Abstract

The subject of the present dissertation concerns the vibronic and vibrational transitions, generated in complex media by interaction with the ultrashort light pulses. The phenomenon, which always accompanies this interaction is the Impulsive Stimulated Raman Scattering. Two experiments, visualizing this process in different media, have been designed and performed. Herewith their results along with the interpretation are presented. The first experiment depends on the supercontiuum generation in diamond crystal. The sample is subjected to highly intense off-resonant light pulses. It is known that the self-phase modulation, self-steepening and interaction with photoionized free carriers are processes, which play the main role in the supercontinuum generation. In the present thesis the influence of the intrapulse stimulated Raman scattering on the evolution of the pulse in time and space is investigated. It has been found that the crystal lattice vibrations only weekly affect the pulse spectrum, yet the change is observed and-discussed in dependence of the pulse fluence. It manifests itself mainly by a peak lying apart from the central pulse frequency at a distance equal to the vibrational frequency of the diamond crystal. The interpretation of the experimental results is supported by numerical simulation. For this purpose the three dimensional nonlinear envelope equation is solved with the split-step Fourier method. The model includes the effects of refractive index dispersion, diffraction, self-phase modulation and its saturation, self-steepening, photoionization, interaction with free-carriers and stimulated Raman scattering. The results of modeling agree well with the experimental observations.

The second experiment concerns the Time-Resolved Femtosecond Stimulated Raman Scattering studies of the trans-β-apo-8’-carotenal molecule. The experimental setup has been constructed for this purpose. The time resolution of the setup is better than 100 fs and the frequency resolution is about 25 cm^{-1}. The trans-β-apo-8’-carotenal molecule was excited electronically to the S_2 state and its relaxation, through S_1 state to the ground state was observed by measuring the FSRS spectra of the C=C symmetrical stretching vibration. The Raman line corresponding to S_2 state decays within 120 fs after excitation, then a long living line at the frequency corresponding to the optically forbidden S_1 state appears. During the first 500 fs this line is negative, which is attributed to the transient vibrational inversion of population in S_1 electronic level. Later the line becomes positive and then it decays with the lifetime of S_1 level. At the same time the frequency of the line up-shifts. The theoretical model, describing the dynamics of the molecular system, has been created in quantum mechanical approach based on the formalism of projection operators. The results of the experiment are analyzed by comparison with the time resolved signal obtained from theoretical calculations. It is shown that the predictions of the numerical model agree well with the experimental results. The main conclusion of the present thesis is, that no additional electronic level (apart of S_0, S_1 and S_2) is required for explanation of the experimentally observed decay of the excited S_2 state, instead the existence of a set of vibrational sublevels of the S_1 state has been proposed to take part in its energy redistribution.