

Random Laser Simulation - COMSOL Multiphysics

1. Part 1 - Fluorescence simulation

1.1 Diffusion equation in Helmholtz form (COMSOL pre-set physics setting)

The problem is divided in two steps: the excitation of the fluorescent material by the absorption of the energy of the incident photons (Equation (1)), and the spontaneous emission when it is returning to the ground state (Equation (2)).

The diffusion equation is used to model propagating light in a medium and find its fluence value.

Equation (1):

$$\varphi_x(r) - \nabla D_x(r) \nabla \varphi_x(r) + \mu_{ax}(r) \varphi_x(r) = S_x(r) \quad (1)$$

$\varphi_x(r)$ is the fluence rate of the propagating light in a medium, $S_x(r)$ is the source term, $D_x(r)$ is the diffusion coefficient and $\mu_{ax}(r)$ is the absorption coefficient.

The calculated result of $\varphi_x(r)$ in the first step is used as initial condition (excitation light, source term) to the following step that describes the spontaneous emission defined by Equation (2)

Equation (2):

$$\varphi_m(r) - \nabla D_m(r) \nabla \varphi_m(r) + \mu_{am}(r) \varphi_m(r) = \mu_{af} \gamma_m \varphi_x(r) \quad (2)$$

where $\varphi_m(r)$ is the fluence rate of the emission light in a medium, μ_{af} is the absorption coefficient at the excitation wavelength, $\varphi_x(r)$ is the excitation light fluence rate obtained after solving the equation (1) and γ_m is the fluorescence yield fraction. In the Equation (2) the diffusion and absorption coefficients are defined according to the material chosen to be the active medium and its source term is the fluence rate for the fluorescence emission (which will be later used as a parameter of the electromagnetic field that will be scattered by the nanoparticles to achieve the non-coherent feedback already studied experimentally)

In COMSOL Multiphysics there is a pre-set physics setting for Helmholtz equations. To make things easier we use this preset and determine the Diffusion equations (1 and 2) in their Helmholtz forms (3-5):

$$\nabla(-c \nabla u) + au = f \quad (3)$$

Where $u = \varphi_x$, $c = D$, $a = \mu_{ax}$ and $f = S$.

Equations 1 and 2 can be written in their Helmholtz form as

$$\nabla(-D \nabla \varphi_{ex}) + \mu_a \varphi_{ex} = S \quad (4)$$

and

$$\nabla(-D^i \nabla \varphi_{em}^i) + \mu_a^i \varphi_{em}^i = \mu_a^{\text{ex}} Y^i \varphi_{ex} \quad (5)$$

respectively.

The subscript i represents the emission wavelength, em and ex are related to the emission and excitation light sources.

1.2 Definitions / Parameters (summary)

From equations 1 and 2:

S_x – source term (incident light intensity)

μ_{ax} / μ_{af} – absorption coefficient of the medium; absorption coefficient of the fluorescent material at the excitation wavelength

$D_x = \frac{1}{3}(\mu_{ax} + \mu_s^1)$ – diffusion coefficient

$\mu_s^1 = SA \left(\frac{\lambda}{1000} \right)^{-sp}$ – reduced scattering

γ_m – fluorescence yield fraction (quantum yield, wavelength dependent – later used for the parametric sweep)

φ_x / φ_m – fluence rate of excitation light (incident propagating light); fluence rate of the spontaneous fluorescence emission (or $u, u2$ – as determined in COMSOL)

1.3 Geometry

The block of air surrounding the experiment, in which the first Helmholtz equation is solved to find the fluence of the excitation light, has (0.1m x 0.1m x 0.1m) dimensions. The fluorescent material dimensions are determined to match the experimental setup where the fluorescent dye is excited inside a 4mL (10mm path length, 40mm height) micro quartz cuvette cell – (0.01m x 0.01m x 0.04m).

The first Helmholtz equation light source is defined by a point source positioned one scattering length inside the boundary. This is the source option that better simulate the excitation light provided from the 2.5mm beam diameter Nd:YAG laser. Point is set at (-0.05x0x0), but it can also be set below the YZ cuvette center to better simulate the real experiment (where the excitation beam hits the cuvette a little bit under its center).

