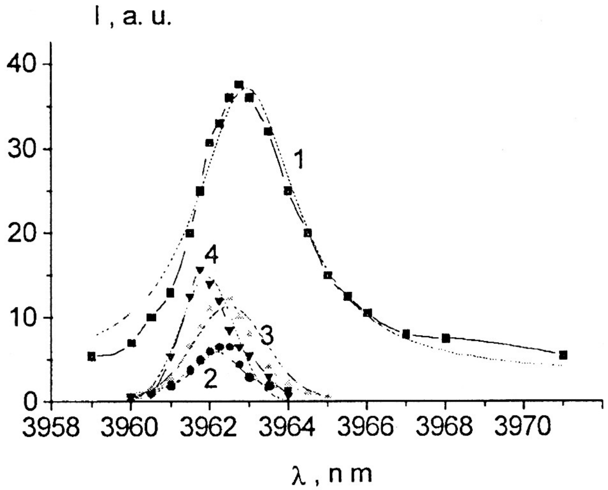

Fig.2. The XeCl laser produced AlI 396.15nm emission line profiles at delays 3 (2), 20 (3) and 46 μs (4) after the ablating Nd:YAG laser pulse and the AlI 396.15nm line profile from the Nd:YAG laser produced plasma in 150 Topp He atmosphere at the Nd:YAG laser irradiance of 500MW/cm^2 and the XeCl laser radiation intensity of 60MW/cm^2.

electrons and multicharged ions is turned to plasma consisting of atoms (molecules) and one-charged ions (predominantly in ground states). The processes that govern the ablation plume-gas dynamics in this stage are chemical kinetics of the ablated species with background gas, material or thermal diffusion in background gas, and cluster/particulate formation (condensation) as plasma cools.

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References

Schriever G., Mager S., Naweed A., Engel A., Bergman K., and Lebert R.: 1998, *Appl. Optics* **37,** 1243.

Ohyanagi T., Miyashita A., Murakami K., and Yoda O.: 1996, *Jpn.J. Appl.Phys.* **35**, 3436.

Burakov V.S., Bokhonov A.F., Naumenkov P.,A., Nedel'ko M.I., and Tarasenko N.V.: 1998, *Zh. Prikl. Spectrosk.* **65,** 426.