Simulation of Reverse Saturable Absorption

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Abstract

Optical limiting materials have been investigated for many years to protect eyes and sensors from high power lasers. Most materials exhibit saturable absorption, i.e., they exhibit strongest absorption for low intensities with decreasing absorption as the intensity increases. This is caused by 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.
This poster describes a simulation of RSA in a solution of C_60 molecules using the COMSOL Multiphysics® software. The optical processes in C_60 can be modeled through a five-level system. The energy states include three levels of the singlet state, coupled to two levels of an excited triplet state. Figure 1 displays this five-level energy model. Electronic states are shown by horizontal lines, while vibronic states are represented by horizontal lines. S0, S1, and S2 represent singlet states while T1 and T2 represent triplet states. Photon absorption is represented by vertical arrows, and decay processes are represented with curved arrows.
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 the COMSOL® software, 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.

Figure 2 shows the population of the excited state S1 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 the COMSOL® software 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.

Reference

[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).

Figures used in the abstract

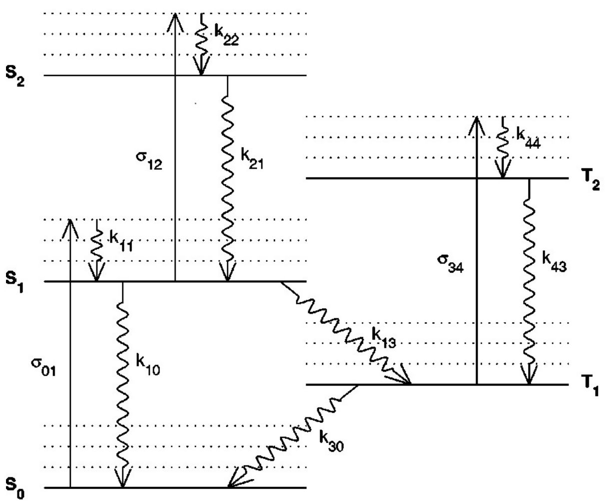

Figure 1: Five-level energy model

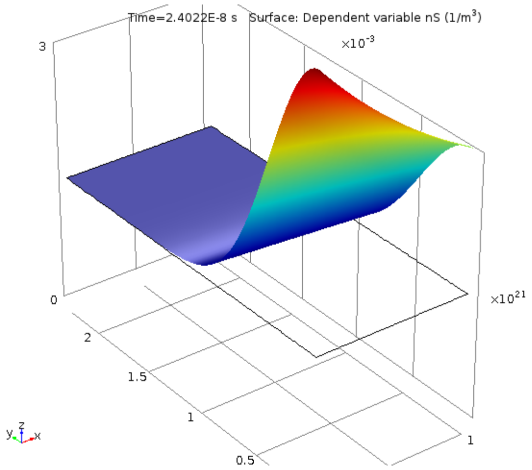

Figure 2: Population of first excited singlet state

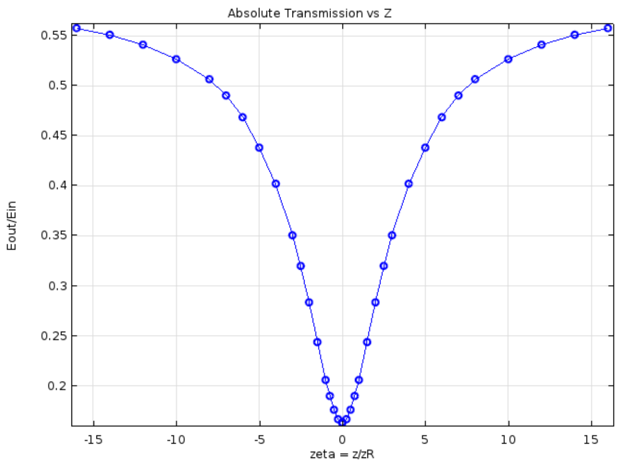

Figure 3: Z-scan simulation