

SATURATION OF ABSORPTION LINES

Last Revision: R. A. Schumacher May 2006
ver. 1.1

I. INTRODUCTION

When any sort of radiation passes through a scattering medium, the thickness of the medium is often assumed to be “thin enough” that essentially every incoming particle has equal probability of interacting with any particle in the scattering medium. However, when the material of the scattering is thick, or if the interaction probability is large, then the beam of incident radiation is significantly attenuated as it passes through. This can lead to “saturation” of the transmitted radiation energy spectrum. If the interaction probability is energy dependent, so is the attenuation, as will be discussed here. Saturation shows up in the absorption line intensity in the Mössbauer effect, and similarly in the line shape of the Optical Pumping experiment.

II. RADIATION ATTENUATION BY A “THICK” SCATTERER

Consider a beam of particles incident on a slab of material that can scatter and/or absorb the particles in the beam. The general arrangement is shown in Figure 1.

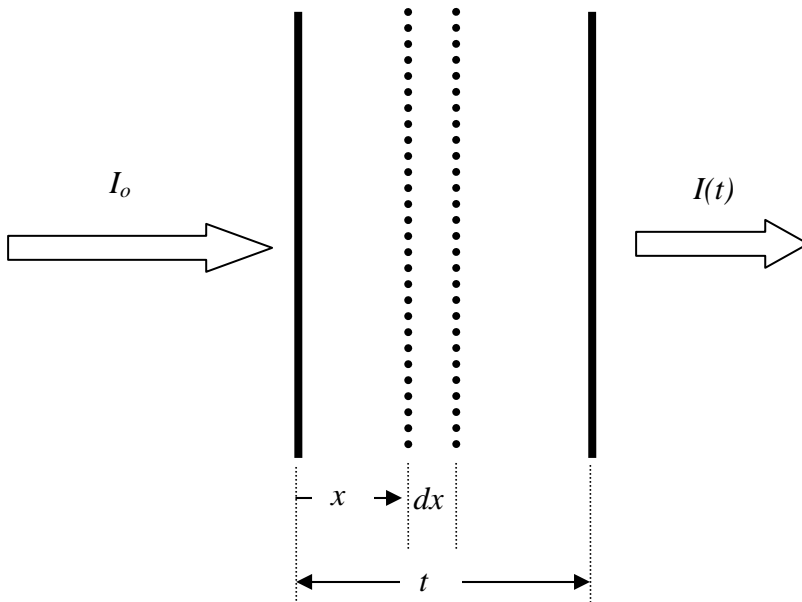


Figure 1: An incident beam of particles of intensity I_0 traverses a material of thickness t which absorbs or scatters some of the beam. The emerging beam has intensity $I(t)$.

The incident beam could be massive particles such as protons or electrons, and the intensity could then be specified as flux of particles per unit time. Or, the beam could be electromagnetic radiation, construed as photons and measured as photons per unit time. Alternatively, a classical electromagnetic wave can have some initial intensity measured

in, for instance, Joules per second. In the forward direction the transmitted beam intensity depends on the thickness of material, t , as well as the interaction strength between the beam and the material. The incident beam has an energy-dependent distribution of intensity $I(E)$ which we will specify below. Designate the interaction strength in terms of a total cross section, $\sigma(E)$, which is an energy dependent function with units of area. It specifies the effective area for one atom of the material scattering one beam particle into any direction other than the forward direction. This area can be larger than or smaller than the geometric area of an atom as seen by the beam, but it can be thought of as being of the order of the geometric area. In atomic physics where the size of the electron cloud matters, this is on the order of 10^{-20} m^2 , while for nuclear physics this is on the order of 10^{-30} m^2 . The question we ask is: what is the intensity distribution of the beam after traversing the material of thickness t ?

When the beam has passed through thickness x , the decrease in beam intensity dI in the next layer of material dx is given by the total cross section for interaction, σ , times the number of scatterers per unit area in the layer of material. This is written

$$dI(x) = -I(x)\sigma\left(\frac{\rho N_o}{W} dx\right) \quad (1)$$

where ρ is the density of the material (grams per cm^3), N_o is Avogadro's number (particles per mole), W is the atomic or molecular weight of the material (grams per mole). The quantity in parentheses is therefore the number of scatterers per unit area (particles per cm^2), and multiplying this by σ yields essentially the fraction of incident particles that will scatter in the layer of material. This equation has the solution, for an absorber of total thickness t ,

$$I(t) = I_o e^{-\sigma \frac{\rho N_o}{W} t} \quad (2)$$

where I_o is the incident beam intensity in particles per unit time. You might worry that the exact distribution of particles across the face of the absorber matters, but if you think it through you will see that this is irrelevant. Thus, the beam intensity is attenuated exponentially, and the rate of attenuation depends on the thickness of the material, its density, and on the strength of the interaction (*i.e.* the cross section). Next we consider the explicit energy dependence of the cross section and source energy since this is relevant in our measurements.

