Lineshape calculations

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Introduction

- Spectroscopy is a unique tool for nonintrusive plasma diagnostics.
- Light $\rightarrow$ discrete spectrum (energy/wavelength and intensity) $\rightarrow$ line width and shift $\rightarrow$ line shape
- plasma $\rightarrow$ electron temperature $\rightarrow$ density or ion temperature/velocity $\rightarrow$ + electromagnetic fields
- Line broadening also affects the radiation transfer and, hence, the level populations in non-optically-thin plasmas.

What is lineshape? Probability to emit (or absorb) photon of a given energy/frequency/... Usually, normalized to unity:

$$\int L(\omega)\,\mathrm{d}\omega = 1$$

One talks about emission or absorption lineshapes, respectively. Often (but not always!) they are the same.

Entities and units

Frequently used entities and units.

- frequency ν [Hz]
- angular frequency $\omega = 2\pi\nu$ [rad/s]
- wavenumber $\sigma \equiv \bar{\nu} = \nu/c$ $[\mathrm{cm}^{-1}]$, $[\mathrm{R}_\infty]$
  $(1\,\mathrm{R}_\infty \approx$ 109,737 $\mathrm{cm}^{-1})$
- energy $E = \hbar\omega$ [eV], [Ry]
  $(1\,\mathrm{Ry} \approx 13.606\,\mathrm{eV})$
- wavelength $\lambda = 1/\sigma$ [Å], [nm]
  (1 nm = 10 Å)

Units: good and bad

$X\,\mathrm{eV} \approx 8065.5\,X\,\mathrm{cm}^{-1} \approx 2.4180 \times 10^{14}\,X\,\mathrm{Hz} \approx$
$1.5193 \times 10^{15}\,X\,\mathrm{rad/s} \approx 12{,}398\,X^{-1}\,$Å

Line-broadening effects are usually much smaller than the unperturbed values, $\delta X \ll X$. Then:

$\delta X\,\mathrm{eV} \approx 8065.5\,\delta X\,\mathrm{cm}^{-1} \approx 2.4180 \times 10^{14}\,\delta X\,\mathrm{Hz} \approx$
$1.5193 \times 10^{15}\,\delta X\,\mathrm{rad/s} \approx 12{,}398\,\delta X/X^{2}\,$Å

Expressions involving λ's are unnecessarily complex and ugly.

Atomic physics is all about energy, not wavelength! Hamiltonian, Lagrangian—all have units of energy.

Below, I will often use word “energy” for ω ($\hbar \equiv 1$; atomic units).

Atomic physics phenomena

- Motion of radiator (Doppler effect)
- Magnetic field (Zeeman effect)
- Electric field (Stark effect)

Doppler effect

"Static" Doppler shift:

$$\omega_0 \to \omega = \omega_0 + \frac{v_{\shortparallel}}{c}\omega_0\,; \;\; \omega - \omega_0 = \frac{v_{\shortparallel}}{c}\omega_0\,.$$

$v_{\shortparallel}$ — projection of the radiator velocity toward the observer. For brevity, I omit the $\shortparallel$ subscript below.

In a plasma, the particles move with different velocities. Hence,

$$L(\omega)\,\mathrm{d}\omega = P(v_\omega)\,\mathrm{d}v$$

$P(v_\omega)$ — probability to find radiator moving at such a v_ω that the Doppler-shifted photon energy is ω.

$$L(\omega) = P(v_\omega)\left[\left.\frac{\mathrm{d}\omega}{\mathrm{d}v}\right|_{v=v_\omega}\right]^{-1}$$

Thermal Doppler broadening

Thermal motion → Maxwellian distribution of v:

$$P_v(v)\,\mathrm{d}v = \sqrt{\frac{M}{2\pi kT}}\exp\left(-\frac{Mv^2}{2kT}\right)\mathrm{d}v\,,$$

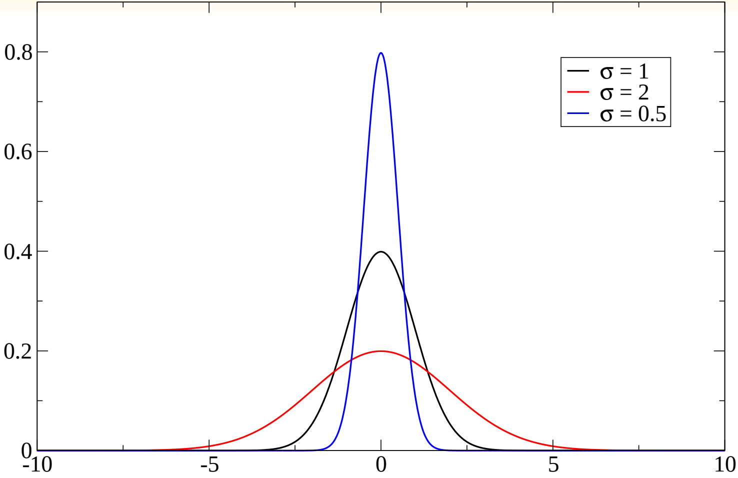
$$L(\omega) = \frac{1}{\sqrt{2\pi}\sigma_D}\exp\left[-\frac{(\omega-\omega_0)^2}{2\sigma_D^2}\right]\,.$$

