Laser and Particle Beams (2003), *21*, 65–71. Printed in the USA.

DOI: 10.1017/S0263034603211137

Study of optical emission from laser-produced plasma expanding across an external magnetic field

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(RECEIVED 21 March 2002; ACCEPTED 21 October 2002)

Abstract

The laser-induced plasma obtained from the liquid target and expanding across a steady magnetic field has been studied using atomic emission spectroscopy. The line emission from the plasma was enhanced (>1.5 times) in the presence of a magnetic field, whereas background emission decreases. Enhancement in line intensity was found to be mainly a function of plasma beta (β). An increased rate of three-body recombination in plasma particles due to the cooling of the plasma during its expansion and an increase in effective plasma density as a result of its confinement seems to be the reason behind this enhancement.

Keywords: Laser-produced plasma; Laser-induced breakdown spectroscopy; Magnetic confinement; Plasma emission

1. INTRODUCTION

The interest in the dynamics of laser-produced plasma (Dawson, 1964) has become important due to its technological application in various field of research such as materials science (Chrisey & Hubler, 1994), chemical physics (Zel'divich & Raizer, 1966), and plasma physics, particularly inertial confinement fusion (Ripin *et al.*, 1987; Radziemski & Cremers, 1989; Peyser *et al.*, 1992; Kilkenny *et al.*, 1994). Investigation of laser-produced plasma expanding across an external magnetic field is the subject of current interest in many laboratories (Suckewer & Fishman, 1980; Suckewer *et al.*, 1985; Rai *et al.*, 1998; Pisarczyc *et al.*, 1992; Pisarczyc & Kasperczuk, 1999). The dynamics of plasma across magnetic field is mainly governed by the β (ratio of the kinetic energy of the plasma to the magnetic energy) of the plasma. In the case of low β, the magnetic field diffuses into the bulk plasma, whereas high β plasma punches its way across the magnetic field. Finally, expansion of plasma stops in the presence of a magnetic field (magnetic confinement of plasma) as the plasma kinetic energy becomes equal to the displaced magnetic energy. Streaming and counterstreaming plasma flow dominate the plasma dynamics with an evolution of structures and density fluctuations indicating the presence of instabilities in the plasma (Ripin *et al.*, 1987; Peyeser *et al.*, 1992)

Any change in the physical properties of plasma in the presence of a magnetic field affects its emission characteristics. The effect of pulsed and steady magnetic fields on the emission from the laser-produced plasma has been studied by many authors, where emission wavelength ranges from X rays to the visible region in different experimental conditions (Suckewer & Fishman, 1980; Suckewer *et al.*, 1985; Mason & Goldberg, 1991*a*; Mason & Goldberg, 1991*b*; Pisarczyc *et al.*, 1992; Rai *et al.*, 1998; Pisarczyc & Kasperczuk, 1999). Magnetic confinement of plasma has been utilized successfully to enhance the gain of an X-ray laser (Suckewer & Fishman, 1980; Suckewer *et al.*, 1985) as well as the analytical characteristics of various low-energy plasma sources used for elemental analysis, particularly laser-induced breakdown spectroscopy (LIBS; Mason & Goldberg, 1991*a*,*b*; Rai *et al.*, 2001). Laser-induced breakdown spectroscopy utilizes a high-intensity laser to generate a luminous microplasma from different types of targets (solids, liquid, and gases) to study their optical emission properties from the constituent atoms/ions in the plasma (Multari *et al.*, 1996; Yueh *et al.*, 2000; Samek *et al.*, 2000). A 2–5 times enhancement in the optical emission has been reported under the effect of an ~8.5-T pulsed magnetic field, where it is supposed that the kinetic energy of the

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plasma is transformed into thermal energy, which helps in heating the plasma (Mason & Goldberg, 1991*b*). The generation of a high-intensity magnetic field and its synchronization with the experimental system is a difficult task and needs to be simplified using a steady magnetic field. For this purpose, the study of the emission process under the effect of low and steady magnetic fields is required, which is very little known.

