Diagnostics of Magnesium-Aluminum alloy plasmas produced by laser induced breakdown spectroscopy

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Abstract
In this work, the Al-Mg alloys plasma plume that produced under vacuum (10^{-1} torr) by Nd:Yag Laser was studied. the plasma spectra were analyzed by comparing the atomic lines of Al and Mg lines with standard lines. The effect of laser energies on spectral lines produced by laser ablation were investigated using optical spectroscopy. several plasma characteristics like electron temperature, electron density, Debye’s length, plasma frequency and number of particles in the “Debye’s Sphere” were calculated in vacuum. The results show the electron temperature and electron density increase in magnesium, aluminum and magnesium :aluminum alloy targets. It was found that the lines intensities at different laser peak powers increase when the laser peak power increases then decreases when the power continues to increase.

Keywords: Vacuum; Nd: YAG laser Laser-induced plasma; Plasma characteristics

Introduction
Laser-induced breakdown spectroscopy (LIBS) is an analytical technique based on the application of one or more high power laser pulses on a reduced region of the sample surface, promoting ablation and excitation of the specimen with the formation of a transient plasma. In the technique, the emitted radiation is associated with the sample chemical composition and is monitored by means of an appropriate detection system (wavelength selector and detector)[1]. Due to the advantages of LIBS,
including short measurement time, versatility in sample types, minimal-preparation with no pretreatment or digestion procedures required, non-destructive or minimally-destructive to the sample, as well as simultaneously completing multi-element analysis, LIBS has been reported to be an attractive technology for fingerprint analysis\cite{2,3}. Optical Emission Spectroscopy (OES) has been used for years to determine the plasma parameters such as electron density \(n_e\), electron temperature \(T_e\), Debye length \(\lambda_D\), Debye number \(N_D\) and plasma frequency \(f_D\)\cite{4}. The electron density in general, specifies the state of thermo-dynamical equilibrium of the plasma, while the temperature determines the strength of the different distribution functions describing the plasma state. The electron temperature of plasma was calculated using Boltzmann plot method\cite{5}: \[
\ln \left( \frac{\lambda_{ji} \, \omega_{ji}}{\hbar c A_{ji}} \right) = -\frac{1}{kT} \left( E_j \right) + \ln \left( \frac{N}{U(\theta)} \right) \tag{1}
\]
where \(\omega_{ji}\) is the relative intensity (in arbitrary units) of the emission line between the energy levels \(i\) and \(j\), \(\lambda_{ji}\) its wavelength (in nanometres), \(A_{ji}\) is the degeneracy or statistical weight of the emitting upper level \(i\) of the studied transition, and \(A_{ji}\) is the transition probability for spontaneous radiative emission from the level \(i\) to the lower level \(j\). Finally, \(E_j\) is the excitation energy (in eV) of level \(i\), \(k\) is the Boltzmann constant, \(N\) state population densities.

The measurement of the electron density \(n_e\) through Stark broadening effect requires a line which is free from self-absorption\cite{6}: \[
\omega_s = \left( \frac{\lambda^2}{2 \omega_0^2} \right) N_r \tag{2}
\]
Debye’s length can be calculated by the formula\cite{7}:
\[\lambda_D = \left( \frac{\varepsilon_0 k_B T_e}{n_e e^2} \right)^{1/2} \approx 7.43x10^{-2} \left( \frac{T_e (eV)}{n_e} \right) \frac{1}{\varepsilon_0} \tag{3}\]
where \(k_B\) is Boltzmann’s constant, \(T_e\) is the plasma temperature, \(e\) is electron charge and \(n_e\) is the electron density. Plasma frequency can be given as\cite{7}:
\[
\omega_{pe} = \left( \frac{n_e e^2}{m_e \varepsilon_0} \right)^{1/2} \tag{4}\]
where \(\varepsilon_0\) is the electric constant, \(m_e\) is the electron mass, \(n_e\) is the electron number density and \(e\) is electron charge.