This is a Gaussian with the standard deviation $\sigma_D = \sqrt{\frac{kT}{Mc^2}}\omega_0$.

$$\mathrm{FWHM} = \sqrt{8\ln 2}\,\sigma_D \approx 7.72\times 10^{-5}\omega_0\sqrt{\frac{T(\mathrm{eV})}{M(\mathrm{a.m.u.})}}$$

$$\mathrm{Peak}\times\mathrm{FWHM} = 2\sqrt{\frac{\ln 2}{\pi}} \approx 0.94$$

Gaussian profiles



Quasistatic/statistical broadening

Average over an ensemble of radiators (atoms/molecules) is called “statistical” or “quasistatic” (QS) broadening.

Thermal Doppler broadening is an example of such a broadening. Recall:

$$L(\omega) = P(v_\omega) \left[\left. \frac{\mathrm{d}\omega}{\mathrm{d}v} \right|_{v=v_\omega} \right]^{-1} .$$

More generally,

$$L(\omega) = P(\alpha_\omega) \left[\left. \frac{\mathrm{d}\omega}{\mathrm{d}\alpha} \right|_{\alpha=\alpha_\omega} \right]^{-1} .$$

Here, α is a property of the radiator or its surrounding causing energy shift (for example, electric field due to neighbour plasma particles); when $\alpha = \alpha_\omega$ the radiator emits/absorbs photons at ω instead of ω_0.

Microfield distributions

Neglecting interactions between particles, the probability to find normalized electric field $\beta \equiv F/F_0$ is given by [Holtsmark, 1919]

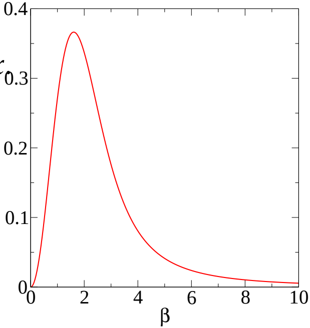
$$H(\beta) = \frac{2}{\pi}\beta \int_0^\infty x \sin(\beta x) \exp(-x^{3/2}) \mathrm{d}x,$$

$$F_0 = 2\pi \left(\frac{4}{15}\right)^{2/3} Z_p e N_p^{2/3}$$

is the Holtsmark normal field.

$\beta \ll 1 \Rightarrow H(\beta) \propto \beta^2$

$\beta \gg 1 \Rightarrow H(\beta) \propto \beta^{-5/2}$



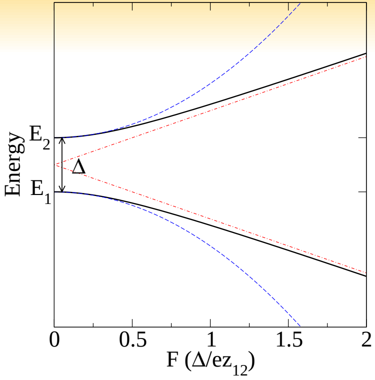
Note 1: similar distribution for gravitating masses [Chandrasekhar and von Neumann, 1942].

Note 2: plasma coupling alters the distribution.

Linear & quadratic Stark effect

Consider a two-level system.

Perturbation due to the electric field $\vec{F} = (0, 0, F)$:



$$V = -\vec{d}\vec{F} = ezF = \begin{pmatrix} 0 & ez_{12}F \\ ez_{12}F & 0 \end{pmatrix}$$

