Simulating Cavity Transmission Spectra

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In these notes, we will work through simulations of the reflection and transmission spectra for materials sandwiched inside planar Fabry-Pérot (FP) optical cavities. We will work through treatment of the intracavity sample as a Lorentz oscillator. We also summarize transfer matrix methodology for treatment of more complex multilayer cavity devices. For the FP cavity expressions we largely follow the derivation used in Chapter 3 of "Quantum Electronics for Atomic Physics" by Warren Nagourney. For the treatment of the complex index of refraction, permittivity, and Lorentz oscillator model, we follow these notes from MIT OpenCourseWare and this technical note from Horiba. For transfer matrix methodology, much more information can be found in Chapter 2 of Macleod's "Thin-Film Optical Filters," available online through the Princeton Library here. Papers by Pettersson et al. and Peumans et al. are also good references which summarize these results. The McGehee Group at Stanford has made some well-documented Matlab code available here.

1 The two-mirror Fabry-Pérot cavity

Let's consider a cavity with two mirrors labeled 1 and 2 (Fig. 1), spaced a distance L apart. We assume that mirrors 1 and 2 are lossless, with reflection and transmission amplitude coefficients of r_1, t_1 and r_2, t_2, respectively. We will eventually assume that these two mirrors are identical (e.g. $r_1 = r_2$ and $t_1 = t_2$) but for the moment these indices are useful for bookkeeping. We will place an absorptive material inside the cavity, which has its own amplitude transmission coefficient of t, and a frequency-dependent refractive index $n(\nu)$.

Note that these lowercase r and t coefficients capture the effects that the mirrors have on the *amplitudes* of fields of light, E. Once we start to talk about *intensities*, I, we will also find it useful to define the intensity transmission and reflection coefficients:

$$R \equiv r_1^2 = r_2^2 \tag{1}$$

$$T \equiv t_1^2 = t_2^2 \tag{2}$$

where, since we are neglecting mirror losses,

$$T + R = 1 \tag{3}$$

indicating that when light hits a mirror, 100% of its power is either transmitted or reflected.

Let's now discuss briefly how to work with the amplitude reflection and transmission coefficients. The transmitted amplitude of light is equivalent when approached from either side of an interface.

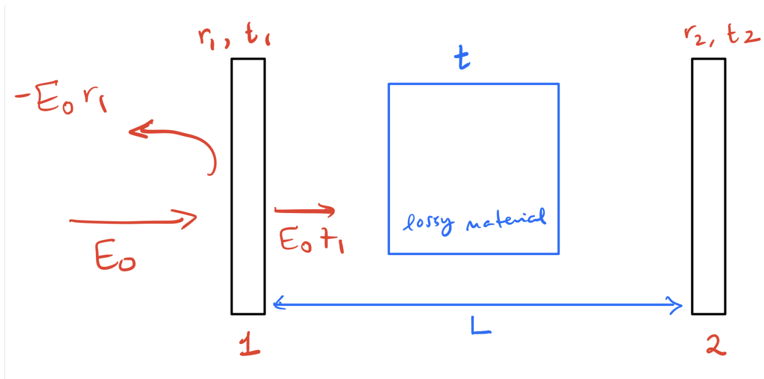

Figure 1

As shown in Fig. 1, when an incoming field with amplitude E_0 strikes mirror 1 from the left, the transmitted field amplitude is $E_T = +E_0 t_1$. The same is true if mirror 1 is approached from the right. For reflection, on the other hand, the amplitude coefficient has opposite sign when approached from either side of an interface. See Nagourney Section 3.2 for more details on how this arises. Here, we define our system such that when E_0 approaches either mirror from outside the cavity (e.g. from the left of mirror 1 or the right of mirror 2), the reflected amplitude is $E_R = -E_0 r_1$. If we approach either mirror from inside the cavity, however, the reflected amplitude is $E_R = +E_0 r_1$. These sign conventions are arbitrary, but it's important to be consistent.

Another important point is that as light traverses the cavity, it accrues a phase shift, δ. To derive an expression for δ, let's consider monochromatic light with vacuum wavelength λ, and a wavelength of $\lambda_n = \lambda/n(\nu)$ in our intracavity medium, with refractive index $n(\nu)$. For each integer multiple of λ_n that the light travels inside the cavity, it accrues a phase shift of 2π. Therefore, for one round trip through the cavity where the light travels a distance of $2L$, the total phase shift can be expressed as:

$$\delta = 2\pi \cdot \frac{2L}{\lambda_n} + 2\phi = \frac{4\pi L n(\nu)}{\lambda} + 2\phi = \frac{4\pi L n(\nu)\nu}{c} + 2\phi \tag{4}$$

where c is the speed of light, and ϕ is the phase change accrued upon a single reflection from each mirror. For our purposes, ϕ can be neglected, since it will simply serve to shift all of our cavity resonances by a common frequency offset. We also neglect the Gouy phase here for the same reason.

2 The reflected field amplitude

After we send our initial E_0 field amplitude into the cavity, the total reflected amplitude E_R can be constructed by considering the interference of all possible field trajectories that have made any integer number of passes through the cavity. Consider, as sketched in Fig. 2:

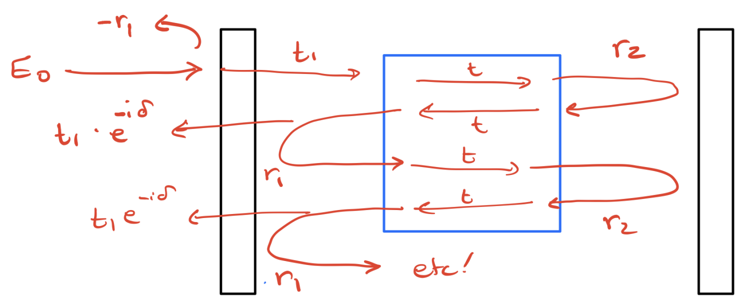

Figure 2

$$
\begin{aligned}
E_R = &- E_0 \cdot r_1 && \text{0 passes} \qquad (5)\\
&+ E_0 \cdot t_1 \cdot t \cdot r_2 \cdot t \cdot t_1 \cdot e^{-i\delta} && \text{1 pass} \qquad (6)\\
&+ E_0 \cdot t_1 \cdot t \cdot r_2 \cdot t \cdot r_1 \cdot t \cdot r_2 \cdot t \cdot t_1 \cdot e^{-2i\delta} && \text{2 passes} \qquad (7)\\
&+ \ldots
\end{aligned}
$$