In the present experiment, we have studied the emission properties of the laser-produced plasma from the liquid target (aqueous solution of Mn) in the presence of a low and steady magnetic field in order to enhance the emission (increase the sensitivity of LIBS for various applications) as well as to better understand the physical processes taking place in the plasma, where plasma after expansion and cooling down behaves as a low beta plasma.

2. EXPERIMENT

The schematic diagram of the experimental setup for recording the laser-induced breakdown emission from a liquid target is shown in Figure 1a. A Q-switched frequency doubled Nd:YAG laser (Continuum Surelite III) that delivers energy of ~400 mJ in 5 ns time duration was used in this experiment. The laser was operated at 10 Hz during this experiment and was focused on the target (center of the liquid jet) using an ultraviolet (UV)-grade quartz lens of 20 cm focal length. The same focusing lens was used to collect and collimate the optical emission from the laser-induced plasma. This collimated beam passes through a dichroic mirror and was then collected by the combination of two UV-grade quartz lenses of focal length 100 and 50 mm to couple the emission to an optical fiber bundle. The fiber bundle consists of a collection of 80 single fibers of 100 μm core diameter. The rectangular exit end of the optical fiber was coupled to an optical spectrograph (Model HR 460, Instrument SA Inc., Edison, NJ) and used as an entrance slit. The spectrograph was equipped with a 3600 l/mm diffraction grating of dimension 75 mm $\times$ 75 mm. A 1024 $\times$ 256 element intensified charge coupled device (ICCD; Princeton Instrument Corporation, Princeton, NJ) with a pixel width of 0.022 mm was attached to the exit focal plane of the spectrograph and used to detect the dispersed light from the laser-produced plasma. The detector was operated in gated mode with the control of a high-voltage pulse generator (PG-10, Princeton Instruments Corporation, Princeton, NJ) and was synchronized to the laser output. Data acquisition and analysis were performed using a personal computer. The gate delay time and gate width were adjusted to maximize the signal-to-background (S/B) and signal-to-noise (S/N) ratios, which are dependent on the emission characteristics of the elements as well as on the target matrix. To increase the sensitivity of the system, 100 single-pulse spectra were integrated to get one spectrum and 10 such spectra were recorded to reduce the standard deviation.

A Teflon nozzle of diameter ≤1 mm was used with a peristaltic pump (Cole-Parmer Instrument Co.) to form a laminar liquid jet. The laser was focused on the jet such that the direction of laser propagation was perpendicular to the direction of the liquid jet. The laser was focused ~15 mm below the jet exit, where the liquid flow was laminar. However, the extent of laminar flow was decided by the speed of the pump. The liquid jet was aligned in a vertically downward direction. The aqueous solution of manganese (10 ppm)

