

## Recent Advances in Stark Line Broadening Calculations and their Applications to Precise Spectroscopy of Pulsed Plasmas

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A new method for the calculation of the spectral line broadening in plasma has been developed and implemented. The main idea of the method is to numerically simulate the motion of the plasma particles, both ions and electrons, and use the resulting time-dependent field to evaluate the emitter oscillating function, the Fourier transform of which provides the spectral line shape. We applied this method to the analysis of dipole-forbidden line shapes used for the determination of plasma properties.

### Introduction

Measurements of light emission and absorption spectra of atoms and ions is one of the most important tools in plasma diagnostics. The data derived from the spectral measurements, however, can be very complicated and, usually, there is no straightforward way to infer the plasma parameters. Thus, plasma spectra have to be simulated and compared to the measured line profiles in order to determine the plasma parameters. In the course of a few decades, several numerical methods for lineshape calculations have been suggested and successfully applied for various cases. However, the existing methods are unsatisfactory in some aspects. The separation of the particle perturbations into the ion and electron parts (e.g. [1, 2]) can not be done, in general, without a loss of accuracy. Other methods restrict themselves to the limiting cases of either isolated lines or the degenerate case of hydrogen- or helium-like configurations (e.g. [3]). Further, addition of any anisotropic perturbation (like plasma waves or external magnetic fields) is in general either impossible or results in cumbersome calculations.

### Method

The shape of a spectral line is calculated in three main steps. First, the perturbing fields are simulated by the Particle Field Generator (PFG). PFG produces time-dependent series of the electric field  $\vec{F}_p(t)$  induced on emitters by the rest of charged plasma particles, where the index  $p$  corresponds to the  $p$ -th emitter. It is done by a computer simulation of the motion of the Coulomb-interacting plasma particles, both emitters and perturbers. The fields can also include external (e.g., due to the plasma oscillations) electric or magnetic fields. Then, using this field as a perturbation, the emitter oscillating function is calculated. Finally, evaluating the power spectrum of the emitter oscillating function gives the spectral line shape.

The Hamiltonian of the atomic system is a sum of the unperturbed Hamiltonian  $H_0$  plus a time-dependent perturbation  $V$ :

$$H = H_0 + V(t). \quad (1)$$

In general, the perturbation is due to the plasma electric field (simulated by the PFG) and/or external electric and magnetic fields. We find the time-development operator  $U(t)$  (acting in the same operator space as  $H$ ) by solving the equation

$$idU(t)/dt = [H_0, U(t)] + V(t)U(t) \quad (2)$$

or, in the interaction representation,

$$id\bar{U}(t)/dt = V(t)\bar{U}(t). \quad (3)$$

The time evolution of the dipole operator  $D(t)$  is then obtained using the time-development operator:

$$D(t) = U(t)^\dagger D(0)U(t) \quad (4)$$

The Fourier transform of the dipole operator

$$\vec{D}(w) = \int_0^\infty dt \exp(-i\omega t) \vec{D}(t) \quad (5)$$

is further used to calculate the line spectrum. In the dipole approximation, the intensity of the emitted light polarized along  $\vec{e}_\lambda$  is given by the following expression:

$$I^\lambda(w) = \frac{1}{2\pi} \sum_{i,f} \omega_{fi}^4 |\vec{e}_\lambda \cdot \langle \vec{D}_{fi}(\omega - \omega_{fi}) \rangle|^2, \quad (6)$$

where  $i$  and  $f$  run over the initial and final levels, respectively,  $\omega_{fi} = E_i - E_f$  is the level energy difference, and the angle brackets denote an averaging over several runs of the code (which corresponds to the averaging over a set of emitters).

## Application

Measurements of forbidden lines is an efficient tool for detecting electric fields in plasma, since the forbidden line amplitude strongly depends on the electric field strength. However, the line shapes of forbidden transitions are more difficult to calculate in the framework of the traditional methods. While the usually dominating effect of the pressure broadening, the impact width, is the same for the allowed and forbidden transitions, the quasistatic contribution to the line shape strongly depends on the type of transition.

The analysis presented herein addresses part of the activities directed towards the development of high-resolution active-beam spectroscopy [4]. The probing beam is produced by applying a laser pulse to a lithium sample, producing an expanding, partially ionized lithium beam. In order to diagnose the lithium beam conditions, the laser induced fluorescence (LIF) technique is applied. A 15-ns pulse of a tunable laser is used for optical pumping of the 4p level, which, due to collisions with the plasma electrons, lead to populating the 4d and 4f

levels. The strong 2p–4d (dipole-allowed) and weak 2p–4f (dipole-forbidden) transitions are observed.

