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Spectroscopy Diagnostic of Laser Intensity Effect on Zn Plasma Parameters Generated by Nd: YAG Laser

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Abstract

The creation and characterization of laser-generated plasma are affected by laser irradiance, representing significant phenomena in many applications. The present work studied the spectroscopy diagnostic of laser irradiance effect on Zn plasma features created in the air by a Q-switched Nd: YAG laser at the fundamental wavelength (1064nm). The major plasma parameters (electron temperature and electron density) have been measured using the Boltzmann plot and the Stark broadening methods. The value of electrons temperature ranged from 6138–6067 K, and the electron density in the range of 1.4×10^{18} to 2×10^{18} cm^{-3}, for laser irradiance range from 2.1 to 4.8×10^{8} (W/cm^{2}). The McWhirter criterion for preserving a local thermodynamic equilibrium (LTE) condition is confirmed in this study. Furthermore, other fundamental plasma parameters, such as Debye length (λ_D), number of particles in Debye sphere (N_D), and plasma frequency (ω_p), were measured. We found that all plasma parameters are affected by laser intensity.

Keywords: Zn plasma, Spectroscopy diagnostic, Plasma parameters, LET.

التشخيص الطيفي لتأثير شدة الليزر على معاملات بلازما الزنك المتولدة بواسطة ليزر النديميوم ياك

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الخلاصة

يتأثر توليد وتوصيف البلازما المنتجة بالليزر بأشعاع الليزر، مما يمثل ظاهرة مهمة في العديد من التطبيقات. في العمل الحالي، تمت دراسة التحليل الطيفي لتأثير إشعاع الليزر على خصائص بلازما الزنك التي تم إنشاؤها في الهواء بواسطة ليزر النديميوم ياك عند الطول الموجي الأساسي (1064 نانومتر). تم قياس معلمات البلازما الرئيسية (درجة حرارة الإلكترون وكثافة الالكترون) باستخدام طرق مخطط بولتزمان و توسيع ستارك. تراوحت قيمة درجة حرارة الإلكترونات بين 6138−6067 كلفن، وتراوحت كثافة الإلكترون من 1.4×10^{18} إلى 2×10^{18} (سم$^{-3}$) ، في مدى إشعاع الليزر من 2.1 إلى 4.8×10^{8} (واط/سم ^{2}). تم تأكيد معيار McWhirter للحفاظ على حالة توازن ديناميكي حراري محلي (LTE) في هذه الدراسة. بالاضافة الى ذلك، تم قياس معلمات البلازما الأساسية الأخرى، مثل طول ديباي (λ_D)، وعدد الجسيمات في كرة ديباي (N_D)، وتردد البلازما (ω_p). وجدنا أن جميع معاملات البلازما تتأثر بشدة الليزر.

1. Introduction

Laser-Induced Breakdown Spectroscopy (LIBS) is a versatile methodology that analyses

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the sample in its original form without changing its chemical composition[1]. LIBS is an emission spectroscopic technique used to characterize the elements in materials, and it has a wide range of applications. A plasma plume is formed when a high-intensity laser pulse is ignited on a sample's surface, ablating it. The material's ablated mass is in the nanograms to micrograms range [2]. LIBS is based on studying spectral lines emitted by the plasma generated when a sample surface is irradiated with a high-intensity focused laser beam, resulting in dense hot plasma at the surface. The resulting plasma contains atoms and ions in various excited states that emit radiation when they transit to lower energy states [3]. Optical emission from laser-generated plasmas has been studied extensively in recent years, and the atomic and ionic line emission was estimated using the laser interaction with the sample target [4]. A spectrometer was used to record plasma emissions, analyzing them to determine the elements' identities and concentrations. Diagnostics of plasma may be performed by estimating plasma parameters, such as electron temperature and density. The plasma characterization is affected by laser parameters (wavelength, laser intensity, pulse duration, time delay), experimental conditions, and target properties [2,3,4]. Many experimental research is available in literature discussing the plasma parameters, including laser energy effect, wavelength dependency, spatial and temporal behaviour. Afgan *et al.* (2018) [5] performed a spectroscopic characterization of thallium plasma using an Nd: YAG laser at 1064 nm. Plasma temperature and electron density had been evaluated with laser irradiance. Wang *et al.* (2018)[6] studied the effect of laser energy and wavelength on detecting trace elements in alloy steel at 1064 nm and 532 nm wavelengths.