III. CALCULATING THE TRANSMISSION SPECTRUM AS A FUNCTION OF ENERGY

The cross section for absorption is energy dependent. For many physical systems in both quantum and classical physics, there is a preferred "natural" energy (or equivalent frequency) at which the system absorbs. The response of such systems is "resonant" as a function of energy, and it can be shown that the response follows a Lorentzian functional

form as given in Eq (3). In this discussion, the cross section for absorption is maximal at an energy E_o , and the cross section as a function of energy is given by

$$\sigma(E, E_o) = \sigma_o \frac{1}{\pi} \frac{\Gamma/2}{(E - E_o)^2 - (\Gamma/2)^2} \quad (3)$$

where the parameter Γ is the full width of the distribution at half its maximum height at the energy E_o . In this equation σ_o is the overall scale factor for the cross section, in units of area, and the $1/\pi$ factor results in a distribution normalized to unity when integrated over all energies, E . Note that in nuclear and atomic physics a ‘‘Lorentzian’’ distribution is equivalently called a ‘‘Breit-Wigner’’ distribution. In atomic and nuclear physics the width parameter Γ is related via the Heisenberg Uncertainty Principle to the *lifetime* of the state, that is, to how long it lives on the average before decaying. In classical physics it is related to the dissipative or frictional damping strength acting on the oscillation, and hence again to the lifetime of the oscillatory motion.

Next we suppose that the incoming beam of particles *also* is characterized by a population of particles that have a distribution of energies that is Lorentzian. For the M6ssbauer experiment this is indeed the case, since the source is a collection of nuclei that are just isomer and Doppler shifted with respect to the absorber. We write the source’s energy distribution as

$$I_s(E, E_v) = I_o \frac{1}{\pi} \frac{\Gamma/2}{(E - E_v)^2 - (\Gamma/2)^2} \quad (4)$$

where we assume that the width Γ parameter is the same as for the absorber, I_o is the energy-integrated source intensity in particles per second, and E_v is the energy for peak intensity for the source. Considering that in M6ssbauer spectroscopy the source energy is isomer shifted by amount δE_i and Doppler shifted by a velocity, V , we could write

$$E_v = E - \delta E_i + E_o \frac{V}{c} \quad (5)$$

but for this discussion we will stick to using E_v . Now we can construct an expression for the transmitted energy distribution $I(E_v, E_o, E)$ which combines the effect of the energy distribution of the source with that of the absorption in the material:

$$I(E_v, E_o, E) = I_s(E, E_v) e^{-\sigma(E, E_o) \frac{\rho t N_o}{W}} \quad (6)$$

Notice how the cross section is now written in a way that emphasizes that it, too, is a function of E and E_o , and that what multiplies the cross section in the exponent to make it dimensionless is the reciprocal of the effective number of particles per unit area, namely $(W/\rho t N_o)$. We do not really know or care what energy, E , any given particle in the beam has. In the M6ssbauer experiment we only know the centroid of the Doppler-shifted energy distribution characterized by E_v . Thus, we want to know the distribution

of transmitted energies as a function of E_V only, so we proceed to integrate over all possible values of E .

$$\begin{aligned}
 I(E_V, E_o) &= \int_0^{\infty} I_S(E, E_V) e^{-\sigma(E, E_o) \frac{\rho t N_o}{W}} dE \\
 &= \int_0^{\infty} I_o \frac{1}{\pi} \frac{\Gamma/2}{(E - E_V)^2 - (\Gamma/2)^2} e^{-\sigma_o \frac{1}{\pi} \frac{\Gamma/2}{(E - E_o)^2 - (\Gamma/2)^2} \frac{\rho t N_o}{W}} dE \quad (7)
 \end{aligned}$$

Thus, we have a convolution integral of the source energy distribution with the absorption energy distribution. Again, $I(E_V, E_o)$ represents the energy distribution of the beam coming out of the absorbing material as a function of the centroid energy of the source energy distribution E_V and the absorber energy distribution E_o . We can guess that this integral will have a maximum when these two energies are the same, and this is indeed the case. There are now three cases we can consider when trying to evaluate this integral.

