Shift and width of the Balmer series Hα line at high electron density in a laser-produced plasma

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Abstract
Time-resolved profiles of the Balmer series Hα line emitted by a laser-produced hydrogen plasma have been measured to determine the shift and width for electron densities from below 10^{18} to above 10^{20} cm^{-3} at an average temperature of 28000 K. Fits of the profiles that allow for self-absorption in the plasma yield shifts and widths that are consistent with experiments on lower density and cooler gas-liner pinch plasmas. The width scales as \( N_e^{0.70 \pm 0.03} \) and the shift as \( N_e^{0.92 \pm 0.03} \) between \( 8.7 \times 10^{17} \) and \( 1.4 \times 10^{20} \) cm^{-3}. Hα shifts monotonically and nearly linearly to the red with increasing density under the reported conditions. A comparison to theory calculations using exact potentials for \( ^2H \) shows that an intrinsic asymmetry becomes significant only in the upper limit of this range when a satellite develops in the far red wing.

Keywords: line shapes, widths, shifts, optical spectra, shock waves, atomic and molecular data

1. Introduction

Observations of the broadening of the hydrogen Balmer-series lines, supported by a well-tested theoretical framework, are used widely as a diagnostic tool for determining the electron density in laboratory and astrophysical plasmas (Stehlé et al 2005, Koester 2010). The first line of the series, Hα, is useful at the highest electron densities and has been a subject of special theoretical and experimental effort.

The standard theory of broadening by collisions with electrons and ions (Griem 1964, 1974, 1997) when applied to Hα is now known to be in agreement with carefully diagnosed experiments on several different laboratory plasmas for electron densities at least as high as \( 10^{18} \) cm^{-3} (Wiese et al 1972, Büscher et al 2002, Griem 2000, Griem et al 2005). However, broadening of Hα at densities above \( 10^{19} \) cm^{-3} approaches a difficult experimental regime where line asymmetry, background continuum, and radiative transport through the optically thick plasma all become significant. Consequently in some instances careful assessment of the experiment and the validity of proposed alternatives to the standard theory have led to conclusion that if deviations from the standard theory occur, they are above \( 10^{19} \) cm^{-3} (Fligh et al 2003, Oks 2002, Griem et al 2005).

However, high quality data for Hα from gas-liner pinch experiments up to \( 10^{19} \) cm^{-3} (Büscher et al 2002, Böddeker et al 1993) disagree with outside statistical error with standard theory models for width and shift (Griem 1974, 1988), and with other calculations (Büscher et al 2002, Alexiou and Leboucher-Dalimier 1999). Griem’s examination of Stark broadening theory and available experimental data for dense plasmas (Griem 2001a, 2001b, Griem et al 2005) noted that a comparison between theory and experiment could be problematic because of the difficulty of interpreting line profiles that were intrinsically asymmetric to determine the shift of the
line in the experimental work, and of the implementation of Debye shielding in the theory. In a conference paper on a laser-plasma experiment, for example, Bock reported shifts that were smaller than the standard theory results extrapolated to high density, but with shift-to-width ratios of the order of 0.1 consistent with the standard theory (Bock et al 2001).

Given this analysis, computationally intensive numerical simulations for high density conditions offer an alternative theoretical method to test the fundamental physics and approximations used to explain Stark broadening, leading to the conclusion that densities above $10^{19} \text{ cm}^{-3}$ may enter a regime where standard theory breaks down (Demura 1988, Hallek 1990, Gigosos and Cardenoso 1996, Gigosos et al 2003).

New experiments that could be compared with both standard theory and with numerical simulations should be useful for reaching the goals of refining methods for plasma diagnostics in extreme conditions, and confirming theoretical calculations for astrophysical applications (Steblé et al 2005, Demura et al 2008, Tremblay and Bergeron 2009, Koe- ster 2010). Here we report measurements of Hα for electron densities in excess of $10^{20} \text{ cm}^{-3}$, conditions under which the line shifts more than 100 Å to the red as it broadens to more than 1000 Å full width.

