Plasma properties of pulsed laser ablation of GaAs*

Man Bao-Yuan[1], Wang Xiang-Tai[2], Hu Xie-Rong[1], Guo Xiang-Xin[2] and Miao Yong[2]
([1] *Infrared and Remote Sensing Laboratory, Shandong University, Jinan 250100;*
[2] *Department of Physics, Shandong Normal University, Jinan 250014*)

Abstract Simultaneous time and space-resolved emission spectrum from a Nd: YAG laser-ablated plasma on a GaAs target has quantitative information on the generation and the propagation of species in the plasma at 1 atm or lower pressures. The experimental results show that the characteristics of the species in plasma are strongly influenced by the ambient pressure. The time-of-flight distributions of Ga atoms exhibit twin-peak structure, which can be explained by recombination process. The mean projected ranges of As^+ ions in the atmosphere of different densities were calculated according to Ziegler' theory model and the slowing down processes of them were simulated by using a Monte Carlo simulation program TRIM90. The calculated results are consistent with the experimental.

Keywords Laser plasma, Time-resolved spectroscopy, Recombination, Projected range.

1 Introduction

Laser ablation of material has proved to be an useful technique and has been widely used in the field of scientific research, industry, medical treatment and so on.[1–4] Pulsed Laser Deposition (PLD) is a new and powerful technique for preparing super conducting thin films.[5–8] The interaction of the laser with the target material involves complex processes. At present, it is confronted with some difficulties to describe this fast process by using existing physical models. Therefore, it is essential to perform more experiments for better understanding of the interaction between the pulsed laser and material. Because laser ablation can be influenced to a great extent by ambient atmosphere and pressure,[9–12] the study on the process is of great interest in dynamic study of laser ablation and practical use such as preparation of films by using PLD. In the present work, a time and space resolved diagnostic technique was used to study the emission spectrum from the plume produced by a pulsed laser on a GaAs target irradiated at 1 atm or lower pressure. The time-of-flight (TOF) distributions of Ga atoms in plasma include information on the velocities of the Ga atoms and show the twin-peak structures, which can be explained by the recombination process. A theory model given by Ziegler *et al*[13] was used to calculate the projected ranges for 1~15 keV As^+ ions ejecting into ambient gas at different pressures. Moreover a popular Monte Carlo simulation program TRIM90 was used to simulate the slowing down process of the As^+ ions. Calculated results give the reason of disappearance of As^+ signal at an atmospheric pressure.

2 Experimental arrangement

The arrangement of the experimental apparatus is schematically shown in Fig.1. The pulsed laser was reflected by three prisms M1, M2 and M3 (used as optical delay path), and then was focused onto the polished GaAs target surface by a quartz focus lens L1 (f=6.3 cm). The spot size of the laser beam is 0.33 mm in diameter. The GaAs sample was positioned on a sample holder in a chamber pumped by a mechanical pump. The chamber and lens L1 were mounted on two-dimensional ($x-y$ directions) movable plates, respectively. The y direction is in the laser beam axis. In the x direction, two cylindrical lenses (L2 and L3) were used to image (1:1) the plasma optical emission from the plane, which has a distance d from the surface of the target, onto the entrance of the slit of a ISA(HR-320) spectrum analyzer. The dispersed emission was subsequently detected by a 10 ns gated, image-intensified optical multichannel analyzer (PARC OMA III). An accurate trigger delayer, which was synchronistically

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triggered by the electric Q-Switched signal produced from the Quanta Ray DCR-3 laser device, was used to control the delay time, at which the spectrum was recorded. By changing the distance d and the delay time t_d, one can get the space time-resolved emission spectra.