III.A The absorber is vanishingly thin.

In the limit that the exponent is small, that is, when the dimensionless combination of factors $\alpha \equiv (\sigma_o \rho t N_o / W)$ is vanishingly small, then the outgoing flux goes to I_o . This is achieved either by the absorber being exceedingly thin ($t \rightarrow 0$) or the interaction being exceedingly weak ($\sigma_o \rightarrow 0$). Recall that we constructed our function out of a Lorentzian lineshape that is normalized to unity. That is

$$I(E_V) = \int_0^{\infty} I_o \frac{1}{\pi} \frac{\Gamma/2}{(E - E_V)^2 - (\Gamma/2)^2} dE = I_o \quad (8)$$

Do this integral by making a trigonometric substitution to reduce it to a simpler form. We will not do this integral explicitly here, and strictly speaking it is true only if the limits of integration are taken from minus infinity to infinity. For us, the values of E_V and Γ are in a range such that Eq (8) is perfectly valid as written.

III.B The absorber is thin.

When we have the dimensionless combination $\alpha \equiv (\sigma_o \rho t N_o / W) \ll 1$, we expand the exponential to first order. This results in an integral that can be done. We have

$$I(E_V, E_o) = I_o \left\{ 1 - \sigma_o \frac{\rho t N_o}{W} \int_0^{\infty} \left(\frac{1}{\pi} \frac{\Gamma/2}{(E - E_V)^2 - (\Gamma/2)^2} \right) \left(\frac{1}{\pi} \frac{\Gamma/2}{(E - E_o)^2 - (\Gamma/2)^2} \right) dE \right\} \quad (9)$$

This expression, Eq (9), contains the convolution of two Lorentzian lineshapes. This integral can be done by first Fourier transforming the integrand, doing the resultant

integral by contour integration in the complex plane, and then Fourier transforming the result back. The result is

$$I(E_v, E_o) = I_o \left\{ 1 - \left(\sigma_o \frac{\rho t N_o}{W} \right) \frac{1}{\pi} \frac{\Gamma}{(E_o - E_v)^2 - (\Gamma)^2} \right\} \quad (10)$$

What is remarkable is that the convolution of the two Lorentzians results in another Lorentzian with a width which is the sum of the widths of the two components. In this case we assumed Γ was the same for both, so the $(\Gamma/2)$ factors in Eq (9) become just Γ in Eq (10). (More generally, if we were to convolve Lorentzians with widths Γ_1 and Γ_2 , then the resultant would be a Lorentzian with $\Gamma = \Gamma_1 + \Gamma_2$.)

Thus, in the limit that the absorber is “thin”, in the sense that $\alpha \ll 1$, the amount of attenuation of the incident beam is linearly related to the thickness of material, t , and the strength of the interaction parameterized by σ_o .

III.C The absorber is not thin.

For thicker absorbers it is no longer valid to expand the exponential in Eq (7), and we resort to numerical integration to evaluate the expression for various values of $\alpha \equiv (\sigma_o \rho t N_o / W)$. The cross section is largest at the peak of the Lorentzian lineshape, so the attenuation is largest there as well. Away from the peak there is a smaller cross section and hence less absorption. This leads to “saturation” of the lineshape. The Lorentzian gets “squashed” at the peak and less so at the sides. Qualitatively, there is a limit to how much the system can absorb: when all the incident radiation is absorbed, there is nothing left and the transmitted intensity cannot dip below zero.

This is illustrated in Figure 2, which shows ten curves for which the values of α differ sequentially by factors of 2. The Lorentzians are centered at 1000 in arbitrary units (could be energy or frequency), and the nominal full width at half maximum for the source and absorber is always 10 units. Thus, the FWHM expected for the convoluted curves is 20 units in the thin absorber limit. The values of α are 10, 20, 40, ..., 5210. It is immediately apparent that the “height” and “width” of the curves are strongly affected by absorption. Table 1 lists the values of α , the ratio of the peak heights to the height of the smallest peak, and the computed effective FWHM for each curve. The curves are progressively “wider” even though the idealized width is always 20 units. This is the result of the peaks getting badly saturated.

