Modeling of pump-probe spectra at high excitation intensity

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Polarization of an arbitrary order can be calculated by expanding density operator in powers of interaction with the excitation field [1]. The lowest order nonlinear optical signal that is generated in isotropic media is third order. At this order, the one exciton states and excited state energy transfer can be observed.

When excitation dynamics are followed at the lowest (third) power of interaction to excitation field, dependence on excitation intensity is often ignored. This dependence can be important as laser pulse intensity is one of parameters that is tuned for better signal-noise ratio. At high excitation intensity exciton-exciton annihilation (EEA) takes place [2].

Nonlinear exciton equations (NEE) [3] were used for calculations in this work. We expanded this equation system with terms that are higher than third order and added secular Redfield relaxation and phenomenological EEA terms. Then Pump-probe spectra were calculated at various excitation intensities and delay times with these equations (Fig. 1).

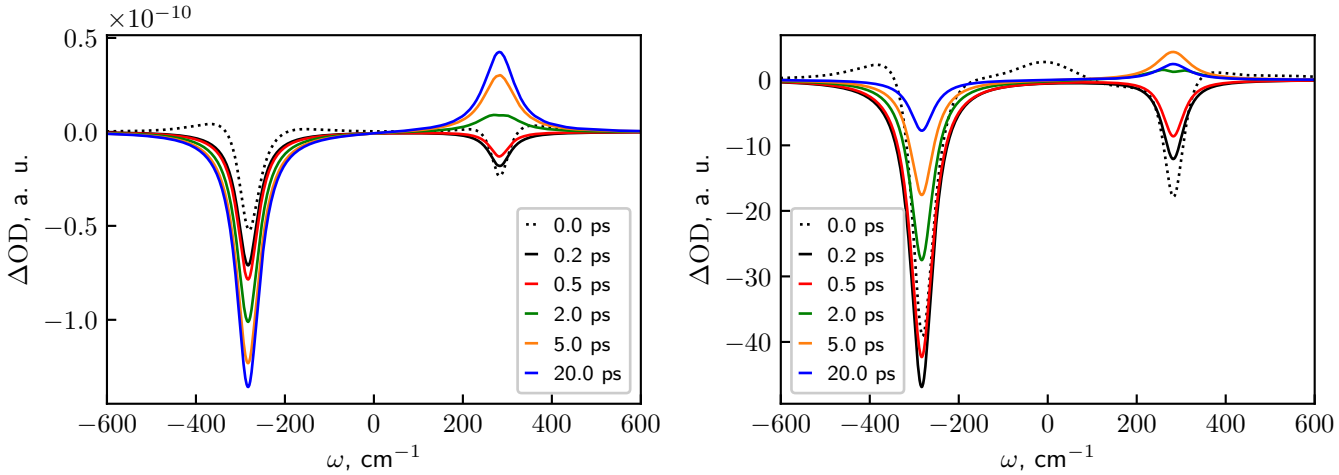

Fig. 1 Calculated pump-probe spectra of molecular dimer at low (left) and high (right) excitation intensities. Legend shows delay times between pulses.

Calculations show that at low excitation intensity the pump-probe spectra shows relaxation and at high excitation intensity nonexponential decay is observed which can be associated with EEA. Thus our expanded equation behave as should be expected.

REFERENCES

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