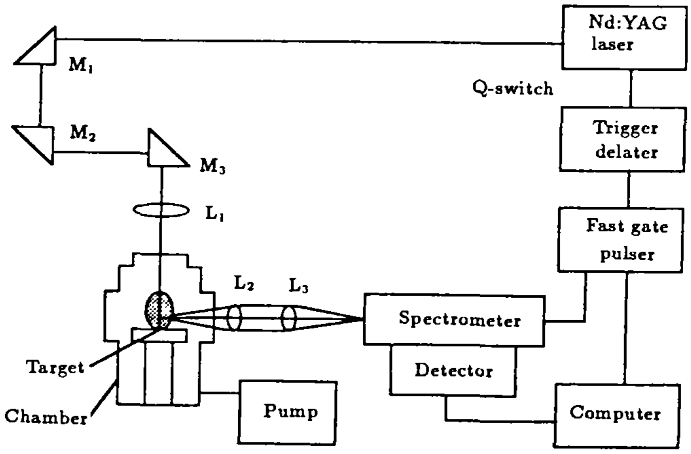

Fig.1 Schematic diagram of the experimental setup

3 Results and discussion

Fig.2 is the temporal evolution of the spectra obtained in the wavelength range 395∼435 nm at $d = 0.75$ mm. The laser power density is 9×10^9 W·cm^{-2}, and the ambient pressures are (a) 1 atm and (b) 5 Pa respectively. From Fig.2(a), it is observed that an intense continuum emission is produced instantaneously after the pulse laser shots the target surface and then gradually disappears at about 200 ns following the laser pulse shot. As known, the continuum is related to the strong collision between the free electrons or the excited atoms or ions (free-free emission, known as the bremstrahlung), and the recombination of electrons with ions (free-bound emission)[14]. Thus, it can be concluded that there must be a lot of electrons, excited atoms and ions in the ejected vapor near the laser ablated surface area. The process of collisions between the free electrons or excited atoms or ions must take place, and the recombination of electrons with ions is existed as well. At about 30 ns the spectrum of N(II) line at 399.5 nm superposed on the continuum emerges, it also quickly disappears because of the recombination of electrons with ions. At first, the emitted species density is very high, N^+ ions will be impacted frequently by other species, so the line is broadened. With the increase in the delay time, the line becomes narrower. Since the processes of the N(II) line at 399.5 nm and the continuum exist in the same interval, it can be deduced that the N^+ ions are resulted from collisions between nitrogen atoms and species in plasma. From Fig.2(b), it is observed that the signal of N^+ ions disappears at 5 Pa because of the reduction of the density of nitrogen atoms. In Fig.2(a) and (b), it can also be found the spectra of the Ga(I) lines at 403.3 nm and at 417.2 nm. Fig.3 shows the TOF curves of excited Ga atoms on a GaAs sample irradiated by the pulse laser at a flux of 9×10^9 W·cm^{-2}. These curves were obtained by integrating the Ga(I) 403.3 nm line. Because the concentration of an electron state of the excited atoms or ions is proportional to the intensity of the spectral line, the recorded changes in the intensity of the spectral lines at different delay time give the evolution information of the excited Ga atoms in the ejected vapor.

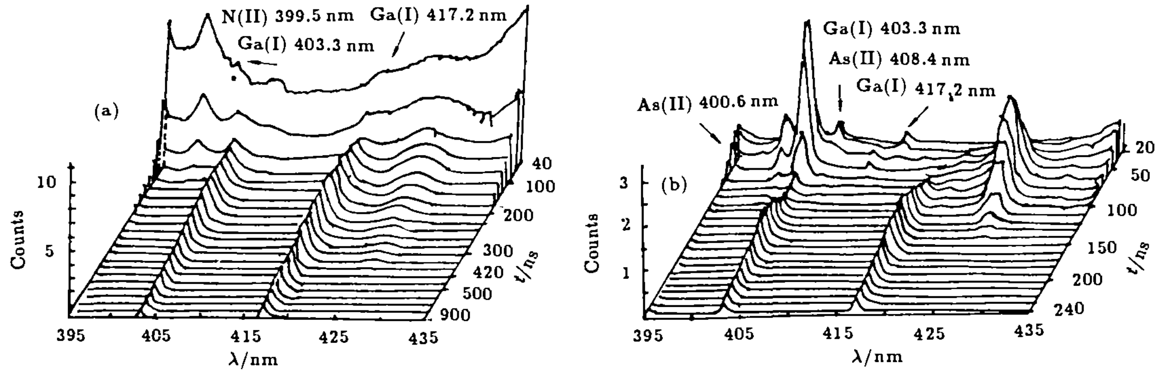

Fig.2 The temporal evolution of the spectra in wavelength ranging from 395 nm to 435 nm, recorded at d=0.75 mm. The ambient pressures are (a) 1 atm (b) 5 Pa, respectively

