Breakdown of the line-space concept in Stark broadening of spectral lines by plasmas

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A study is presented that proves that the standard model for calculation of line shapes, e.g., transitions from nl to $n'l'$ must be modified to include levels from higher principal quantum number manifolds, $n''>n'$. This becomes particularly important in the study of transitions without central components, exhibiting a central minimum, as the effect of the higher n'' states is to cause a filling in of the central minimum. An analytic derivation is provided and detailed calculations are performed to illustrate the effect.
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INTRODUCTION

In the study of the Stark broadening of spectral line shapes, the role of the low-lying resonance series members of hydrogen has been critically important since these transitions are easily observable and they possess simple enough atomic structures to allow detailed calculations [1,2]. Thus we find numerous detailed studies of the lines $L\alpha$, $L\beta$, $H\alpha$, $H\beta$. In these studies there have been many effects discussed, such as the electron broadening operators, the ion microfield effect, the quadratic Stark effect, and the quadrupole interaction [1,2], as well as the role of ion dynamics (see, e.g., [3,4] and references therein). In most studies the upper manifolds of these transitions, i.e., the subset of the atomic states that are included in the broadening calculations, are limited to include states of the same principal quantum number (PQN). Thus, for example, when one calculates the $L\beta$ transition the broadening of the upper state is usually limited to the interactions from the $n=3$ manifold. For arbitrary spectral series, this approximation is known as the concept of the line space (also known as the double-atom space), which is restricted to the direct product of the upper and lower manifolds.

In this paper we demonstrate that going beyond this approximation dramatically affects the *central* area of line profiles. This leads to the need for the revision of a variety of the experimental data, especially while searching for manifestations of ion dynamics in the line center. The present study shows that a basic assumption used in the standard line broadening breaks down and makes the central feature of the profiles very susceptible to the effect due to higher-lying states. Although the effect of higher-lying states has been incorporated in some codes [5–7], it was *counterintuitive* to believe that the effect would occur at the central part of the profiles. This paper proves that this does in fact occur by computational example (employing essentially the same code as in [5,7]) and then explains the counterintuitive results analytically.

In the following we first show a detailed numerical calculation of the $L\beta$ transition emitted from neutral hydrogen immersed in a plasma at a temperature of 3 eV and an electron density of 2×10^{16} cm^{-3}. This is a standard experimentally attainable plasma condition and thus provides an ideal test case. The detailed calculation is performed for two cases: one uses the standard selection criteria for the perturbing states, i.e., only the $n=3$ levels, while the other uses the $n=3$ levels *and* levels from higher PQN states. This shows the effects of the inclusion of the higher levels.

We next discuss the reason why the inclusion of the higher levels affects the central portion of the line profile. This discussion is carried out using a model of the upper manifold represented in a Stark basis set. The analytic model not only shows agreement with the calculations but also shows that there will be a temperature-dependent aspect to the effect.

Finally, we show that the effect of the higher PQN is important in previously observed spectra and note that the effects are significant even when the central region of the line is not a local minimum. To illustrate this we compare our theoretical results with the experimental results [5] on hydrogen Balmer series, and comment on the size of this effect in the detailed calculations of the $P\alpha$ transition ($n=3$ to 4) of He II measured and calculated over several order of magnitude.

CALCULATIONS OF THE $L\beta$ LINE

We illustrate that the standard assumption used in spectral line broadening, that the perturbations due to PQN beyond that in the transition are incorrect, lead to discrepancies in the line core. To do this we present calculations of the $L\beta$ transition of hydrogen. We select this transition as it is the simplest atomic structure that has a minimum in the central part of the line profile and allows ease of comparison with analytic estimates that are contained the next section. Further, by using a member of the Lyman series we are ensuring that the lower state broadening is obviated. In any other type of transition, for example $H\beta$, there would be complications due to lower state effects.