The purpose of this work is to investigate the effect of laser irradiance on LIBS emissions, electron temperature, density, and other basic plasma parameters that depend on T_e and n_e using emission spectroscopy-based characterization techniques.

2. Materials and Methods

Figure 1 shows the experimental setup of the LIBS. In this experiment, plasma is generated utilizing nanosecond Q-switched Nd: YAG laser at a fundamental wavelength of 1064 nm with 10 ns pulse duration, 6 Hz repetition frequency, and laser intensity ranging from 2.1 to 4.8×10^{8} W/cm^{2}. The purity of the target was 99.9999% Zinc (Zn nanoparticle). The Zn powder was pressed into pellets by applying 80 MPa pressure using a hydraulic press for 10 min. The laser beam was focused, utilizing a quartz lens with a 10 cm focal length, on the target's surface at atmospheric pressure. Optical fibre, with a 50 μm diameter core, collected the laser-generated plasma light emissions from the Zn target surface. The fibre was placed 1 cm above the sample surface; it was connected to a Surwit (S3000-UV-NIR) spectrometer to analyze the plasma emissions.

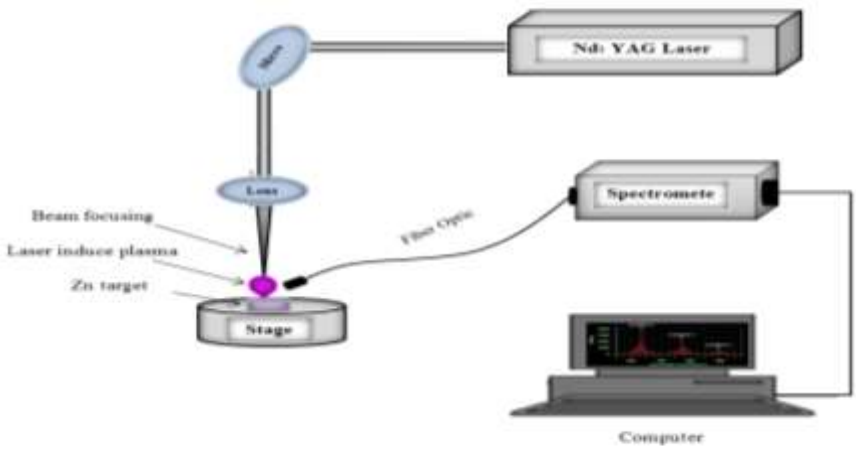

Figure 1- Schematic diagram of LIBS system.

3. Results and Discussion

3.1. Spectroscopy diagnostics

The Zn plasma generated by Nd: YAG laser was characterized by optical emission spectroscopy. The effect of laser intensity on plasma parameters was studied; laser irradiance is one factor affecting the intensity of plasma emission. When excited species decay in the plasma, optical emissions are produced represented by the emission spectra of Zn plasma. The results of Zn plasma optical emission lines were calibrated with NIST database software [7] to estimate the plasma characteristics, as listed in Table 1.

Table 1- Spectroscopy parameters of the spectral lines emitted from Zn plasma corresponding to the NIST database [7].

Spectral lines	λ (nm)	Transitions	g_iA_{ij} (s^{-1})	E_j (eV)
Zn II	276.3	$3d^9 4s^2\ ^2D_{3/2} \rightarrow 3d^{10} 5p\ ^2P_{3/2}$	7.6×10^6	14.629
	491.1	$3d^{10} 4d\ ^2D_{3/2} \rightarrow 3d^{10} 4f\ ^2F_{5/2}$	1.09×10^9	14.195
	602.1	$3d^{10} 5p\ ^2P_{1/2} \rightarrow 3d^{10} 5d\ ^2D_{3/2}$	3.00×10^8	14.195

The intensity of the LIBS signal increased as the laser intensity increased at a laser irradiance range of 2.1 to 4.8×10^8 (W/cm^2), as shown in Figure 2. As laser irradiance increases, the plasma's absorption of laser light increases, leading to an increase in ablation rate and a rise in spectral line intensity [8]. The Zn plasma spectra covered the region from 250 to 700 nm, including the emission of 9 spectral lines upon excitation by the 1064 nm wavelength at different laser irradiance. The spectral lines consist of 4 ionic lines for Zn II at 255.7, 276.3, 491.1, 602.1 nm, and 5 atomic lines for Zn I at 334.5, 466.5, 468.0, 636.2, 647.9 nm.

