This Ph.D. thesis is focused on the study of optical nonlinearities and dynamics of excited charge carriers in monocrystalline diamond, nanocrystalline diamond and silicon. The dynamics of high density carriers in bulk diamond is investigated in detail (the transition from excitons and free carriers to electron-hole liquid or plasma). We study the picosecond dynamics of electron-hole liquid condensation using several techniques of time-resolved optical spectroscopy and demonstrate its evaporation by femtosecond laser pulses. We also propose two new optical techniques for measurement of lifetime, diffusion coefficient and surface recombination velocity of excitons in diamond. The results obtained by these techniques are described theoretically using diffusion equation and compared with the results obtained by the transient grating diffraction measurement. Further we study two- and three-photon absorption and nonlinear refractive index in diamond.

In nanocrystalline diamond we study the second and third harmonic generation and its physical origin. In superlattices of silicon nanocrystals in SiO_2 matrix we investigate the nonlinear transient absorption dynamics and carrier diffusion.