These calculations assume that the plasma is in local thermodynamic equilibrium, which is not critical to the conclusion but makes the calculations of the those ingredients of the line-shape formalism simpler. The line-shape computa-

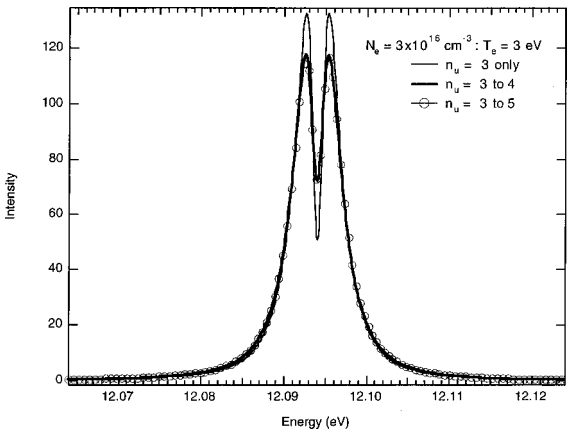

FIG. 1. Profiles of the $L\beta$ line of hydrogen calculated at $N_e = 3\times10^{16}$ cm^{-3} and $T=3$ eV. The upper manifold includes $n=3$ levels (thin line); $n=3$ and 4 levels (bold curve); $n=3$, 4, and 5 levels (line with the circles).

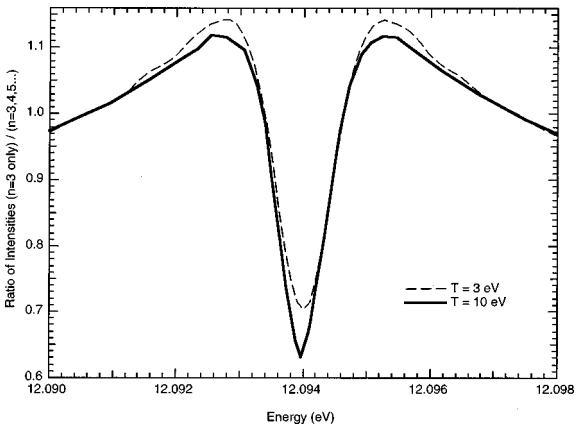

FIG. 2. Ratio of the $L\beta$ line profile calculated for the standard $n=3$ level only case to that calculated with the upper state manifold including all states from $n=3$ to $n=10$ for $N_e=3\times10^{16}$ cm^{-3}. Dashed line, $T=3$ eV; solid line, $T=10$ eV.

tion of the emission including a large number of PQN, as we are interested in here, requires an efficient method for the treatment of large level arrays interacting with a perturbing Stark field. A single radiative transition of a highly ionized emitter may, typically, involve at least 20 states in the lower and 50 states in the upper subspace. In addition, the probability distribution of the plasma ion microfield perturbation commonly requires 50 or more ion microfields, and this results in a basis that can involve more than 50 000 states. The model used here has been formulated to permit calculations of the spectra emitted by an arbitrarily complex atomic structure perturbed by very general plasma environments [3]. The computational approach to modeling the shape of the spectral lines emitted, used in the following, begins with a consideration of the time-dependent coupling of the emitting ion with the plasma environment. The first step in the procedure is to remove the time dependence in the plasma-emitter interaction. This is performed through two assumptions. First, the perturbing plasma ions are considered to be stationary and, second, the effect of the plasma electrons on the emitting ion is taken to be perturbative in nature due to their short collision durations with respect to the average lifetime of the emission process. In the standard theory of Stark broadening of spectral lines, this is customarily referred to as the quasistatic ion, impact-electron approximation. The result is a spectral line shape with separate inhomogeneous ion and homogeneous electron contributions. The line profile becomes a pure sum of independent electron impact broadened static Stark components.

In Fig. 1 we show the $L\beta$ transition of hydrogen emitted from a plasma with a temperature of 3 eV and a density of 3×10^{16} cm^{-3}. First, the thin solid line profile arises when the upper manifold is restricted to the $n=3$ levels of hydrogen, as in all other calculations to date. Second, the bold curve is when the upper manifold includes the $n=4$ states in addition to the $n=3$ states, and the line with the circles is calculated with the $n=3$, 4, and 5 states included. Note a large change in the profile near line center: the dip is reduced substantially by the inclusion of the higher n states. The intensity at line center is increased by 40% when the higher PQN are included. But one can easily observe that there is a small additional effect due to the addition of levels in the $n=5$ manifold. Calculations were carried out for the limiting case and it was found that the differences in the line profiles when PQN up to $n=10$ were included differed by $<3\%$ from those when the $n=3$ and 4 principal quantum numbers were included.

Further, we performed similar calculation, as above, but changing only the temperature to 10 eV. The idea here is to map the trend of the modification due to the change in temperature. In all respects the qualitative differences in the profiles due to the inclusion of the higher n states are the same in the 10 eV as can be seen in Fig. 1 for the 3 eV case. There are, however, quantitative differences and these are shown in Fig. 2. In Fig. 2 we show the ratio of the line profile for the standard $n=3$ level only case to the line profile calculated with the upper state manifold including PQN 3 to 10 states. This line profile ratio is then shown for two different plasma temperatures, 3 and 10 eV. It is seen that the inclusion of the higher n levels dramatically increases the intensity at the unperturbed wavelength: by 42% at $T=3$ eV and by 59% at $T=10$ eV. Further, one observes that the effect of the inclusion of the higher n levels is larger as the temperature increases.