This is an infinite geometric series, which we can simplify as

$$
\begin{aligned}
E_R &= -E_0 \cdot r_1 + E_0 \cdot t_1^2 \left[t^2 r_2 e^{-i\delta}\right] + E_0 \cdot t_1^2 \left[t^4 r_1 r_2^2 e^{-2i\delta}\right] + E_0 \cdot t_1^2 \left[t^6 r_1^2 r_2^3 e^{-3i\delta}\right] + \ldots && (8)\\
&= -E_0 \cdot r_1 + E_0 \cdot \frac{t_1^2}{r_1} \left[t^2 r_1 r_2 e^{-i\delta} + t^4 r_1^2 r_2^2 e^{-2i\delta} + t^6 r_1^3 r_2^3 e^{-3i\delta} \ldots\right] && (9)\\
&= -E_0 \cdot r_1 + E_0 \cdot \frac{t_1^2}{r_1} \sum_{k=1}^{\infty} \left[t^2 r_1 r_2 e^{-i\delta}\right]^k && (10)
\end{aligned}
$$

Since the terms within the sum above are all less than 1, we can use what we know about geometric series. For $|r| < 1$

$$
\sum_{k=0}^{\infty} r^k = \frac{1}{1-r} \qquad \rightarrow \sum_{k=1}^{\infty} r^k = \frac{1}{1-r} - 1 \tag{11}
$$

So:

$$E_R = -E_0 \cdot r_1 + E_0 \cdot \frac{t_1^2}{r_1}\left[\frac{1}{1-t^2 r_1 r_2 e^{-i\delta}} - 1\right] \tag{12}$$

$$= -E_0 \cdot r_1 + E_0 \cdot \frac{t_1^2}{\cancel{r_1}}\left[\frac{\cancel{1} - (\cancel{1} - t^2 \cancel{r_1} r_2 e^{-i\delta})}{1-t^2 r_1 r_2 e^{-i\delta}}\right] \tag{13}$$

$$= -E_0 \cdot r_1 + E_0 \left[\frac{t^2 t_1^2 r_2 e^{-i\delta}}{1-t^2 r_1 r_2 e^{-i\delta}}\right] \tag{14}$$

$$= E_0 \left[\frac{t^2 t_1^2 r_2 e^{-i\delta}}{1-t^2 r_1 r_2 e^{-i\delta}} - r_1 \cdot \frac{1-t^2 r_1 r_2 e^{-i\delta}}{1-t^2 r_1 r_2 e^{-i\delta}}\right] \tag{15}$$

$$= E_0 \left[\frac{t^2 r_2 (t_1^2 + r_1^2) e^{-i\delta} - r_1}{1-t^2 r_1 r_2 e^{-i\delta}}\right] = \boxed{E_0 \left[\frac{t^2 r_2 e^{-i\delta} - r_1}{1-t^2 r_1 r_2 e^{-i\delta}}\right]} \tag{16}$$

where in the last step we used $t_1^2 + r_1^2 = 1$.

3 The circulating field amplitude

We can carry out a very similar calculation for the field circulating inside the cavity. We will consider only the intracavity field propagating towards the right, in the region to the left of the lossy medium:

$$E_C = E_0 \cdot t_1 \tag{17}$$

$$+ E_0 \cdot t_1 \cdot t \cdot r_2 \cdot t \cdot r_1 \cdot e^{-i\delta} \tag{18}$$

$$+ E_0 \cdot t_1 \cdot t \cdot r_2 \cdot t \cdot r_1 \cdot e^{-i\delta} \cdot t \cdot r_2 \cdot t \cdot r_1 \cdot e^{-i\delta} \tag{19}$$

$$+ \ldots$$

$$= E_0 \cdot t_1 \left[1 + t^2 r_1 r_2 e^{-i\delta} + t^4 r_1^2 r_2^2 e^{-2i\delta} + \ldots\right] \tag{20}$$

$$= E_0 \cdot t_1 \sum_{k=0}^{\infty} \left[t^2 r_1 r_2 e^{-i\delta}\right]^k \tag{21}$$

$$= \boxed{E_0 \left[\frac{t_1}{1-t^2 r_1 r_2 e^{-i\delta}}\right]} \tag{22}$$

4 The transmitted field amplitude

Calculating the transmitted field is straightforward given the circulating field amplitude. For each term in the sum of interfering waves, the field makes one additional pass through the lossy intracavity medium and the second cavity mirror. So:

$$E_T = E_C \cdot t \cdot t_2 = \boxed{E_0 \left[\frac{t_1 t_2 t}{1 - t^2 r_1 r_2 e^{-i\delta}} \right]} \tag{23}$$

5 The transmitted *intensity*

When we use a square-law detector in the laboratory, we are measuring the intensity of fields of light rather than their amplitudes. So it is useful to convert the amplitude quantities we've discussed so far into intensities for the reflected, circulating, and transmitted fields of light in our FP cavity. In general, intensity is related to the amplitude as:

$$\frac{I}{I_0} = \left| \frac{E}{E_0} \right|^2 \tag{24}$$

For cavity transmission in particular:

$$\begin{aligned} \frac{I_T}{I_0} &= \left| \frac{E_T}{E_0} \right|^2 = \left| \frac{t_1 t_2 t}{1 - t^2 r_1 r_2 e^{-i\delta}} \right|^2 & (25) \\ &= \frac{t_1^2 t_2^2 t^2}{|1 - t^2 r_1 r_2 e^{-i\delta}|^2} & (26) \\ &= \frac{t_1^2 t_2^2 t^2}{(1 - t^2 r_1 r_2 e^{-i\delta}) \cdot (1 - t^2 r_1 r_2 e^{+i\delta})} & (27) \\ &= \frac{t_1^2 t_2^2 t^2}{1 - t^2 r_1 r_2 (e^{+i\delta} + e^{-i\delta}) + t^4 r_1^2 r_2^2} & (28) \\ &= \frac{t_1^2 t_2^2 t^2}{1 - t^2 r_1 r_2 \cdot 2\cos\delta + t^4 r_1^2 r_2^2} & (29) \\ & & (30) \end{aligned}$$

We can simplify this expression by assuming that our two mirrors are identical ($r_1 = r_2$, $t_1 = t_2$), and using $r_1^2 = R$, $t_1^2 = T$.

