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ON DENSE PLASMA LINE SHAPES

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INTRODUCTION

Computer simulations have been widely used in studying dense plasma properties¹ including the local field properties important in spectral line broadening calculations.² We will review here a more recent use of simulation, possibly less familiar to this audience, where the time dependent ionic microfield generated by computer simulation of a plasma is used directly as a time dependent external potential for the evolution of the electronic structure of an ion. This permits calculation of the dipole correlation function and thus line shapes with the inclusion of ion dynamic effects.

Some of the first line shape results calculated this way, by Stamm and co-workers,³ were questioned on: the usual concerns of numerical and statistical accuracy since this depends on how long an ion microfield time series is used for computing the dipole correlation function; the importance of omitted effects such as fine structure splitting, natural lifetimes, and Doppler broadening; the significance of the results to density diagnostics which depend on the line wings where dynamic effects are least important.⁴ Some of these questions have been settled. Subsequent calculations have established the statistical accuracy of the original work.⁵ The method has been extended to include effects such as natural lifetimes and fine structure splitting which dominates some lines.⁶ Questions on the importance of ion dynamics to specific line widths or radiation redistribution problems are still being decided. The correct inclusion of ion dynamic

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effects is, nonetheless, crucial in many instances and the method discussed here, although more time consuming than the usual line shape calculation, is the best available way of doing this. Results obtained this way are already being used to benchmark quicker, more analytic methods.⁷

We begin by repeating the formulae used in line shape calculations and from these give a discussion of when ion dynamic effects should be important. As an illustration, some now classic experimental results (although not for a dense plasma) are presented. We then use simulation results as a further illustration. Although done for realistic plasma conditions these are intended for demonstration rather than experimental comparison since in order to isolate ion dynamic effects complicating, but theoretically straightforward effects such as fine structure splitting and Doppler broadening are not included. Finally we discuss Stark-Doppler coupling.

LINE SHAPE FORMALISM

The starting point in most line shape studies is the expression for the power spectrum of dipolar radiation

$$P(\omega) = \frac{4\pi\omega^4}{3c^3} S(\omega) \quad (1)$$

where the line shape function

$$S(\omega) = \frac{Re}{\pi} \int_0^\infty e^{i\omega t} \langle e^{ik \cdot r(t)} \vec{d}(t) \cdot \vec{d}(0) \rangle dt \quad (2)$$

In the following $\vec{d}$ refers to the dipole operator for a single radiator as coherence effects are not considered. The $\langle \rangle$ denotes an ensemble average over plasma conditions and the Doppler term, $e^{ik \cdot r(t)}$, will be considered only in the last section when we discuss its correlation with the electronic dipole terms.

Although more general cases can be treated with somewhat more effort, for radiative transitions to the ground state the expression for the dipole correlation function may be simplified to

$$C(t) \equiv \langle \vec{d}(t) \cdot \vec{d}(0) \rangle = \sum_u \sum_{u'} \rho_u e^{i\omega_{ul}t} \vec{d}_{ul} \cdot \vec{d}_{lu} \langle u_{uu'}(t) \rangle \quad (3)$$

where u and u' denote upper states for the particular line and l the ground state and the $\vec{d}_{ul}$ etc. are dipole matrix elements. The time

development operator for the emitter $U(t)$ satisfies

$$i \frac{\partial U(t)}{\partial t} = [\vec{d} \cdot (\vec{E}_{ion}(t) + \vec{E}_{elec}(t))] U(t) \quad (4)$$

assuming only dipolar coupling between the emitter electronic states and the total microfield of the plasma due to both electrons and other ions. Presently available time dependent plasma simulations give no information about the electronic component of the microfield. The main approximation in these calculations is to treat this electronic microfield component separately using the impact approximation which leads to

$$i \frac{\partial U}{\partial t} = (\vec{d} \cdot \vec{E}_{ion}(t) + \phi) U(t). \quad (5)$$

For hydrogenic emitters evaluations of the electron collision operator ϕ are available.⁸

It remains to average the time development operator $U(t)$ by solving the above equation for a representative ensemble of ion microfields. This is then used in equations 2 and 3 to give the line shapes. For the results we show below these were produced by standard molecular dynamics computations using systems of a hundred or so particles and periodic boundary conditions. Other methods have been used for weakly coupled plasmas where the use of periodic boundary conditions would require very large systems.

