Evidence of plasma polarization shift of Ti He-α resonance line in high density laser produced plasmas

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Abstract. A spectroscopic study of the He-α (1s² ¹S₀ - 1s2p ¹P₁) line emission (4749.73 eV) from high density plasma was conducted. The plasma was produced by irradiating Ti targets with intense (I ≈ 1x10¹⁹ W/cm²), 400nm wavelength high contrast, short (45fs) p-polarized laser pulses at an angle of 45°. A line shift up to 3.4±1.0 eV (1.9±0.55 mÅ) was observed in the He-α line. The line width of the resonance line at FWHM was measured to be 12.1±0.6 eV (6.7±0.35 mÅ). For comparison, we looked into the emission of the same spectral line from plasma produced by irradiating the same target with laser pulses of reduced intensities (≈10¹⁷ W/cm²): we observed a spectral shift of only 1.8±1.0 eV (0.9±0.55 mÅ) and the line-width measures up to 5.8±0.25 eV (2.7±0.35 mÅ). These data provide evidence of plasma polarization shift of the Ti He-α line.

1. Introduction

The subject of spectral line broadening and shift has been studied extensively, theoretically and experimentally both (see for example [1-22]). Its diagnostic characteristic has its relevance to plasma under extreme conditions and hence of interest to researchers actively pursuing the Inertial Confinement Fusion (ICF) scheme of harnessing the thermonuclear energy [23] and also to astrophysicists [24]. Spectral lines emitted from a system may be broadened and shifted due to interaction of the emitter with the free electrons in the system. When averaged over time, this interaction produces an excess of negative charge around the nucleus of the emitting ion. This can lead to overlapping orbits of the bound electrons of the emitter and, hence, partially shields the nuclear charge. Consequently, the energy level structures of the emitting ion in a plasma environment are shifted depending on the electron density of plasma. Such a shift is usually termed as plasma polarization shift (PPS). Recently, a generalized analytical approach (valid for any ion and quantum numbers) has been developed to calculate energy level due to plasma polarisation shifts [25].
Since the line shift depends strongly on the initial state of the emitter, particularly its principal quantum number, we do not find many studies about the low lying lines, for example the α and β resonance lines. Majority of the studies concentrated on low Z plasmas. With the availability of high contrast ultra-short laser pulses and the highly efficient curved crystals, however, such studies have been recently reported. Saemann et al [16] have reported a red polarization shift (~3.7±0.7 eV) and 20 eV for the Al Ly-α and He-β line respectively. Renner et al [18, 22] have presented a comprehensive study of the Al Lyman series (α-η) and He-α indicating a strong dependence of the shift on the density. The observed shifts for the Ly-α and Ly-β lines were too small to draw a definite conclusion about their agreement with the quantum mechanical impact theory [9]. Woolsey et al [19] have reported a wavelength shift of about 4mA of He-β line in Ar plasma. Their results indicate a systematic red shift with increasing density. Andiel et al [21] have very recently reported line shifts for Ly-α Ly-β, He-β and He-γ line in aluminium plasma generated by irradiating buried layer of Al in carbon of varying thickness using 395 nm, 150 fs pulses with plasma densities approaching 5x10^{23} cm^{-3}.

Despite of numerous theoretical and experimental progresses in understanding the spectral line shifts in dense plasmas, the subject remains controversy. Although the existence of x-ray line shifts in dense laser produced plasmas is now widely accepted [17, 19, 26], large red shifts at near solid density [16, 27] are sometimes questioned due to overlapping satellite transitions and uncertainties in wavelength markers [22, 28].

We report here on an experiment where we have observed a line shift of ~3 eV in He-α resonance spectral line from Ti, to our knowledge shift of the He-α line from the highest Z element reported so far, by focusing high contrast frequency doubled (400 nm), 45 fs p-polarized pulses on 12.5 micron thick high purity titanium (Ti) foils.

2. Experimental

We carried out the experiment at the Rutherford Appleton Laboratory using the terawatt short pulse laser facility ASTRA. The system is capable of delivering half a joule of infra-red (800nm) pulses with duration of 45(±5) fs. The output pulse is accompanied by a pre-pulse with a contrast of 10^{-7} reaching the target 13ns before the arrival of the main pulse. In addition, the main pulse is superimposed on a residual uncompressed CPA pedestal and ASE. We used a third order auto-correlator in 800nm wavelength to measure the pulse contrast of the beam and it was found to be 10^{-5} at 1.5 ps before the arrival of the main pulse.

