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Comparison of the polarisation of line and continuum emission in a laser produced plasma

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Abstract. Anisotropic emission from a laser-produced aluminium plasma emission has been studied using time and polarisation resolved spectroscopy. The time-resolved degree of polarisation of emission from a bound – bound transition in Al^+ (466.3 nm) was compared to the time-resolved degree of polarisation from nearby continuum emission. The results are interpreted in the framework of radiative recombination and statistical imbalances in the populations of magnetic sub-states. It is found that the continuum emission is more strongly polarised than the line emission. Thus, this technique is a valid alternative to gated LIBS.

1. Introduction

Plasma polarisation spectroscopy (PPS) has been shown to be a powerful diagnostic technique which reveals information about the electron distribution function (EDF) or a population imbalance within the magnetic sub-shells of the atoms and ions within the plasma [1]. The technique of PPS has been applied to laser produced plasmas where partial polarisation has been observed in the x-ray and visible spectral ranges [2,3]. Importantly, a particular application of PPS known as polarisation-resolved laser induced breakdown spectroscopy (PRLIBS) exploits this partial polarisation in order to enhance the signal to background ratio over traditional LIBS measurements [4]. In various wide-ranging studies [4–6], it was shown that the continuum radiation tends to have a greater degree of polarisation than the line emission. For this reason, placing a polariser in front of a spectrometer results in lower background continuum, thus increasing the signal-background ratio which is a figure of merit for LIBS. The origin of the polarisation in this study was postulated to be an anisotropic electron distribution function caused by steep density gradients which result in polarized recombination radiation [6]. Similarly, studies on the visible emission from an Al plasma showed that the line emission can also be partially polarized [3]. In this study, the degree of polarisation in the Al^{2+} doublet at 569.6 and 572.3 nm was found to be on the order of 3 %. The origin of the polarisation in this study is attributed to an inherent population imbalance in the magnetic sub-shells of the ionic system arising from anisotropic recombination of higher charged ions. This study was extended in [7] where the degree of polarisation of the Al^{2+} doublet was measured with time resolution and in a variety of background gas pressures. Here, the degree of polarisation was found to oscillate with time and its origin was attributed to anisotropic electron distribution functions caused at the plasma – gas interface. In this article, we extend the studies of [6,7] and make a time-resolved comparison of the degree of polarisation of a spectral emission line in an Al plasma (the Al^+ line at 466.3 nm) and continuum emission (at 456 nm). The study thus elucidates on the nature of the polarized emission from a laser produced plasma.

2. Experimental

Figure 1 shows the experimental setup. An Nd:YAG laser (Spectron SL800, 14 ns pulse width, 1064nm wavelength) was focused onto a pure Al target at normal incidence to create a laser plasma. The on target laser fluence was measured to be ~450 J/cm^2). The target was mounted onto a precision x-y-z translation stage to ensure a fresh target after each laser shot. The plasma was imaged onto the entrance slit of a Czerny-Turner spectrometer (Chromex 500is) which was coupled to an ICCD camera (Andor i-Star) with a time resolution of ~10 ns and a mean spectral resolution of 0.17 nm. A Wollaston prism was placed in the beam path of the plasma imaging system. This created to orthogonally polarized images of the plasma on the slit. The optic axis of the experiment (defined here to mean the axis of quantization) was chosen to be normal to the target. Thus, the Wollaston prism was aligned to produce images polarized parallel and perpendicular to this (and parallel to the entrance slit of the spectrometer). The system was calibrated to account for a polarisation dependent efficiency in the spectrometer. This was achieved by imaging a small pinhole illuminated with unpolarized light through the system. This allowed for correction of the polarisation sensitivity. The degree of polarisation was then calculated from the resultant spectra from:

$$\boldsymbol{P} = \frac{\boldsymbol{I}^{para} - \boldsymbol{I}^{Perp}}{\boldsymbol{I}^{para} + \boldsymbol{I}^{Perp}}$$

Where I^{para} is the intensity of the light polarized normal to the target and I^{perp} is light polarized parallel to the slit.

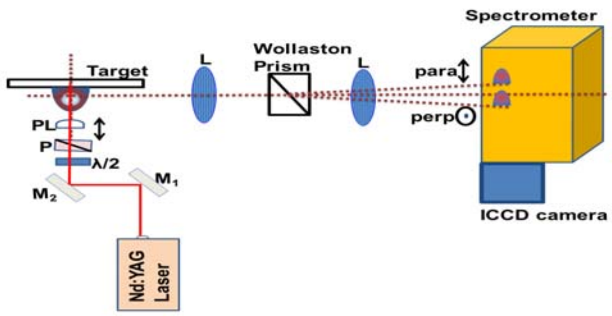

Figure 1. Schematic of experimental setup (M, mirror; λ/2, half wave plate; P, polariser; PL, planoconvex lens; and L, lens).