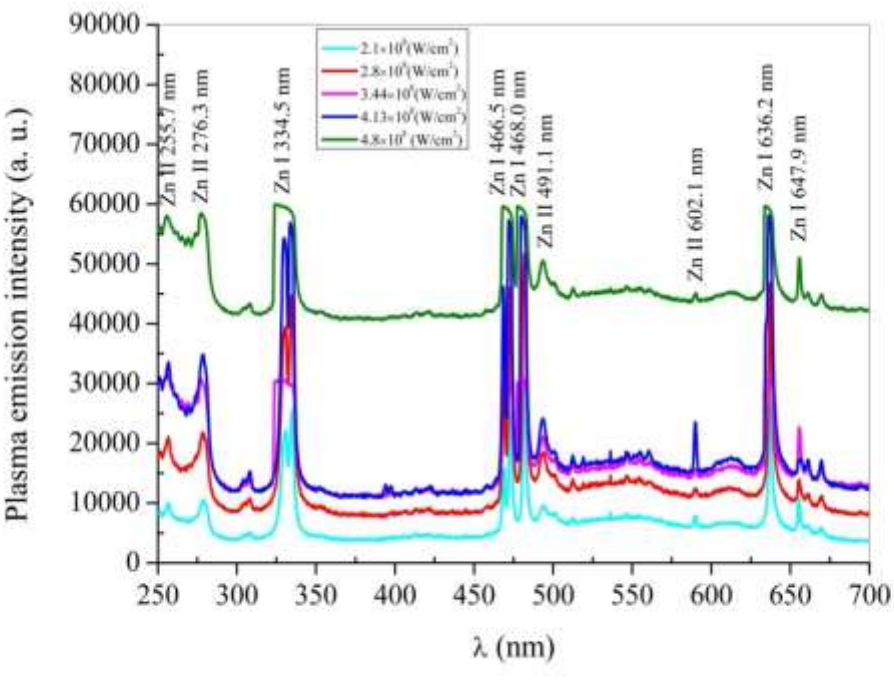

Figure 2- Zn plasma emission spectra induced by 1064 nm laser at different laser irradiance

3.2. Laser plasma characterization

The spectral lines released by Zn plasma can estimate plasma parameters such as electron temperature (T_e), electron density (n_e), plasma frequency (ω_p), Debye length (λ_D), and Debye sphere (N_D). T_e and n_e are essential parameters for understanding plasma excitation and atomic ionization processes.

3.3. Electron temperature

The electron temperature of the Zn plasma was measured using the Boltzmann plot method as shown in Equation (1) [9], assuming local thermodynamic equilibrium (LTE) condition:

$$Ln(\lambda_{ji} I_{ji} / g_j A_{ji}) = -(E_j / k_B T) + C \quad \text{…….. (1)}$$

Where: λ_{ji} is the laser wavelength, g_i is the statistical weight, A_{ji} is the probability of transition, E_j is the upper energy obtained from a standard spectrum of NIST database [7]. Three ionic lines were used to apply the Boltzmann plot method, as listed in Table 1 for Zn plasma. The electron temperature was calculated from the slope of the graph of $Ln(\lambda_{ji} I_{ji} / g_j A_{ji})$ versus the energy E_j (eV), as shown in Figure 3. The slope of the straight line was equal $(-1/k_B T)$.