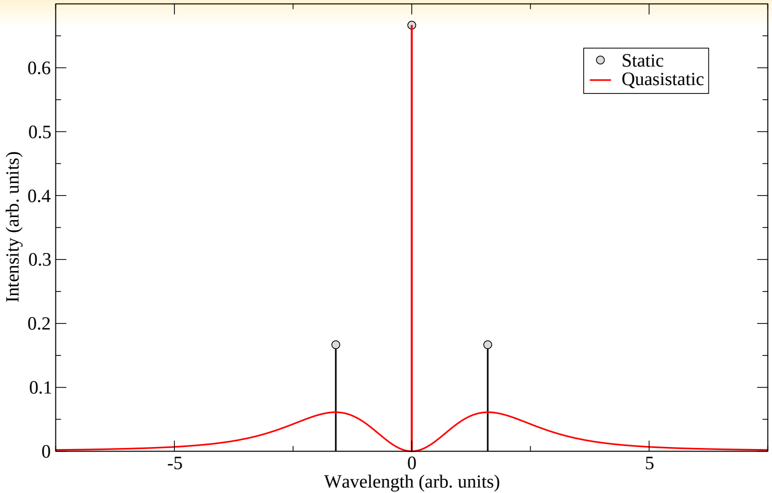
$$H = H_0 + V = \begin{pmatrix} E_1^0 & ez_{12}F \\ ez_{12}F & E_2^0 \end{pmatrix}$$

$$|H - E| = 0 \Rightarrow E_{1,2} = (E_1^0 + E_2^0)/2 \pm \sqrt{(\Delta/2)^2 + (ez_{12}F)^2}$$

$$ez_{12}F \ll \Delta \Rightarrow E_{1,2} \approx E_{1,2}^0 \pm (ez_{12}F)^2/\Delta \Rightarrow \text{quadratic effect}$$

$$ez_{12}F \gg \Delta \Rightarrow E_{1,2} \approx \pm\, ez_{12}F \Rightarrow \text{linear effect}$$

Stark effect of Ly-α



Time-dependent perturbation

Power spectrum of a physical quantity evolving with time $f(t)$ is given via its Fourier transform:

$$I(\omega) \propto |\mathcal{F}(f)|^2 = \left| \int_{-\infty}^{+\infty} f(t) e^{-i\omega t} \, \mathrm{d}t \right|^2$$

Dipole radiation:
Electric field $\vec{F}$ in an EM wave is proportional to the dipole moment $\vec{d}$ of radiator:

$$\vec{F}(t) \propto \vec{d}(t)$$

(For classical and quantal harmonic oscillators alike; for the latter, $\vec{d} \to \vec{d}_{if}$, for transition between states i and f.)

$$I(\omega) \propto \left|\vec{d}_\omega\right|^2 \text{, with } \vec{d}_\omega \equiv \int_{-\infty}^{+\infty} \vec{d}(t)\, e^{-i\omega t} \, \mathrm{d}t$$

Example: Natural broadening

Radiative decay $|i\rangle \to |f\rangle$, characteristic time = A_{if} (Einstein coefficient). Decay rate is $\Gamma \equiv A$.

$$|\Psi_i(t)|^2 \propto e^{-\Gamma t};\ \Psi_i(t) \propto e^{i\omega_i t - \Gamma t/2},\ t \geq 0\,.$$

If $|f\rangle$ is stable (GS), $\omega_i = w_0$, $\langle i|\, d\, |f\rangle \propto \Psi_i(t) \propto e^{i\omega_0 t - \Gamma t/2}\eta(t)$ ($\eta(t)$ - Heaviside unit step function). Then

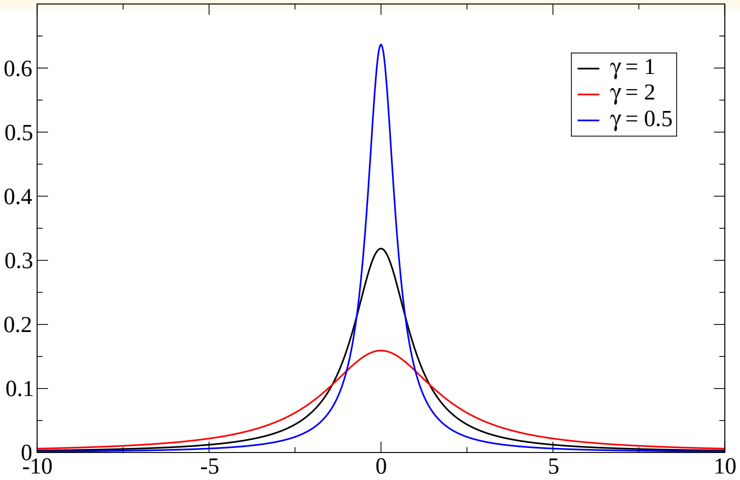
$$d_\omega \propto \mathcal{F}\left[e^{i\omega_0 t - \Gamma t/2}\eta(t)\right] = \frac{1}{i(\omega - \omega_0) + \Gamma/2}\,,$$

$$I(\omega) \propto |d_\omega|^2 \propto \frac{1}{(\omega - \omega_0)^2 + (\Gamma/2)^2}\,.$$