In Fig.3, the lines obtained at 1 atmospheric pressure last for a long time up to 2000 ns, which is much longer than the duration of the excited state of Ga atom. So it can be concluded that Ga atoms are generated and excited constantly during traveling to the laser source. Moreover, the lines last for much longer at 1 atm than at 5 Pa, the reason for this result is that, at 1 atmospheric pressure and the present laser intensity, a Laser-Supported Detonation (LSD) wave was ignited[15]. Because of the ignition of the LSD wave, the temperature of the plasma was higher and the duration was longer. Right now, ejected Ga atoms were excited constantly by absorbing energy from the heated plasma. As the ambient pressure decreases to 5 Pa, the LSD wave was hardly ignited, so the integrating intensity of the Ga(I) 403.3 nm lines was weaker and the duration was shorter than at 1 atm. However, the ejected distance of Ga atom at 5 Pa is longer than at 1 atmosphere pressure due to the reduction of air resistance. In addition, one can find the twin peak distributions of Ga atoms from Fig.3, but cannot find those at 1 atmospheric pressure. Moreover, in Fig.3, the twin-peak distribution was obtained only at d >0.5 mm. The most plausible explanation is that the twin-peak distribution stems from two different origins of Ga atoms in plasma. One kind of Ga atoms comes directly from the target surface and have larger TOF time delays corresponding to the second peak. The other kind of Ga atoms generated away from the target surface is due to recombination between electrons and Ga ions, which occur earlier giving rise to the first peak. Because the time corresponding to the second peak increases with the distance d more quickly than that to the first peak, the time difference between the two peaks increases with the distance d. In the vicinity of the target surface, the time difference is so small that the two peaks can not be separated. When d increases to 0.5 mm, the two peaks begin to be separated, but the first peak is much smaller than the second. At d = 1.0 mm, the twin-peak distribution is very obvious, and the first peak is greater in intensity but narrower in width than the second. Another reason for finding only one peak in the vicinity of the target surface is that no the recombination exists near the surface. The recombination process occurs only at d >0.5 mm, and the intensity of the first peak for the process is the greatest at d=1.0 mm. At 1 atmospheric pressure it is difficult to separate the two peaks because the distribution of TOF is very wide. We also observed that the twin-peak structure occurs only at d=1.0 mm for the TOF distribution of Ga atoms at 1000 Pa, and the two peaks is very closed. Each peak of the TOF curves corresponds to a delay time. Dividing the distance between two position by the corresponding delay time difference gives the average velocity of excited Ga atoms, which can be approximately regarded as the velocity at the center of the two positions (see Table 1).

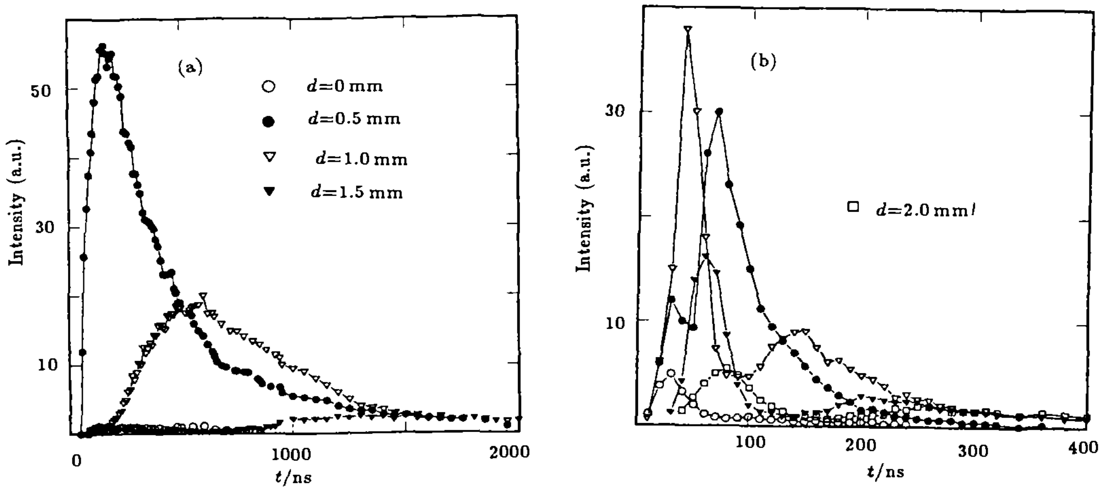

Fig.3 The TOF curves obtained from Ga(I) 403.3 nm line at different d and at the pressures of 1 atm (a) and 5 Pa (b)

Table 1 The traveling velocities of the excited Ga atoms at different positions and pressures

d/mm	Velocities of peaks/m·s^{-1}		
	1 atm	5 Pa	
		Peak(I)	Peak (II)
0.75	1.4×10^3	5.0×10^4	1.7×10^4
1.25	5.1×10^2	5.0×10^4	1.0×10^4
1.75	–	5.0×10^4	7.1×10^3

From Table 1, it can be found that the velocities for the first peak of the excited Ga atoms are constant at the position 0.75~1.75 mm away from the target surface, and the velocity is greater than those for the second peak. The velocities for second peak decrease with increase of the distance from the surface. Moreover, the velocities at 1 atm decrease with increase of d more rapidly than in vacuum. The velocities of Ga atoms may be slowed down by the collisions between air particles and the ejected Ga atoms, and this deduction is consistent with the conclusion given by Sappy.[16] Two As(II) lines at 400.6 nm and at 408.4 nm can be found in Fig.2(b) , but disappear in Fig.2(a). The spectrum of the two lines is very obvious as the ambient pressure decreases to 10^3 Pa.