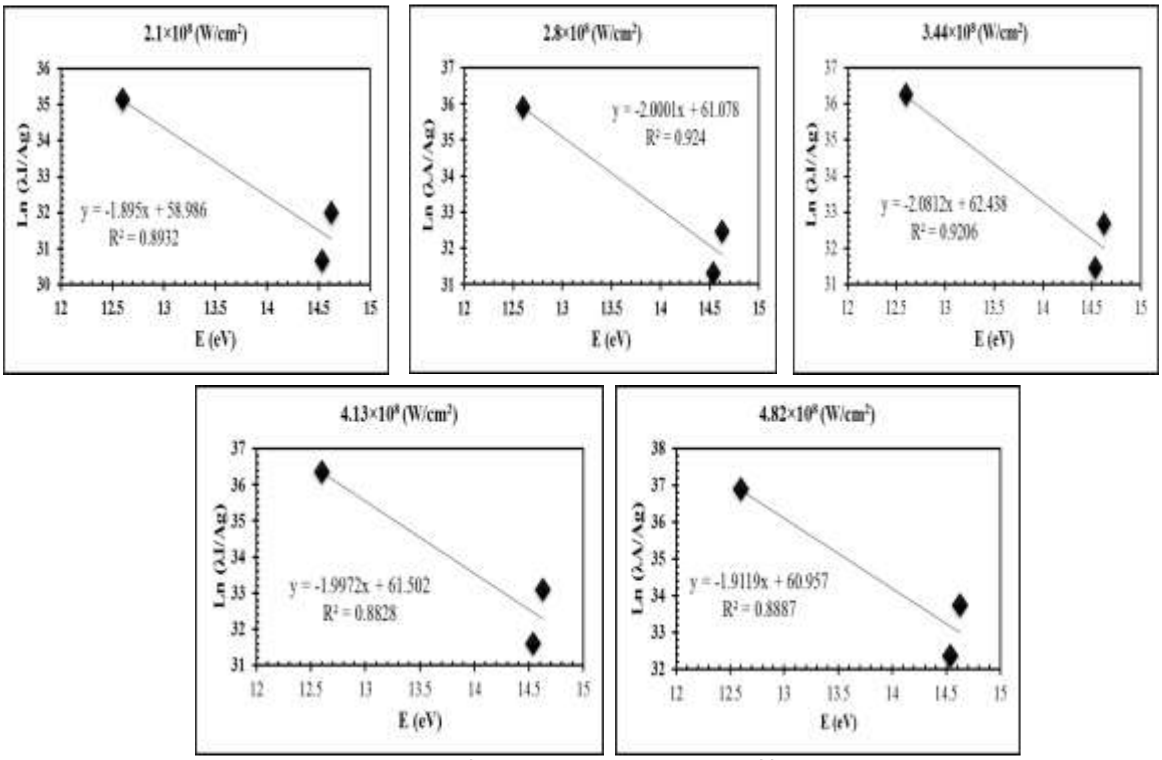

Figure 3-Boltzmann plots for the Zn plasma at different laser irradiance

The electron temperature for Zn plasma was calculated depending on laser intensity, ranging from 2.1 to 4.8×10^8 (W/cm^2). The value of T_e range was (6138-6067) K, as tabulated in Table 2.

Table 2-Data analysis from the Boltzmann plots with the linear fitting of R^2 and electron temperature at different laser intensity

Laser intensity ×10^8 (W/cm^2)	R^2	Slope	Te (K)
2.1	0.8932	-1.895	6138
2.8	0.924	-2.0001	5800
3.44	0.9206	-2.0812	5574
4.13	0.8828	-1.9972	5808
4.82	0.8887	-1.9119	6067

Figure 4 shows a nonlinear change in electron temperature against laser irradiance. The electron temperature dropped with increasing the laser intensity and then increased. When plasma reaches maximum expansion velocities, thermal energy is rapidly converted to kinetic energy, leading the plasma temperature to drop; this finding agrees with that of Ahmed et al. [10].

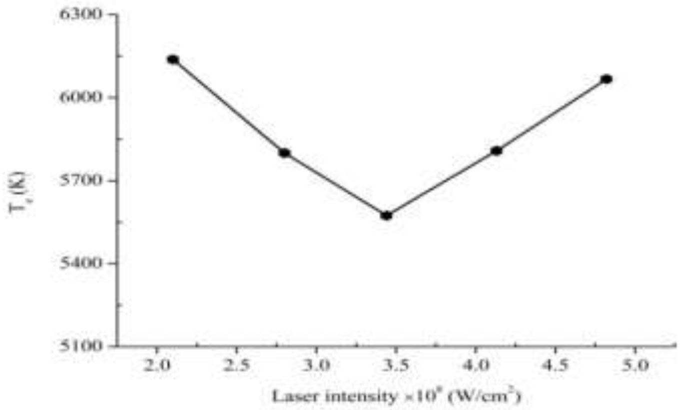

Figure 4-The T_e at different laser intensity

3.4. Electron density

The laser-generated Zn plasma's emission spectra showed broadened lines. Collisions between the released atom and charged particles result in Stark broadening, which is the primary effect on these emission spectra; consequently, the electron density may be calculated using the widths of the spectral lines through the relation[11]:

$$n_e\ (cm^{-3}) = \left(\frac{\lambda_{FWHM}}{2\omega}\right) \times\ 10^{16} \qquad (2)$$

Where: n_e is the electron density, λ_{FWHM} is the Stark full-width at half-maximum (FHWM), and ω is the electron impact parameter.

