Time-resolved aluminium laser-induced plasma temperature measurements

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Abstract. We seek to characterize the temperature decay of laser-induced plasma near the surface of an aluminium target from laser-induced breakdown spectroscopy measurements of aluminium alloy sample. Laser-induced plasma are initiated by tightly focussing 1064 nm, nanosecond pulsed Nd:YAG laser radiation. Temperatures are inferred from aluminium monoxide spectra viewed at systematically varied time delays by comparing experimental spectra to theoretical calculations with a Nelder Mead algorithm. The temperatures are found to decay from 5173 ± 270 to 3862 ± 46 Kelvin from 10 to 100 μs time delays following optical breakdown. The temperature profile along the plasma height is also inferred from spatially resolved spectral measurements and the electron number density is inferred from Stark broadened Hβ spectra.

1. Introduction
Time-resolved laser-induced breakdown spectroscopy (LIBS) is a valuable experimental tool for determining key plasma parameters, such as the temperature and electron number density. The laser-induced plasma created following breakdown has multiple applications, including use in forensics, geophysics, and, of particular interest, combustion diagnostics. For the purposes of studying combustion, aluminium LIBS studies are of significant as aluminium is commonly found in solid rocket propellants and high explosives, where the goal is commonly to infer temperature [1, 2]. Determinations of the electron number density of aluminium laser-induced plasma also proven to be useful in diagnostic applications, such as with aluminum alloys [3, 4]. In previous time-resolved, aluminium laser ablation studies, the temperature of laser-induced plasma were determined from Boltzmann plots of aluminium atomic emissions and were found to be in the 4000-4250 Kelvin range [1, 5]. The plasma temperature may also be determined from the temperature dependence of diatomic molecular spectra such as aluminium monoxide (AlO). Temperature results from previous studies were found to be 4000 Kelvin at 50 μs [6] and 4880 Kelvin at 45 μs [7] from AlO emissions. Electron densities were also inferred in reference [6] and were found to be \(10^{23} \text{ m}^{-3}\). In this work, we seek to use AlO diatomic molecular emissions to characterize the temperature decay of laser-induced plasma near the surface of an aluminium target in laboratory air over a broad range of times following optical breakdown using time-resolved LIBS measurements. Inferences are also made on the temperature distribution along the plasma height and the electron number density.
2. Experiment
Time-resolved spectral measurements are used to characterize the temperature decay of laser-induced plasma initiated near the surface of an aluminium target. Laser-induced plasma are formed by tightly focusing laser radiation from a Q-switched, Nd:YAG laser operating in its fundamental mode with a 12 nanosecond pulse width and an energy per pulse of 190 mJ. Line of sight spectral measurements are made with a Jobin-Yvon HR640 spectrometer with 0.1 nm spectral resolution coupled to gated, intensified detectors. An intensified, linear diode array coupled to an optical multichannel analyzer (Princeton Instruments 1460) was used to collect AlO spectra from 10 to 100 μs time delays in 5 μs steps with gate widths of 5, 10 and 20 μs for 10-20, 20-100, and 70-100 μs delays, respectively. A gated ICCD was used to infer the temperature dependence along the plasma height at a time delay of 60 μs with a 6 μs gate width. Time resolution was achieved by synchronizing the laser repetition rate with the measurement rate of the gated detectors with the use of waveform, pulse, and delay generators. Reference 8 provides a detailed description of the apparatus [8]. Prior to analysis, all spectra were properly calibrated for detector response and background sensitivity.

