Hydrogen Balmer-α broadening in dense plasmas

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This work presents a theoretical analysis of experimental results for the hydrogen Balmer-α line in dense plasmas, with electron densities between 2×10^{18} and 9×10^{18} e/cm^3. A simulation of *both* electrons and ions is employed to produce reliable theoretical widths. These results are essentially in agreement with standard theory results and, for the most part, disagree with the experimental results. Consequently, either mechanisms not accounted for in the theoretical results (such as quadrupoles) are more important than previously thought at these densities, or else there is a problem in the experimental data (such as a possible reabsorption, which is not ruled out by the experimental data). [S1063-651X(99)00208-1]

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I. INTRODUCTION

Because of its simple atomic structure and astrophysical importance, hydrogen is a well-studied element in Stark broadening of spectral lines, and widely used in diagnostic applications. Usually these studies and applications are concerned with the low and medium density plasmas (electron density $n\leqslant10^{17}$ e/cm^3). In a recent paper [1], experimental data for the hydrogen H_α line were reported for densities between 2.44×10^{18} e/cm^3 and 9.27×10^{18} e/cm^3, and temperatures of 7–10 eV. These results were obtained from a gas-liner *z* pinch, and the plasma was independently diagnosed by Thomson scattering. These results were compared with standard theory (ST) theoretical results [2,3] in Ref. [1] and serious discrepancies were found. Since the understanding of hydrogen broadening in such plasma conditions is crucial, especially in view of current interest in broadening for denser plasmas, it is important to resolve these discrepancies. For this purpose we present in this work benchmark theoretical results for hydrogen line broadening at the conditions of this experimental study. The new theoretical results presented here are obtained by means of a joint simulation of electrons and ions. The use of this most sophisticated method, which has *always* until now given excellent agreement with experimental data, is needed to remove any ambiguities, namely,

(i) Griem [4] has maintained that the issue of dropping the interference terms in the impact approximation is still unresolved [5]. Although in the author's opinion the line-broadening community has long sided with Voslamber's opinion [5] that the interference terms should not be dropped, as was evidenced by the complete lack of implementation of such dropping of the interference terms in line-broadening codes, for the present calculation this question does not even arise, as the interference terms arise in a perturbative impact calculation and the present results are fully nonperturbative, for electrons as well as ions. We note that Oks has recently given an analytic proof [6] that the interference terms should *not* be dropped.

(ii) Questions regarding the validity of the impact approximation and the perturbative impact approximation used in the standard theory results, compared to the experimental data in [1] at these high densities, also do not arise as the impact approximation is *not* employed.

(iii) Questions regarding a possible electron-ion coupling at these high densities, as found in the ''convergent theory'' [7] and also expected theoretically [8,9] also do not arise in the present treatment, which employs a *joint* electron-ion simulation and, hence, whatever coupling effects there may be are already included in the calculation.

(iv) Questions regarding deviations from quasistatic behavior for the ions, since we are dealing with a line with an unshifted central component. Such lines ''are not influenced by static fields and clear-cut validity criteria for the static-ion regime have proven elusive'' [10]. It is found in retrospect and may perhaps have been expected that the standard theory assumption of quasistatic ions is valid for the parameters of the experiment considered here. However, the criterion for the validity of the quasistatic approximation is that the half width at half maximum (HWHM) of the line be much larger than $\langle v\rangle/\langle\rho\rangle$, with $\langle v\rangle$ and $\langle\rho\rangle$ as a typical velocity and impact parameter for the perturbers that contribute most to the broadening. In practical calculations these are not known *a priori*, so one takes them equal to the average velocity and impact parameters, respectively. This criterion is then seen to be not too helpful due to the presence of the ''much larger than;'' as a factor of 10 may still not be enough [11,9]. This is due to the fact that impact parameters, significantly smaller than the mean interionic spacing, often give the dominant contribution to broadening (see the discussion) and, in agreement with the quotation in Ref. [10], render the *a priori* determination of the validity of the quasistatic approximation difficult. We should point out at this stage that the HWHM must be computed by including *all* broadening mechanisms, including electrons and, if they are important, dynamic ions.