The electron density was calculated from spectral lines of Zn II at 602.1 *nm* for various laser irradiance. The electron density ranged from 1.4 to $2\times10^{18}cm^{-3}$, as stated in Table 3. Figure 5 shows that the electron density increases as laser intensity increases. The results demonstrated the slow variation of n_e; this behaviour is due to the plasma shielding effect, i.e., plasma reflection of laser light. Because the experiment was conducted at atmospheric pressure, the ionization process was limited by the plasma shielding effect of air, reducing the efficiency of the laser energy available for mass ablation.

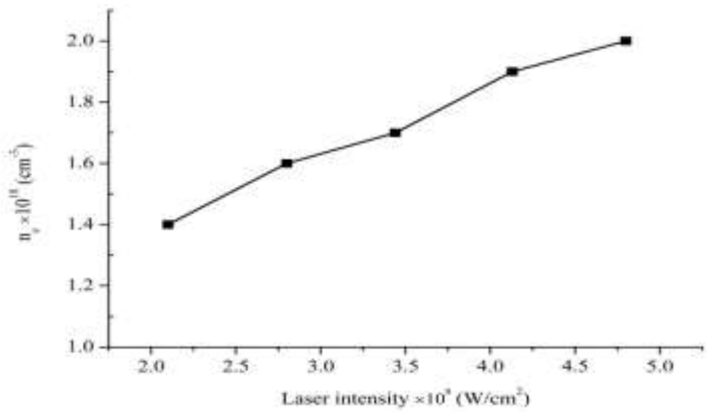

Figure 5-The electron density of Zn plasma at different laser intensit

3.5. The verification of local thermodynamic equilibrium (LTE)

Local Thermal Equilibrium (LTE) means that particles have Maxwellian energy distributions, and collisional processes dominate radiative processes in laser-generated plasma.

The condition that determines the critical electron density required to achieve McWhirter criterion is [11]:

$$N_e \geq 1.6 \times 10^{12} \Delta E^3 T^{1/2} \qquad \ldots..(3)$$

Where: T_e and N_e are the temperature and density of the electron, respectively, ΔE is the energy gap between the upper and lower energy levels of the corresponding spectral line. Due to the relatively high electron density, $N_e \geq 10^{16}$ (cm^{-3}) [8], it was assumed that the plasma is in a state of local thermal equilibrium (LTE) and that the species distribution has a Boltzmann form. In our study, the maximum value of electron temperature was 6138 K, and the energy difference between the top and the bottom level was 2.518 eV. The electron density measured using the Stark broadening method was 2.11×0^{18} cm^{-3}. The electron density measured using the Mcwhirte criterion was 7.8×10^{16} cm^{-3}, and this value is far less than the electron density measured using the Stark broadening method. As a result, the local thermal equilibrium (LTE) system was stated throughout our study.

3.6. Characterization of other plasma parameters

In order to better characterize the Zn plasma, the influence of laser intensity on other fundamental plasma parameters such as Debye length (λ_D), Debye sphere (N_D), and plasma frequency (ω_p) was determined using T_e and n_e. λ_D, N_D, and ω_p are given by equations 4, 5, and 6, respectively [12, 13].

$$\lambda_D = \left(\frac{\varepsilon_0 k_B T_e}{n_e e^2}\right)^{\frac{1}{2}} \qquad \ldots..(4)$$

$$N_D = \frac{4\pi}{3} \lambda^3{}_D n_e \qquad \ldots..(5)$$

$$\omega_p = \left(\frac{n_e e^2}{\varepsilon_0 m_e}\right)^{1/2} \qquad \ldots..(6)$$

Where: ε_0 is the permittivity of free space, k_B is Boltzmann constant, e is the charge of the electron, n_e, is the electron density, and m_e is the mass of the electron. The obtained results are listed in Table 3, indicating that λ_D and N_D were reduced with the increase of laser intensity. On the other hand, ω_p increased with increased laser irradiance.

Table 3- Plasma parameters for the Zn plasma plum induced by 1064 nm laser at various laser irradiance.

Laser intensity $\times 10^8$ (W/cm^2)	FWHM (nm)	$n_e \times 10^{18}$ (cm^{-3})	$\lambda_D \times 10^{-5}$ (cm)	$N_D \times 10^3$ (cm^{-3})	$\omega_p \times 10^{12}$ (Hz)
2.1	1.80	1.4	0.465	0.569	10.434
2.8	2.15	1.6	0.412	0.473	11.403
3.44	2.25	1.7	0.395	0.435	11.665
4.13	2.50	1.9	0.382	0.439	12.296
4.82	2.70	2	0.376	0.451	12.779

Figure 6 shows the relationship between Debye length (λ_D) and laser intensity; as the laser intensity increased λ_D became shorter because more light was absorbed in the material. This result agrees with that of Aadim et al. (2021) [13].

