Linewidth broadening from short laser pulses

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https://bartmcguyer.com/notes/note-3-ShortPulseBroadening.pdf

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TL;DR: Probing an atomic or molecular transition with laser light for a short duration broadens the measured linewidth of the transition.

This note calculates how a spectroscopic lineshape broadens for a transition probed by a gated laser pulse. The approach is a little different than that in common textbooks.[1,2] This phenomenon is very important in linewidth measurements because too short of a pulse artificially broadens a transition under study. It's related to the transit-time broadening of a fast atomic beam crossing a perpendicular laser beam with finite area.[1,2] Short pulses are often used in experiments with laser-cooled atoms and molecules.

I. SPECTRUM OF A LASER PULSE

Suppose we have a laser with a carrier frequency f_{c} such that the electric field $\mathbf{E}$ at a given point in the laser beam is

$$\mathbf{E}(t) = \mathbf{E}(0)\sin(2\pi f_{\mathrm{c}} t). \tag{1}$$

What is the spectrum, or distribution of frequencies f, of this laser? To answer this, first note that the Fourier transform of some function of time, say $S(t)$ where t has units of seconds, is given by

$$\widehat{S}(f) = \mathcal{F}[S(t)] = \int_{-\infty}^{\infty} e^{-i2\pi f t} S(t) dt, \tag{2}$$

where f is a temporal frequency in Hz. If we assume our laser is continuous and is on for all time (!), then this Fourier transform computes a spectrum describing how an army of other ideal, always-on continuous lasers (with complex frequencies) could reproduce its pulse. The result is then

$$S(t) = \sin(2\pi f_{\mathrm{c}} t) \quad \longrightarrow \quad \widehat{S}(f) = \frac{1}{2i}\left[\delta(f - f_{\mathrm{c}}) - \delta(f + f_{\mathrm{c}})\right], \tag{3}$$

where $\delta(x)$ is a Dirac delta function. The spectrum contains only f_{c} and its negative, since $S(t)$ is real. This result follows from using $\sin(2\pi f) = (e^{i2\pi f} - e^{-i2\pi f})/(2i)$ and noting that $\mathcal{F}[e^{i2\pi f_{\mathrm{c}} t}] = \delta(f - f_{\mathrm{c}})$.

Now, what happens if the laser is pulsed so that it lasts only a finite duration T in time? Let's introduce a new function,

$$\mathrm{rect}(x) = \begin{cases} 1 & |x| < 1/2 \\ 1/2 & |x| = 1/2 \\ 0 & |x| > 1/2 \end{cases}, \tag{4}$$

so we can describe the time-dependence of the laser's electric field as

$$S(t) = \mathrm{rect}(x/T)\sin(2\pi f_c t). \tag{5}$$

To tackle this, note that the Fourier transform has the useful property that $\mathcal{F}[e^{i2\pi gt}H(t)] = \widehat{H}(f-g)$, and that $\mathcal{F}[\mathrm{rect}(at)] = \mathrm{sinc}(f/a)/|a|$, where

$$\mathrm{sinc}(x) = \frac{\sin(\pi x)}{\pi x}. \tag{6}$$

(Note that $\mathrm{sinc}(x)$ has two common definitions, but this definition is correct here.) Together, these give the result:

$$\widehat{S}(f) = \frac{|T|}{2i}\left\{\mathrm{sinc}[T(f-f_c)] - \mathrm{sinc}[T(f+f_c)]\right\}. \tag{7}$$

Therefore the laser pulse broadens the peaks at f_c and $-f_c$ because of the finite duration T. Note that $\lim_{T\longrightarrow\infty} T\,\mathrm{sinc}(xT) = \delta(x)$, so we recover the previous result in the limit of an infinite pulse.

II. LINEWIDTH BROADENING

Now suppose we use our pulsed laser from above to probe a transition. What lineshape and linewidth should we expect?

There are different ways to approach this problem. Let's use a simple picture with Einstein coefficients following Hilborn.[3] Here, the induced absorption rate

$$W^i_{12} = N_1 B^\omega_{12}\int g(\omega)\rho(\omega)d\omega, \tag{8}$$

where N_1 is the number of molecules in the ground state, B^ω_{12} is an Einstein B coefficient of induced absorption, $g(\omega)$ is a normalized lineshape function satisfying $\int g(\omega)d\omega = 1$, and $\rho(\omega)$ is the energy density per angular frequency at $\omega = 2\pi f$. For our laser, the energy density is proportional to the square of the spectrum,

$$\rho(\omega) \propto |\widehat{E}(\omega)|^2 \propto |\widehat{S}(\omega/2\pi)|^2, \tag{9}$$

and is normalized such that $\int \rho(\omega)d\omega = I/c$, where I is a unidirectional irradiance (or "intensity") and c is the speed of light. The square of the spectrum is

$$|\widehat{S}(f)|^2 = \frac{T^2}{4}\left\{\mathrm{sinc}[T(f-f_c)]^2 + \mathrm{sinc}[T(f+f_c)]^2 + 2\,\mathrm{sinc}[T(f+f_c)]\mathrm{sinc}[T(f-f_c)]\right\}. \tag{10}$$