Area-normalized lineshape is a Lorentzian:

$$L(\omega) = \frac{1}{\pi}\frac{\Gamma/2}{(\omega - \omega_0)^2 + (\Gamma/2)^2}\,.$$

Lorentzian profiles



Voigt profiles

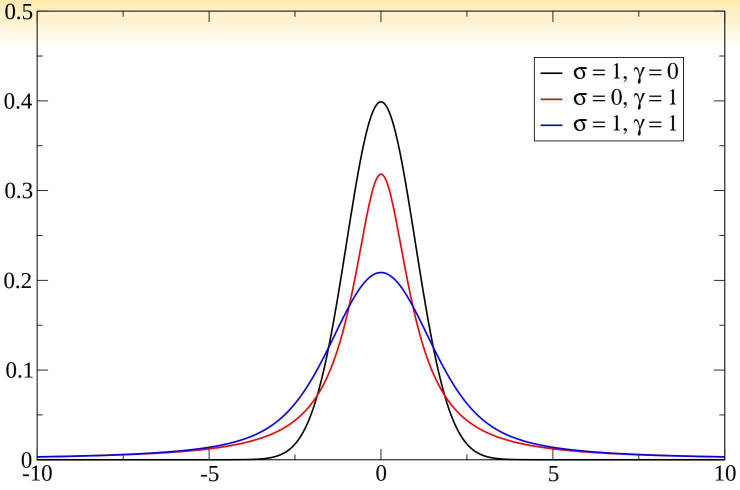
Convolution of a Lorentzian and a Gaussian:

$$\mathcal{V}(x;\gamma,\sigma) \equiv \mathcal{L}(x;\gamma) * \mathcal{G}(x;\sigma) = \int_{-\infty}^{\infty} \mathcal{L}(x';\gamma)\mathcal{G}(x-x';\sigma)\,\mathrm{d}x'$$

With a $\sim 1\%$ accuracy,

$$\delta\omega_{\mathcal{V}} \approx \left[\left(\frac{\delta\omega_{\mathcal{L}}}{2}\right)^2 + \delta\omega_{\mathcal{G}}^2\right]^{1/2} + \frac{\delta\omega_{\mathcal{L}}}{2}$$

Voigt profiles



The wings of Voigt are determined by the Lorentzian contribution.

Formal theory of line broadening I

$$I(\omega) \propto \left|\vec{d}_\omega\right|^2, \text{ with } \vec{d}_\omega \equiv \int_{-\infty}^{+\infty} \vec{d}(t)\, e^{-i\omega t}\, \mathrm{d}t$$

Using the cross-correlation theorem $\mathcal{F}(f)\mathcal{F}(g) = (2\pi)^{-1}\mathcal{F}(f * g)$:

$$\left|\vec{d}_\omega\right|^2 = \frac{1}{2\pi}\int_{-\infty}^{+\infty} C(t)\, e^{-i\omega t}\, \mathrm{d}t\,,$$

where $C(t)$ is the dipole auto-correlation function:

$$C(t) \equiv \int_{-\infty}^{+\infty} \vec{d}^*(\tau)\cdot\vec{d}(\tau + t)\ \mathrm{d}\tau\,,$$

or (using $C(t) = C(-t)$)

$$I(\omega) \propto \frac{1}{\pi} Re \int_{0}^{+\infty} C(t)\, e^{-i\omega t}\, \mathrm{d}t$$

Formal theory of line broadening II

Now we need to average over an ensemble of radiators:

$$I(\omega) \propto \frac{1}{\pi} Re \left\{ \int_0^{+\infty} C(t)\, e^{-i\omega t}\, \mathrm{d}t \right\}_{\mathrm{av}} = \frac{1}{\pi} Re \int_0^{+\infty} \{C(t)\}_{\mathrm{av}}\, e^{-i\omega t}\, \mathrm{d}t$$

However, $C(t)$ is already “averaged” by time, recall

$$C(t) \equiv \int_{-\infty}^{+\infty} \vec{d}^*(\tau) \cdot \vec{d}(\tau + t)\ \mathrm{d}\tau \simeq \sum_\tau \vec{d}^*(\tau) \cdot \vec{d}(\tau + t)$$

From the ergodicity argument (time average = ensemble average), it suffices to keep only one term in the infinite sum, e.g. at $\tau = 0$:

$$\{C(t)\}_{\mathrm{av}} \simeq \left\{ \vec{d}^*(0) \cdot \vec{d}(t) \right\}_{\mathrm{av}}$$

$$I(\omega) \propto \frac{1}{\pi} Re \int_0^{+\infty} \left\{ \vec{d}^*(0) \cdot \vec{d}(t) \right\}_{\mathrm{av}} e^{-i\omega t}\, \mathrm{d}t$$