We employed a 0.6 mm thick type-I KDP crystal to frequency double the 800 nm (IR) beam to a 400 nm (blue), s-polarized beam with a conversion efficiency of 20%. A beam splitter, reflecting the blue and transmitting the IR, was used at 45° to relay the blue beam onto the target. The IR part of the beam was dumped at the back of the splitter. Any IR component reflected off the beam-splitter was further reduced by two dielectric mirrors in the beam-path. With this arrangement, interaction of high contrast clean pulses with the target surface was made possible [29]. The motorized controlled target orientation mechanism ensured the interaction of p-polarized pulses with the target at different angles.

The blue beam (400nm) was focused onto the surface of the target surface at different angles from the normal to the target with the help of an f/2.5 off-axis silver-coated parabola plane. An 8-bit CCD coupled with a x40 microscope objective recorded the focal spot at different offsets using the low energy mode of the laser. The full width at half maximum (FWHM) at the best focus was measured to be ~2 μm with energy content of about 35%. A retro-alignment system was in place by collinearly injecting a diode laser via one of the dielectric mirrors that allowed us to find the best focal spot whenever the target was moved to a fresh position. A 10 μm wire was then inserted into the focal plane and the reflection of focal spot from this was transmitted back to the alignment system and an image of the focal spot formed on a CCD camera equipped with an objective. In this way, using the very small amount of backscattered light even from smooth target surfaces, the position of best focus onto the foil could be identified when the target was moved 1mm between shots.

The focal spot on target was varied by moving the parabola off the best focus position along the line-of-focus by a known amount with the help of a micro-controller, towards the target and away from the target (referred to as positive off-set and negative off-set respectively). With the negative off-set the incident beam converges inside the target while in case of the positive off-set the focus lies before the target and a divergent beam interacts with the target. A small leakage of the 400 nm light through the second dielectric mirror in the incident beam was used to monitor the energy of the blue beam in each shot. This was achieved through a
photodiode coupled to an integrating sphere. An IR filter was employed to cut out 800 nm light. At the best focus position of the parabola, the intensity of the blue beam on the target reached a maximum of $\sim 10^{19}$ W/cm².

To ensure interaction of the beam with a fresh surface of the target, after every shot, the target was moved by 1 mm with an external computer controlled micro-drive. For every shot, before moving the parabola to the desired offset position, the beam was first imaged through the retro system at the best focus position of the parabola. The parabola was protected from the plasma debris by placing a thin glass pellicle in front of the parabola.

The time integrated single shot line emission spectra from Ti plasma, in the range of 4400 eV to 5000 eV, were recorded on individual shots with a Von-Hamos (VH) spectrometer coupled with an x-ray CCD system [30,31]. The spectrometer comprises a cylindrically curved (5 cm radius of curvature), 25 mm wide and 15 mm arc-length, LiF (200) crystal having a 2d lattice spacing of 4.027 angstrom, on a micro-controlled base stage in a lightproof metal box. The line of sight of the spectrometer (crystal centre and the source position) made an angle of $\sim 68^\circ$ with the horizontal plane.

We used $\lambda(K_\alpha_2)_{\text{ref}} = 2752.195$ mÅ (4504.92 eV) and $\lambda(K_\beta)_{\text{ref}} = 2513.96$ mÅ (4931.83 eV) from the NIST site [32] as our wavelength marker. With profile fitting (see the following section), total spectral resolution of the spectrometer is estimated to be $\lambda/\delta\lambda = 1350$. With the given set-up, one pixel on the CCD corresponds to an interval of $\approx 0.65$ eV ($\approx 0.35$ mÅ).