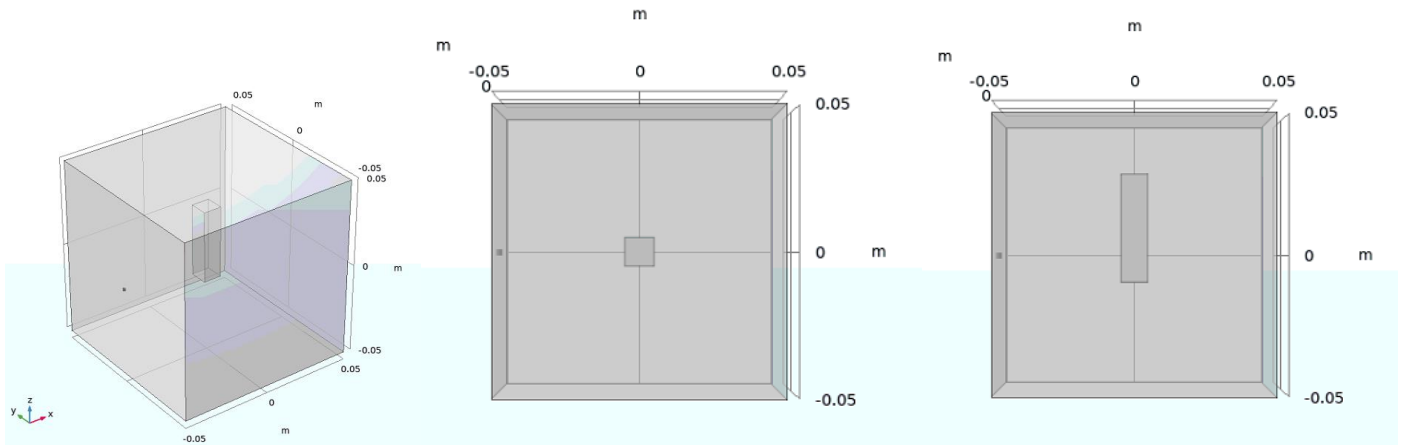


Figure 1

1.4 Solution Domains & Boundary Conditions

Both equations (1) and (2) are set to be solved in all domains (domain 1 and 2, corresponding to the block of air and the cuvette, respectively), since light entering through the point source is propagating through all the geometry. Flux/Source for Helmholtz equation (1) is defined as 0 and there is a second source (point source) defined at our third domain (which corresponds to the point previously set). The source for Helmholtz equation (2) is only solved for the cuvette (domain 2) and defined as $\mu_{af} \gamma_m \phi_x$ (the result from eq. (1) multiplied by the optical parameters to define the absorbed portion of the excitation light that propagates through air and hits the fluorescent material).

The dependent variables from equations (1) and (2) are defined as u and $u2$, respectively.

For the boundary conditions there is a pre-set Zero Flux 1 condition that states the fluence rate is zero at the selected boundaries. This is not correct since we should be also simulating reflections and other effects induced by the refractive index mismatch. A Robin-type boundary condition is used instead to model a realistic problem. After setting the correct parameters it looks like we have two boundary conditions on the left menu (both Zero Flux and Flux/Source), however if we check closely all the boundary selections for the Zero Flux are overridden (this means the other boundary condition is being assigned).

1.5 Mesh

The tetrahedral mesh chosen was set to be finer at the air-cuvette/dye boundaries and point source entry, with basic optimization levels (avoiding only inverted curved elements). Bigger maximum element sizes were set everywhere else (fine/normal, according to the pre-set COMSOL settings).

If we set the mesh too coarse the diffusion equation numerical solution is most likely wrong due to discretization errors; setting the mesh to be too fine everywhere will make the computation take too long (specially later with more studies and the parametric sweep function).

