Simulation of Reverse Saturable Absorption

N. K. Bambha[1]

1. US Army Research Laboratory, Adelphi, MD, USA

Introduction: Optical limiting materials have been investigated for many years to protect eyes and sensors from high power lasers. These materials must be able to absorb or scatter high intensity laser radiation, while transmitting low intensity light. Most materials exhibit saturable absorption, which means that they exhibit strongest absorption for low intensities, with decreasing absorption as the intensity increases. The cause of this decrease in absorption is the depletion of the ground state population at higher intensities. Several materials, however, display reverse saturable absorption (RSA) [1], which arises when the absorption cross section of an excited state is greater than the absorption cross section of the ground state. RSA has been observed in a number of organic-metallic molecules, including the fullerene C_60.

•Computational Methods: The optical process is modeled with simple rate equations coupled to a propagation equation for the optical intensity. The equations are as follows, with I representing the optical intensity, Ni representing the density of states in each level, hv the photon energy, sigma_ij the absorption cross section for level i to level j, and t_ij the decay time for a transition from level i to level j:

dI/dz = -(sigma_01*N0 + sigma_12*N1 + sigma_34*N3)*I
dN0/dt = -sigma_01*N1*I/hv + N1/t_10 + N3/t_30
dN1/dt = -sigma_12*N1I*/hv + N2/t_21 - N1*(1/t_10 + 1/t_13) + sigma_01*N0*I/hv
dN2/dt = sigma_12*N1*I/hv - N2/t_21
dN3/dt = -sigma_34*N3*I/hv + N4/t_43 + N1/t13 - N3/t30
dN4/dt = sigma_34*N3*I/hv - N4/t_43

•From these equations, it can be seen that the intensity I depends on the populations Ni, and the Ni depend on the intensity I, so this problem is well suited to a multiphysics modeling program. In addition, the absorption of laser light can cause heating, which is also well modeled in COMSOL. Heating will affect the propagation through a change in the index of refraction of the material.

•The equations above were simulated in COMSOL, using a laser pulse with a Gaussian temporal profile and Gaussian spatial profile. One common experiment that is performed to characterize nonlinear materials is the z-scan [2], in which the sample is translated through the focus, and measurements are made of the transmission. In this simulation, an open aperture experiment was modeled, in which all the light is captured after it travels through the sample. In the z-scan, the optical intensity is highest when the sample is at the focus of the laser, and in an RSA material the transmission will be the lowest at this point.

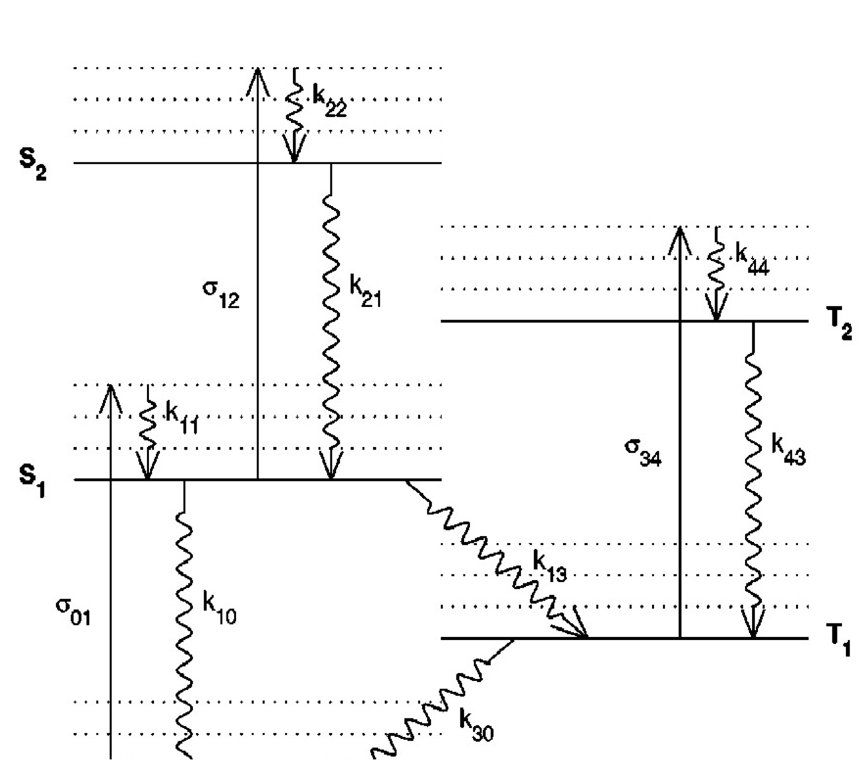

Figure 1. Energy level diagram

•Results: Figure 2 shows the population of the ground state as a function of radial position and propagation direction for the ground state N0 at time t=0 corresponding to the peak of the laser pulse for a z-scan position of z=0. Using a parameter sweep in COMSOL using the z-position of the sample as the parameter, the z-scan experiment was simulated. This is shown in Figure 3. This z-scan matches well with experimental data.