3. Time and polarisation resolved spectroscopy of the 466 nm line of Al^+

Figure 2a shows time and polarisation resolved spectroscopy of the Al^+ line at 466.3 nm (corresponding to the transition 4p $^1P^0_1 \rightarrow 3p^2\ ^1D_2$). It is clear from Figure 2 that, at early times, the emission is dominated by broadband continuum. At later times, line emission starts to appear. Figure 2b shows the calculated degree of polarisation at a wavelength corresponding to continuum emission (456 nm) and a wavelength corresponding to line emission (466.3 nm). The following is clear from the data: The continuum emission is more strongly polarised than the line emission, the emission is polarised along the direction of the quantisation axis (plasma expansion direction), and the degree of polarisation varies weakly over time.

This result appears to support the hypothesis of both [3] and [5] and thus we propose the following framework: Steep density gradients between the plasma and the background gas create an anisotropic electron distribution function. The continuum emission, which is comprised of either free-bound or free-free transitions is polarised due to this anisotropy. The free-free transitions can occur due to Bremsstrahlung emission which is where an electron is accelerated / decelerated in the vicinity of an ion. This qualitatively should have no preferred polarisation because the electrons are not decelerated in any preferred direction. However, free-free emission can also occur by rapid deceleration of electrons due to the background gas. These electrons should be preferentially decelerated in the direction of plasma expansion and hence be polarised along this direction. The weak polarisation of the line emission is explained using the framework of [3]. The emission of the line at 466.3 nm can arise due to excited singly charged aluminium being produced in the formation of the plasma and, again, there is no reason to assume that this has a preferred polarisation direction. Bound-bound transition emission can be polarised if there is an imbalance in the magnetic sub-levels of the upper state in the transition. The second way the line emission at 466.3 nm can occur is due to recombination of electrons with higher charged ions to create excited lower charged ions. Thus, a free electron is captured by doubly ionized aluminium ion to produce a singly charged ion in an excited state. The free electrons that participate in this process are presumably aligned due to their deceleration in a preferred direction by the background gas and thus populate a preferred magnetic sub-level upon their capture. This leads to a non-zero degree of polarisation of the line emission.

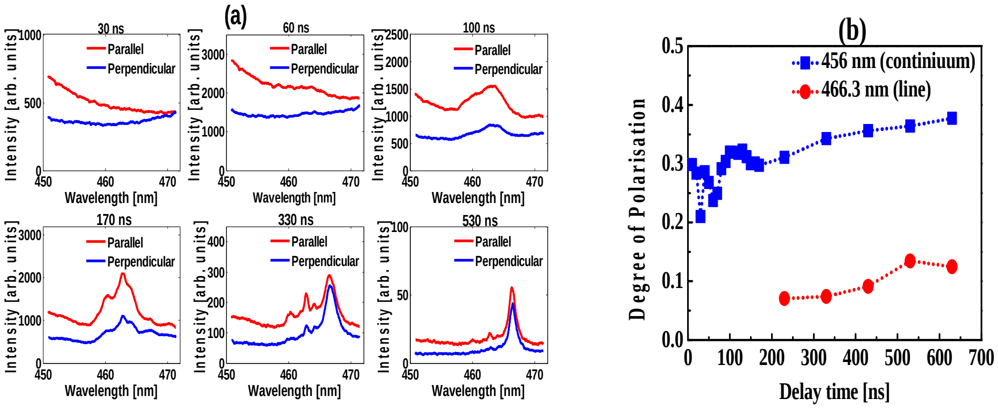

Figure 2. (a) time and polarisation resolved spectra of an Al^+ (at 466.3 nm) at a laser fluence of 452 J/cm^2 and (b) degree of polarisation as a function of gate delay.

There is one last point to be made which is that the degree of polarisation seems to vary weakly over time. One potential application of this is using plasma polarisation spectroscopy as an alternative to gated spectroscopy in LIBS. This has been alluded to in the past and expanded upon here. It seems that the continuum can be suppressed with the use of a polariser in front of the spectrometer with little loss to the intensity of the line emission.

4. Conclusion

We have compared the continuum and line emission from a laser produced plasma in terms of their respective degrees of polarisation using time resolved polarisation spectroscopy. It was found that the continuum emission is more strongly polarized than the line emission. We interpret the results in the framework of recombination of electrons with an anisotropic velocity distribution.

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