2. Experimental methods

For the work reported here we used a table-top laser-produced pure-hydrogen plasma which has been described previously (Kielkopf 1995, 2001, Kielkopf et al 2002, 2004). A compact five-way high vacuum stainless steel cross with 4 cm diameter arms was the target chamber. Two orthogonal optical paths, O-ring sealed with optical quality fused silica windows, provided for laser excitation, beam dump, emission spectroscopy, and imaging diagnostics. The chamber was evacuated with an oil-free turbomolecular pumping system, and then filled at room temperature with 99.999% H₂ gas to static pressure of 600 Torr as measured with a calibrated capacitance manometer.

A Continuum Surelite II 1064 nm Nd:YAG Q-switched laser provided 6 ns, 390 mJ, pulses at 10 pulses s⁻¹ with a uniform Gaussian beam nominally 6 mm in diameter. The beam was focused into the chamber by a 10 cm focal length well-corrected lens to a diffraction-limited spot (in a vacuum). With H₂ present at pressures above 400 Torr, there was no significant transmitted laser energy, and a bright linear plasma appeared displaced toward the laser from the vacuum focal point. Limitations on laser power and gas fill pressure were imposed for safety during the experiment runs discussed here (Kielkopf et al 2002).

As shown in figure 3 of (Kielkopf 1995), images integrated over several microseconds reveal that a cylindrical plasma develops around the focal line. Close inspection of these images and spatially time-resolved diagnostics show that the laser self-focused in front of the vacuum focal point, and threaded through the volume around the optical axis, creating a short line only a few μm in diameter within which the laser energy was absorbed. The visually impressive ‘spark’ that resulted was remarkably stable in position and average length. As we reported in (Kielkopf 1995, 2001), the emission resulted from a cylindrical shock wave that traveled outward from the axis. It dissociated the molecular gas as it propagated, and then in ionization equilibrium excited the atomic H gas to emit a spectrum from the region of peak atomic density corresponding to the upper state of the spectral line in question. Following the laser pulse and the fast propagation of the shock front a hot bubble of atomic H surrounded by cold H₂ was left in its post-shock wake. The fascinating dynamics of the bubble was described by (Kielkopf 2001), but for our purposes only the earliest times of emission from the shock front itself are of interest.

Spectra were obtained by focusing an image of the plasma observed through the cross along an axis orthogonal to the laser excitation axis, onto the slit of a 0.25 m digitally-controlled aberration-corrected Czerny–Turner grating spectrometer. The instrument was optimal for measurements in the Hα spectral region, with a 1200 groove mm⁻¹ grating blazed at 7000 Å and a scanning range extending from the ultraviolet through the near-IR. Wavelengths were calibrated by comparison to a low-pressure Hg–Ar lamp. The selected slit width gave an instrumental width of 10.2 Å measured from the narrow lines of the calibration spectra. The relatively low resolution used here is an appropriate match for the very broad lines produced by the source. Light was detected with a fast (response <1 ns) InSb PIN photodiode and a Stanford Instruments SR250 integrating amplifier with gate width of 5–10 ns after a controlled delay from the initiating laser pulse. Although the gate was triggered electronically by detecting the infrared light from the laser, the reference time for the physical delay zero point was the first detectable emission from the plasma. Given the gate width and the response time of the detector, the uncertainty in the delay times is of the order of 5 ns, which is comparable to the duration of the laser pulse. This arrangement and the calibrations used were similar to those reported for measurements at higher neutral gas density by (Kielkopf et al 2002), except that here we employed a higher speed diode sensor, and narrower gate widths, to record spectra at the earliest times possible for these plasmas.

We note that there were no spectroscopically detectable impurities other than weak C I emission in the vacuum ultraviolet. At the early gate times the spectra are nearly featureless continua apart from a very broad Hα feature. With increasing delay time the Hα line sharpens and shifts toward its usual, perturbation-free, wavelength, and higher lines of the Balmer series appear. We have shown that this source is a temporally evolving LTE plasma for which conditions as a function of time may be modeled using shock wave theory (Kielkopf 1995, Kielkopf et al 2004). Based on these models, this clean table-top laser plasma source reached an electron density of more than $10^{20} \text{ cm}^{-3}$ shortly after the nanosecond time-scale pulsed laser initiated the plasma. Subsequent time-resolved observations selected electron densities down to the order of $10^{18} \text{ cm}^{-3}$, thus overlapping the currently assured region of line shape theory validity. The models show a
temperature in the laser-plasma source that is intermediate between that of wall-stabilized arc and the gas-liner pinch experiments, but an electron density that is an order of magnitude higher. These laboratory conditions are uniquely interesting because they are tunable by selecting the delay time and span those found in stellar white dwarf atmospheres (Koester 2010).