Mesh Parameters	General Mesh	Free Tetrahedral 1 (point source)	Free Tetrahedral 2 (cuvette)
Maximum Element Size	0.008	0.0055	0.0055
Minimum Element Size	0.001	4E-4	4E-4
ME Growth rate	1.45	1.4	1.4
Curvature Factor	0.5	0.4	0.4
Resolution of NR	0.6	0.7	0.7

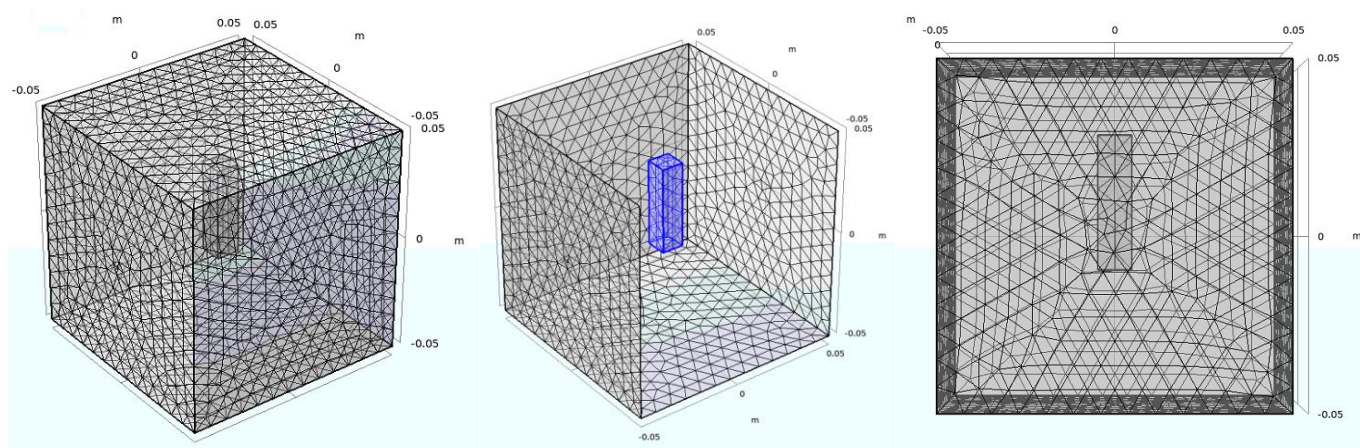


Figure 2

1.6 Results

Our final solution for this first simulation is contained in the variables u and u_2 , as defined before.

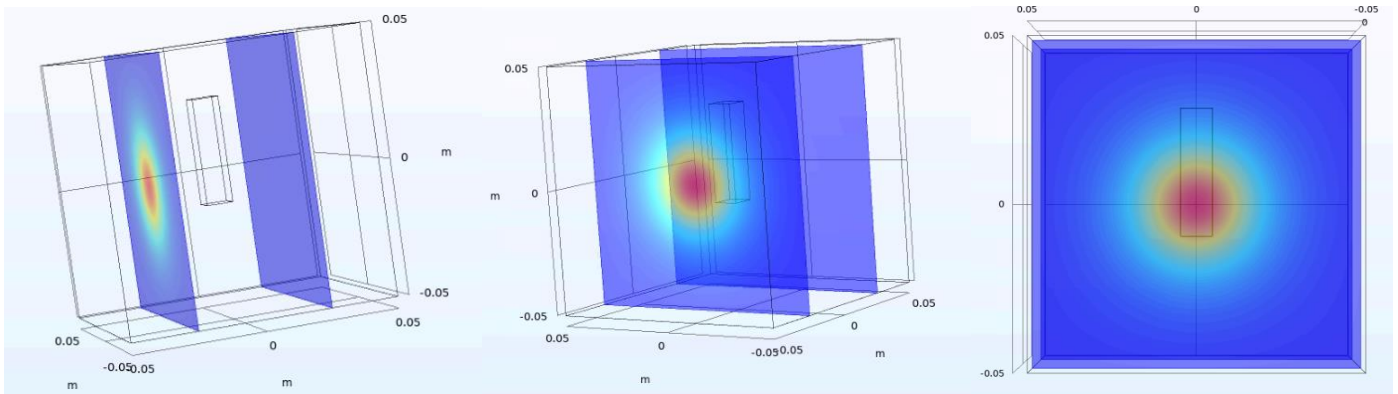


Figure 3

Fig. 3 shows the solution for the first diffusion equation (φ_x, u), which corresponds to the fluence rate of the incident light. As stated before, this is the excitation light that will be partially absorbed by the sample and used as an initial condition for the second diffusion equation. The excitation light does not go through the sample (as can be seen in the multi-slice type graph above) since the block of fluorescent dye is considered to be solid.