Due to the low density of the beam, the line shape measurements should be very precise and the analysis should accurately take into account all other effects that may contribute to the total line shape. The instrumental response function of the spectroscopic diagnostic system was evaluated by measuring line spectra of the Ne and Hg calibration lamps. The Doppler broadening was derived from the Li I 2p–3d transition spectrum by deconvolving the instrumental function from the total line shape (the 2p–3d transition is significantly less sensitive to the Stark broadening than 2p–4d, and its width, for plasma densities below  $10^{14} \text{ cm}^{-3}$ , is only affected by the Doppler broadening). The Doppler distribution thus obtained corresponded to a  $\sim 2 \text{ eV}$  Maxwellian. The broadening, however, is not thermal, since the plasma is essentially collisionless. Rather, it represents a range of transverse velocities in the plasma plume. The longitudinal velocity distribution of Li I was obtained using time-of-flight measurements with laser tagging (as in [5]), giving a narrow  $(7 \pm 1) \times 10^5 \text{ cm/sec}$  distribution. The mean value is thus consistent with the transverse velocity distribution. Assuming that Li II ions have a similar velocity distribution, we conclude that in the reference frame of the moving neutrals, the plasma ions are very slow, with an effective "temperature" of less than 0.1 eV.

To analyze the 2p–4d experimental data (see Fig. 1), we calculated the Stark-broadened spectrum and then consequently convoluted it with the instrumental and the Doppler broadening functions. The ion and electron temperatures were chosen to be 0.05 and 0.4 eV, respectively.  $T_i$  was so chosen based on the velocity distribution analysis discussed above. The choice of the electron temperature was obtained from independent collisional-radiative calculations [6] showing that hotter electrons would quickly ionize the 4d and 4f levels, resulting in an absolute intensity of the 2p–4d transition that is lower by an order of magnitude than the measured intensity (we note that the dependence of the line shape on the electron temperature is rather weak). The LS coupling was neglected, since the fine splitting of the 2p–4d and 2p–4f lines is significantly smaller than the sum of the instrumental and the Doppler broadenings. We varied the electron density in order to achieve a good fit to the experimental data. However, it was found that no plasma conditions can explain the experimental data. Low densities can not produce strong enough forbidden component, whereas high densities broaden the lines too much, resulting in filling up the dip between the allowed and forbidden components. Varying either  $T_e$  or  $T_i$  or both of them was unsuccessful, too. Thus, it appears evident that a low-frequency field should be present. Since in our case  $T_i \ll T_e$ , the ion-acoustic plasma waves were an acceptable candidate. Ion-acoustic waves in denser laser-produced plasmas have been observed earlier (e.g. [7]). Therefore, we added a harmonic perturbation of the form  $A_o \sin(\omega_{pi} t)$  (an exact spectrum of the waves is perhaps unimportant since  $\omega_{pi}$  is well below the line width).

In spite of the addition of another parameter in the fitting, a rather high accuracy determination of the plasma parameters was possible. It was found that  $n_e = (5 \pm 1) \times 10^{13} \text{ cm}^{-3}$  and  $A_o = (3 \pm 0.5) \text{ kV/cm}$  provide a good fit to the measured spectrum. Higher densities, as

was mentioned above, broaden the lines too much, whereas lower values require  $A_o$  well above typical plasma field amplitudes.

## Summary

A novel method for the calculation of the spectral line broadening in plasma has been developed and implemented. Based on the method, a lineshape analysis of dipole-forbidden transitions was presented and proved to be a sensitive tool for plasma-parameter investigations. It was demonstrated that reliable conclusions on the plasma parameters can be drawn even in the low-density ( $n_e < 10^{14} \text{ cm}^{-3}$ ) regime. For the first time, a line-shape diagnostics with a sub-microsecond time resolution was successfully applied to such low-density plasma. The presence of low-frequency waves in the laser-produced dilute plasma was inferred. The high-accuracy results were achieved in spite of the line spectrum being significantly influenced by the instrumental and Doppler broadenings.

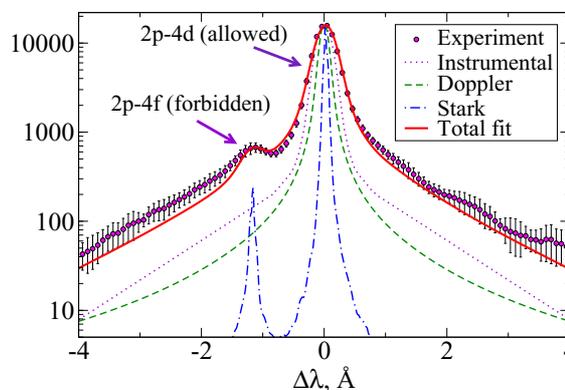


Figure 1: The measured and the calculated Li I 2p-4d and 2p-4f line shapes.  $T_e = 0.4 \text{ eV}$ ,  $T_i = 0.05 \text{ eV}$ ,  $n_e = 5 \times 10^{13} \text{ cm}^{-3}$ , and  $A_o = 3 \text{ kV/cm}$ . Shown are the different contributions to the total line shape.

## Acknowledgments

This work was supported in part by the German-Israeli Foundation, the Israeli Science Foundation Grant No. 6858, and Sandia National Laboratories (USA).

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