ANALYTIC MODEL

The Hamiltonian of a hydrogen atom in a static field F is

$$H=H_0+zF. \tag{1}$$

In our analysis we use the basis of the parabolic wave functions (WF's) and the atomic units $\hbar=e=m_e=1$ (unless specified). In parabolic quantization the energy is given by $E_{n\alpha}=3nqF/2$, where $q\equiv n_1-n_2$ and $\alpha\equiv(q,m)$ with n_1,n_2,m being parabolic quantum numbers. We consider the Hamiltonian in two manifolds.

The first manifold $M_1(n)$ is a subspace corresponding to a particular PQN n ($n=n_a$ for the upper level or $n=n_b$ for the lower level). We note that the direct product $M_1(n_a)\otimes M_1(n_b)$ is the so-called ''line space'' used for calculation of line shapes (see, e.g., [1]). The Hamiltonian is diagonal in M_1.

The second manifold M_2 is an extended space including all levels with the PQN from $n=1$ to $n=n_{\max}$, where $n_{\max} > n_a$. The diagonalization of H in M_2 results in WF's $\Psi_{n\alpha}$ that are superpositions of the unperturbed parabolic WF's $\Psi^{(0)}_{n\alpha}$: $\Psi_{n\alpha}=\Sigma_{n'\alpha'} C^{n\alpha}_{n'\alpha'}\Psi^{(0)}_{n'\alpha'}$, where $|C^{n\alpha}_{n'\alpha'}| \ll 1$ for $(n'\alpha') \neq (n\alpha)$.

For the static Stark effect there is a perturbation series in terms of $n^4F \ll 1$, so that $|C^{n\alpha}_{n'\alpha'}| - n^4F \ll 1$, $(n'\alpha') \neq (n\alpha)$. Employing instead of F its average F_1 over the Holtsmark distribution

$$F_1 \equiv \langle F \rangle \approx 8.8N^{2/3}, \tag{2}$$

we obtain

$$|C^{n\alpha}_{n'\alpha'}| \sim n^4F_1 \approx 1.1\times 10^{-5} n^4 [N \ (\mathrm{cm}^{-3})/10^{16}]^{2/3}. \tag{3}$$

For $n=3$ and the ion density $N=3\times 10^{16}\ \mathrm{cm}^{-3}$ we get $|C^{n\alpha}_{n'\alpha'}| \sim 2\times 10^{-3}$, indicating that higher PQN would seem to be unimportant. Thus, although the relative difference between intensities f_1 and f_2 of some Stark component $n_a\alpha \leftrightarrow n_b\beta$ calculated in M_1 and M_2 seems to be of the order $\Delta f/f = 2|f_1-f_2|/(f_1+f_2) \sim |C^{n\alpha}_{n'\alpha'}|$, the simulation resulted in the $\Delta f/f$ on the order of 50%.

This apparent discrepancy can be resolved by three observations: (i) the higher PQN cause dipole forbidden central components to appear; (ii) the central component will be substantially narrower than the allowed lateral components; and (iii) the lateral components have small contributions at line center.

In more detail, first, the higher PQN mixing, which occurs for M_2 basis results in a central component of a nonzero intensity, which is controlled by the following matrix element: $|z^{110}_{000}|^2 \approx |C^{110}_{200}(z^{200}_{000})^{(0)} + C^{110}_{000}(z^{020}_{000})^{(0)}|^2 = |2C^{110}_{200}(z^{200}_{000})^{(0)}|^2 \approx 4K^4F^2|(z^{200}_{000})^{(0)}|^2$. Here K is the absolute value of the constant of the linear Stark effect averaged over all components of the radiative transition $n_a \leftrightarrow n_b$:

$$K = (3/2)\langle |n_a q_\alpha - n_b q_\beta| \rangle \approx (n_a^2 - n_b^2)/2. \tag{4}$$

This is true for any hydrogen line that would not have a central Stark component in the limit $F \to 0$, which constitutes 50% of all hydrogen lines.