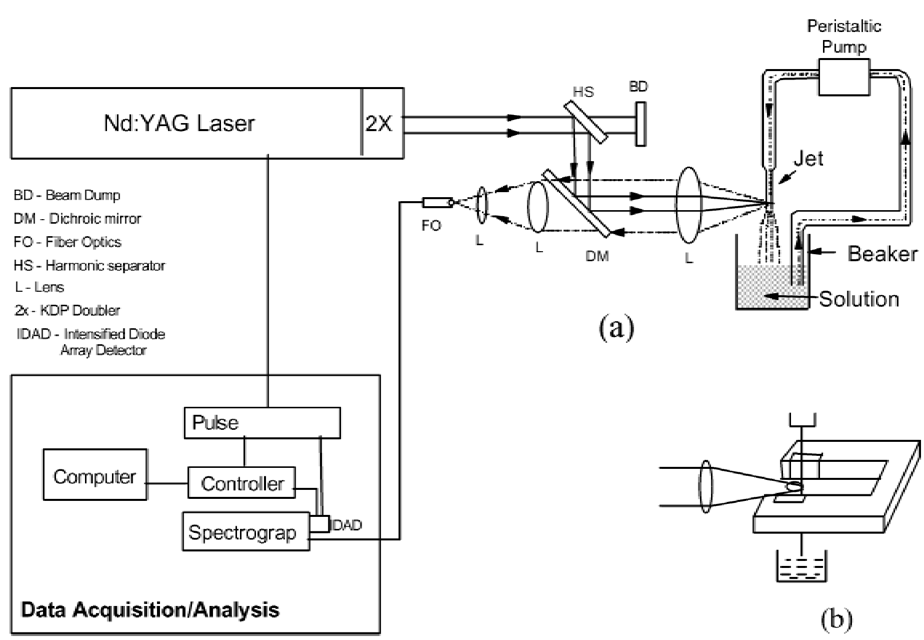

Fig. 1. (a) Schematic diagram of experimental setup for recording laser-induced breakdown emission. (b) Location of liquid jet in between the magnetic poles.

https://doi.org/10.1017/S0263034603211137 Published online by Cambridge University Press

in 2% HNO_3 was used as the target in the form of liquid jet. This solution was prepared from the standard solution of 1000 ppm available commercially in 2% HNO_3. Aqueous solution of manganese was chosen as the sample in this experiment because it has optical characteristics similar to technetium, a radioactive material found during the processing of nuclear waste. Manganese was used for all the laboratory experiments for increasing the sensitivity of LIBS for detecting the technetium.

Two rare earth (neodymium and samarium cobalt) permanent magnets of size $12.5 \times 12.5 \times 3.12$ mm were used for generating the steady magnetic field during this experiment. Both the magnets were held in a mild steel (MS) structural arrangement separated by ~5 mm. This arrangement provided a horseshoe type magnet with an ~0.5 T magnetic field in between the poles. The magnetic field was measured using a hall probe. The magnet system was held in such a way that the liquid jet passed vertically between and at an equal distance from the two poles (Figs. 1b and 2). The laser was focused on the jet such that the plasma plume expanded in a nearly uniform magnetic field. These experiments were performed for two magnetic field geometries: (1) when the north pole of one magnet was facing the south pole of the other (Fig. 2a) and the field lines were passing straight from one pole to the other (N-S configuration), and (2) when similar poles from both the magnets were facing each other (Fig. 2b) and making a cusp magnetic field geometry (N-N configuration). In the first case, the plasma expanded across a nearly uniform magnetic field present between the two poles, whereas in the second case, the plasma was confined in a nearly zero magnetic field produced by field lines forming cusp geometry. The MS bars holding the magnets were wrapped by adhesive and insulating tape so as to avoid direct interaction of laser or plasma particles with the MS bar.

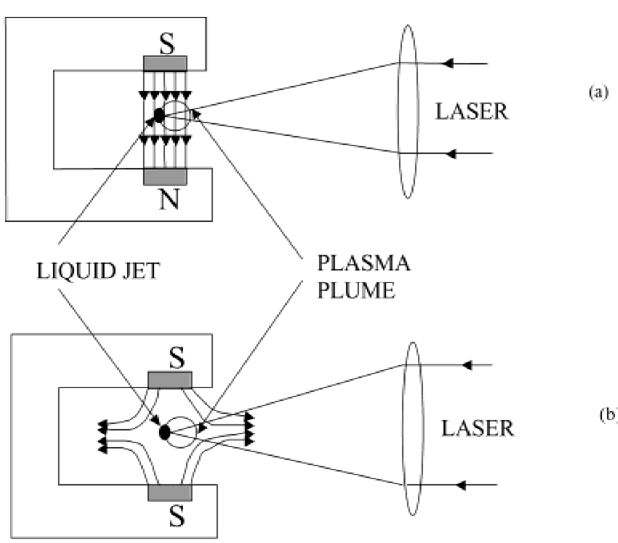

Fig. 2. Schematic diagram (top view) of the experimental setup in the presence of. magnets. (a) Opposite poles of magnets facing each other (linear field). (b) Same pole of the magnets facing each other (cusp field).