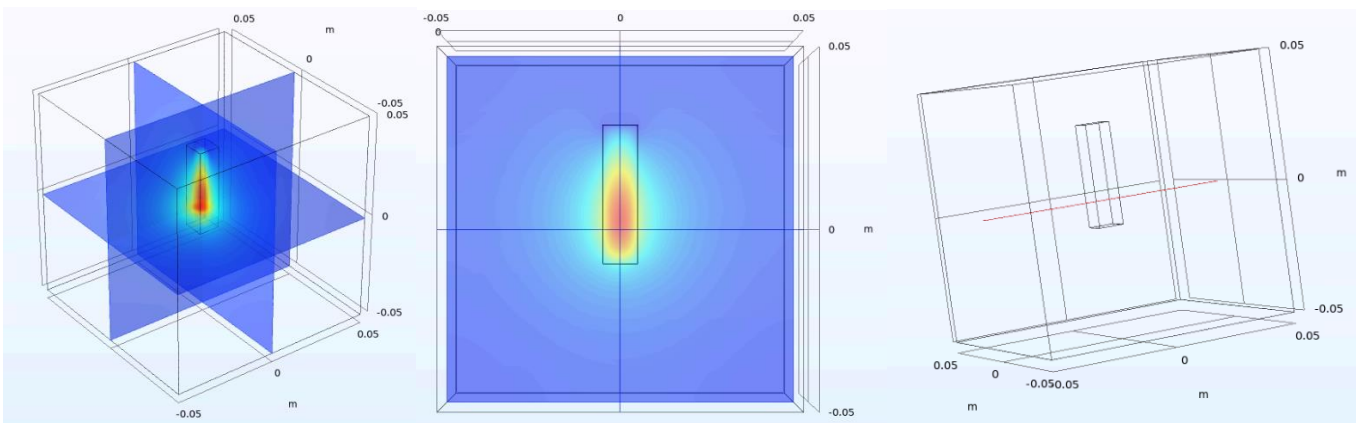


Figure 4

The solution to the second diffusion equation (φ_m, u_2) can be seen in the figure above. It corresponds to the fluence of the fluorescence light emitted from the sample. Fig. 5 (below) is a 2D line plot that shows the fluence rate as a function of the x-axis of the system (represented by the red segment in figure 4c). As expected, the fluorescence is symmetrical to the sample along its axis and peaks at its center.

These results are independent of the excitation and emission light wavelengths, which are only being controlled externally with the parameters. Next steps will show how to create a wavelength dependency to make the simulation more realistic, with Rhodamine (the chosen fluorescent dye) having its maximum quantum yield at $\sim 532\text{nm}$ and maximum emission at $\sim 560\text{nm}$.

2 Wavelength Dependence – parametric sweep

To create a wavelength dependence in our system for both excitation and emission spectra we need to define a parameter as a function of the wavelength. We chose the fluorescence yield fraction (γ_m) since it naturally depends on the wavelength and multiplies the fluence rate of the fluorescence light in the right side of the equation, making it easier to model our material emission spectra. Remember that μ_{af} is the absorption coefficient at the excitation wavelength and will now be a constant value since we are only exciting our fluorescent dye with 532nm light.

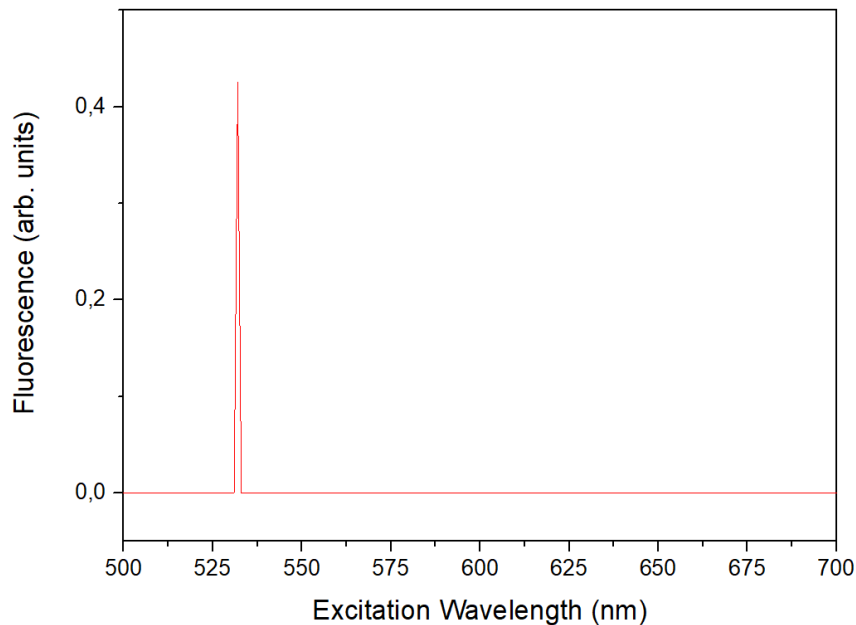
$$\varphi_m(r) - \nabla D_m(r) \nabla \varphi_m(r) + \mu_{am}(r) \varphi_m(r) = \mu_{af} \gamma_m \varphi_x(r)$$