$$\frac{I_T}{I_0} = \frac{T^2 t^2}{1 + R^2 t^4 - 2Rt^2 \cos\delta} \tag{31}$$

Finally, we can tweak this expression to capture the dependence of the cavity transmission spectrum on a few important experimental parameters. From the Beer-Lambert Law, we know that the intensity of light transmitted through the lossy intracavity medium decays exponentially with its absorption coefficient $\alpha(\nu)$ and the pathlength through the sample. Assuming the sample fills the cavity, so the pathlength is L, we can state

$$t^2 = e^{-\alpha(\nu)L} \tag{32}$$

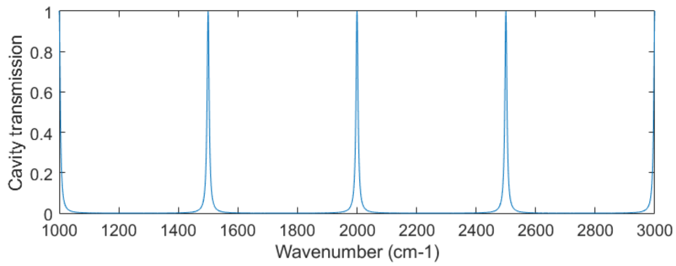

Figure 3

We will also include our expression for δ from Eqn. 4, to give:

$$\boxed{\frac{I_T}{I_0} = \frac{T^2 e^{-\alpha(\nu)L}}{1 + R^2 e^{-2\alpha(\nu)L} - 2Re^{-\alpha(\nu)L}\cos\left(\frac{4\pi L n(\nu)\nu}{c}\right)}} \tag{33}$$

It is from the frequency dependence of the refractive index, the absorption coefficient, and the round-trip phase shift that the interesting spectral properties of the Fabry-Pérot cavity emerges. The cavity transmission spectrum for an empty cavity with $R = 0.95$, $T = 0.05$, $L = 10\,\mu$m, $\alpha = 0$, and $n = 1$ is shown in Fig. 3.

The positions of the cavity transmission maxima occur when $\cos\left(\frac{4\pi L n(\nu)\nu}{c}\right)$ is maximized, which occur when

$$\frac{4\pi L n(\nu)\nu_m}{c} = 2\pi m \tag{34}$$

$$\rightarrow \nu_m = \frac{c}{2n(\nu)L} \cdot m \tag{35}$$

where m is an integer. These modes would be perfectly evenly spaced were it not for the frequency dependence of n, which causes dispersion. The spacing between neighboring resonances is called the free spectral range (FSR), and is given by

$$\text{FSR} = \frac{c}{2n(\nu)L} \tag{36}$$

Another useful cavity parameter is the finesse, $\mathcal{F}$, which is a measure of how lossy the resonator is. $\mathcal{F}$ is defined as the ratio between the FSR and the full-width-at-half-maximum cavity resonance linewidth, $\Delta\nu$, and can be expressed for a lossless cavity as

$$\mathcal{F} = \frac{\text{FSR}}{\Delta\nu} \approx \frac{\pi\sqrt{R}}{1 - R} \tag{37}$$

Note that the FSR is constrained solely by the cavity geometry and length, the finesse is constrained by the quality of the mirrors, and the cavity linewidths depend on these two parameters. Wee Nagourney Ch. 3 for more thorough discussion of these parameters and detailed derivations.

6 The complex refractive index

We now have a good understanding of the optical properties of an empty Fabry-Pérot optical cavity. We will next consider how to treat an intracavity molecular sample. In particular, how do we model the refractive index $n(\nu)$ and absorption coefficient $\alpha(\nu)$ of an arbitrary sample?

It is instructive to review what the refractive index actually captures about a material. Let's consider first the complex refractive index, $\bar{n}$

$$\bar{n}(\nu) \equiv n(\nu) + i\kappa(\nu) \tag{38}$$

which is composed of a real component, $n(\nu)$, what we usually think of as the "refractive index" and the imaginary component $\kappa(\nu)$, also called the "extinction coefficient." $\kappa(\nu)$ is closely related to $\alpha(\nu)$, as we will see in a moment. $n(\nu)$ captures the *phase delay* of light as it traverses the medium, while $\kappa(\nu)$ captures the *attenuation* of the amplitude of that light.

To see these parameters in action, let's consider a beam of light traversing our material in the z direction with a sinusoidal electric field represented by the real part of a complex exponential

$$E(z,t) = \text{Re}\left[\overrightarrow{E_0} \cdot e^{i(\bar{k}z - \omega t)}\right] \tag{39}$$

Here $\bar{k}$ is the complex wavevector given by

$$\bar{k} = \frac{2\pi}{\lambda_n} = \frac{2\pi\bar{n}}{\lambda} = \frac{2\pi(n+i\kappa)}{\lambda} \tag{40}$$

where again, λ_n is the wavelength of light in the material, and λ is the vacuum wavelength. Plugging this expression for $\bar{k}$ back into Eqn. 39, we find

$$E(z,t) = \text{Re}\left[\overrightarrow{E_0} \cdot e^{i(2\pi(n-i\kappa)z/\lambda - \omega t)}\right] \tag{41}$$

$$= e^{-2\pi\kappa z/\lambda} \cdot \text{Re}\left[\overrightarrow{E_0} \cdot e^{i(2\pi n z/\lambda - \omega t)}\right] \tag{42}$$

From inspection of Eqn. 42, it is clear that as we traverse a distance z into the material, the electromagnetic field decays exponentially with κ, while n leads only to a change in the phase of the wavefront. We can relate κ to the absorption coefficient α with reference to the Beer-Lambert law. The attenuation of the *intensity* of the light goes like the field squared, and therefore:

$$e^{-\alpha z} = \left[e^{-2\pi k z/\lambda}\right]^2 \tag{43}$$

$$\rightarrow \alpha = \frac{4\pi\kappa}{\lambda} = \frac{4\pi\kappa\nu}{c} \tag{44}$$

7 The complex dielectric constant

It is also convenient to discuss the permittivity, or dielectric constant, of the sample, ϵ, which has units of F/m. We will relate this parameter to the complex index of refraction. Permittivity is a measure of the electric polarizability of a material under an applied field. ϵ has a frequency dependence because the polarization of the material cannot change instantaneously under an oscillating field. Instead, the system can experience phase delays near resonances or that generally increases with frequency.

We usually consider the permittivity of the system relative to that of the vacuum:

$$\epsilon_r \equiv \frac{\epsilon}{\epsilon_0} \tag{45}$$

which is a dimensionless complex quantity.