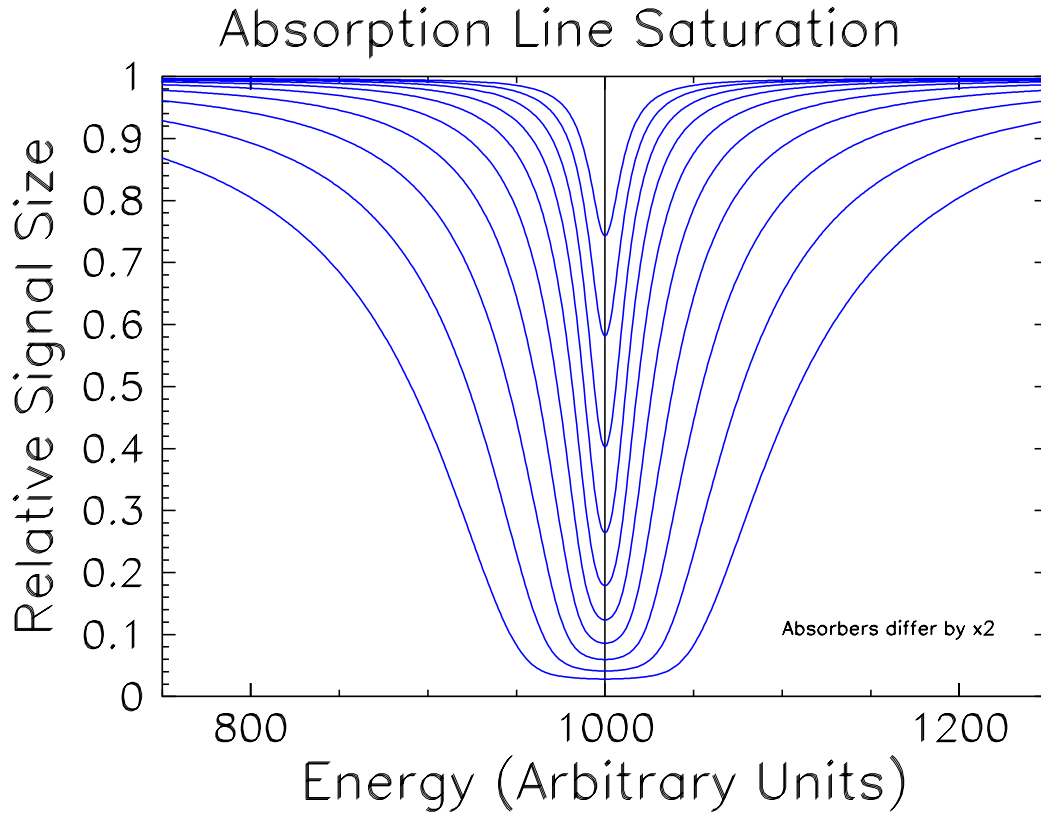


Figure 2: Lineshapes for absorption using ten values of the absorption coefficient α which differ in stepwise by factors of 2. Each curve has a nominal FWHM of 20 units. Peak 1 is the smallest one, Peak 10 the largest; cf. Table 1.

Peak Number	α	Peak Ratio	Width (FWHM)
1	10	1.00	20
2	20	1.63	22
3	40	2.32	26
4	80	2.86	32
5	160	3.19	44
6	320	3.41	60
7	640	3.56	82
8	1280	3.66	114
9	2560	3.73	158
10	5120	3.78	222

Table1: Numerical computation of the degree of absorption and apparent broadening of a Lorentzian lineshape for ten values of the absorption coefficient α . Each of the peaks has an idealized FWHM of 20 units.

IV. APPLICATION TO MOSSBAUER SPECTROSCOPY

The Zeeman splitting of the iron-57 nuclear energy levels results in six absorption lines of differing strength. Figure 3 shows a simulated spectrum for the six peaks which we will call A, B, C, D, E, and F, labeling from left to right. Peaks A, B, and C are drawn in the intensity ratio A:B:C = 3:2:1 which arises from an iron foil which is *not* polarized by an external magnetic field. These ratios are a result of the angular momentum coupling among the $I = \frac{1}{2}$ ground state, the $I = \frac{3}{2}$ excited state, and the $L=1$ dipole field of the absorbed (or emitted) photon. It can be shown via the Clebsch-Gordon algebra that the various transitions occur in this intensity ratio when averaging over all direction of photon absorption (or emission). Peaks D, E, and F are drawn in the intensity ratio D:E:F = 1:4:3 which arises when the absorbing foil is magnetized perpendicular to the direction of the incoming gamma rays. Thus the incoming gamma rays are absorbed (or emitted) specifically at 90° . The dipole radiation pattern for an oriented dipole differs by a factor of 2 depending on whether the change in m value of the nucleus is 0 or ± 1 ; this is reflected in the m value of the photon field. The steps of this calculation are not in the scope of this discussion.