QUALITATIVE DISCUSSION OF DYNAMIC EFFECTS

Perhaps the fullest discussion of the effects of fluctuations on line shapes appears in the NMR literature.⁹ Here we only want to indicate the time scales involved by considering a simplified model for equation 5

$$i \frac{\partial U(t)}{\partial t} = \vec{\mu} \cdot \vec{F}(t) U(t) \quad (6)$$

where μ is a scalar rather than an operator. The averaged solution to this equation

$$\langle U(t) \rangle = \left\langle e^{i \int_0^t \vec{\mu} \cdot \vec{F}(s) ds} \right\rangle \quad (7)$$

simplifies when $\vec{F}(s)$ is almost constant during the times t where $U(t)$ is non-negligible. The averaging then reduces to an average over initial field values. In this quasi-static limit

$$\langle U(t) \rangle_{Q.S.} = \int P(P) e^{-i\vec{\mu} \cdot \vec{P}t} dP \quad (8)$$

A rough upper limit for the time scale over which the field is constant is the field autocorrelation time. If the quasi-static $\langle U(t) \rangle$ is negligible for times larger than this then dynamic effects are not important. Applying this to a dense plasma we may use the inverse of the ion plasma frequency as an estimate for the field autocorrelation time. Using a Gaussian as an approximation to the distribution of the field along $\vec{u}$ equation 8 gives

$$\langle U(t = \omega_{ion}^{-1}) \rangle_{Q.S.} \approx e^{-\frac{\mu^2 \langle P^2 \rangle}{6^2 \hbar^2} \omega_{ion}^{-2}} \quad (9)$$

If the exponent in this equation is large then $\langle U(t) \rangle$ is small for times such that the ion microfield changes significantly and the line in question is not strongly affected by ion dynamics.

The dipole moment μ for a hydrogenic emitter increases as the square of the upper level principal quantum number (n) and decreases with the nuclear charge Z . $\langle P^2 \rangle$ may be estimated as $Z^2 \text{perturber}^4 / r_{ion}^4$ where r_{ion} is the ion sphere radius for the perturbing plasma. Combining these with the expression for the plasma frequency shows that this exponent (R) scales as

$$R \sim \frac{n^4 M_{ion}}{Z^2} \rho_{ion}^{1/3} \quad (10)$$

Ion dynamic effects are thus reduced for the higher members of the Lyman series, because the dipole matrix elements increase. They also drop with ion density since the increase in the field strength magnitude more than compensates for the increasing plasma frequency. Although there is no explicit temperature dependence in this expression R will decrease with temperature since the ionization state Z increases. Not surprisingly, ion dynamic effects are predicted to be largest for the Lyman- α or β lines of highly ionized emitters in low mass perturbers.

The above formula provides only a qualitative estimate. Before turning to simulation results we show an early experimental indication of the importance of ion dynamic effects. This experiment is not strictly relevant to the above discussion, since the time dependence of the field is produced by the motion of the emitter rather than by the

motion of the perturbers as we assumed above for a dense plasma, but the results are still instructive.

EXPERIMENTAL OBSERVATION OF ION DYNAMIC EFFECTS

One of the earliest observations of ion dynamic effects were for the Lyman spectra of hydrogen or deuterium impurities in an arc plasma of singly ionized argon.^{10,11} The measured line spectra (crosses in fig. 1) differed considerably from theoretical predictions which assumed static ions (solid line in fig. 1). For example the Lyman- α width was over twice theoretical estimates and the central dip in Lyman- β markedly