The complex index of refraction can be expressed in terms of the relative permittivity and its magnetic analog, the relative permeability μ_r:

$$\bar{n} = \sqrt{\epsilon_r \cdot \mu_r} \tag{46}$$

For nonmagnetic materials and optical frequencies, it is a good assumption that $\mu_r = 1$. Therefore

$$\epsilon_r = \bar{n}^2 = (n + i\kappa)^2 = n^2 - \kappa^2 + 2in\kappa \tag{47}$$

We will define the real and imaginary parts of ϵ_r such that $\epsilon_r = \epsilon_1 - i\epsilon_2$. Therefore

$$\epsilon_1 = n^2 - \kappa^2 \tag{48}$$

$$\epsilon_2 = -2n\kappa \tag{49}$$

It is straightforward to rearrange this system of equations to obtain n and κ in terms of ϵ_1 and ϵ_2. Note that

$$\epsilon_1^2 + \epsilon_2^2 = (n^2 - \kappa^2)^2 + 4n^2k^2 \tag{50}$$

$$= n^4 + \kappa^4 - 2n^2\kappa^2 + 4n^2\kappa^2 \tag{51}$$

$$= n^4 + \kappa^4 + 2n^2\kappa^2 = (n^2 + \kappa^2)^2 \tag{52}$$

$$\rightarrow n^2 + \kappa^2 = \sqrt{\epsilon_1^2 + \epsilon_2^2} \tag{53}$$

And therefore, combining Eqn. 53 with Eqns. 48 and 49 we arrive at

$$n = \sqrt{\frac{\sqrt{\epsilon_1^2 + \epsilon_2^2} + \epsilon_1}{2}} \tag{54}$$

$$\kappa = \sqrt{\frac{\sqrt{\epsilon_1^2 + \epsilon_2^2} - \epsilon_1}{2}} \tag{55}$$

Eqns. 54 and 55 are general, regardless of the system under study or the functional form of the permittivity. We will next consider a specific model system to help construct the complex permittivity of a molecular sample.

8 The Lorentz oscillator model

When light strikes a sample, its oscillating electromagnetic field can drive the motion of nuclear or electronic charges in the material, giving rise to vibrational or electronic optical transitions. The Lorenz oscillator model treats the motion of nuclei or electrons in response to the driving field as a damped harmonic oscillator.

While we won't work through all the details here, it is illuminating to write down the equation of motion of such a system. The amplitude of displacement $\overrightarrow{r}(t)$ of a particle with charge Z and mass m can be related to the external time-dependent electromagnetic field $\overrightarrow{E_0}(t)$ by the differential equation:

$$m \cdot \frac{d^2\overrightarrow{r}}{dt^2} + m \cdot \Gamma_0 \cdot \frac{d\overrightarrow{r}}{dt} + m \cdot \omega_t^2 \cdot r = -Z\overrightarrow{E_0}(t) \tag{56}$$

where the first term on the left hand side represents the force due to acceleration, the second term represents a viscous damping force with damping factor Γ_0, and the third term is a restoring force given by Hooke's law where ω_t is the resonant frequency of the oscillator. The right hand side represents the total force felt on charge Z by the field.

If the external field takes the form $\overrightarrow{E_0}(t) = \overrightarrow{E_0} \cdot e^{i\omega t}$, we can make the assumption that the displacement of the particle has a similar time-dependent form $\overrightarrow{r}(t) = \overrightarrow{r} \cdot e^{i\omega t}$, where $\overrightarrow{r}$ is complex and will depend on the field's driving frequency ω.

By plugging this functional form of $\overrightarrow{r}(t)$ into our differential equation Eqn. 56, we find

$$m\left[(-i\omega)^2 \cdot \overrightarrow{r} \cdot e^{i\omega t}\right] + m\Gamma_0\left[i\omega \cdot \overrightarrow{r} \cdot e^{i\omega t}\right] + m\omega_t^2 \cdot \overrightarrow{r} \cdot e^{i\omega t} = -Z\overrightarrow{E_0} \cdot e^{i\omega t} \tag{57}$$

$$= \overrightarrow{r}\left[-m\omega^2 + im\omega\Gamma_0 + m\omega_t^2\right] = -Z\overrightarrow{E_0} \tag{58}$$

$$\rightarrow \boxed{\overrightarrow{r} = \overrightarrow{r}(\omega) = \frac{-Z\overrightarrow{E_0}}{m(\omega_t^2 - \omega^2) + im\omega\Gamma_0}} \tag{59}$$

We ultimately want relate this expression for field-induced displacement to the dielectric constant of the material, which we can subsequently relate to the complex index of refraction. The induced dipole moment $\overrightarrow{\mu}$ is related to both the displacement and the polarizability of the material $\alpha(\omega)$ by

$$\overrightarrow{\mu} = -Z \cdot \overrightarrow{r}(\omega) = \alpha(\omega) \cdot \overrightarrow{E_0} \tag{60}$$

$$\rightarrow \alpha(\omega) = \frac{-Z \cdot \overrightarrow{r}(\omega)}{\overrightarrow{E_0}} \tag{61}$$

If we consider an ensemble of N oscillators per unit volume, the polarization per unit volume $P(\omega)$ allows us to relate the polarizability $\alpha(\omega)$ to the susceptibility $\chi(\omega)$ by:

$$P(\omega) = N \cdot \alpha(\omega) \cdot E(\omega) = \epsilon_0 \cdot \chi(\omega) \cdot E(\omega) \tag{62}$$

$$\rightarrow \chi(\omega) = \frac{N \cdot \alpha(\omega)}{\epsilon_0} \tag{63}$$

$\chi(\omega)$ is closely related to the relative dielectric constant by $\epsilon_r = 1 + \chi(\omega)$. So we now can write:

$$\epsilon_r = 1 + \chi(\omega) = 1 + \frac{N\alpha(\omega)}{\epsilon_0} = 1 + \frac{N}{\epsilon_0} \cdot \frac{-Z\overrightarrow{r}(\omega)}{\overrightarrow{E_0}} \tag{64}$$

$$= 1 - \frac{NZ}{\epsilon_0\overrightarrow{E_0}} \cdot \frac{-Z\overrightarrow{E_0}}{m(\omega_t^2 - \omega^2) + im\omega\Gamma_0} \tag{65}$$

$$= 1 + \frac{NZ^2}{m\epsilon_0}\left[\frac{1}{(\omega_t^2 - \omega^2) + i\omega\Gamma_0}\right] \tag{66}$$