Formal theory of line broadening III

Generalization for multiple components of transition(s) and level populations (via density matrix ρ):

$$C(t) = Tr\left[\left\{\vec{D}^{\dagger}(0) \cdot \vec{D}(t)\rho\right\}_{\rm av}\right]$$

Note 1: one often sees in the literature

$$C(t) = Tr\left[\vec{D}^{\dagger}(0) \cdot \vec{D}(t)\rho\right]$$

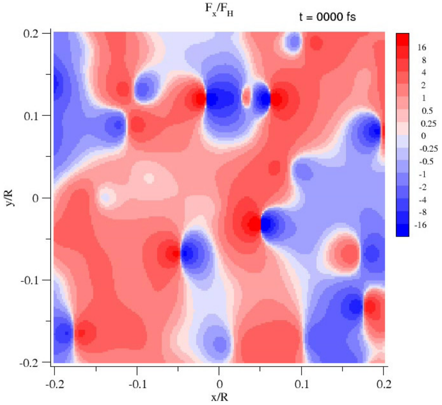
This expression, strictly speaking, is only valid in context of further averaging over an ergodic ensemble!

Note 2: $C(t)$ is sometimes called auto-correlation function of the light amplitude.

Formal theory of line broadening IV

$$C(t) = Tr\left[\left\{\vec{D}^{\dagger}(0)\cdot\vec{D}(t)\rho\right\}_{\rm av}\right]$$

It looks easy, but $\vec{D}(t)$, $\{\}_{\rm av}$, and even ρ are, in principle, results of complex N-body plasma dynamics where there is no strict separation between the radiators and the "bath".



Broadening of isolated lines

According to [Baranger, 1958], a $u \to \ell$ transition assumes Lorentzian shape with FWHM defined by

$$w = N_e \int_0^\infty vF(v)\mathrm{d}v \left(\sum_{u' \neq u} \sigma_{uu'}(v) + \sum_{\ell' \neq \ell} \sigma_{\ell\ell'}(v) + |f_u(v) - f_\ell(v)|^2 \right),$$

where $F(v)$ is the (Maxwellian) electron velocity distribution, $\sigma_{ik}(v)$ is impact cross section from i to k, and $f_k(v)$ is elastic scattering amplitude.

$$w \equiv w_{\mathrm{in}} + w_{\mathrm{el}}$$

The inelastic part:

$$w_{\mathrm{in}} \equiv N_e \int_0^\infty vF(v)\mathrm{d}v \left(\sum_{u' \neq u} \sigma_{uu'}(v) + \sum_{\ell' \neq \ell} \sigma_{\ell\ell'}(v) \right)$$

or

$$w_{\mathrm{in}} = \sum_{u' \neq u} \langle \sigma_{uu'} N_e v \rangle_v + \sum_{\ell' \neq \ell} \langle \sigma_{\ell\ell'} N_e v \rangle_v$$

Broadening of isolated lines (cont.)

The derivation assumes the broadenings of Stark-coupled levels do not overlap; hence the name "isolated" lines.

Typically (except for near-threshold energies), $w_{\mathrm{el}} < (\mathrm{or} \ll)\, w_{\mathrm{in}}$.

$$w \approx w_{\mathrm{in}} = \sum_{u' \neq u} \langle \sigma_{uu'} N_e v \rangle_v + \sum_{\ell' \neq \ell} \langle \sigma_{\ell\ell'} N_e v \rangle_v$$

Compare to the natural broadening:

$$w = \sum_{\text{relevant phenomena}} (\text{time of life of } |\mathrm{u}\rangle)^{-1} + (\text{time of life of } |\ell\rangle)^{-1}$$

Generalizing to other impact mechanisms (ionization, recombination of any kind),

$$w \approx \sum_{\text{all mechanisms}} \text{depop. rate of } |\mathrm{u}\rangle + \sum_{\text{all mechanisms}} \text{depop. rate of } |\ell\rangle$$

"Standard theory" of line broadening

In the "standard theory" (ST), the ions are usually given a [quasi]static role, while electrons are dynamic (impact):

$$I(\omega) = \frac{1}{\pi} \operatorname{Re} Tr \int_0^\infty dF_i W(F_i) \{ d^\dagger [i\omega - iH_s(F_i) + \phi_e(F_i)]^{-1} d \}_{av}$$

Alternatively, if applicable, ions may be treated in the impact approximation, like electrons ($\phi_e \to \phi_e + \phi_i$):

$$I(\omega) = \frac{1}{\pi} \operatorname{Re} Tr \{ \Delta_d [i\omega - iH_0 + \phi_e + \phi_i]^{-1} \}_{av}$$

Intermediate cases?