3. RESULTS AND DISCUSSIONS

The optical emission from the laser-produced plasma from the liquid target having manganese as a trace element was recorded in the absence and the presence of the magnetic field. Plasma emission was collected in the direction opposite to the laser propagation, which provided a spatially integrated intensity from the plasma plume. Initially, the plasma had high density and temperature during and just after the laser pulse, which is why emission from the plasma is dominated mainly by Bremsstrahlung and black body radiation forming continuum spectra. LIBS spectra of magnese (10 ppm) were recorded at 10-μs gate delay to avoid the intense background emission from the hot plasma, which decays very fast with delay time. Figure 3 shows Mn spectra recorded in the absence and the presence of the magnetic field at a laser energy of 140 mJ. An enhancement of about 1.5 times in the intensity of the Mn lines was found in the spectra in the presence of the magnetic field as compared to no magnetic field. A different feature was observed when the experiment was performed at the laser energy of 280 mJ (Fig. 4) as compared to the laser energy of ~140 mJ. Both the intensity of Mn line and background emission decreased in the presence of the magnetic field at laser energy of 280 mJ. A plot of the variation of the intensity of Mn (403.03 nm) at various laser energy in the absence and the presence of the magnetic field is shown in Figure 5 (Rai *et al.*, 2001). It was found that the intensity of Mn lines increases linearly with the laser energy. A log-log plot of data follow a power law variation with a slope of ~1.85 in the absence of the magnetic field. Similar power law variation has been reported by many authors, having slopes ranging from 1.5 to 2.5 in the case of X-ray emission from the laser-produced plasma recorded in vacuum (Bleach & Nagel, 1978). There is an indication of signal saturation toward the higher laser energy side in the present experiment (Fig. 5), whereas no saturation was observed in plasma emission (X-ray) during the experiment performed in vacuum (Rai

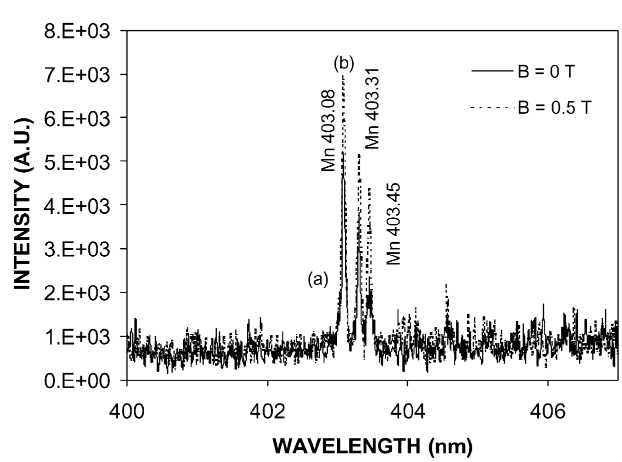

Fig. 3. LIBS spectrum of manganese recorded at laser energy of 140 mJ. (a) No magnetic field ($B = 0$ T). (b) Linear magnetic field ($B = 0.5$ T).

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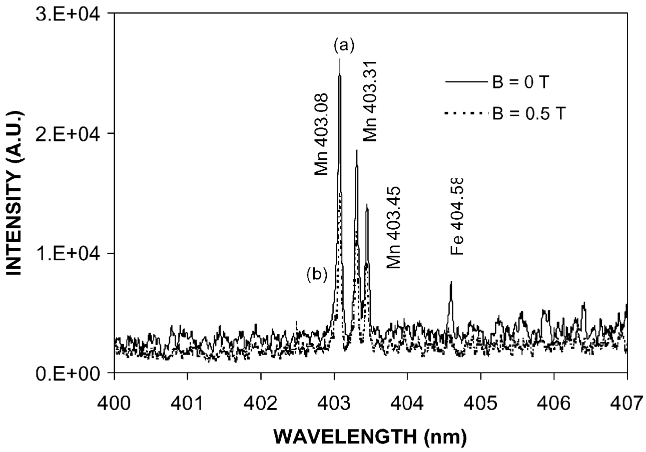