The prefactor $NZ^2/m\epsilon_0$ is usually defined as the square of the "plasma frequency" ω_p, which is an innate property of the material and represents the resonant frequency with which the ensemble of charges oscillates collectively. We can therefore write the dielectric constant of a Lorentz oscillator as:

$$\boxed{\epsilon_r = 1 + \frac{\omega_p^2}{(\omega_t^2 - \omega^2) + i\omega\Gamma_0}} \tag{67}$$

We will now massage Eqn. 67 into a somewhat more useful form and also consider the possibility of multiple resonant frequencies. We first define the low and high frequency limits of ϵ_r:

$$\epsilon_s = \epsilon_r(\omega \to 0) = 1 + \frac{\omega_p^2}{\omega_t^2} \tag{68}$$

$$\epsilon_\infty = \epsilon_r(\omega \to \infty) = 1 \tag{69}$$

We can therefore rewrite Eqn. 67 as

$$\epsilon_r = \epsilon_\infty + \frac{(\epsilon_s - \epsilon_\infty) \cdot \omega_t^2}{\omega_t^2 - \omega^2 + i\Gamma_0\omega} \tag{70}$$

where we have abstracted the plasma frequency away into ϵ_∞. The factor $\epsilon_s - \epsilon_\infty$ encodes the oscillator strength. ϵ_∞ can be higher than 1 for real systems where resonances of the system lying at higher frequencies are not explicitly taken into account. Recalling that the relative dielectric constant is related to the index of refraction by $\epsilon_r = n^2$, we can say $\epsilon_\infty = n_{bg}^2$, where n_{bg} is a constant real component of the background refractive index, independent of frequency.

If we want to treat multiple resonant frequencies of the system, we assume that their contributions to the dielectric function add linearly. So for M resonant frequencies ω_j we have

$$\epsilon_r = n_{bg}^2 + \sum_{j=1}^{M} \frac{A_j \cdot \omega_j^2}{\omega_j^2 - \omega^2 + i\Gamma_j\omega} \tag{71}$$

where A_j represents the unitless oscillator strength for the j^{th} resonant frequency, and Γ_j represents its damping factor, which also turns out to be the full-width-at-half-maximum resonance linewidth.

We can split Eqn. 71 into its real and imaginary components:

$$\epsilon_r = n_{bg}^2 + \sum_{j=1}^{M} \frac{A_j \cdot \omega_j^2}{\omega_j^2 - \omega^2 + i\Gamma_j\omega} \cdot \left[\frac{\omega_j^2 - \omega^2 - i\Gamma_j\omega}{\omega_j^2 - \omega^2 - i\Gamma_j\omega}\right] \tag{72}$$

$$= n_{bg}^2 + \sum_{j=1}^{M} \frac{A_j\omega_j^2(\omega_j^2 - \omega^2) - iA_j\omega_j^2\omega\Gamma_j}{(\omega_j^2 - \omega^2)^2 + \omega^2\Gamma_j^2} \tag{73}$$

$$\to \epsilon_1 = \mathrm{Re}[\epsilon_r] = n_{bg}^2 + \sum_{j=1}^{M} \frac{A_j\omega_j^2(\omega_j^2 - \omega^2)}{(\omega_j^2 - \omega^2)^2 + \omega^2\Gamma_j^2} \tag{74}$$

$$\to \epsilon_2 = -\,\mathrm{Im}[\epsilon_r] = \sum_{j=1}^{M} \frac{A_j\omega_j^2\omega\Gamma_j}{(\omega_j^2 - \omega^2)^2 + \omega^2\Gamma_j^2} \tag{75}$$

Eqns. 74 and 75 can now be plugged into Eqns. 54 and 55 to obtain the index of refraction and extinction coefficient of an arbitrary Lorentz oscillator.

9 Some numerical examples

We now have all the pieces in place to take the empty cavity modeled in Fig. 3, and place a molecular sample described as a Lorentz oscillator inside it.

Let's set up an oscillator with a single resonant frequency $\omega_1 = 2000$ cm^{-1}, linewidth $\Gamma_1 = 10$ cm^{-1}, oscillator strength $A_1 = 1 \times 10^{-4}$, and $n_{bg} = 1$. The dielectric constant, index of refraction, and extinction coefficient are plotted as a function of frequency near ω_1 in Fig. 4 using the expressions we've just derived. We then plug $n(\nu)$ and $\alpha(nu)$ into Eqn. 33 to obtain the transmission spectrum of a resonant Fabry-Pérot cavity, assuming the same cavity parameters we used previously ($R = 0.95$, $T = 0.05$, $L = 10\,\mu$m).