The treatment used is thus seen to give definitive results with much more physics included than any other treatment thus far. Based on these results, which essentially confirm the ST results [3], we re-examine the assumptions in both experiment and calculations.

II. BRIEF DESCRIPTION OF THE CALCULATIONS

The calculation employs the collision-time statistics method [12] to generate ''relevant'' perturbers with the correct distribution of impact parameters, velocities, and times of closest approach. In other words, the perturbers are treated as independent quasiparticles interacting with the emitter, but not with themselves. This is a standard assumption in line-broadening theory and practice, and only very few works have employed molecular dynamics for very simple lines. As already mentioned, *both* electrons and ions are simulated this way. Using these randomly generated perturbers and their time-varying electric field based on a straight line trajectory assumption, we solve the Schrödinger equation in the stochastic field produced by the moving electrons and ions. In the simulation, the perturbing ions are taken to be singly ionized helium ions, although in the experiment the ion abundance was not determined. The ion perturber composition does not make much of a difference, as ions are quasistatic for these conditions. For example, at a density of 4.84×10^{18} e/cm^3 and a temperature of 8.4 eV, if we assume deuterium instead of He perturbers, the calculated width only changes by 3.5%. A key advantage of the collision-time statistics method is that it guarantees the recovery of the correct impact limit. These impact parameters, velocities, and times of closest approach are, in turn, used to generate random electric fields by assuming that the perturbers (electrons and ions) move in prescribed straight line trajectories. We then solve the Schrödinger equation for the time evolution of the atomic wave functions in the presence of this time-dependent random field. The quantity computed is the autocorrelation function (AF) $C(t)$, which is a linear combination of products of matrix elements (one matrix element between upper level states and one matrix element between lower level states) of the time evolution operator $U(t)$. Its Fourier transform gives the line profile. The Gigosos *et al.* group theoretical method [13], valid only for hydrogen in the dipole approximation with no quenching and without an account of fine structure splitting is employed, since it results in a significant reduction of run time by solving a completely equivalent system of differential equations with lower system dimensions (4×4 per level).

$N=400$ configurations were used in the calculations, and convergence was checked by comparing the $N/4$, $N/2$, $3N/4$, and N configuration AFs. It was found that convergence was never an issue in these calculations.

III. RESULTS

Table I compares the experimental results with theoretical predictions, both by the ''standard theory'' (ST) [1] and the present simulations. In the simulation results we quote the ''semiclassical dipole'' contribution by the joint action of electrons and ions followed (in parentheses) by a strong-collision Lorenz-Weisskopf estimate for the contribution of perturbers with impact parameters $<9a_0$, with a_0 as the Bohr radius. That is, the quantity $\{1-S_aS_b^\dagger\}$ with S as the S matrix, and the subscripts a and b denoting the upper and the lower levels, is replaced by unity for smaller impact parameters. The total width should be between the first value and the sum of the two. In principle, the total width could be as large as the sum of the first value quoted and twice the strong collision contribution, corresponding to a choice of $\{1-S_aS_b^\dagger\}=2$, which is the absolute maximum value allowed by unitarity. However, such a choice would correspond to the extremely unlikely scenario, where one S matrix remains equal to unity, while the other one is -1 for *all* velocities and impact parameters. Nonperturbative calculations show that the S matrices (and also their product) oscillate very strongly at small impact parameters, so this scenario is extremely unlikely.

TABLE I. FWHM (Å) experiment [1] vs theory.

Electron density (10^{18} e/cm^3)	T (eV)	FWHM (Å) Expt.	ST	Simulation
2.44	7	153±21	78	74.5(+29)
3.44	7.6	182±24	104	100 (+41.5)
4.84	8.4	187±36	134	122.7 (+61.6)
7.08	9.2	228±64	180	154.4(+94)
9.27	10	245±54	224	183.4 (+127.98)

The choice of the cutoff at $9a_0$ was made to ensure that the perturbers do not penetrate the atomic wave function extent and, hence, monopole interactions do not enter the picture. In contrast to ST, no unitarity cutoff is required since the calculation is fully nonperturbative, and unitarity is never violated.