3. Results
To infer the temperature as function of time following optical breakdown, spectra collected with intensified, linear diode array were fit to theoretical calculations of the AlO \( B^2\Sigma^+ \rightarrow X^2\Sigma^+ \Delta v = 0 \) transition. Comparisons were made with a Nelder-Mead fitting algorithm which was utilized for its ability to fit multiple parameters simultaneously, including a variable baseline offset. The Nelder-Mead algorithm is a minimization method in which a geometric simplex is reduced in size until a satisfactory tolerance level is reached [9, 10]. The theoretical spectra are calculated from tables of accurately calculated line strengths, where the line strength is calculated in its factorized form from the electronic transition strength, the H"onl-London factor, and Franck-Condon factor [11, 12, 13]. The baseline offset of all fitted AlO spectra were quadratic in nature and errors in the inferred temperatures were found by considering variations of the spectral resolution and baseline offset. Analysis showed that the error in the inferred temperatures was in the 45-75 Kelvin range, with the exception of the inference made at 10 μs with an error of 236 Kelvin. This resulted from the presence of H\( \beta \) superposition spectra. The results of the temperature analysis are shown in Figure 1 with an exponential decay trend. The trend line shows that the plasma temperature decay slows at later times as was expected. To infer the temperature dependence along the plasma height, spatially resolved spectra with 0.013 mm resolution were fit using the Nelder-Mead algorithm. The temperature at the center of the plasma was found to be 5780 ± 236 Kelvin at 5.2 mm slit height and decreased to 5396 ± 315 Kelvin at 6.5 mm. The temperature was found to increase above the plasma edge to a value of 5820 ± 230 Kelvin at a slit height of 9.1 mm. The larger temperatures and errors are likely result from non-optimal plasma and detector conditions; however, the temperature profile suggests that aluminium particles are combusting in a plume of above the plasma, given the ignition temperature of aluminium is 2750 Kelvin and the inferred temperatures from the linear diode experiment.

At time delays of 10, 15, and 20 μs following breakdown, superposition of Balmer series H\( \beta \) spectra with AlO measurements were observed. The H\( \beta \) spectra may be extracted and used to infer the electron number density from the Stark broadened emission. The line width of the spectra is taken to be due to Stark effects and is inferred by fitting a Lorentzian line profile to the H\( \beta \) spectra. The Stark widths are used with the empirical theory of Konjević [14] and the convergent theory of Oks [15] to infer the electron number density, \( N_e \). To determine the \( N_e \) with the theory of Oks, published data were fit to determine the \( N_e \) vs. \( H_\beta \) width dependence [16]. Figure 2 shows the fitted H\( \beta \) spectrum from the 10 μs delay along with its corresponding Stark width and electron number density. The noticeable dip in the residual in Figure 2 near 486.5 nm
is due to the subtraction of the fitted AlO spectra and represents the 1-1 vibrational peak from the transition of interest. Also indicated in the figure is the 0.5 nm H\textsubscript{\beta} peak separation. Though not considered in this work, this peak separation may also be used to infer the electron density \cite{16}. The inferred Ne are found to be 4.5 ± 0.5, 3.8 ± 0.5 and 3.0 ± 0.6 × 10\textsuperscript{22} m\textsuperscript{-3}. Errors in the inferred Ne result from AlO superposition influences.

4. Conclusions
The goal of this work was to characterize the temporal temperature decay of laser-induced plasma near the surface of an aluminium target and was accomplished with time-resolved LIBS measurements. A Nelder-Mead fitting algorithm was employed to infer the temperature from the experimental spectra and errors were determined from spectral resolution and baseline offset variations. The temperatures were found to decrease as a function of time from 5173 ± 290 to 3862 ± 46 Kelvin from 10 to 100 \(\mu\)s time delays. The decay of the temperature was also found to slow at later time delays. Spatially resolved AlO measurements were used to infer the temperature along the height of the plasma and showed the temperature increases above the plasma edge. The electron density of the plasma was inferred from measurements of H\textsubscript{\beta} spectra superimposed on AlO measurements at time delays preceding 20 \(\mu\)s and were found to decrease from 4.5 ± 0.5 to 3 ± 0.5 × 10\textsuperscript{22} m\textsuperscript{-3}. Observations of the slowing of the temperature decay, the temperature increase above the plasma, and H\textsubscript{\beta} superposition spectra suggest that combustion occurs. Future work will seek to further quantify the presence of combustion in aluminium laser-induced plasma from spatially resolved AlO spectral measurements.

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