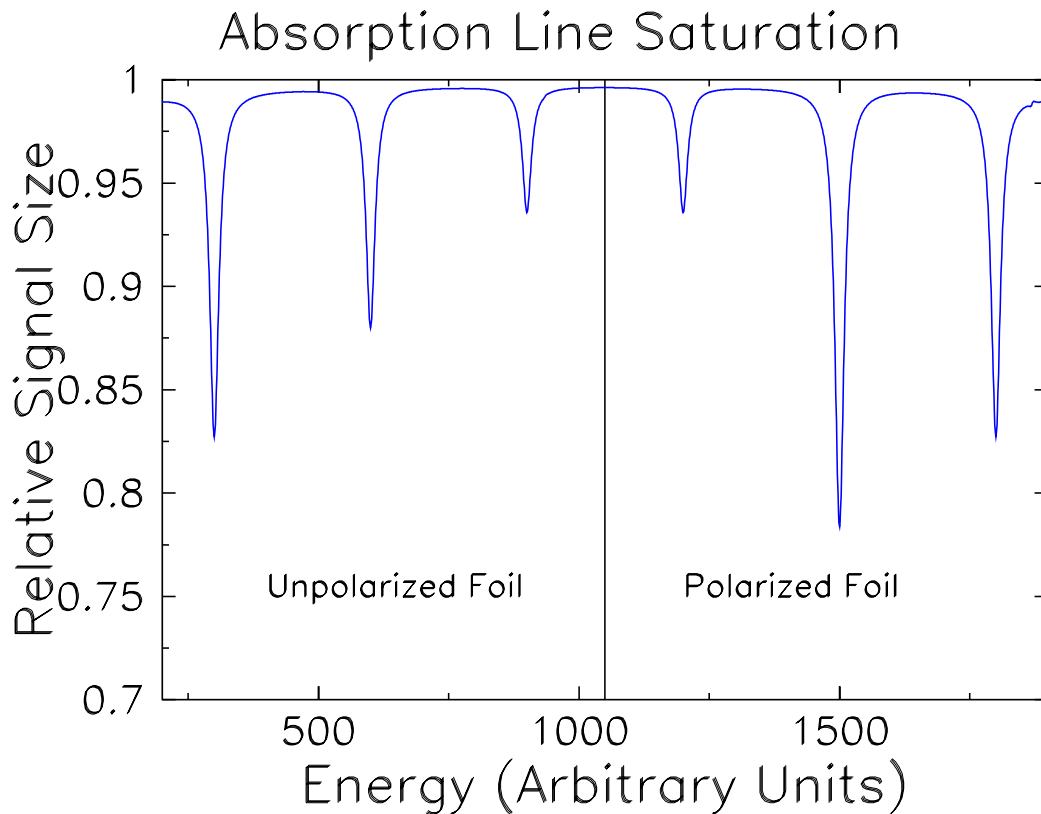


Figure 3: The lineshape for the Zeeman effect seen in ^{57}Fe showing six resonant absorption lines with relative weights 3:2:1:1:4:3 reading from left to right. Absorption is very weak. Note that the vertical scale has a large offset from zero.

For Figure 3 the values of α ranged from 20 to 80. Figure 4 illustrates what happens to the distribution of line intensities when all values of α are increased by a factor of 10. Now the saturation effect on the relative line intensities is very apparent. The outer peaks are very similar in magnitude and their ratio to the central peaks is less than 2. This roughly agrees with what is seen with a 0.050 mm (“2 mil”) thick absorbing foil.