These results are in agreement with the ST results and, at least for the lowest two densities considered, systematically lower than the experimental values.

IV. DISCUSSION

In view of these theory-experiment discrepancies, we must conclude that either there is an additional broadening mechanism that is not taken into account in the theoretical calculations, or that the experimental data have a systematic error that leads to larger widths. Possible candidates for the first scenario are quadrupole interactions, which are not included in the calculations. Quadrupole terms had been estimated in Ref. [3] and turn out to be quite small, though their estimated accuracy of ±50% for the quadrupole contribution has been questioned [14]. Other than that, for high densities, there are no effects known to cause an increase in line widths that are not accounted for in the calculations. The inclusion of perturber-perturber interactions [which would necessitate a molecular dynamics (MD) simulation, compared to the independent particle model used here], although clearly essential at very high densities, is not expected to result in any substantial changes here. For example, at the highest density, 34% of the width is contributed by impact parameters less than half the mean interparticle spacing (and, of course, larger than $9a_0$), and 74% by impact parameters less than the mean interionic distance. However, this remains to be checked by molecular dynamics calculations. Such calculations must use the correct collision-time statistics method.

The possibility that the problem is in the experimental data needs to be addressed also. In particular, the experimental data show a plateau at the density region 3.44–4.84 $\times10^{18}$ e/cm^3, where the width increases by just 2.5%. Moreover, this plateau is followed by a normal increase in

experimental widths at higher densities. The error bars for the line widths in this plateau are large enough as to cloud the very existence of the plateau, but would require an unlikely scenario to do that. Indeed, it is only if we take the sum of the observed width and the error bar for the upper density and their difference for the lowest, that the ratio of the widths is the same as the density ratios. We should note that the observation of this plateau is inconsistent with any kind of theoretical prediction or understanding of Stark broadening. If the problem lies in the experimental data, the most likely origin is reabsorption. This is hard to determine experimentally, especially in a pulsed discharge, and great care was taken in the experiment to address this issue. In the experiment a concave mirror was placed behind the plasma column on the optical axis of the detection system, so that the mirror's center of curvature would be at the center of the plasma column. Hence, for optically thin lines, one should observe a doubling of the observed line intensities. Experimentally, this factor is 1.86 due to mirror reflectivity and adjustment. The average ratio for the densities larger than 2×10^{18} e/cm^3 is 1.7, rather than 1.86. However, in Fig. 6 of Ref. [1], the uncertainties are such that this ratio is by no means certain. For example, the intensities with and without the mirror could be almost equal to within experimental error. In addition, this ratio is not nearly the same for all experimental data points. Thus, the ratio is 1.7 for the points corresponding to densities above 2×10^{18} e/cm^3, and 1.5 if we exclude the result at 4.84×10^{18} e/cm^3. In other words, while the results of the optical thickness check are consistent with an optically thin scenario, they do not rule out an optically thick scenario either. Hence, one might need to reconsider to what extent reabsorption increases the observed widths. We should note that the lower density results were obtained in the decompression phase, where the optical length is larger and, hence, is not true that reabsorption would be more pronounced for the higher densities.

V. CONCLUSIONS

The observed theory-experiment discrepancy is an important issue that needs to be resolved because understanding a system with a simple atomic structure like hydrogen at high densities is crucial, as spectral lines at high density are receiving increasing attention [15]. It is the *first* instance where simulation calculations do not agree with recent experimental data. Thus, to proceed with confidence, we need to clear up results that are a potential gray area. In fact, the experimental results discussed in this work are the only thorn in the subject of hydrogen line broadening, which is thought to be completely understood as long as we do not venture into the strongly coupled regime. Further efforts, both experimental and theoretical, are needed to settle the origin of the observed discrepancies along the lines suggested above.

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