Second, the profile $S_0(\Delta\omega)$ of the forbidden central component is significantly narrower than the profile $S_L(\Delta\omega)$ of the rest of the line (here the suffix L stands for ''lateral''). Indeed, for densities $N \ll 10^{19}\ \mathrm{cm}^{-3}$ and $n_a \leqslant 6$, the half-width at half maximum (HWHM) for the central component is controlled by the electron impact broadening while the HWHM for the rest of the line is controlled by the linear Stark broadening in the ion microfields, so that $\mathrm{HWHM}_0 \sim \gamma \ll \mathrm{HWHM}_L \sim KF_1$, where γ is the electron impact width averaged over all Stark components of the line and F_1 was defined by Eq. (2). Therefore the intensity at the maximum of the profile $S_0(\Delta\omega)$ is $S_0^{\max} \sim S_0(0) \sim \langle f_0 \rangle/\gamma$ while the intensity at the maximum of the profile $S_L(\Delta\omega)$ is $S_L^{\max} \sim 1/(KF_1)$. Consequently the ratio becomes $S_0^{\max}/S_L^{\max} \sim (KF_1/\gamma)\langle f_0 \rangle \gg \langle f_0 \rangle$.

Third, the maximum of $S_L(\Delta\omega)$ is displaced by $\delta\omega \sim KF_1$. So, the profile $S_0(\Delta\omega)$ reaches its maximum at $\Delta\omega \approx 0$, where $S_L(0) \ll S_L^{\max}$. Thus we need to compare the values of $S_0^{\max}$ and of $S_L(0)$.

To estimate $S_L(0)$ we approximate the profile $S_i(\Delta\omega)$ of each lateral component as a convolution of the Lorentzian and the Holtsmark distribution,

$$S_i(\Delta\omega) \approx f_i \pi^{-1} \int_0^\infty dF \ W(F)\gamma/[\gamma^2 + (\Delta\omega - K_iF)^2], \tag{5}$$

where K_i is the Stark constant of the individual component and f_i is the relative intensity of the individual component (relative to the intensity of any allowed component averaged over the same line, so that $\langle f_i \rangle = 1$). Employing the inequality $\gamma \ll KF_1$ we obtain $S_L(0) = \Sigma_{i=1}^{i_{\max}} S_i(0) \sim \gamma F_1^{-2} \Sigma_{i=1}^{i_{\max}} f_i K_i^{-2} \sim \gamma F_1^{-2}/K$, where we used the fact that $i_{\max} \sim K$. The electron impact width γ here can be approximated as $\gamma \sim \gamma_s[0.5 + \ln(\Omega_{\mathrm{We}}/\omega_{\mathrm{pe}})] \approx \gamma_s \ln(K^{-2}T^2/N)$, where $\Omega_{\mathrm{We}} \sim T/K$ is the electron Weisskopf frequency, ω_{pe} is the plasma electron frequency, and γ_s is the strong collision contribution: $\gamma_s \sim K^2N/T^{1/2}$.

To estimate $S_0^{\max}$ we set $K_i=0$ in Eq. (5) and multiply the integrand by the factor $f_0 \sim 4K^4F^2$:

$$S_0^{\max} - 4K^4 \int_0^\infty dF \ W(F)F^2/\gamma. \tag{6}$$

Physically, the integration should be truncated at the field strength F_m, where spectral lines merge, i.e., at $F_m \sim K^{-5/2}$. The primary contribution to the integral originates from the fields $F \sim F_m$. However, at these fields, the splitting between the adjacent Stark sublevels $\omega(F) \sim K^{1/2}F$ exceeds the plasma electron frequency ω_{pe} for densities $N_e < (16\pi n_a^8)^{-1}$ (the latter inequality translates into the usual units as $N_e < 2\times 10^{19}\ \mathrm{cm}^{-3}$ for $n_a=3$ and as $N_e < 2\times 10^{18}\ \mathrm{cm}^{-3}$ for $n_a=4$). Under the condition $\omega(F) \sim K^{1/2}F > \omega_{\mathrm{pe}}$ the electron impact width becomes field-dependent and can be estimated as $\gamma(F) \sim \gamma_s\{0.5 + \ln[\max(\Omega_{\mathrm{We}}/\omega(F),1)]\} \approx \gamma_s\{0.5 + \ln[\max(K^{-3/2}T/F,1)]\}$, where we keep the strong collision constant 0.5 since here it can be comparable to the second term. Using the fact of the relatively weak dependence of γ on F, the integral in Eq. (6) can be estimated as follows:

$$\begin{aligned} S_0^{\max} &\sim [4K^4/\gamma(F_m)] \int_0^{F_m} dF \ W(F)F^2 \\ &\sim [4K^4/\gamma(F_m)] F_1^{3/2} F_m^{1/2} \sim 4K^{11/4} F_1^{3/2} \gamma_s \\ &\quad \times \{0.75 + \ln[\max(KT,1)]\}. \end{aligned}$$