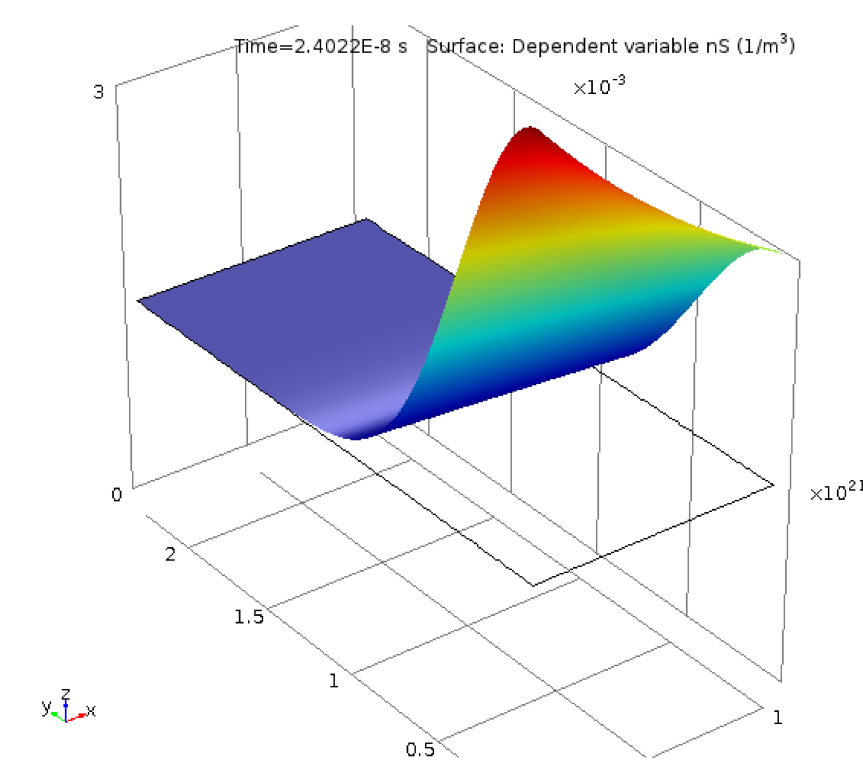

Figure 2. Ground state population as function of radial position and propagation direction.

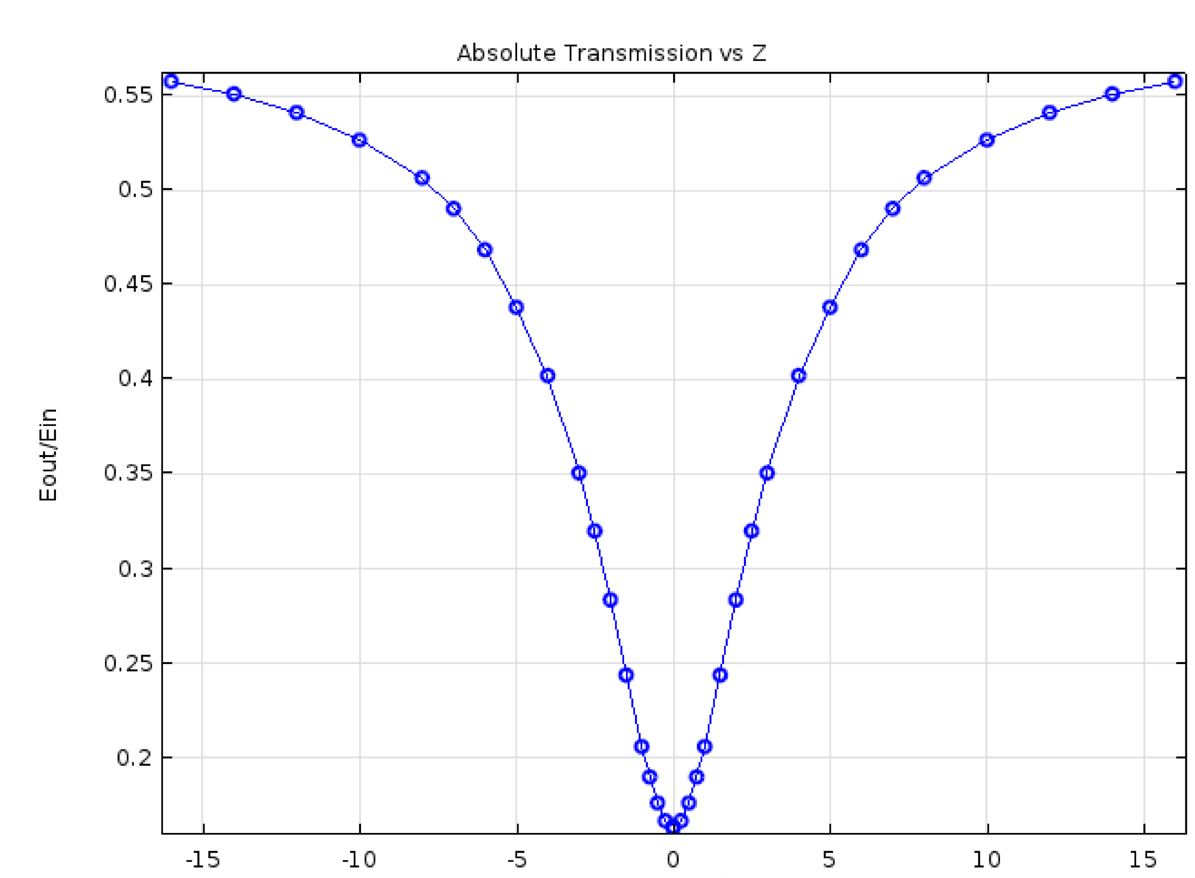

Figure 3. Z-scan simulation

Conclusions: COMSOL is well suited for modeling the dynamics of reverse saturable absorption. Absorption of light also cause heating, which can affect the refractive index. This effect could be coupled into a future model.

References:

1. Tutt, L. W. & Kost, A. Optical limiting performance of C60 and C70 solutions. Nature 356, 225-226 (1992).
2. M. Sheik-Bahae et al., "Sensitive measurement of optical nonlinearities using a single beam", IEEE J. Quantum Electron. 26 (4), 760, (1990).

Excerpt from the Proceedings of the 2016 COMSOL Conference in Boston