The modeling of the plasma described previously (Kielkopf 1995) has been incorporated into a Mathematica (Wolfram Research, Inc. 2008) program that is also used to predict the propagation of the shock front and the electron density at later times when it can be measured through the H β linewidth, thus adding confidence to its use as a tool to establish the electron density and temperature immediately following the laser pulse. An example is shown in figure 1 where the calculated electron density $N_e$ and the density of the Hα $n = 3$ upper state, $N_3$, as a function of distance off the axis of the plasma for two representative times after the laser pulse are plotted. The upper state density is sharply peaked, and since the most intense emission arises from the radial zone where there are the most atoms, it selects a well-defined electron density. For each selected delay time, there is a corresponding density and temperature under which Hα is emitted. This region is also close to the maximum in electron density at that time, so emission from other nearby zones will also be representative of nearly the same electron density. Each wavelength that is measured at a given delay has at least 30 laser shots averaged to make the recorded data point. Although there are shot to shot variations in spatial pattern along the focal axis due to the random nature of the threading of the laser beam and self-focusing, the radial extent, and thus the comparison to the cylindrical model, appears to be stable. This is particularly obvious when the post-shock bubble is imaged by probing the hot gas with a second delayed laser, as was done in experiments reported by (Kielkopf 2001). The hot bubble was uniform from shot to shot, proving that the deposition of energy was consistent. There is, however, a gradient in the deposition along the axis because the energy which is available decreases as the laser propagates. We see this in a slight ellipticity of the resulting bubble, and ultimately in the development of gas flow back toward the laser. Thus we have to regard the model as providing an estimate of the average electron density and temperature in the region responsible for Hα emission. While it is not as quantitatively precise as the excellent measurements of laboratory standards diagnosed by several methods, it offers an exceptional range of electron density and an impurity-free environment.

Table 1 lists the different delay times for which we acquired data, the calculated electron densities, and the plasma temperature. The uncertainty in $N_e$ given in the table is the range expected for the gate width. At the earliest times the plasma is changing very rapidly and the uncertainty in density due to integration over time is greatest. The uncertainty given does not reflect an uncertainty in the plasma theory. In this regard we know that at later times, when the density is low enough for Hβ to be used as an independent diagnostic, the measured and computed densities are in good agreement. The theory also predicts the expansion of the shock front for delays much longer than those used here, which we note were consistent with schlieren measurements in other experiments.

3. Spectra

The experiment operated with the excitation laser running at 10 Hz. At each wavelength the gated signal at a selected delay time was averaged with analog electronics over 30 laser pulses and then digitized at 16-bit accuracy and stored for subsequent processing. For a given delay, each complete recorded spectrum covered the Hα profile into the continuum on both sides of the line. Typical spectra at three different delay times are shown in figure 2.

A cursory examination of the spectra reveals that, as expected, at short delays the Hα line is very broad and shifted to the red. It is also apparent that especially at short delays,
the plasma is optically thick close to the unperturbed Hα line center. The reversal arises from the temperature gradient and the presence of $n = 2$ atoms along the optical path, and a determination of the intrinsic line width and shift has to take this into account. This behavior is not unexpected, and it occurs often in dense hydrogen plasmas. It is unusual, however, to see it appear in a dynamic sequence of spectra such that the self-reversal from cool hydrogen outside the emitting region can be isolated by modeling the entire profile to obtain an emission line width from the hot dense inner plasma at the shock front.

To extract useful information from these complex line shapes, after calibration for wavelength, the spectra were fitted with an expression of the form

$$
F(\lambda) = f \frac{w^2}{(\lambda - \lambda_0 - d)^2 + w^2} \\
\times \exp \left( -a \frac{w_0^2}{(\lambda - \lambda_0 - d)^2 + w_0^2} \right) \\
+ b + c(\lambda - \lambda_0)
$$