Now it is necessary to solve several variations of our model to find its properties along a range for excitation and emission light. Instead of manually changing the parameter value and re-solving each time, we perform a parametric sweep that allows us to change the parameter through a specified range. Implementing the sweep makes our simulation take a lot longer since it will solve both Helmholtz equations for each new parameter value in the whole geometry, and in order to have a precise fluorescence intensity as a function of the wavelength graph we need to vary the wavelength in small steps.

Our initial wavelength value was set to be 500[nm] for both studies, the step was set to 2[nm] and the final wavelength value is 700[nm].

2.1 Defining excitation wavelength

It is known that Rhodamine6G is a fluorescent compound with an excitation peak at 532nm. To model the excitation light response, we manually determined values for a δ -function that equals zero everywhere except $\lambda=532\text{nm}$. Multiplying the second diffusion equation right side by this function we guarantee that we only have a positive non-zero fluence rate for the fluorescence light when the excitation light is at 532nm wavelength. Below we can see the graph of the δ -function.



The numerical value of our function was defined with OriginLab v9.0. The iteration algorithm chosen was Lavenberg Marquardt, which determined our function as an amplified gaussian with infimal standard error for every numerical value.

The function and its numerical values are shown below:

$$y = y_0 + A e^{-0.5 \left(\frac{x-\lambda}{w} \right)^2}$$

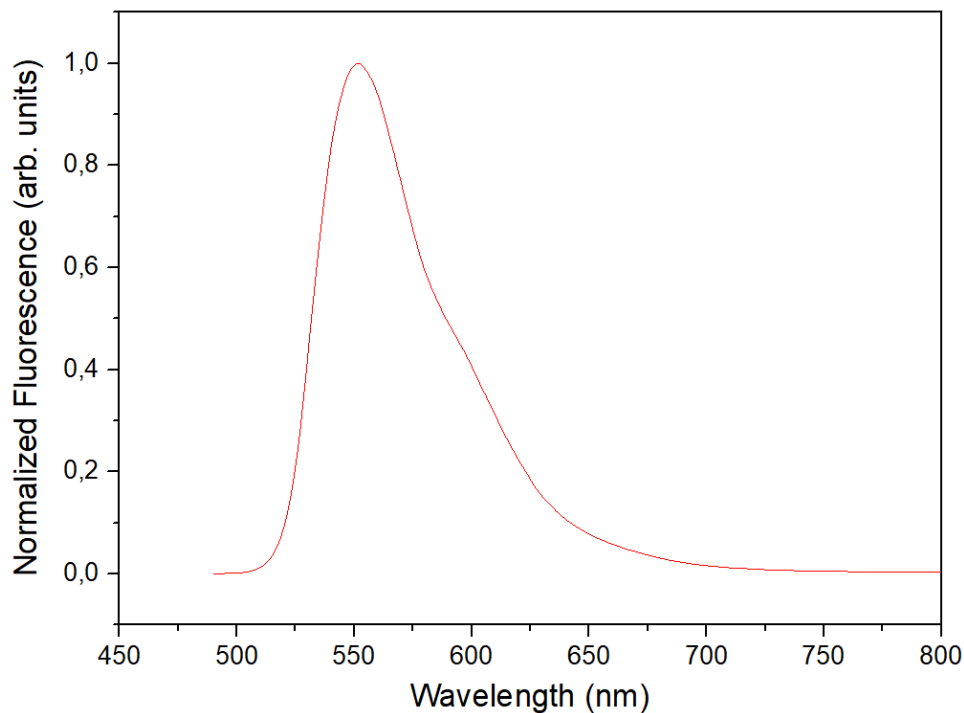
y_0	1
x	532
w	0.10391
A	10000
FWHM	0.24469