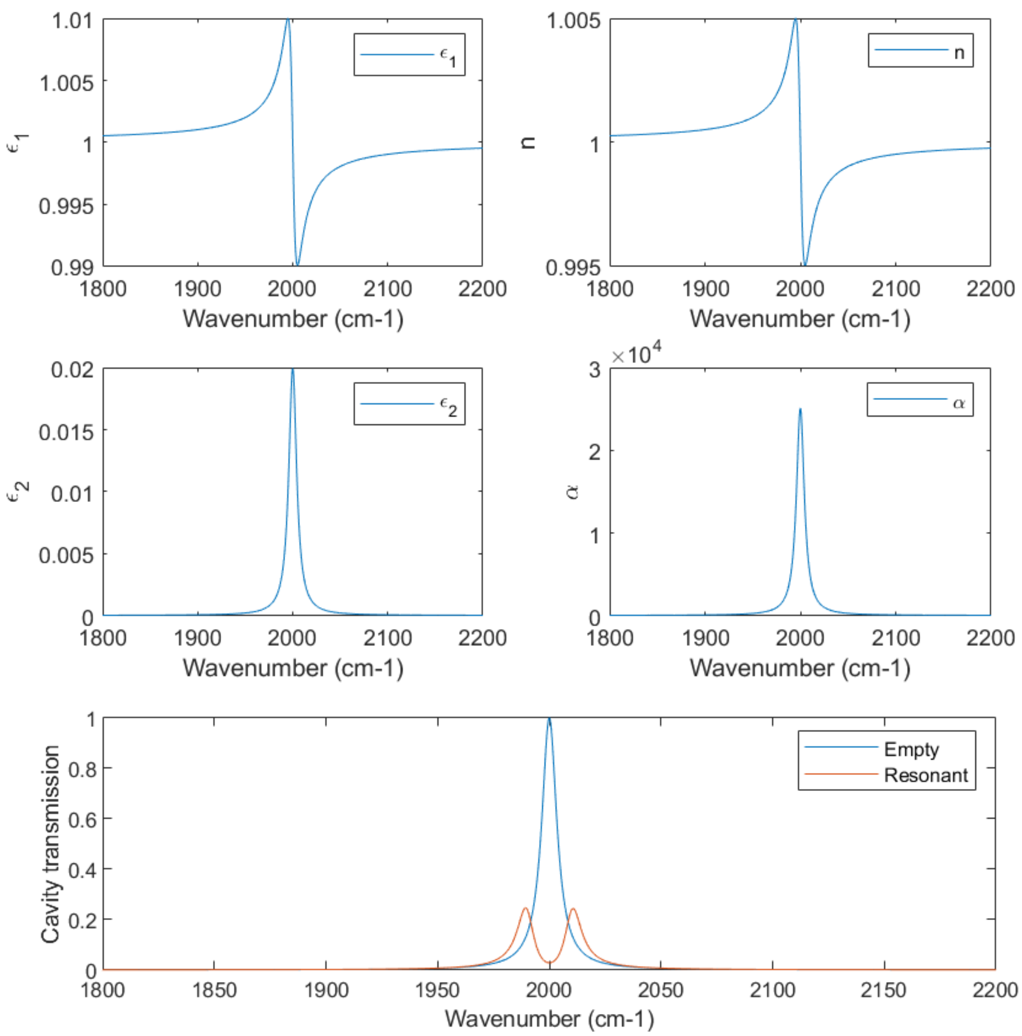

Figure 4

Fig. 5 repeats the same simulation carried out with various values of A_i:

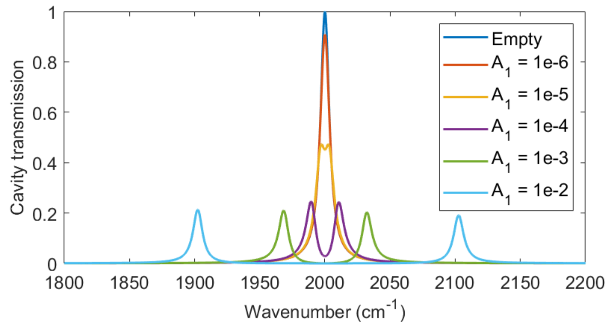

Figure 5

And Fig. 6 show the same simulation with $A_i = 1 \times 10^{-4}$ with the Lorentz oscillator frequency systematically detuned from the cavity resonance, resulting in the appearance of asymmetric transmission peaks.

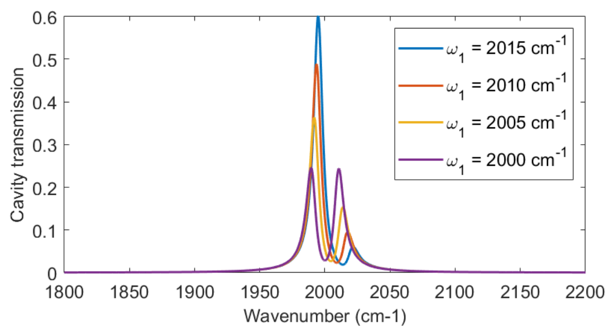

Figure 6

One can also simulate tilting the cavity by angle θ by rescaling our expression for the wavevector $\bar{k}$ in Eqn. 40 by $\cos(\theta)$, which causes the transmission peaks to move towards higher energies (shorter wavelengths) as θ increases. The somewhat non-intuitive reasoning for these frequency shifts are discussed further here and here. A simulated dispersion spectrum for a tilt-tuned planar Fabry-Pérot cavity is shown in Fig. 7.

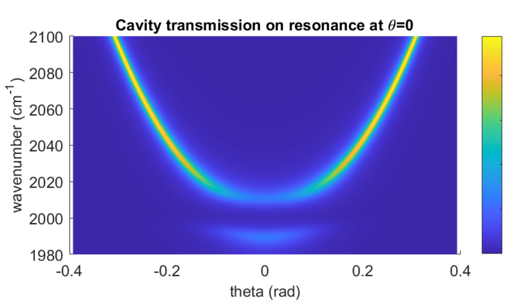

Figure 7

10 The transfer matrix method for multilayer devices

What we have worked through thus far is valid for treating the cavity transmission spectrum of an ideal two-mirror Fabry-Pérot cavity containing one slab of intracavity material with refractive index $\bar{n}$. In reality, we will often be dealing with cavities containing many thin film layers, e.g. dielectric Bragg mirrors or more intricate multilayer cavity samples. As we worked through for the Fabry-Pérot example, treating the reflected, intracavity, and transmitted cavity fields requires the consideration of an infinite series of interfering reflections. A multilayer stack will have several internal interfaces between media with different indices of refraction, each producing a reflected and transmitted beam when lights strikes the interface. There are therefore going to be nested infinite series of interfering beams due to all these interfaces. This seems like a difficult problem! The "transfer matrix" (TM) method is an elegant way to perform the analyses we need while largely bypassing explicit treatment of these infinite series.

In brief, the TM method tells us that if we know the input field into the multilayer stack, we can use Maxwell's equations to determine how the total field propagates through the medium and across each interface, based on simple continuity conditions for the field. We won't go through the detailed derivation of these methods here, and will just summarize the important results.

Let's assume that we have a plane wave of light incident along the x axis from the left which impinges on a multilayer structure composed of m layers. Each layer is composed of a homogeneous, isotropic material, and has label j, a thickness d_j, a wavelength-dependent complex index of refraction $\bar{n}_j = n_j + i\kappa_j$, and a dielectric constant $\epsilon_j = \epsilon_{1,j} - i\epsilon_{2,j} = \bar{n}_j^2$. To the left of the stack (region $j = 0$) and to the right of the stack (region $j = m + 1$) we assume a transparent ambient material (e.g. air or glass substrate). At any point inside layer j, the optical electric field is composed of complex exponential components counter-propagating along the x-axis, $E_j^+(x)$ and $E_j^-(x)$. The sum of these $E_j^+(x)$ and $E_j^-(x)$ components represent the total field inside layer j. In reality, these net fields arise from the infinite series of reflected and transmitted beams within the device.