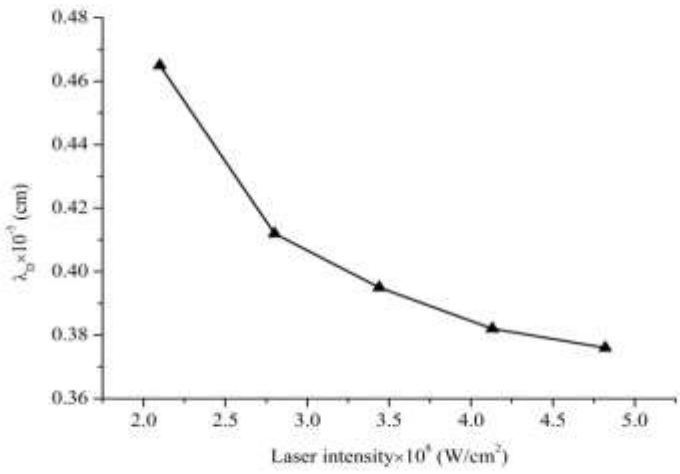

Figure 6- Debye length of Zn plasma at different laser intensity

The number of particles in Debye's sphere (N_D) at various laser intensities is shown in Figure 7. As a function of laser intensity, N_D and λ_D exhibit a similar behavior due to their direct relationship, and these results agree with those of Maryam and Kadhim (2021) [14].

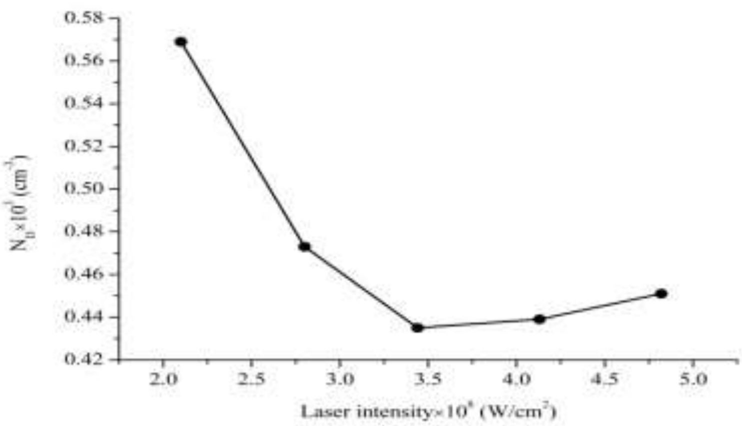

Figure 7- Debye number of Zn plasma at different laser intensity

Figure 8 shows that the plasma frequency (ω_p) increased as the laser intensity increased because higher irradiance produces more ablated material, which leads to more plasma species.

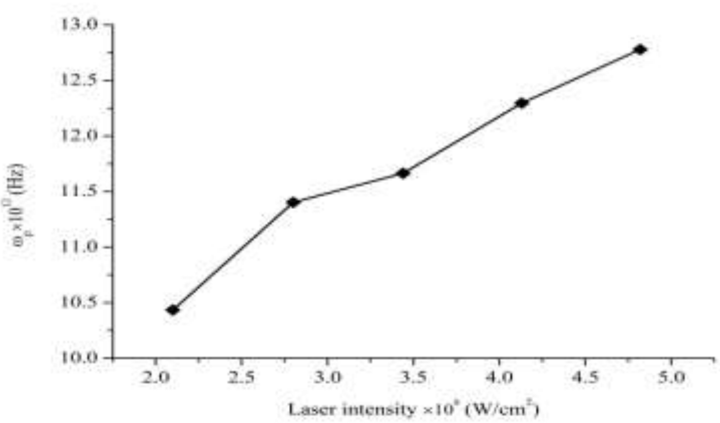

Figure 8- Plasma frequency of Zn plasma at different laser intensity

4. Conclusion

The effect of laser intensity on the Zn plasma parameters was studied. Changes in laser intensity were observed to significantly affect optical emission spectra, electron temperature, electron density, and other basic plasma parameters. There was a nonlinear change in electron temperature with laser intensity for Zn plasma; electron temperature fluctuation was due to plasma expansion. The broadening of spectral lines appeared with increased laser intensity due to collisions of the emitted atom with charged particles causing the Stark line broadening, and this broadening was related to electron density. In contrast, a linear increase in electron density with laser intensity was found due to mass ablation rate increase.

Acknowledgments

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