Thus the ratio $S_0^{\max}/S_L(0)$, characterizing the relative increase of the intensity at the very center of the line due to the inclusion of the higher n levels, becomes

$$\begin{aligned} \varepsilon \equiv S_0^{\max}/S_L(0) &\sim 8.1\times 10^3 N^{1/3} T/\{K^{1/4}[0.5 \\ &\quad + \ln(\max(KT,1))]\ln(K^{-2}T^2/N)\}, \end{aligned} \tag{7}$$

where K is given by Eq. (4).

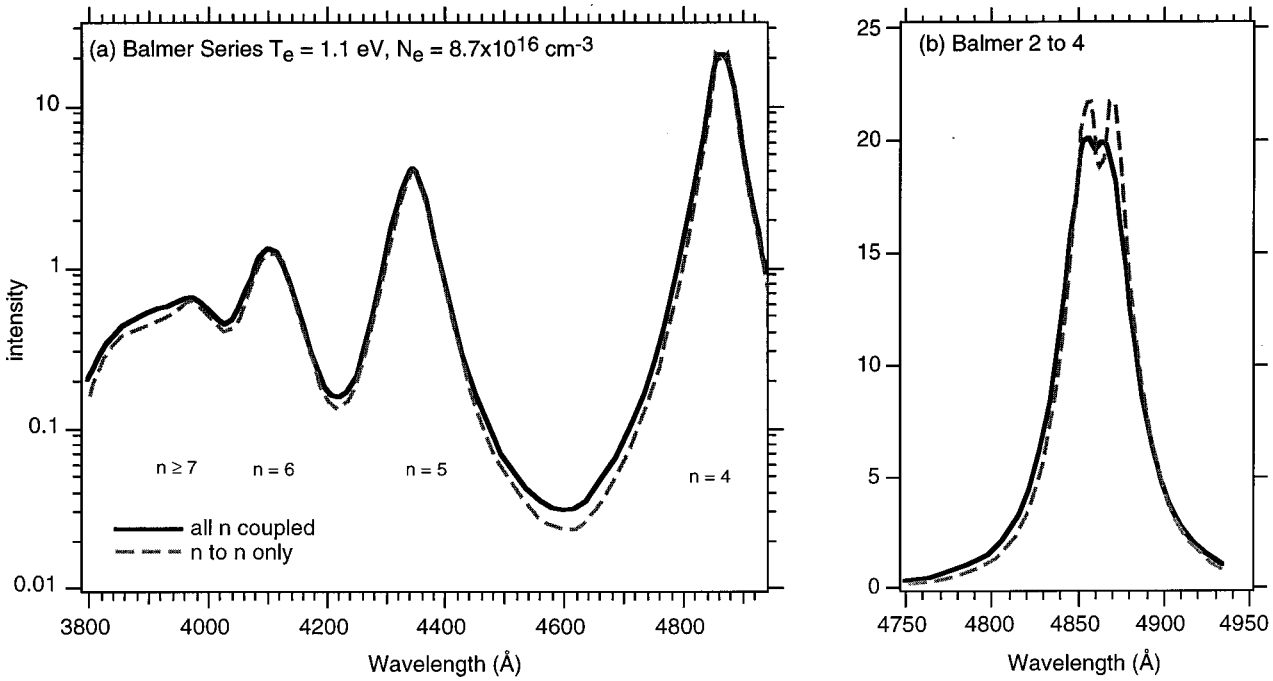

FIG. 3. Calculations for the Balmer series starting from the $H\beta$ line for the conditions of the experiment [8] ($N_e = 8.68\times10^{16}$ cm^{-3}, $T = 1.122$ eV). Dashed line, each upper state manifold is limited to its own principal quantum number; solid line, all principal quantum number states included in the upper manifold.