The incoming and reflected fields in region 0 to the left of the stack, $E_0^+(x)$ and $E_0^-(x)$, are related to the fields in region $m+1$ to the right of the stack, $E_{m+1}^+(x)$ and $E_{m+1}^-(x)$ by a 2×2 scattering matrix S:

$$\begin{bmatrix} E_0^+ \\ E_0^- \end{bmatrix} = S \begin{bmatrix} E_{m+1}^+ \\ E_{m+1}^- \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix} \cdot \begin{bmatrix} E_{m+1}^+ \\ E_{m+1}^- \end{bmatrix} \tag{76}$$

As we are only considering light impinging from the left, we assume $E_{m+1}^- = 0$, and therefore

$$E_0^+ = S_{11} \cdot E_{m+1}^+ \tag{77}$$

$$E_0^- = S_{21} \cdot E_{m+1}^+ \tag{78}$$

We are ultimately in search of the reflection and transmission coefficients for the entire layered structure, which we can express in terms of elements of the scattering matrix:

$$r = \frac{E_0^-}{E_0^+} = \frac{S_{21}}{S_{11}} \qquad R = |r|^2 \tag{79}$$

$$t = \frac{E_{m+1}^+}{E_0^+} = \frac{1}{S_{11}} \qquad T = |t|^2 \tag{80}$$

So how do we construct this scattering matrix? We build it up in pieces by relating the fields $E_j^{\pm}$ and $E_k^{\pm}$ across each jk interface with an interface matrix I_{jk}, and by treating the propagation of the field through layer j by the layer matrix L_j.

First, we will use an interface matrix I_{jk} that encodes the Fresnel coefficients for reflection and transmission of the total field at an interface. These Fresnel coefficients are derived in MacLeod Ch. 2 for those interested.

$$\begin{bmatrix} E_j^+ \\ E_j^- \end{bmatrix} = I_{jk} \begin{bmatrix} E_k^+ \\ E_k^- \end{bmatrix} \tag{81}$$

$$I_{jk} = \frac{1}{t_j k} \begin{bmatrix} 1 & r_{jk} \\ r_{jk} & 1 \end{bmatrix} \tag{82}$$

$$r_{jk} = \begin{cases} \dfrac{q_j - q_k}{q_j + q_k} & s\text{-polarized} \\ \\ \dfrac{\bar{n}_k^2 q_j - \bar{n}_j^2 q_k}{\bar{n}_k^2 q_j + \bar{n}_j^2 q_k} & p\text{-polarized} \end{cases} \tag{83}$$

$$t_{jk} = \begin{cases} \dfrac{2q_j}{q_j + q_k} & s\text{-polarized} \\ \\ \dfrac{2\bar{n}_j \bar{n}_k q_j}{\bar{n}_k^2 q_j + \bar{n}_j^2 q_k} & p\text{-polarized} \end{cases} \tag{84}$$

$$q_j = \bar{n}_j \cos\phi_j = \left[\bar{n}_j^2 - n_0^2 \sin^2\phi_0\right]^{1/2} \tag{85}$$

Here we account for the light traveling through the ambient material with refractive index n_0 before hitting the stack at an angle of incidence ϕ_0, with either s or p polarization. ϕ_j represents the angle of refraction into layer j.

For the simplest case of normal incidence,

$$r_{jk} = \frac{\bar{n}_j - \bar{n}_k}{\bar{n}_j + \bar{n}_k} \tag{86}$$

$$t_{jk} = \frac{2\bar{n}_j}{\bar{n}_j + \bar{n}_k} \tag{87}$$

The layer matrix L_j encodes the absorption and change in phase of the field of light as it propagates across layer j, where again d_j is the thickness of layer j:

$$L_j = \begin{bmatrix} e^{-i\xi_j d_j} & 0 \\ 0 & e^{i\xi_j d_j} \end{bmatrix} \tag{88}$$

$$\xi_j = \frac{2\pi}{\lambda} q_j \tag{89}$$

The total scattering matrix is then built up as a product of these interface and layer matrices

$$S = \left(\prod_{j=1}^{m} I_{(j-1)j} L_j \right) \cdot I_{m(m+1)} \tag{90}$$

It is therefore quite straightforward to calculate the transmission spectrum of an arbitrary multilayer stack of thin films by calculating S according to Eqn. 90 and plugging the relevant matrix element into Eqn. 80.

11 Calculating the intracavity field with TM

We can also use the scattering matrix S to calculate the amplitude of the internal electric field within each layer as a function of distance into the stack. We can break S into components:

$$S = S'_j L_j S''_j \tag{91}$$

where

$$S'_j = \begin{bmatrix} S'_{j11} & S'_{j12} \\ S'_{j21} & S'_{j22} \end{bmatrix} = \left(\prod_{k=1}^{j-1} I_{(k-1)k} L_k \right) I_{(j-1)j} \tag{92}$$

$$S''_j = \begin{bmatrix} S''_{j11} & S''_{j12} \\ S''_{j21} & S''_{j22} \end{bmatrix} = \left(\prod_{k=j+1}^{m} I_{(k-1)k} L_k \right) I_{m(m+1)} \tag{93}$$

$$\tag{94}$$

and

$$\begin{bmatrix} E_0^+ \\ E_0^- \end{bmatrix} = S'_j \begin{bmatrix} E_j'^+ \\ E_j'^- \end{bmatrix} \tag{95}$$

$$\begin{bmatrix} E_j''^+ \\ E_j''^- \end{bmatrix} = S''_j \begin{bmatrix} E_{m+1}^+ \\ E_{m-1}^- \end{bmatrix} \tag{96}$$

Here $E_j'^{\pm}$ and $E_j''^{\pm}$ encode the electric field amplitudes inside layer j at the $(j-1)j$ and $j(j+1)$ interfaces.