Fig. 1. The top graph shows the experimental (xxx) and theoretically predicted (---), for static ions, L_α , L_β , L_γ lines shapes for hydrogen in singly ionized Ar at $T = 15,500$ °K and $n_e = 2 \times 10^{17} \text{ cm}^{-3}$. The lower graph shows the decreased emitter motion effect when deuterium (•) is substituted for hydrogen (-). This figure is reproduced from references 10 and 11.

reduced. It was convincingly shown that these differences were due to ion motion when deuterium was substituted for hydrogen. As mentioned these effects are due to the fluctuating field caused by emitter motion and relate more to the Stark-Doppler coupling discussed in the last section than to the effects of perturber motion discussed above. These effects could have been anticipated. The measured half-width for the Lyman- α line shown in fig. 1 is $\sim 2 \times 10^{12} \text{ sec}^{-1}$ corresponding to times for the dipole correlation function of $\sim .5 \times 10^{-12} \text{ sec}^{-1}$. From the plasma conditions of a temperature of 15,000 °K and an ion density of $9.3 \times 10^{17} \text{ cm}^{-3}$ the hydrogen emitter at mean thermal velocity traverses an ion sphere radius in $\sim 10^{-12} \text{ sec}$ so the ion field can not be treated as constant for the relevant dipole correlation times. Recent calculations,¹² some of them using computer simulation, now agree well with the spectra in fig. 1.

COMPARISON OF STATIC AND DYNAMIC LYMAN SPECTRA

The central object to be obtained from simulation for use in spectral studies is the time history of the local field (other things such as field gradients could also be obtained for studying quadrupolar effects) and the emitter trajectory, for use in Doppler broadening. Figure 2 shows a short section of the microfield time history for a moderately coupled plasma of hydrogenic argon ions. The three field components and the magnitude are displayed as functions of $\omega_{\text{ion}} t$ along with the distributions and field correlation obtained from much longer time histories. A noticeable feature here is the strong collision near the start of the time series, $\omega_{\text{ion}} t \sim .6$. An ensemble of such time histories is then used in equation 5 to compute the averaged time evolution matrix.

In terms of the dipole correlation function the main effect of ion microfield fluctuations is to reduce correlations for times beyond an inverse ion plasma frequency. As a simple example, for a Lyman- α line the correlation, in the quasi-static approximation, at a given field strength (also assuming the electron broadening is the same for all components) is

$$C(F, t) = \frac{d_z}{3} \left\{ 2 + \cos \left(\frac{D_z t F}{\hbar} \right) \right\}$$

where $D_z = (200 | d_z | 210)$. If this is averaged with an assumed Gaussian distribution for the field component distribution it becomes

Fig. 2. Typical ion microfield time series from a simulated Ar^{+17} plasma at $T = 10^7$ °K and $n_e = 1.5 \times 10^{23} \text{ m}^{-3}$. All field values are in units of Z/r_{ion}^2 . The distributions for a field component, $P(F_x)$, and the field magnitude, $P(F)$, are shown to the side and the field autocorrelation function below. The time scales are $\omega_i t$.

$$C(t) = \frac{e\phi t}{3} \left[2 + [1 - (\omega_s^2 t)^2] e^{-\frac{\omega_s^2 t^2}{2}} \right]$$

where $\omega_s \equiv D_e \sqrt{\langle F^2 \rangle / 3}$. If the electron broadening is weak the effect of ion dynamics on the unshifted component can be dramatic as shown in fig. 3 where this simplified quasi-static result is compared to a typical case including ion dynamics. The effect for Lyman- β , fig. 3b, is similar but smaller.

Fig. 3. Schematic dipole correlation functions for L_a and L_g . The dashed lines are for static ions. The solid lines are typical of the reduced correlation time due to ion motion.

Fig. 4. Comparison of static (---) and dynamic (—) dipole L_a dipole correlation functions and line shapes for 1% Ar⁺¹⁷ in H⁺¹ and 100% Ar⁺¹⁷ at $T = 10^7$ °K and $n_e = 1.5 \times 10^{23} \text{ cm}^{-3}$.