Fig. 4. LIBS spectra of manganese at laser energy of 280 mJ (gate delay and gate width 10 μs). (a) No magnetic field ($B = 0$ T). (b) Linear magnetic field ($B = 0.5$ T).

et al., 1998). This shows that the presence of air at atmospheric pressure around the plasma confines it and increases the effective density of the plasma, which may be enhancing the self-absorption of the emission, coming out from the plasma, leading to an observation of saturation in the signal (Treytl *et al.*, 1971). The presence of the magnetic field (Fig. 5) shows an enhancement in the LIBS intensity toward the lower and intermediate energy side, whereas it decreases toward the higher energy side. A similar variation was noted whether a linear magnetic field (N-S) or a cusp magnetic field (N-N) geometry was used. However, a little change (increase) was observed in the case of the cusp geometry rather than the linear field geometry. The maximum enhancement in signal was found for the laser energy of ~140 mJ. The variation in manganese line emission intensity with the laser energy in the presence of a magnetic field (Fig. 5) clearly shows the presence of two slopes (a higher slope in the low-energy side and a lower slope toward the higher energy side). This change in the slope (decrease in emission intensity) toward the higher laser energy in the presence of a magnetic field may be the result of a strong saturation in the emission intensity. During the saturation, emission intensity was found less in the presence of the magnetic field in comparison to its absence. The decrease in the slope (emission) or the presence of saturation in the plasma emission in the presence of a magnetic field indicates the loss of plasma energy, which may be due to the opening of a new channel of loss in the content of plasma energy in the presence of a magnetic field. The generation of instabilities and high-energy particles in the plasma along with self-absorption of the emission by the plasma may be the process for loss of plasma energy. It is important to identify the most probable process for enhancement in emission in the lower laser energy regime and the loss in energy content of plasma, which decreases the plasma emission toward the higher laser energy in the presence of a magnetic field.

Generally, the presence of a magnetic field confines the plasma and increases the effective density of the plasma in the confinement region. An increase in the plasma density due to magnetic confinement was verified experimentally with a liquid sample (Rai *et al.*, 2002). The electron density of 5.47×10^{16} cm^{-3} and 9.54×10^{16} cm^{-3} was inferred in the absence and the presence of the magnetic field with the help of a Stark broadening measurement in H_α emission. An increase in the density has nearly a factor of ~1.74, which confirms that the enhancement in the emission is due to an increase in the plasma density as a result of magnetic confinement.

The continuum spectrum observed here (10-μs gate delay) is dominated by the matrix effect, that is, the atomic, ionic, and molecular emission from hydrogen, oxygen, nitrogen, and hydroxyl radical (OH). Figure 6 shows that background emission increases with an increase in the laser energy, which is expected because the higher laser intensity will enhance the ablation and ionization through the enhanced heating of the plasma. The presence of the magnetic

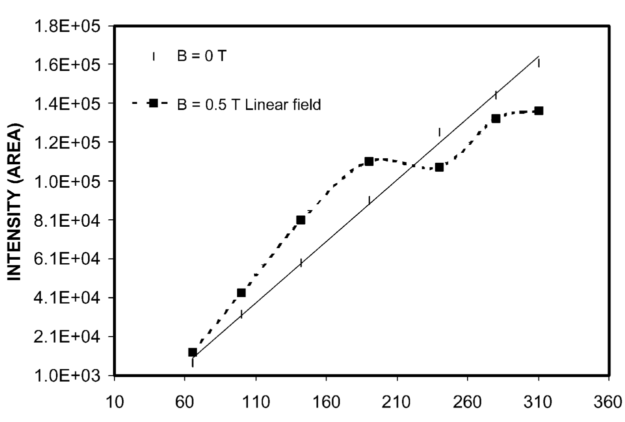

Fig. 5. Variation in the line emission intensity at $\lambda \sim 403.076$ nm (gate delay and gate width 10 μs) with laser energy in the absence and presence of a magnetic field (Rai *et al.*, 2001).