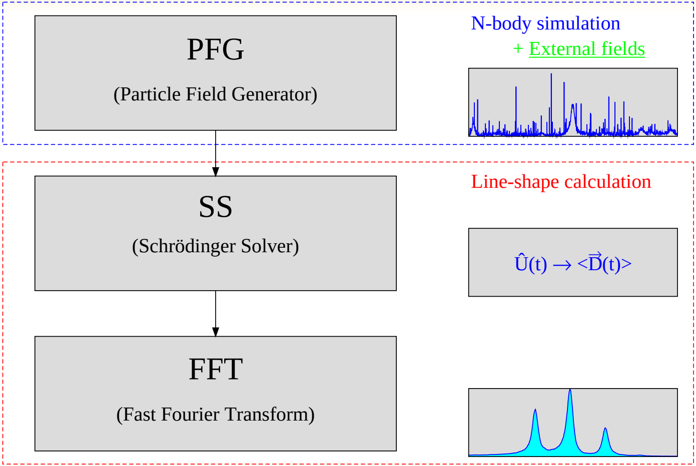
Computer simulations

The closest to *ab initio* calculations; since [Stamm and Voslamber, 1979].

The shape of a spectral line is calculated in three steps:

- The perturbing fields are simulated using the Particle Field Generator (PFG), by calculating the motion of a finite number of interacting electrons and ions (of a few types).
- Using this field as a perturbation, the emitter oscillating function is calculated by the Schrödinger Solver (SS).
- The power spectrum of the emitter oscillating function is evaluated using the Fast Fourier Transformation (FFT) method, giving the spectral line profile.

Computer simulations (cont.)



Computer simulations (cont.)

The Hamiltonian of the atomic system:

$$H = H_\circ + V(t).$$

The perturbation $V(t)$ is due to the plasma electric field (simulated by the PFG) and external electric and magnetic fields. We solve the Schrödinger equation

$$id\Psi(t)/dt = H\Psi(t)$$

using the time-development operator U in the interaction representation:

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

Computer simulations (cont.)

The evolution of the dipole operator $D(t)$ is then obtained:

$$\vec{D}(t) = U(t)^{\dagger}\vec{D}(0)U(t).$$

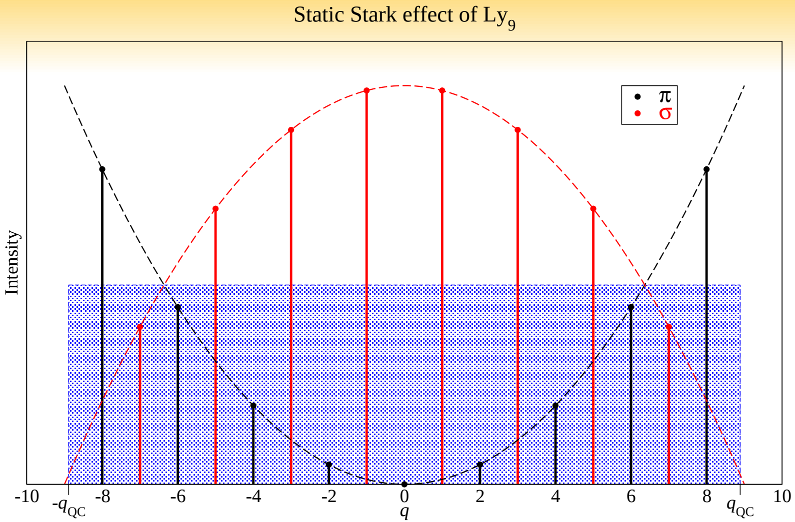
The Fourier transform of the dipole operator $\vec{D}(\omega)$ is further used to calculate the line spectrum:

$$I^{\lambda}(\omega) \propto \sum_{i}\sum_{f}\omega_{fi}^{4}|\vec{e}_{\lambda}\cdot\langle\vec{D}_{fi}(\omega)\rangle|^{2}.$$

The angle brackets denote an averaging over several runs of the code (which corresponds to the averaging over an ensemble of emitters).

Computer simulations are accurate, but very time consuming.

Quasi-contiguous (QC) approximation



Intensities of the π and σ components form two parabolae, which, on average, can be substituted with a simple rectangular shape.

