Coherent excitation of vibrational levels using ultra short pulses

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INTRODUCTION

The purpose of this study was to develop a model of the coherent excitation of the first few vibrational modes in the electronic ground state of the molecule. The model will be used in combination with an optimization algorithm to optimize a population in a specific vibrational level. We used two approaches to do this, in the one model we used Von Neumann's equations and the other the Optical Bloch equations (OBE's). In this poster presentation the Optical Bloch model was used to do the simulations.

VON NEUMANN AND OPTICAL BLOCH EQUATIONS

For both the cases we have two Hamiltonians within the model we consider, the one describes the molecule unperturbed and the other one that is called the interaction Hamiltonian [1]

$$\hat{H}_E = \begin{bmatrix} E_1 & 0 & \cdots & 0 \\ 0 & E_2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & E_N \end{bmatrix}$$

$$\hat{H}_I = e\boldsymbol{D} \bullet \boldsymbol{E}_0 \Re[\varepsilon(t)]$$

An element of the interaction Hamiltonian looks like

$$I_{ab} = eE_0 X_{ab} \Re[\varepsilon(t)]$$

Here *e* is electron charge, E_0 is the amplitude of the electric field, X_{ab} is the dipole moment between level a and b, and then lastly ε(t) is

$$\varepsilon(t) = e^{-\alpha t^2} e^{-i\omega_L t}$$

Here α is given in term of the full width at half the maximum as

$$\alpha = \frac{4\ln 2}{FWHM^2}$$

And ω_L is the center frequency of the laser pulse.The Von Neumann's equations for the system can be written in the following manner

$$\frac{d\rho_{ab}}{dt} = \frac{-i}{\hbar}\sum_{l=1}^{N}\left(I_{al}\rho_{lb} - \rho_{al}I_{lb}\right) - i\rho_{ab}\omega_{a,b}$$

Now the Optical Bloch equations for the same system can be written as

$$\frac{d\rho_{ab}}{dt} = \frac{-i}{\hbar}\sum_{l=1}^{N}\left(\rho_{al}I_{bl}e^{-i\omega_{b,l}t} - \rho_{lb}I_{al}e^{i\omega_{a,l}t}\right)$$

In both cases we have that

$$\omega_{a,b} = \omega_a - \omega_b$$

Since both equations describe the same situation where a pulse interacts with a molecule we can choose whether we want to work with either Von Neumann's equations or the OBE's. In our work chose to work with the OBE's.

SIMULATIONS

The equations of a 9 level system were set up and solved for particular values. The molecule under investigation is not a real molecule and it served only as a test for the model. We can depict the molecule as follows

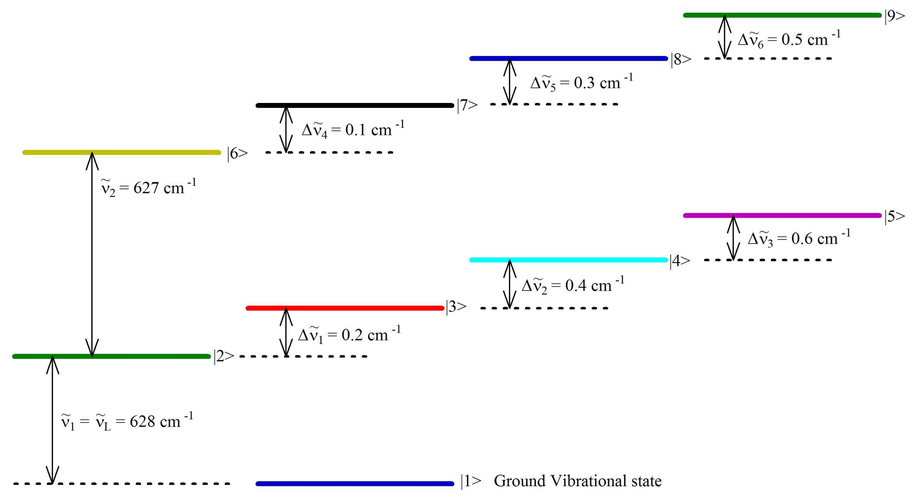

Figure 1: A vibrational mode of a hypothetical molecule with wave number differences indicated for the vibrational states.

A full width at half maximum (FWHM) of 10ps, fluence of the pulse (F_{pulse}) of 15 J/m^2 and center wavelength of 16μm was used for the simulation.

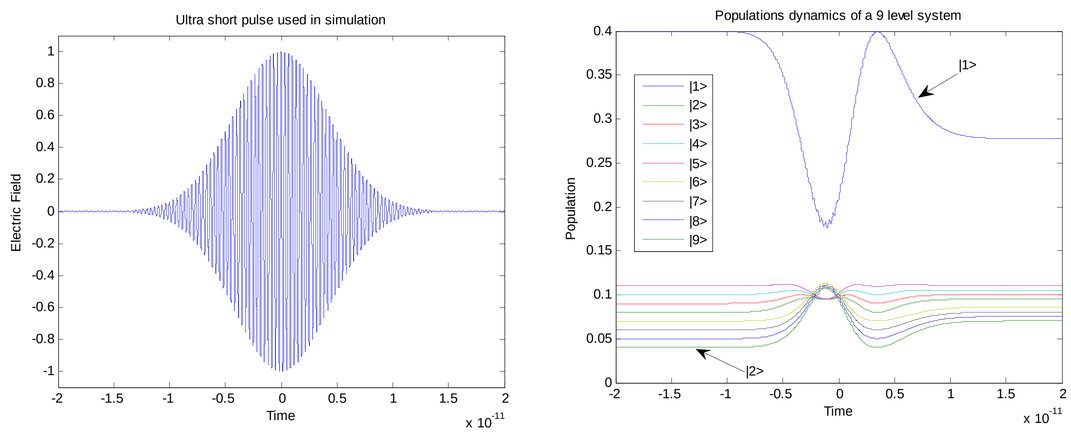

Figure 2: The USP (left) and the population dynamics of the nine level system (right).

CONCLUSION

In our work so far we have simulated how ultra short pulses interact with hypothetical molecules. Simulations of real molecules and the development of an optimization algorithm will follow in our work. The need of an optimization algorithm is to find, if it is possible pulse such that after the interaction with the molecules the population will only be in a certain vibrational state for instance in state |9> of figure 1. Typical optimization algorithm we may consider is a genetic algorithm.

BIBLIOGRAPHY

1. Loudon Rodney. 1983. The Quantum Theory of Light Second edn. Oxford University Press