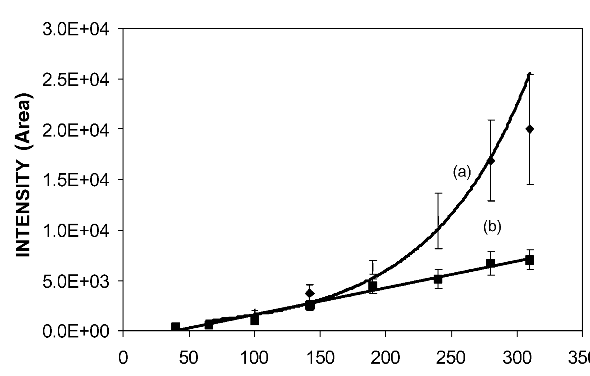

Fig. 6. Variation in background emission intensity with laser energy (gate delay and gate width ~10 μs). (a) No magnetic field ($B = 0$ T). (b) Linear magnetic field ($B = 0.5$ T).

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field reduces the background emission towards the higher laser energy side, whereas it remains the same towards the lower laser energy side. Background emission is mainly related to the plasma temperature. Therefore, any decrease in background emission in the presence of a magnetic field indicates a fast decrease in the plasma temperature. It seems that the increased plasma density due to the magnetic confinement enhances the plasma cooling, which may take place either through enhanced particle collision in the plasma or by the radiative cooling process or by the density-fluctuation-induced diffusion of the plasma particles (Liewer, 1985).

The study of the temporal evolution of atomic emission in the absence as well as in the presence of a magnetic field (Fig. 7) was performed by recording emission spectra at different gate delays, ranging from 5 to 45 μs. It was noted that the emission intensity was high at lower gate delays, whereas it decayed exponentially with an increase in the gate delay. A similar variation was noted whether it was recorded in the absence or in the presence of a magnetic field except that the signals in the presence of a magnetic field were higher. The signal enhancement was found maximum near a gate delay of 5 μs. The emission from the plasma was dominated by Bremsstrahlung continuum emission due to the high plasma temperature, below a gate delay of 5 μs. However, the plasma emission in the presence of a magnetic field decreases fast towards higher gate delay due to a decrease in plasma density as a result of recombination and the loss due to the diffusion process. It seems that the rate of recombination of electrons and ions increases as a result of an increase in effective plasma density due to the magnetic confinement and a decrease in plasma temperature due to its expansion (Dawson, 1964). This seems to be the main reason behind an enhancement in emission in the presence of a magnetic field. However, the maximum enhancement will be decided by the balance between the rate of recombination of plasma particles and the loss of plasma particles or neutral atoms from the emission region as a result of various diffusion processes. Either a little or negligible enhancement in the emission below a 5-μs gate delay was due to high density and high temperature of the plasma, which makes the plasma β high in the presence of a magnetic field. These experimental observations are in qualitative agreement with the simple expression giving the ratio of plasma emission in the presence (I_2) as well as in the absence (I_1) of a magnetic field, which is derived considering an increase in plasma density under magnetic confinement (Rai *et al.*, 1999):

$$\frac{I_2}{I_1} = \left(1 - \frac{1}{\beta}\right)^{-3/2} \left(\frac{t_1}{t_2}\right), \tag{1}$$

where $\beta = 8\pi nkT_e/B^2$, and t_1 and t_2 are the emission times of plasma in the absence and presence of a magnetic field. Equation (1) clearly indicates that, in principle, the enhancement in emission intensity can be increased even to more than two times by maintaining the plasma β close to 1. This may be possible by increasing the value of the steady magnetic field as well as by avoiding the loss of atoms due to various diffusion processes (minimization of instabilities in the plasma). Considering complex plasma evolution during this experiment, it is very difficult to explain the process properly. However, a detailed numerical modeling including three-dimensional MHD, atomic kinetics and radiation transfer, is required for this purpose.