Turning now to some numerical results, fig. 4 shows the correlation functions and line shapes for a plasma of hydrogenic argon in hydrogen ion perturbers and for pure hydrogenic argon at the same temperatures (10^7 °K) and electronic density ($1.5 \times 10^{23} \text{ cm}^{-3}$). The difference between the static approximation (dashed line) and the results including dynamics (solid line) is similar to the qualitative comparison in fig. 3. The difference is larger for the hydrogen ion perturbers since the ion microfield due to these light mass perturber fluctuates more rapidly. The same trend is shown in fig. 5 for the Lyman- β line shapes. We caution again that the omitted fine structure splitting is important for these lines. (This is included in ref. 6.)

The effect of temperature, perturber mass, electron density and other variations on dynamic effects has also been studied.⁵

Fig. 5. Comparison of static (---) and dynamic (—) L_{β} line shapes at $T = 10^7$ °K and $n_e = 1.5 \times 10^{23} \text{ cm}^{-3}$.

STARK DOPPLER COUPLING

So far we have only considered the dipole correlation $\langle \vec{d}(t) \cdot \vec{d}(0) \rangle$ rather than $\langle e^{ik \cdot r(t)} \vec{d}(t) \cdot \vec{d}(0) \rangle$ which includes Doppler effect. It is usually assumed that the Doppler term is independent of the dipole correlation and the final line shape comes from convoluting the two together. For this the free particle limit of

$$\langle e^{ik \cdot r(t)} \rangle \rightarrow \begin{cases} e^{-k^2 t^2 / 2\beta m} & \text{free particle limit} \\ e^{-3k^2 Dt} & \text{diffusion limit} \end{cases} \quad (11)$$

is used. The diffusion limit is valid only for times $t_{\text{ion}} \sim 10$ and is probably never relevant for dense plasma spectral line broadening.

Figure 6 shows these two limits for an Al^{+12} plasma at $T = 2.7 \times 10^6$ °K and electron density of $4 \times 10^{21} \text{ cm}^{-3}$ so the coupling parameter $\Gamma \sim 1$. The time limit beyond which the Lyman- α dipole correlation function is negligible, t_a , is seen to be in the free particle (dashed line) regime.

This does not mean that Stark-Doppler coupling is negligible. The experiments discussed in section 4 being a dramatic counterexample! For times sufficiently small that the initial velocity is unchanged we can write

$$\langle e^{ik \cdot r(t)} \vec{d}(t) \cdot \vec{d}(0) \rangle = \int e^{ik \cdot v_0 t} \langle \vec{d}(t) \cdot \vec{d}(0) | v_0 \rangle P(v_0) dv_0 \quad (12)$$

where a dipole correlation function conditional on the (presumed constant) initial velocity of the emitter has been introduced which is integrated over the (Maxwellian) distribution of initial velocities. Intuitively, faster moving emitters will see more rapidly fluctuating fields and thus have a more rapidly decaying dipole correlation. Figure 7 shows this effect for the same plasma as in fig. 6. For this case the more rapidly moving ions (dashed line) do show a faster decay of the dipole correlation than the slower moving ones (solid line) but the effect is slight since most of the field variation is due to the motion of the perturbing ions as can be seen by comparing with the static ion case (dot dashed line). For the arc plasma case discussed in section 4 most of the field variation is due to the motion of the light emitter in the heavy perturbers and a plot similar to fig. 7 would show much larger variation presumably.

Fig. 6. Mean squared displacement of Al+12 ions (o) at $T = 2.7 \times 10^6$ °K and $n_e = 4 \times 10^{21} \text{ cm}^{-3}$. The dashed line is for free particles and the solid line is the diffusion limit. The time t_a indicates where the L_a dipole correlation function for this system become negligible.

Fig. 7. The L_a dipole correlation function for slow (—) and fast (---) Al+12 emitters, (same conditions as fig. 6), compared to the static ion approximation (-o-).

We hope this review has given some indication of the types of plasma spectra questions which can be addressed with this method. Most of the specific results used as illustrations here are from research done in collaboration with Roland Stamm and Carlos Iglesias.

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