Quasi-contiguous (QC) approximation

Therefore:

$$I_n(\omega) = \begin{cases} \frac{I_n^{(0)}}{2\alpha_n F/\hbar} & \text{for } |\hbar\omega| \leq \alpha_n F \\ 0 & \text{for } |\hbar\omega| > \alpha_n F, \end{cases}$$

where $I_n^{(0)}$ is the total line intensity, and α_n is the linear-Stark-effect coefficient:

$$\alpha_n = \frac{3}{2}(n^2 - 1)\frac{ea_0}{Z}.$$

Generalization for $n' > 1$:

$$\alpha_{nn'} = \frac{3}{2}(n^2 - n'^2)\frac{ea_0}{Z}.$$

QC approximation: quasistatic shape

Convolution with a microfield distribution $W(F)$:

$$I(\omega) = I_{nn'}^{(0)} \int_{\hbar\omega/\alpha_n}^{\infty} \frac{W(F)dF}{2\alpha_{nn'}F/\hbar} \equiv I_{nn'}^{(0)} L_{\rm qs}(\omega),$$

or, with the reduced field strength $\beta = F/F_0$ and detuning $\bar{\omega} = \omega/\Delta_0$,

$$L_{\rm qs}(\bar{\omega}) = \frac{1}{2} \int_{\beta}^{\infty} \frac{H(\beta)}{\beta} {\rm d}\beta,$$

where

$$H(\beta) = W(F)/F_0$$

and

$$\Delta_0 = \frac{\alpha_{nn'}F_0}{\hbar}.$$

QC approximation: quasistatic shape

In ideal plasma:

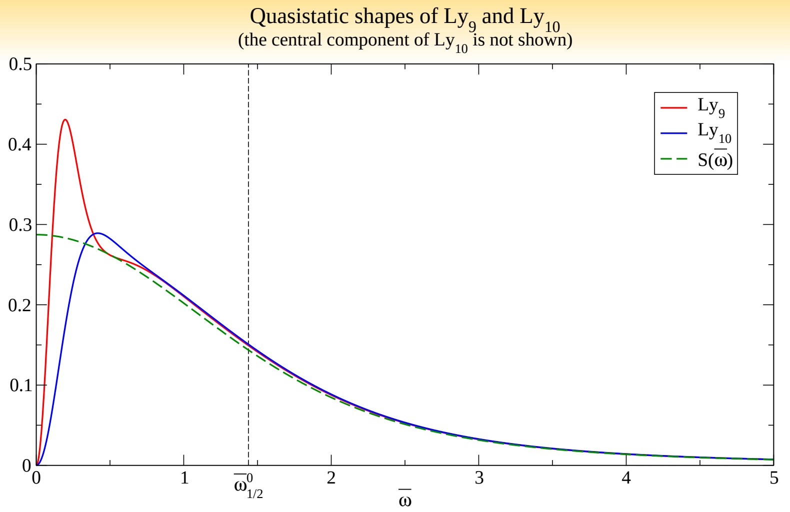
$$L_{\mathrm{qs}}(\bar{\omega}) = S(\bar{\omega}),$$

where the S function is defined as

$$S(\bar{\omega}) = \frac{1}{\pi} \int_0^\infty \cos(\bar{\omega} x) \exp(-x^{3/2}) \mathrm{d}x.$$

[Stambulchik and Maron, 2008]; also corrections due to moderate plasma coupling.

QC approximation: quasistatic shape



(only “blue” half of the symmetric profiles are shown).

QC-FFM I

Frequency-fluctuation model (FFM) [Calisti et al., 2010] accounts for field fluctuations with the typical field frequency

$$w_{\mathrm{dyn}} = \frac{\langle v \rangle}{\langle r \rangle} = \sqrt{\frac{kT}{m_p^*}} \left(\frac{4\pi N_p}{3} \right)^{1/3} .$$

Instead of the quasistatic lineshape $L_{\mathrm{qs}}(\bar{\omega})$, the dynamic one is

$$L(\bar{\nu};\bar{\omega}) = \frac{1}{\pi} \mathrm{Re} \frac{J(\bar{\nu};\bar{\omega})}{1 - \bar{\nu} J(\bar{\nu};\bar{\omega})} ,$$

where

$$J(\bar{\nu};\bar{\omega}) = \int \frac{L_{\mathrm{qs}}(\bar{\omega}')\mathrm{d}\bar{\omega}'}{\bar{\nu} + i(\bar{\omega} - \bar{\omega}')},$$

$$\bar{\nu} \sim w_{\mathrm{dyn}} / \Delta_0.$$

QC-FFM II

Applying FFM to quasi-contiguous lineshape, for ideal one-component plasma one gets:

$$J(\bar{v};\bar{\omega}) = \int_0^\infty \mathrm{d}\tau \exp\left(-\tau^{3/2} - i(\bar{\omega} - i\bar{v})\tau\right) .$$

[Stambulchik and Maron, 2013]; straightforward extension for multi-component non-ideal plasmas.