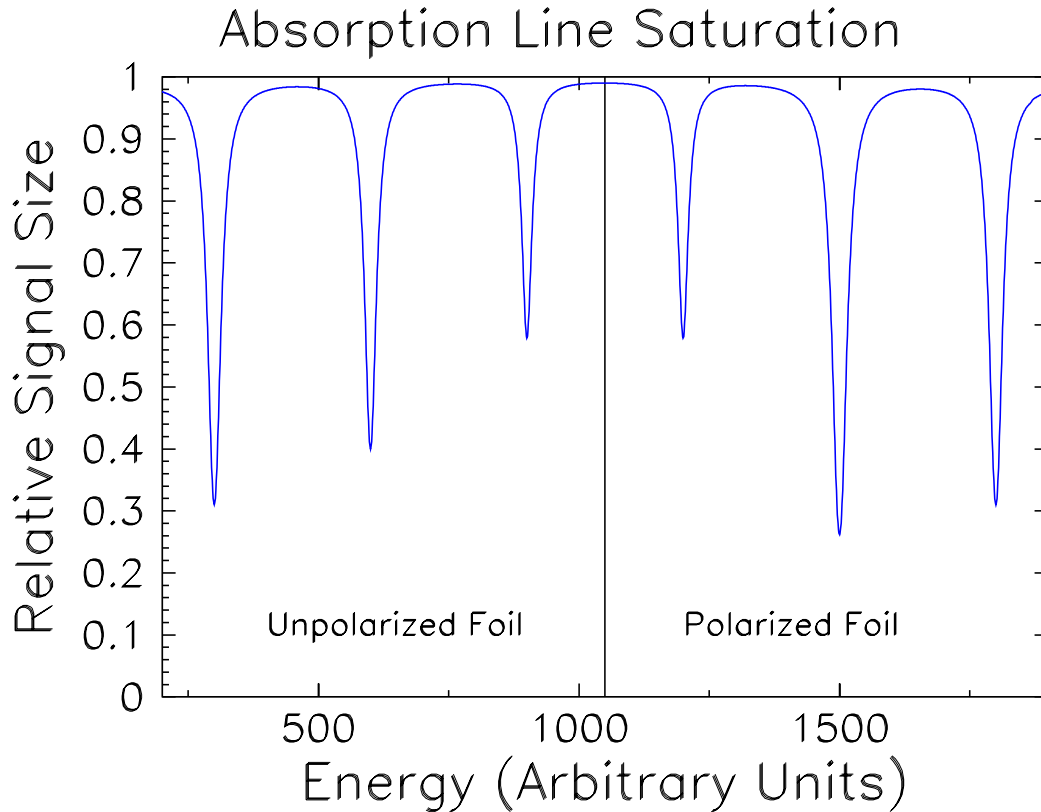


Figure 4: The lineshape for the Zeeman effect seen in ^{57}Fe showing six resonant absorption lines with nominal relative weights 3:2:1:1:4:3 reading from left to right. The vertical scale reaches down to zero. For the “thick” absorber in this illustration a saturation of the relative intensities is seen.

V. APPLICATION TO OPTICAL PUMPING

Once optical pumping is established the Rubidium cell is transparent to the incoming light. The detector receives light that is minimally attenuated, and the signal is as big as it can get. When the RF pulse is applied transverse to the magnetic field direction, optical pumping is destroyed when the corresponding photon energy matches the energy gap between the states in the Zeeman-split hyperfine ground state. At this point the absorption cross section shoots up and the detector sees a reduction in intensity. Experimentally we can sweep either the magnetic field strength or the RF frequency. In

either scenario the lineshape of the absorption spectrum will be affected by the type of saturation discussed above.

The discussion in Section II about attenuation applies to this situation. The energy dependence discussed in terms of a Lorentzian lineshape in Section III is partly applicable to optical pumping. The absorber's lineshape, that is, the resonant absorption of the radiation that induces transitions between the levels in the Rubidium atom is in principle Lorentzian. However, the energy distribution of what we called the "source" can not be expected to be identical to that of the absorbing Rubidium atoms. The RF frequency stability may be much better, for instance, than the natural line width of the atomic states. In Eq (7), one must have some new form for $I_S(E, E_V)$ that we cannot write down in some obvious form. Nevertheless, whatever $I_S(E, E_V)$ turns out to be, it is in principle not difficult to insert it into the equation and perform the numerical integration as before.

VI. CONCLUSION

In conclusion, we have shown how the combined effects of absorption and beam attenuation conspire to distort a Lorentzian line shape, such that both the relative absorption strength between two lines and the width of the lines are masked. Experimentally, the way to combat this difficulty is to work with progressively "thinner" samples in order to make an extrapolation of absorption cross section and line width to the unattainable limit of zero sample thickness.