3. Results and Discussions

Figure 1a shows an integrated lineout of the spectrum obtained by an electronic sum of spectra from 13 data shots individually recorded when Ti foil is irradiated with high contrast 400 nm, 45 fs, p-polarized pulses at 45° angle of incidence. In all these shots, the parabola position is set at the best focus (with an accuracy of $\pm 15 \mu$m). These shots are recorded during the same laser run without letting up the chamber. The spectral interval from the cold K-$\alpha$ up to K-$\beta$ is shown in logarithmic intensity scale. Apart from the K-$\alpha$ and K-$\beta$ line, the resonance line of He-$\alpha$, inter-combination line Y and the K-$\alpha$ satellites (Li, Be, B, C, N, O, F) are also visible. With the presence of K-$\alpha$ and K-$\beta$ line on the spectrum we can use two reference lines almost at the two extremes of the detector and check the accuracy of our wavelength calibration for the intermediate values. K-$\beta$ line can be seen in single shot data taken at larger offset ($\approx 400$ micron) from the best focus position due to lower backround hard x-rays. Although the level of He-$\alpha$ line emission in these cases was too low to be recorded, it was crucial to monitor possible changes in dispersion during the same laser session or from one session to another.

We used Voigt profile fitting to find the central pixel position for the emission lines. In figure 1b-c, K-$\alpha_1$, K-$\alpha_2$ and K-$\beta$ are fitted by means of Voigt profiles taking the line centre position of K-$\alpha_2$ and K-$\beta$ as a reference line on the spectrum. The best fit for K-$\alpha_1$ is obtained with a line centre position of $\lambda(K_{\alpha_1}) = 2748.7$ mÅ (4510.7 eV). This value is in very good agreement (within $0.15$ mÅ or $0.2$ eV) with the reference value of $\lambda(K_{\alpha_1})_{\text{ref}} = 2748.5471$ mÅ (4510.899 eV) [32], indicating an appropriate use of K-$\alpha_2$ and K-$\beta$ as our wavelength marker to find out the intermediate dispersion with a precision well above the values of shifts and widths to be discussed below. Figure 1d shows Voigt profile fits to the experimental profile of He-$\alpha$ and the inter-combination line. The best fit is obtained for the He-$\alpha$ line centre position of at 2612.2 mÅ (4746.3 eV) and FWHM of 7.1 mÅ (12.8 eV). This indicates a red shift of 1.85 mÅ (3.4 eV) for the He-$\alpha$ resonance line 4749.7 eV [32]. We note that the FWHM as obtained from the fits include all sources of broadening (e.g., Stark, Doppler, opacity, line overlapping, instrumental).

Because we see in figure 1a, evidence of inner-shell K-$\alpha$ emission from different ion stages and because we expect fast electron resistive heating, we would like to exclude its effect on the K-$\alpha_2$ spectral position. We compare single-shot spectral data from Ti foil irradiated at the optimum focus of the parabola and at an offset of 100 $\mu$m from the best focus position.
Figure 1: (a) An integrated line-out of the spectral data electronically summed over ~10 shots during the same session. These data are electronically corrected for the background and presentation on a log scale. The red curves show Voigt profile fits to the experimental profile of (b) K-\( \alpha \), (c) K-\( \beta \), and (d) He-\( \alpha \), intercombination line Y and Li-like satellites.

Figure 2: Spectral line out of the data recorded from single shot when Ti foil is irradiated at the optimum focus (a,b) and at an offset of 100 \( \mu \)m (c,d) along with the Voigt profile fits. The spectral band in the vicinity of K-\( \alpha \) is shown in (a, c) He-\( \alpha \) is shown in (b,d).
We compared the spectral positions and widths of the K-α and He-α resonance line emitted from Ti foil irradiated at an offset of ~100 μm (Fig 2c, d) to a shot at the best focus position (Fig 2a, b). These two shots were recorded during the same experimental session without letting-up the chamber and without disturbing the position of the spectrometer. Once again Voigt profile fitting procedure was used, as outlined above, to work out the line centre position. The K-α₂ line centre position in the best focus case is fitted at 110.9 pixel (figure 2a) whereas the 100 μm offset case gives 111.0 pixel. The difference of these values (~ 0.1 pixel) translates into a wavelength difference of 0.04 mÅ or energy difference of 0.06 eV which is about an order of magnitude better than the desired wavelength precision. Furthermore, as can be seen from the insert in figure 2, the difference in the FWHM of the K-α emission in these two cases is also only 0.1 pixel. With two orders of magnitude difference in intensity (~10¹⁹ and ~10¹⁷ W/cm²), our simulation (see below) show a considerable difference in temperature and density. Negligible change in the FWHM and the central pixel position of K-α emission in these two cases demonstrates that the ion stages and resistive heating have negligible effect on the observed K-α₂ wavelength position. Hence, justifying the use of λ(K-α₂)ref = 2752.195 mÅ as a reference line for our wavelength calibration.