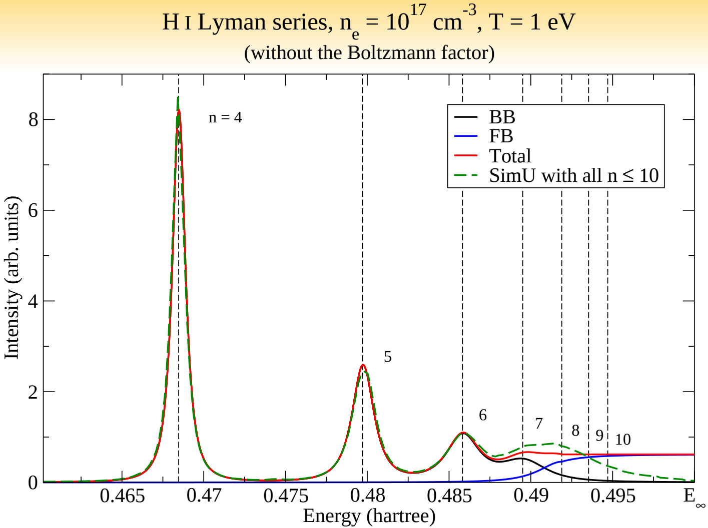
QC-FFM :: High-n series & continuum lowering

The approach (closely following [Griem, 1997]):

- Calculate bound-bound (BB) $n_\ell \to n_u$ shapes for a series of n_u until FWHM exceeds $|E_{n_u} - E_{n_u+1}|$ (the “Inglis-Teller” reasoning).
- (Optional) continue for a few more n_u with the same width.
- Assume the free-bound (FB) edge at the E_{n_u+1} energy.
- Convolve the FB continuum with the last ($n_\ell \to n_u$) BB lineshape.
- Sum up.

No ionization potential depression assumed.

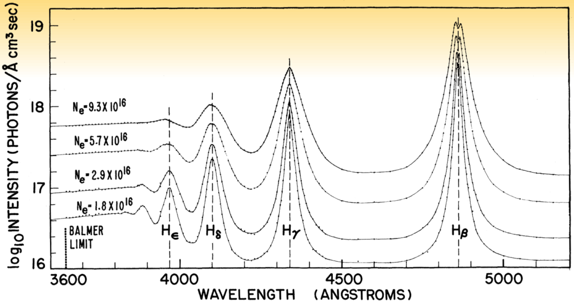
QC-FFM :: High-n series & continuum lowering (cont.)



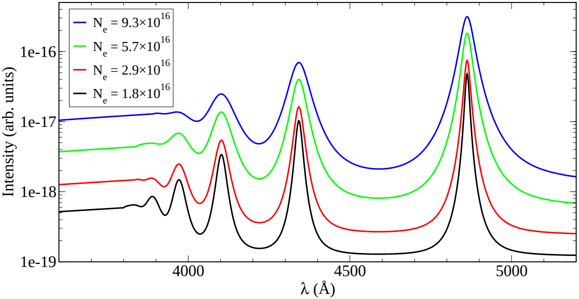
SimU: Hamiltonian with 385 fully interacting states.

SimU CPU time: > 1 month. QC-FFM: $\sim 1\,\mathrm{sec}$ ($\sim \times 3,000,000$).

QC-FFM :: High-n series & continuum lowering example



[Wiese et al., 1972]



$\sim 1\,\mathrm{s}$ CPU time

Data sources and tools

- Stark-B database (isolated lines): `http://stark-b.obspm.fr/`
- [Griem, 1974] appendices (isolated lines)
- Plasma Formulary Interactive (H-like and Rydberg): `http://plasma-gate.weizmann.ac.il/pf`
- NIST Atomic Spectral Line Broadening Bibliographic Database `http://physics.nist.gov/cgi-bin/ASBib1/LineBroadBib.cgi`

Notes on lineshape accuracy

Lineshape calculations are complex; reliably assessing theoretical uncertantities can be more difficult than the calculations themselves. Claimed accuracy should be taken with a grain of salt. Some rules of thumb:

- Accuracy is not mentioned at all — assume factor two or worse;
- Claimed 20%–40% — must be discussed and explained, at least shortly;
- Claimed 10%–20% — there must be a section devoted to accuracy estimates;
- Claimed 3%–10% — accuracy is the subject of the study;

When using published data, pay attention what is meant by the "width"—it could be FWHM or HWHM!

Books and reviews

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