that allows approximately for both the continuum background and the self-absorption through a two-component model. While this analysis cannot separate the effects of self-absorption within the inner radiative shell of the shock, it allows for the cooler outer neutral regions which do leave an obvious signature in the observed profiles. We note that the fits achieved with this simple approach are largely within the scatter of data for each profile. The shifts which are obtained in this fitting process would correspond to those usually found in theoretical calculations by examining the slope of the imaginary part of the exponential damping of the auto-correlation function. Even for an asymmetric line profile, the shift defined this way is readily connected to the physical processes underpinning the observed line shape. Alternatively, in experimental work one may find the shift by locating the peak of the profile, or its bisector at half the peak value or half the area. Defining a shift by the peak position is problematic because the derivative is zero there, and defining it by the bisector depends on how the bisector is defined and how far into the line wings the measurements are made. We use the Lorentzian profile and its $\chi^2$ fit to the data as the most practical solution in this situation, and again note the unambiguous results of the fitting.

Equation (1) was incorporated into the non-linear least squares fitting routine of Grace (Stambulchik 2008), which performs iterative non-linear regression to optimize the eight unknown parameters that describe the profile. The Grace data analysis tool includes fitting routines from MINPACK (Moré et al. 1999), modified to work in that application which offers a graphical interface to the selection of initial parameters, the fitting region, and the convergence. As much of the spectrum as possible was included in the fitting. At longer delays and lower density the background is essentially flat outside the profile. At very short delays the data are noisy and we estimate the region where the profile disappears into the continuum and use that cutoff before fitting. Once approximate solutions are found, the chi-square search settles quickly to a good match to the observed spectra, as shown in figure 2. All of the spectra were treated individually this way, and the automated convergence was tested by trials to explore other possible minima in chi-square. We made use only of the width and shift from the region of the plasma which we could associate with a known electron density and temperature from the plasma model. The full width at half maximum $2w$, the shift $d$, and the width-to-shift ratio $2w/d$ derived at various delays $t$ are given in table 1. Corresponding electron densities $n_e$ and temperatures $T$ are from the theoretical plasma model. Over the observed time delay interval, the plasma emitting Hα decreases in electron density from $1.4 \times 10^{20}$ to $8.7 \times 10^{19}$ cm$^{-3}$, but the temperature drops less dramatically from 34500 K to 20100 K.

The uncertainties reported in the table for width and shift are standard deviations extracted from the non-linear fitting. The instrumental width for the spectrograph would not be significant except at the lowest electron densities, and the wavelength calibration error is a negligible contribution as well. The uncertainties result from a combination of random factors outside our control—the statistical fluctuations in the

| $\Delta t$ (ns) | $N_e$ (cm$^{-3}$) | $T$ (K) | $2w$ (Å) | $d$ (Å) | $d/2w$ |
|----------------|-----------------|---------|----------|--------|--------|
| 08 ± 5         | 1.39 ± 0.22 \times 10^{20} | 34486   | 2166 ± 317 | 373 ± 12 | 0.17 ± 0.03 |
| 20 ± 5         | 3.87 ± 0.66 \times 10^{20} | 30265   | 813 ± 359 | 153 ± 52 | 0.19 ± 0.18 |
| 30 ± 5         | 2.55 ± 0.32 \times 10^{20} | 28535   | 572 ± 68  | 113 ± 14 | 0.20 ± 0.05 |
| 40 ± 5         | 1.90 ± 0.25 \times 10^{20} | 27852   | 432 ± 61  | 93 ± 11  | 0.22 ± 0.06 |
| 50 ± 5         | 1.49 ± 0.13 \times 10^{20} | 27018   | 431 ± 61  | 84 ± 10  | 0.19 ± 0.05 |
| 60 ± 5         | 1.22 ± 0.09 \times 10^{20} | 26299   | 350 ± 60  | 80 ± 9   | 0.23 ± 0.07 |
| 70 ± 5         | 1.03 ± 0.09 \times 10^{20} | 25613   | 257 ± 42  | 65 ± 11  | 0.25 ± 0.09 |
| 80 ± 5         | 8.85 ± 0.47 \times 10^{20} | 25160   | 314 ± 23  | 62 ± 13  | 0.20 ± 0.06 |
| 100 ± 5        | 6.96 ± 0.18 \times 10^{20} | 24562   | 265 ± 63  | 30 ± 21  | 0.11 ± 0.11 |
| 200 ± 5        | 3.30 ± 0.05 \times 10^{20} | 22793   | 139 ± 23  | 12 ± 3   | 0.09 ± 0.04 |
| 400 ± 5        | 1.43 ± 0.01 \times 10^{20} | 20880   | 67 ± 3    | 5 ± 1    | 0.08 ± 0.02 |
| 600 ± 5        | 8.65 ± 0.14 \times 10^{19} | 20133   | 53 ± 9    | 3 ± 1    | 0.06 ± 0.03 |