Similarly, Voigt profile fit was carried out for the He-α resonance line and inter-combination line as was done in figure 1. Figure 2b and 2d show lineouts of the spectra with Voigt fits, in the vicinity of He-α resonance line, from Ti foil irradiated at 45° with p-polarized pulses at the best focus and at an offset of 100 μm respectively. These spectra are from the same shots as above figure 2a and 2c. The vertical dotted lines indicate the un-shifted position of He-α resonance line at 4749.73 eV [33] in both cases. For the optimum focus case, we measure a spectral red shift of 3.4 eV (~2 mÅ) for the He-α resonance line and FWHM of 12.1 eV (6.7 mÅ). These values are similar to the values obtained for the integrated case discussed in figure 1. For the second case of irradiation at an offset of 100 μm from best focus, we measure a spectral red shift of 1.8 eV (0.95 mÅ) and a FWHM of 5.8 eV (2.7 mÅ). These values are almost half of that measured for the optimum focus case. Our data from electronically integrated shots at best focus from Ti foil irradiated at 30° with p-polarized pulses also indicate a shift of about 3.6 eV (2.1 mÅ). Again, very similar to the data of 45° irradiation presented above. Because of the noise in the spectral data, any shift in the Li-like satellite lines could not be ascertained.

We carried out hydrodynamic simulation using HYADES [34] (the code has the facility of electromagnetic wave solver) for the Ti foil irradiated with 400 nm, 45 fs p-polarized pulses at an angle of 45° to the target normal for these two cases in order to estimate the difference in average plasma density as observed with x-ray emission spectroscopy. We observed that the density exceeded 10²⁴ cm⁻³ with a peak temperature above ~3keV for the optimum focus whereas for the defocus case we noticed lower density and temperature well below 1 keV. The hydrodynamic output data from HYADES were post-processed with the collisional radiative model SOBOLEV [35] to simulate the temporal profile of the He-α emission. Our results show that for the optimum focus, the density exceeds 10²⁴ cm⁻³ with a peak temperature above ~3keV whereas for the defocus case a temperature well below 1 keV is observed.

We have independently cross checked the hydro-simulations of emission averaged data with the spectral simulation code MARIA [36-38]. The fits (collisional radiative model including opacity and hot electrons, for further details see [36-38]) of the time integrated data of He-α, the intercombination line Y and the associated dielectronic satellite structure indicate a bulk electron temperature of 0.8-1.0 keV and a density of about (3-10)×10²³ cm⁻³. This is in reasonable agreement with the simulated electron density of 0.5-1×10²⁴ cm⁻³ and an electron temperature of 0.5-1 keV from about t=1–10 ps. Moreover, the particular spectral distribution of the Li-like 1s2121l'-saturates indicates [36] that the electron density in the case of tight focus is near 10²⁴ cm⁻³, whereas for the case of 100 μm defocusing, the electron density is more close to 10²³ cm⁻³. This is also in rather good qualitative agreement with the hydrodynamic simulations.

Another possible source of line shift is the directed Doppler shift as well as differential plasma motion in optically thick plasmas. The maximum red shift due to Doppler effect in our case is ~0.3 eV (0.17 mÅ), This is far less than the observed shift. Similarly, a differential plasma motion of the order of ΔE/E = 6ΔV/c = 0.3 eV/4.7 keV = 6×10⁻² will not lead to a significant shift of an optically thick line profile.

The hydrodynamic simulation shows that the density in case of the optimum focus remains about 3 times to that of the 100μm offset case for the relevant duration of the emission. As lower shifts correspond to lower plasma densities and all possible sources of line shifts are significantly below the observed ones we have a clear evidence of having observed the plasma polarisation shift of He-α of titanium, the highest Z reported so far.
4. Conclusions

To conclude, we have observed a spectral red shift of \(~3.4\text{eV}\) for the He-\(\alpha\) line emission from plasma generated by focussing intense high contrast 45fs, 400nm p-polarized pulses at 45° degree on Ti foil at \(~10^{19}\ W/cm^2\). At lower irradiance of \(~10^{17}\ W/cm^2\), this shift reduces to \(~1.8\text{eV}\). The difference in the peak-shift may be considered as a signature of contribution from the plasma polarization effect and our simulations of foil targets and spectral distributions support this thesis.

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