The number of particles in Debye sphere can be calculated as \cite{7}:
\[N_D = \frac{4}{3} \pi n_e \lambda_D^3 \tag{5}\]

**Experimental part**

Mg, Al, and mix Mg:Al pellets were prepared using equal molar quantities of magnesium and aluminum powders and mix them by mechanical motor with steel balls for 10 minutes. A piston with a pressure of 3.5 tons was used to make a disk of 5 gm mass and 1 cm diameter for each Mg, Al and Mg:Al mix alloy. The samples were bombarded by Nd:YAG pulse laser (9 ns duration time, 10 Hz frequency, and wavelength 1064 nm), at an angle of 45 and the target is normal. The laser was focused on target by using IR Plano convex lens of focal length 10 cm to produce plasma. The emitted spectrum from the surface of the samples was transferred by optical fiber to be analyzed using a spectrometer connected with a computer to study the effect of laser energy and wavelength on the properties of the produced plasma.

**Results and discussions**

Plasma was produce by the laser interaction with Al, Mg and mix Al:Mg alloy targets using Q-switched Nd:YAG under vacuum. A spectrum consists of a number of characteristic spectral lines of a particular atoms and ions. Figures-(1, 2, 3) show the emission spectra of the laser induced from (Al,Mg and mix Al:Mg) targets plasma in vacuum, by Q-switched pulse laser with fundamental wavelength.
Figure 1 - Emission spectra of laser induced aluminum powder target plasma with different laser energies in vacuum.

Figure 2 - Emission spectra of laser induced magnesium powder target plasma with different laser energies in vacuum.
Figure 3: Emission spectra of laser induced on Mg-Al alloy target in a vacuum with different laser energies

The value of $T_e$ is obtained from the Boltzmann plot method using eq(1), as shown in Figures-(4, 5, 6) this requires peaks that originated from the same atomic species and the same ionization stage with data from NIST site, where the electron temperature equal to the invert of slope of fitting line. The fitting equations with the $R^2$ is (a statistical coefficient indicating the goodness of the linear fit)[8]
Figure 4- Boltzmann plot for aluminum powder target with different laser energies in vacuum
Figure 5-Boltzmann plot for magnesium powder target with different laser energy in vacuum.
Electron temperature ($T_e$) was calculated from the slope of fitting line using Eq. (2) and electron density ($n_e$) using stark broadening as shown in Figure-7.
Figure 7- Variation in the signal intensity and width of the Mg (II) lines at 279.55301 nm and at different values of 1064 nm laser for Mg:Al alloy in vacuum.

Figures-(8, 9, 10) shows the variation of $T_e$ and $n_e$ as a function of laser energy. The results of these figures illustrated that the increasing of laser energy shown to increase of electron temperature and the electron density. At higher laser peak energy $T_e$ being near stable and doesn't increase, because the plasma becomes opaque to the laser beam which shields the target, the electron temperatures show a slow linear increasing as the laser peak power increased; This is due to the absorption of laser photon by the plasma[9]. The increasing of $n_e$ with increasing of laser energy attributed to the increasing of electron collisions with the ambient gas.

Figure 8- The variation of $(T_e)$ and $(n_e)$ versus the laser energies for Al powder target in vacuum
Tables-(1, 2, 3) displays the calculated electron temperature \((T_e)\), electron density \((n_e)\), Debye length \((\lambda_D)\) by using equation (3), plasma frequency \((f_p)\) using equation (4) and Debye number \((N_D)\) using equation (5) for Mg, Al and Mg:Al alloy targets at different laser pulse energies. The results shown that the values of plasma frequency increase because of the dependence of plasma frequency on density of electrons \((n_e)^{1/2}\). We can also observe that the plasma frequency increasing with the increase of the laser peak power. The higher values of \(N_D\) due to that its dependence on \(n_e\) and \(T_e\). Also we notice the Debye length increase because Debye length depends on plasma temperature and plasma density (varies directly with \(\sqrt{T_e}\) and inversely with \(\sqrt{n_e}\)). [10].
Table 1- Plasma parameters for Al powder in vacuum with different laser energies.