Just like we defined complex reflection and transmission coefficients r and t for the total scattering matrix S, we can define partial coefficients

$$r'_j \equiv \frac{S'_{j21}}{S'_{j11}} \qquad\qquad t'_j \equiv \frac{1}{S'_{j11}} \tag{97}$$

$$r''_j \equiv \frac{S''_{j21}}{S''_{j11}} \qquad\qquad t''_j \equiv \frac{1}{S''_{j11}} \tag{98}$$

The physical meaning of these partial coefficients is a bit abstract. r'_j represents the fractional amplitude of light that would be reflected from the stack on the $j = 0$ side, considering only interfering contributions up to reflections from the $(j-1)j$ interface. t'_j represents the fractional

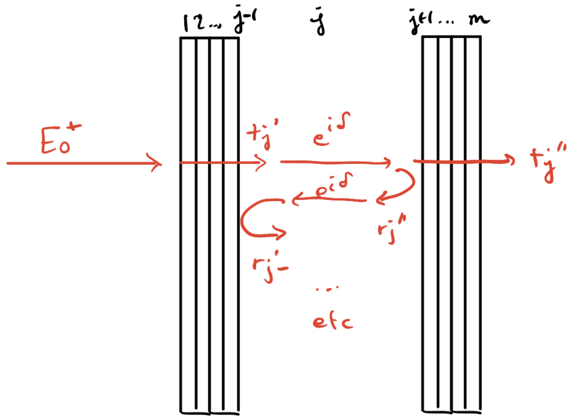

Figure 8

amplitude of light that would be transmitted into layer j from the left, ignoring any interference with light entering layer j from the $j(j+1)$ side.

We can use these partial coefficients to write down an expression for the internal field propagating towards the right in layer j at the $(j-1)j$ boundary. We will consider the infinite series of reflections between the two halves of the stack, as sketched in Fig. 8.

$$E_j^+ = t_j' \cdot E_0^+ \cdot \left[e^{i\xi_j d_j} \cdot r_j'' \cdot r_{-j}' \cdot e^{i\xi_j d_j} + e^{i\xi_j d_j} \cdot r_j'' \cdot e^{i\xi_j d_j} \cdot r_{-j}' \cdot e^{i\xi_j d_j} \cdot r_j'' \cdot e^{i\xi_j d_j} \cdot r_{-j}' + \dots \right] \tag{99}$$

$$= t_j' E_0^+ \sum_{n=1}^{\infty} \left(r_{-j}' r_j'' e^{2i\xi_j d_j} \right)^n \tag{100}$$

$$= E_0^+ \frac{t_j'}{1 - r_{-j}' r_j'' e^{2i\xi_j d_j}} \tag{101}$$

where in the last step we have used the expression for infinite geometric series. $r_{j-}' \equiv -r_j'$, to account for the change in sign for reflections on the internal side of the $(j-1)j$ interface. $e^{i\xi_j d_j}$ represents the phase accrued by one pass through layer j. We can therefore define

$$t_j^+ \equiv \frac{E_j^+}{E_0^+} = \frac{t_j'}{1 - r_{-j}' r_j'' e^{2i\xi_j d_j}} \tag{102}$$

Note that Eqn. 102 is defined at the $(j-1)j$ interface. We can re-express it as a function of distance $x < d_j$ into layer j from the $(j-1)j$ interface by noting that this extra propagation just adds an additional phase factor of $e^{i\xi_j x}$. Therefore:

$$t_j^+(x) = \frac{E_j^+(x)}{E_0^+} = \frac{t_j' e^{i\xi_j x}}{1 - r_{-j}' r_j'' e^{2i\xi_j d_j}} \tag{103}$$

We can follow the same process to derive the field amplitude propagating to the left in layer j at the $(j-1)j$ interface, E_j^-, by noting that E_j^- derives directly from E_j^+, but has undergone two additional complete passes through layer j and one additional reflection off the $j(j+1)$ interface:

$$E_j^- = r_j'' \cdot e^{i2\xi_j d_j} \cdot E_j^+ \tag{104}$$

$$\rightarrow t_j^- \equiv \frac{E_j^-}{E_0^+} = t_j^+ \cdot r_j'' \cdot e^{i2\xi_j d_j} \tag{105}$$

And again, to express t_j^- as a function of propagation distance x into layer j, we just correct the accrued phase by a factor of $e^{-i\xi_j x}$, to reflect the fact that last trip through layer j stopped short of the $(j-1)j$ interface:

$$t_j^-(x) = \frac{E_j^-(x)}{E_0^+} = t_j^+ r_j'' \cdot e^{i2\xi_j d_j} \cdot e^{-i\xi_j x} \tag{106}$$

$$= t_j^+ r_j'' \cdot e^{i\xi_j(2d_j - x)} \tag{107}$$

We are now finally in a position to write down the total electric field in an arbitrary layer j:

$$E_j(x) = E_j^+(x) + E_j^-(x) \tag{108}$$

$$= \left[t_j^+(x) + t_j^-(x)\right] E_0^+ \tag{109}$$

$$= t_j^+ \left[e^{i\xi_j x} + r_j'' e^{i\xi_j(2d_j - x)}\right] E_0^+ \tag{110}$$

and we can rewrite this in terms of the partial transfer matrix elements from Eqns. 97 and 98 as:

$$E_j(x) = \frac{t_j'}{1 - r_{-j}' r_j'' e^{2i\xi_j d_j}} \cdot \left[e^{i\xi_j x} + r_j'' e^{i\xi_j(2d_j - x)}\right] E_0^+ \tag{111}$$

$$= \frac{1}{S_{j11}'} \cdot \frac{1}{1 + \frac{S_{j21}'}{S_{j11}'} \frac{S_{j21}''}{S_{j11}''} \cdot e^{2i\xi_j d_j}} \cdot \left[e^{i\xi_j x} + \frac{S_{j21}''}{S_{j11}''} e^{i\xi_j(2d_j - x)}\right] E_0^+ \tag{112}$$

$$= \frac{e^{i\xi_j x} + \frac{S_{j21}''}{S_{j11}''} \cdot e^{i\xi_j(2d_j - x)}}{S_{j11}' + S_{j21}' \cdot \frac{S_{j21}''}{S_{j11}''} \cdot e^{2i\xi_j d_j}} \cdot E_0^+ \cdot \frac{S_{j11}'' e^{-i\xi_j d_j}}{S_{j11}'' e^{-i\xi_j d_j}} \tag{113}$$

$$= \boxed{\frac{S_{j11}'' \cdot e^{-i\xi_j(d_j - x)} + S_{j21}'' \cdot e^{i\xi_j(d_j - x)}}{S_{j11}' S_{j11}'' \cdot e^{-i\xi_j d_j} + S_{j21}' S_{j21}'' \cdot e^{i\xi_j d_j}} \cdot E_0^+} \tag{114}$$

This expression therefore allows us to calculate the structure of electromagnetic modes propagating in the cavity, using only matrix elements of the relevant partial scattering matrices. If we calculate $E_j(x)$ throughout the stack for frequencies of light that are resonant with the cavity, we can do various useful things, including simulating the nodal structure of these resonant cavity modes and their penetration depth into dielectric mirrors.