With its values determined it becomes clear that multiplying our fluence rate in the second diffusion equation by this function and setting our parametric sweep from 500 to 700 nanometers only gives us non-zero values for the fluorescence at $\lambda=532$ wavelength. Obviously Rhodamine6G also shows lower emission values for excitation wavelengths around 532nm but as a first test of the parametric sweep tool this serves very well. Our final model only depends on the emission wavelength determination which is very well defined and will be show in the next section.

2.2 Defining emission wavelength

Our simulation already finds the amount of light generated for determined excitation energy; this step will only model the fluorescence as a function of the wavelength. For that we also need to make sure that $0 < \gamma_m \mu_{af} < 1$. This way we are not generating more light by multiplying the right side of the equation by a positive number – we are only modeling the fluorescence spectra for some determined fluorescent material.

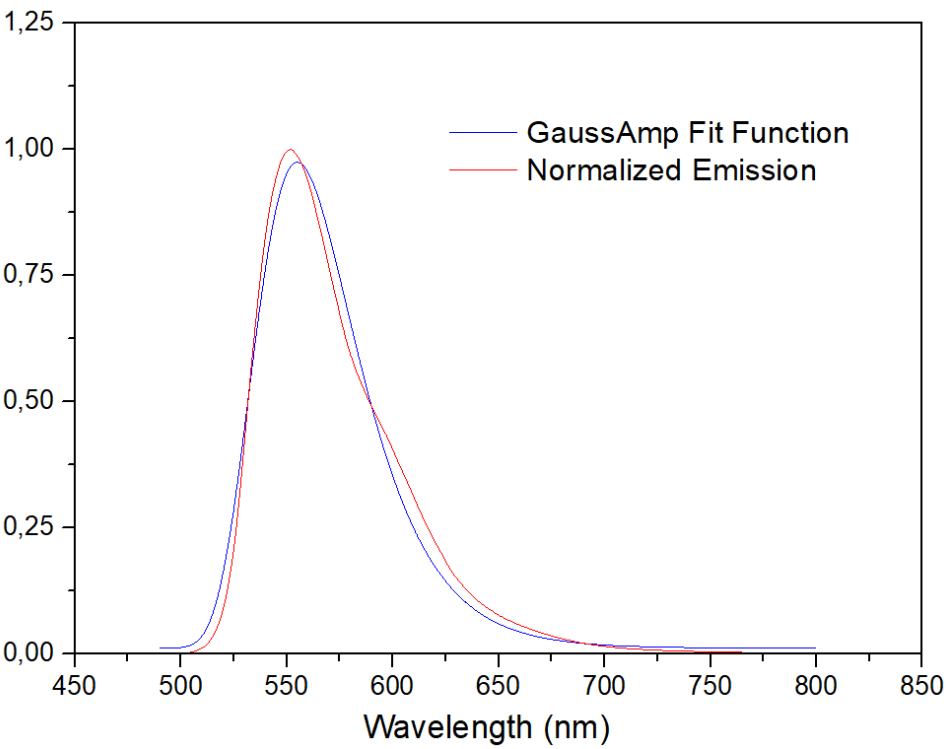
Below is the plot of the Rhodamine6G normalized fluorescence as a function of the wavelength, when being excited by 532nm light from a Nd:YAG laser system. As before, we will find a function of the wavelength that corresponds to this curve and multiply the right side of the equation by it.



The numerical value of our function was once again defined using Origin. The iteration algorithm chosen was Lavenberg Marquardt, which determined our function as a gaussian with the following form, numerical values and plot:

$$y = y_0 + A e^{-0.5 \left(\frac{x-\lambda}{w}\right)^2}$$

y_0	0.02752
x	562
w	25.32906
A	0.91192
FWHM	59.64



2.2.1 Results