<table>
<thead>
<tr>
<th>Laser energy(mJ)</th>
<th>Te (eV)</th>
<th>(n_e \times 10^{16}) (cm(^{-3}))</th>
<th>(f_p \times 10^{12})(Hz)</th>
<th>(\lambda_0 \times 10^5)(cm)</th>
<th>(N_D)</th>
</tr>
</thead>
<tbody>
<tr>
<td>400</td>
<td>0.590</td>
<td>6.368</td>
<td>2.2661</td>
<td>0.2261</td>
<td>3.081</td>
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<tr>
<td>500</td>
<td>0.613</td>
<td>7.075</td>
<td>2.3887</td>
<td>0.2187</td>
<td>3.098</td>
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<td>600</td>
<td>0.621</td>
<td>7.547</td>
<td>2.4670</td>
<td>0.2131</td>
<td>3.250</td>
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<tr>
<td>700</td>
<td>0.811</td>
<td>8.019</td>
<td>2.5429</td>
<td>0.2363</td>
<td>4.429</td>
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<tr>
<td>800</td>
<td>0.908</td>
<td>8.255</td>
<td>2.5800</td>
<td>0.2491</td>
<td>5.343</td>
</tr>
<tr>
<td>900</td>
<td>0.928</td>
<td>8.255</td>
<td>2.5800</td>
<td>0.2491</td>
<td>5.343</td>
</tr>
</tbody>
</table>

Table 2- plasma parameters for Mg powder in vacuum with different laser energies

<table>
<thead>
<tr>
<th>Laser energy(mJ)</th>
<th>Te (eV)</th>
<th>(n_e \times 10^{16}) (cm(^{-3}))</th>
<th>(f_p \times 10^{12})(Hz)</th>
<th>(\lambda_0 \times 10^5)(cm)</th>
<th>(N_D)</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>0.282</td>
<td>8.039</td>
<td>2.546</td>
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<tr>
<td>400</td>
<td>0.290</td>
<td>8.824</td>
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<tr>
<td>500</td>
<td>0.299</td>
<td>9.020</td>
<td>2.697</td>
<td>0.1353</td>
<td>0.935</td>
</tr>
<tr>
<td>600</td>
<td>0.307</td>
<td>9.412</td>
<td>2.755</td>
<td>0.1342</td>
<td>0.952</td>
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<tr>
<td>700</td>
<td>0.306</td>
<td>9.804</td>
<td>2.812</td>
<td>0.1313</td>
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<tr>
<td>800</td>
<td>0.312</td>
<td>10.980</td>
<td>2.976</td>
<td>0.1252</td>
<td>0.903</td>
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Table 3- plasma parameters for Mg:Al alloy target in vacuum with different laser energies

<table>
<thead>
<tr>
<th>Laser energy(mJ)</th>
<th>Te (eV)</th>
<th>(n_e \times 10^{16}) (cm(^{-3}))</th>
<th>(f_p \times 10^{12})(Hz)</th>
<th>(\lambda_0 \times 10^5)(cm)</th>
<th>(N_D)</th>
</tr>
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<td>5.0405</td>
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<td>600</td>
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<tr>
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<td>10.142</td>
<td>5.689</td>
<td>0.2314</td>
<td>0.526</td>
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<tr>
<td>800</td>
<td>0.992</td>
<td>10.377</td>
<td>5.734</td>
<td>0.2298</td>
<td>0.527</td>
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<tr>
<td>900</td>
<td>1.002</td>
<td>10.613</td>
<td>5.799</td>
<td>0.2283</td>
<td>0.529</td>
</tr>
</tbody>
</table>
Conclusions
We can summarize our conclusions as follows:
1- The interaction of laser irradiation with metallic targets is a helpful method to produce plasma plumes consisting of highly concentrated electrons, ions and neutral particles.
2- The spectral lines intensities of the laser induced plasma emission exhibited a strong dependence on the ambient conditions. It is found that the lines intensities at different laser peak powers increase when the laser peak power increases and then decreases when the power continues to increase.
3- The plasma parameters such as temperature and number density are found to increase with the laser irradiance. It is inferred that at first stage the laser vapor interaction is largely due to the inverse bremsstrahlung (IB) process. And their values is different because of the competing effects of the target surface reflectivity and the laser plasma absorption.

References