To simplify things a bit, let's assume that the lineshape function $g(\omega)$ is narrow enough that we only need to care about carrier frequencies $f_c = \omega_c/(2\pi)$ very close to the molecular transition, taken to be centered at $f_0 = \omega_0/(2\pi)$. Although molecules don't care about the sign of the frequency, let's also ignore negative frequencies from now on. With these changes, we can approximate

$$\rho(\omega) \approx \left(\frac{I}{\pi c}\right) \mathrm{sinc}[T(\omega - \omega_{\mathrm{c}})/(2\pi)]^2 \tag{11}$$

where we normalized $\rho(\omega)$ using the property $\int_{-\infty}^{\infty} \mathrm{sinc}(x)^2 dx = \pi$. This functional form agrees with Eq. (3.244) on p. 178 of Ref. 1.

Then our absorption lineshape will be of the approximate form

$$L(f_{\mathrm{c}}) \approx \int_{-\infty}^{\infty} g(\omega)\, \mathrm{sinc}[T(\omega - \omega_{\mathrm{c}})/(2\pi)]^2 d\omega, \tag{12}$$

where we'll ignore normalization here and below. This follows because $L(f_{\mathrm{c}}) \propto W_{12}^{i}$.

A. Zero natural width

For sufficiently small natural width (or sufficiently large broadening), we can approximate

$$g(\omega) \approx \delta(\omega - \omega_0), \tag{13}$$

which gives a lineshape that is just

$$L(f_{\mathrm{c}}) \approx \mathrm{sinc}[T(f_0 - f_{\mathrm{c}})]^2. \tag{14}$$

Plotting $L(f_{\mathrm{c}})$ vs f_{c}, we would see a bump centered around f_0. Numerically, the full width at half maximum (or FWHM) of this function is

$$\mathrm{FWHM} \approx \frac{0.8859}{T}. \tag{15}$$

For reference, a 1 ms pulse gives a FWHM of 886 Hz. Note that this lineshape looks roughly like a Gaussian, but with weak sidelobes.

For comparison, most texts use an uncertainty relation to estimate this broadening, with very different order-unity coefficients: Ref. 1 (p. 176) gives a width of about $1/(2\pi T)$; Ref. 2 (p. 154) gives a width of about $1/T$.

B. Lorentzian natural width

Next, suppose that the lineshape is a Lorentzian,

$$g(\omega) = \frac{\Gamma'/(2\pi)}{(\Gamma'/2)^2 + (\omega - \omega_0)^2}, \tag{16}$$

with an angular FWHM of $\Gamma' = 2\pi\Gamma$.

One way to proceed is to notice that, numerically, a Gaussian is a very decent approximation for $\text{sinc}^2(c)$ within its FWHM range,

$$\text{sinc}[T(f - f\text{c})]^2 \approx e^{-3.98T^2(f-f_c)^2} \propto \frac{1}{\sigma\sqrt{2\pi}}e^{-(f-f_c)^2/(2\sigma^2)} \tag{17}$$

for a Gaussian standard deviation $\sigma = 1/(\sqrt{2 * 3.89}\,T) \approx 0.3585/T$. (These numbers were determined by fitting a Gaussian function to points comprising a sinc^2 function.)

With this approximation,

$$L(f_c) \approx \int_{-\infty}^{\infty} \left(\frac{\Gamma/(2\pi)}{(\Gamma/2)^2 + (f - f_0)^2}\right)\left(\frac{1}{\sigma\sqrt{2\pi}}e^{-(f-f_c)^2/(2\sigma^2)}\right) df \tag{18}$$

$$\propto V(f_0 - f_c; \sigma, \Gamma/2), \tag{19}$$

which is a Voigt profile.[4] The FWHM of a Voigt profile (in Hz) is approximately

$$\Gamma_V \approx 0.5346\Gamma + \sqrt{0.2166\,\Gamma^2 + (2\sigma\sqrt{2\ln 2})^2}, \tag{20}$$

which for our case simplifies to

$$\Gamma_V \approx 0.5346\Gamma + \sqrt{0.2166\,\Gamma^2 + 0.713/T^2}. \tag{21}$$

The limiting cases of this are

$$\lim_{\Gamma \gg 1/T} \Gamma_V \approx \Gamma \tag{22}$$

$$\lim_{\Gamma \ll 1/T} \Gamma_V \approx \frac{0.844}{T}, \tag{23}$$

which agrees pretty closely with the results of the last section.

REFERENCES

[1] D. Budker, D. F. Kimball, D. P. DeMille. *Atomic Physics: An Exploration through Problems and Solutions*, Oxford, 2nd. ed. (2008).

[2] C. J. Foot. *Atomic Physics*, Oxford, (2005).

[3] R. C. Hilborn, "Einstein coefficients, cross sections, f values, dipole moments, and all that", *American Journal of Physics* **50**, 982–986 (1982). Updated arXiv version (2002): `http://arxiv.org/ftp/physics/papers/0202/0202029.pdf`

[4] Wikipedia has a good description of Voigt profiles: `http://en.wikipedia.org/wiki/Voigt_profile`