In order to find the reason of disappearance of As^+ ions signal in Fig.2(a), the traveling range of As^+ ion in the ambient gas is estimated. The energy of As^+ ions can be evaluated to be in the range of 1~15 keV by measuring the velocities of As^+ atoms. We can estimate the range of 1~15 keV As^+ ion traveling in the ambient gas by using the theory given by Ziegler *et al.*[13] The calculated result is shown in Fig.4. Further more, a Monte Carlo simulation program TRIM 90 is used to simulate the slowing down process of As^+ ions (see Table 2). Fig.5 shows the projected range distribution of 10 keV As^+ ions at 1 atm.

Table 2 The projected range of distributions of 10 keV As^+ ions

Pressure	1 atm	10^3 Pa	5 Pa
Range $<R>$/μm	19.6	19600	9.8×10^5
Straggling ΔR/μm	4.9	4900	2.45×10^5

From Fig.5 and Table 2, it can be clearly seen that the background pressure has an important influence on the projected ranges and the range distribution for As^+ ions in plasma induced by laser ablation. At 1 atm the mean projected range and the range straggling are 19.6μm and 4.9μm, respectively and the signals of As^+ ions can not be detected at vertical viewing angle. But at 10^3 Pa, the projected range and the straggling are 19.6 mm and 4.9 mm, respectively and the signal of As^+ ions can be easily detected. The projected range and the straggling increase with decreaes in the pressure. However, as the pressure decreases to

5 Pa, the signal intensity observed at a position far from surface is too small to be detected.

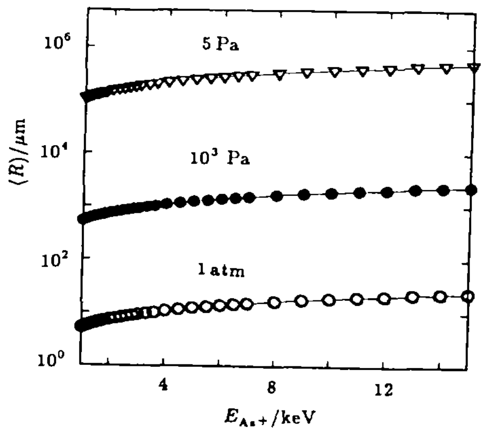

Fig.4 The mean projected ranges for 1~15 keV As^+ ions in air at three kinds of pressures

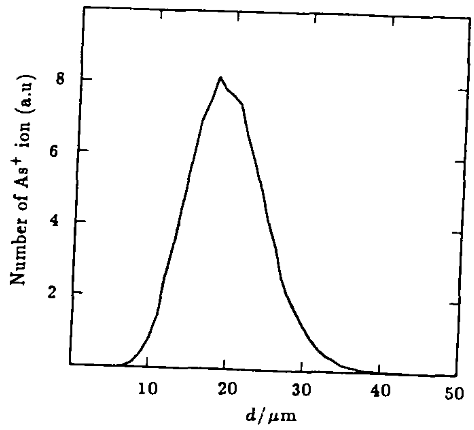

Fig.5 The projected range distribution for As^+ ions in air at 1 atm

4 Conclusion

A time and space resolved diagnostic technique was used to study the emission spectrum from the plume produced by a pulsed laser on a GaAs target irradiated at different ambient pressures. The results show that energetic plasmas were generated at $1.06\,\mu m$ from the surface of GaAs target irradiated at a laser flux $9\times10^9\,W\cdot cm^{-2}$; the generation, propagation velocities and projected ranges show a strong dependence on the ambient pressure. The time evolution of the spectral emission clearly reveals the TOF distribution of Ga atoms with two peaks, which is due to the recombination process. In addition at 1 atm, the projected range of an As^+ ion is so short that the signal cannot be detected at 90° angle to the laser axis because of the strong stopping power of air. At lower ambient pressure the spectrum of As^+ ions can be easily detected, because the projected range is longer while the signal intensity is weaker. Thus, an appropriate ambient pressure should be chosen to make the ejected ions reach to certain area in the control of PLD.

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