Now we use Eq. (7) for the comparison with our simulations for the $L\beta$ line ($n_a = 3, n_b = 1$) at $N = 3\times10^{16}$ cm^{-3}. For $T = 3$ eV, we obtain $\varepsilon \approx 0.17$ as compared to the simulation result $\varepsilon_{\rm sim} \approx 0.42$. For $T = 10$ eV, Eq. (7) yields $\varepsilon \approx 0.27$ as compared to the simulation result $\varepsilon_{\rm sim} \approx 0.59$. In terms of the temperature dependence of the effect, this means that as the temperature decreases from 10 to 3 eV, our model predicts a 36% decrease in the value of ε, which compares well with the corresponding 29% decrease following from the simulations. We next compare the predictions of the model with our simulation for the $H\beta$ line for different plasma conditions. All these comparisons demonstrate that our simple analytical model correctly reproduces the dependence of the effect on the controlling parameters such as the density N, the temperature T, and the average Stark constant K. As for the absolute values of ε, the analytical and simulation results agree broadly, as should be expected from a simplified model designed to illuminate the principal effect.

DISCUSSION AND CONCLUSIONS

The effect of including the higher n states to calculations of the broadening of spectral line shapes emitted from plasmas has been demonstrated to be important to the line core. That the higher n levels make a modification in the core of the profile is somewhat surprising. For example, one would expect that as the profile broadens, with increasing electron density, the Stark components that form the wings on the high-energy side of the profile will interact with Stark components from the low-energy wings of higher n state. Indeed, this does happen and was originally considered to be the dominant mechanism. However, the present study indicates that the mixing of the higher n states providing an allowed radiative path is clearly much more important. In fact, the effect is so dramatic that it clearly constitutes the breakdown of one of the most fundamental concepts in Stark broadening of spectral lines by plasmas—the concept of the line space (or double atom), which for almost 40 years seemed to be inviolable.

Moreover, it should be noted that there are experimental examples in the literature where the effects of the higher n level are important. We note the high-precision experiments [8] where a spectrum of the hydrogen Balmer series was measured with high spectral resolution and independent plasma diagnostics. The important point is that for the conditions of the experiment, temperature of 1.122 eV, and an electron density of 8.68×10^{16} cm^{-3} we find that the contributions to the $H\beta$ line due to the inclusion of higher n states are again substantial in the line core. In Fig. 3 we show the calculations for the Balmer series starting from the $H\beta$ transition for the conditions of the experiment [8]. The dashed line is the calculation performed for all the transitions independently. That is, each upper state manifold is limited to the same PQN. Next the bold curve is the spectrum calculated with the entire set of PQN included in the upper manifold.

There are two interesting aspects of the comparison of the spectra in Fig. 3(a). First, we note that the troughs in between the line centers are modified by the inclusion of the additional PQN in the broadening. This leads to an increase in the intensity in these wing areas. (We note that in the experiments the emissivity due to free-free and the H^- ion mask the trough areas.) Second, we can see that the effect of allowing all the states to broaden the transitions is to modify the line cores.

In Fig. 3(b) we show a detailed view of the Balmer β transition for the same calculations as shown in Fig. 3(a). Here we also see that the inclusion of higher levels increases the intensity at the very center of the line. However, the

relative value of the increase is smaller than in the above simulations for the $L\beta$ line: $\epsilon_{\rm sim}\approx 0.05$. The estimate using Eq. (7) yields $\epsilon\approx 0.12$. Thus our simple analytical model correctly predicts a smaller value of ϵ for the $H\beta$ line in the conditions of the experiment [8] as being primarily due to the lower temperature and the greater average Stark constant K [see Eq. (7)]. As for the absolute value of ϵ, our analytical and simulation results agree in this case again within the factor of 2.5.

In general we note that this effect, due to the inclusion of the higher PQN in the broadening of the upper level, will become only slightly less important as the PQN of the upper level of the transition n_a increases. Indeed, Eq. (7) shows that $\epsilon\propto K^{-1/4}$. In each spectral series, the most intense and practically important line without central components is the β line, for which $n_b=n_a-2$. It follows that $K\sim n_a$ for the β lines so that $\epsilon\propto n_a^{-1/4}$. Thus, Eq. (7) leads to the conclusion that the effect is primarily controlled by the temperature ($\epsilon\propto T$) while having much weaker dependence on the density and on the PQN of the upper level.

We note that the same effect we see for these transitions with line center minima (no allowed central components) will also be operative for lines with line center maxima (allowed central components). The size of the effect will be much less dramatic as the mixing of states that are allowed with other PQN [see Eq. (3)], must compete with the central component.

Finally, we comment on the fact that at least for the hydrogen line profiles the competition between the effects of ion dynamics and the higher PQN should be studied to determine the role of the two effects. This type of calculation is currently difficult due to limitation associated to size in the current generation of ion dynamics formulations. However, when these limitations are overcome it will be important to evaluate the comparative importance of the two effects.

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