The experimental observations from this experiment have been compared with the experimental results reported earlier to better understand the physical processes taking place in the plasma in the presence of magnetic field. An earlier experiment (Rai *et al.*, 1998) was performed by focusing a picosecond laser on a solid target in vacuum and the X-ray emission was recorded, whereas in the present experiment, plasma was formed using a nanosecond laser, focused on a liquid jet at atmospheric pressure. The experimental conditions are entirely different in the two experiments even though there are some similarities in the results. It seems that in the lower laser energy regime, the kinetic energy of the plasma is comparatively low such that most of it is confined by the applied magnetic field ($B = 0.5$ T). Usually, confinement of the plasma increases the effective number of plasma particles in the confinement region. Initially, during or just after the laser pulse, the plasma has a high temperature and high density, and, as a result, a high plasma pressure (nkT_e). The magnetic field applied in this experiment is not sufficient to confine such a high-energy-density plasma. But after expansion for some time, the plasma becomes cool and its density also decreases, resulting in a low plasma pressure. For a constant value of magnetic field, initially plasma will have high β value, and after expansion, plasma β goes down. A lower magnetic field will affect only low β plasma. In fact, the plasma expansion velocity may be zero in the presence of the magnetic field, when it is completely stopped at a certain spatial location, where the plasma pressure (kinetic energy) and magnetic pressure (energy) become equal (near $\beta = 1$). But in reality, plasma cannot be completely stopped

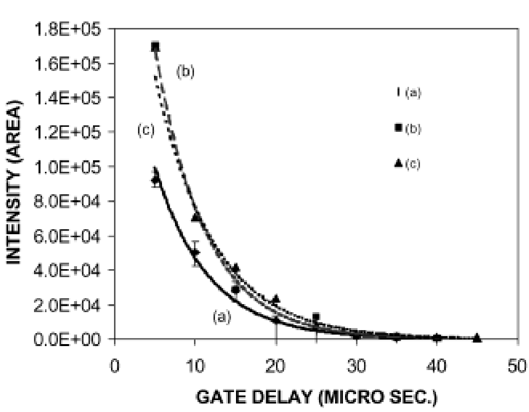

Fig. 7. Variation in line emission intensity at $\lambda \sim 403.076$ nm with gate delay (gate width $\sim$10 μs; Rai *et al.*, 2002). (a) No magnetic field (B = 0 T). (b) Linear magnetic field ($B = 0.5$ T). (c) Cusp magnetic field.

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by the magnetic field, because of its finite resistivity at low plasma temperature (collisional plasma; Bhadra, 1968; Tuckfield & Schwirzke, 1969). Perfect confinement of plasma is possible only when the plasma is fully conducting, which is quite impossible for the present experimental condition. Finally, a part of the plasma will escape from the confinement zone either due to cross-field diffusion as a result of increased collision at low plasma temperature, or due to generation of instability in the plasma. The decrease in the emission intensity slope (Fig. 5) in the presence of a magnetic field toward higher laser energy indicates that part of the absorbed laser energy is being utilized in the generation of instability in the plasma. Another possibility of energy loss may be the generation of high-energy particles (ions and electrons), which can escape from the plasma with a certain amount of kinetic energy. It is also possible that a part of the laser energy is being scattered from the plasma if some parametric instability is present in the plasma. However, the possibility of the presence of a parametric instability is very low at such a low laser intensity (Radziemski & Cremers, 1989). Considering all the conditions, it seems that the change in the slope (decrease in the emission intensity) toward higher laser energy is due to the generation of instability in the plasma and the escape of high-energy plasma particles from it. The self-absorption of emission in the plasma also may contribute toward saturation in the presence or absence of a magnetic field, which is possible as a result of the increased plasma density due to confinement by air in the absence of a magnetic field and due to double confinement by air and a magnetic field in the presence of a magnetic field. The generation of a laser-induced shock, that is, a laser-induced detonation wave and laser-induced ablation pressure also may distort the laminar jet of the liquid sample. It seems that all these factors are important and make their own contribution in decreasing the signal in the presence of a magnetic field toward higher laser energy.