Table 1. FWHM ($2w$) and shift ($d$) of Hα.
photon count, timing jitter, and coupling of the laser to the gas from shot to shot as the spectrograph scans the line profile being the most obvious ones. There are also uncertainties in the electron density associated with each delay, though these are more difficult to quantify. Even in a hypothetically ideal experiment, at the earliest times the finite width of the timing gate means that the plasma is changing its density significantly as data are acquired. The uncertainty given in reflects this change.

4. Discussion

Figures 3 and 4 show these width and shift data together with other recent experimental measurements. While the uncertainty for the new laser plasma data is larger than for some of the other experiments, they extend the available range of in an order of magnitude upward. A monotonic increase of both the width and shift of density is apparent in these new data points. Fitting all experimental data shown in these figures to a power law gives in (Å) ±−21 5 10 5.5 w n e = × ±−(2) and

\[ d = + 1.23 \times 10^{-16} n_e^{0.92} \pm 0.03 \]  

(3)

for \( n_e \) in \( \text{cm}^{-3} \). This is remarkably close to the behavior predicted by standard Stark broadening theory (Griem 1974, 1983) in which, with a weak temperature dependence, the width increases as \( n_e^{2/3} \) and the shift to the red is linear in \( n_e \). The shift-to-width ratio is nominally 0.1 as expected over this range of density. Gigosos has run computer simulations of H\( \alpha \) profiles for densities up to \( 10^{19} \text{ cm}^{-3} \) and found a power law dependence of width on electron density that is the same as observed here (Gigosos et al 2003). The simulations as well as standard theory also show a weak temperature dependence which we neglect in comparison to the strong effect of density over the range considered here.

We also compare the data to a calculation based on the exact \( H^+_2 \) potentials incorporated into the atomic collision framework for line shape calculations but not including electron collision broadening. In the context of neutral and ion atomic spectral line shape literature we refer to this as a ‘unified’ theory in that it computes a complete line profile at all frequencies consistently, and in this case offers a solution for ion broadening which differs from the approximations usually included in standard Stark theories.

Figure 5 compares a theoretical profile with the experimental data corrected for continuum background. It illustrates
how less-broadened hydrogen absorbs at a nearly unshifted frequency, revealing the asymmetric effects of self-reversal on the line shape and confirming the small shift associated with broadening at this density. It also shows the range and significance of the far wing of Hα extending into the near-infrared. A satellite feature due to ion-atom collisions arises that is weak at this density, but at higher densities is expected to create a rapid non-linear shift to the red, beginning at the density of 1020 cm−3 that is weak at this density, but at higher densities is expected to converge with the measurement of Hα, as densities approach 1020 ions cm−3 (Grieg 1983, Gigosos et al 2003). The shifts also follow the trend seen in previous measurements at lower density, at least until the shift is of the order of 100 Å. However, at the highest electron densities for which we could reliably measure the profile, the observed shift falls significantly below the power law extrapolation of lower density experiments and the theory of (Grieg 1983). Profiles due to Hα collisions calculated with the complete molecular potentials of H2+ appear to converge with the measurement of widths and shifts at these elevated densities where significant asymmetry due to ion collisions is expected (Allard et al 1999).

5. Conclusion

We have observed that the Balmer series Hα line broadens and shifts monotonically as electron density increases to more than 1020 cm−3 for temperatures approaching 30000 K. The broadening is generally in agreement with standard theories, and it follows the trend found in simulations for densities below 1019 cm−3 (Grieg 1983, Gigosos et al 2003). The shifts also follow the trend seen in previous measurements at lower density, at least until the shift is of the order of 100 Å. However, at the highest electron densities for which we could reliably measure the profile, the observed shift falls significantly below the power law extrapolation of lower density experiments and the theory of (Grieg 1983). Profiles due to Hα collisions calculated with the complete molecular potentials of H2+ appear to converge with the measurement of widths and shifts at these elevated densities where significant asymmetry due to ion collisions is expected (Allard et al 1999).

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