The possibility of the generation of instability in the plasma is discussed in the light of results obtained from an earlier experiment on a solid copper target using a picosecond laser in the presence of a magnetic field (Rai *et al.*, 1998), which is different than the present experiment performed using a nanosecond-length time duration laser. It has been reported that mainly the ablation threshold reduced when the laser pulse duration was reduced from nanoseconds toward femtoseconds (Gotz & Stuke, 1997). The X-ray pinhole pictures of the laser-produced plasma in the absence and the presence of a magnetic field (0.6 T) indicated that plasma expansion remained smooth in the absence of the magnetic field, where as distorted structures developed in the isointensity contour in the presence of the magnetic field. Generation of these structures was due to the presence of instabilities in the plasma, as evidenced by the observation of density fluctuations in the ion saturation current (Rai *et al.*, 2000) in the expanding plasma. This indicates that the free energy of the expanding (streaming and counterstreaming) plasma was utilized in generation of instability in the plasma. Similarly Chang and Hashmi (1977) have also reported onset of density fluctuation in laser-produced plasma even at 0.02 T magnetic fields. The density fluctuation in plasma can also occur due to the oscillation of magnetic field lines under the effect of the plasma pressure and the restoring force due to the magnetic field lines (Bhadra, 1968; Rai *et al.*, 2000). The Rayleigh–Taylor and Kelvin–Helmholtz instabilities are magnetohydrodynamic instabilities, which can develop in the present experimental situation, where the laser-induced plasma is expanding across an external magnetic field. We expect that the major structure of density fluctuation is similar when plasma is formed at atmospheric pressure (LIBS). Investigation of plasma plume imaging at atmospheric pressure may provide better information. However, a complicated gas dynamic equation has to be solved to better understand the evolution of the plasma plume at atmospheric pressure (Puretzky *et al.*, 1999).

The enhancement in the visible and X-ray emission from the plasma in the presence of a steady magnetic field has various advantages in different fields of research. Particularly, enhancement of optical emission from the plasma has a special implication in the elemental analysis of solid and liquid samples using laser-induced breakdown spectroscopy (Radziemski & Cremers, 1989; Samek *et al.*, 2000; Yueh *et al.*, 2000). This phenomenon has been utilized to improve the limit of detection (LOD) of the trace elements in the liquid (aqueous) solution. It was found that the LOD of manganese was improved from 1.74 to 0.83 ppm in the presence of 0.5 T linear magnetic fields (Rai *et al.*, 2001).

5. CONCLUSIONS

In summary, it was found that the emission from laser-induced breakdown plasma, expanding across the steady magnetic field, shows an enhancement of ~1.5 times at intermediate laser energy range. However, the saturation (decrease) in the signal was noted towards the higher laser energy side in the absence as well as in the presence of a magnetic field. Saturation remains feeble due to the air confinement of the plasma (in the absence of a magnetic field) whereas, it became pronounced in the presence of a magnetic field. No enhancement of plasma emission was found when the plasma beta was high due to the high plasma temperature and density, which indicated that the emission was mainly dependent on the plasma β a function of plasma density, its temperature, and the external magnetic field. A computer simulation of this complex plasma evolution is needed for more information about the process. However, an increase in the rate of recombination of ions and electrons in the plasma at low temperature (due to plasma expansion) and at comparatively high density (due to confinement) seems to be the main process for enhancement of emission. The generation of instabilities and the self-absorption of emission seem to be the reasons behind the observation of a decrease in the slope (emission signal) with the laser power variation. However, it is a matter of further investigation to identify the exact role of instability as well as the self-absorption in the plasma to avoid and minimize this effect. A detailed study of plume imaging in various ambient condition along with its temporal evolution will provide even better understanding about the dynamics of plasma in the magnetic field. Finally a low-intensity steady magnetic field was found more convenient in increasing the emission from the plasma in comparison to a high-intensity pulsed magnetic field.

ACKNOWLEDGMENT

This work is supported by U.S. Department of Energy cooperative agreement no. DE-FC26-98FT 40395.

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