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Contents

Femtosecond Carrier Relaxation in Semiconductor-Doped Glasses

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Semiconductor-doped glasses incorporating small CdS_xSe_{1-x} or $CdSe_xTe_{1-x}$ crystallites in a glass matrix, show strong nonlinear absorption /1/ and large values of the nonlinear optical susceptibility χ_3 /2/. The semiconductor crystallite system currently attracts increasing attention when the semiconductor inclusions are small enough to expect quantum effects.

The semiconductor-doped glassfilters studied (Schott RG 830, RG 715, and RG 645) are characterized by an exponential absorption edge ~ three times less steep than the corresponding bulk semiconductors. The frequency position of the absorption edge varies with composition, but it may as well be influenced by the size of the semiconductor inclusions. No confinement effects are observed in the absorption spectra at 300 K, most probably due to a certain variation of crystallite size.

Femtosecond absorption recovery of all three Schott filter glasses was studied. In the experiment, femtosecond light pulses of ≈ 60 fs duration and λ = 620 nm from a colliding-pulse modelocking dye laser generate an electron-hole plasma with a carrier density $N \approx 3\times10^{17}$ cm^{-3} in the semiconductor inclusions. A weaker probing pulse samples the absorption changes as a function of the delay time between exciting and probing pulses. All data were recorded at room temperature.

Fig.1a,b shows the time-resolved absorption changes for the semiconductor-doped glass filters RG 645 and RG 830. The absorption change ΔA is plotted versus delay time between exciting and probing pulses. The dashed curves are the cross-correlation traces determining time zero. In the RG 830 semiconductor-doped glass (Fig.1b) the energy of the femtosecond laser pulses is well above the bandgap, creating carriers with an excess energy $\Delta E \approx 500$ meV. A fast recovery of the initial bleaching is observed with a time constant of 230 fs. This process can be identified with cooling of the initially hot electron gas to the lattice temperature. A comparison with the expected energy loss rate of the electron gas due to the emission of LO-phonons in crystalline semiconductors /3/ yields similar relaxation times for the absorption recovery. In RG 645 (Fig.1a) the opposite extreme is realized. The excess energy is only slightly larger than the bandgap. The absorption decreases with the integrated pulse intensity. No fast carrier relaxation process is observed since the carriers are injected with low excess energy. Consequently, the temperature of the electron gas will not change and no fast absorption recovery is observed. The bleaching is due to the filling of states in the conduction and valence bands and remains essentially constant over the plotted time.

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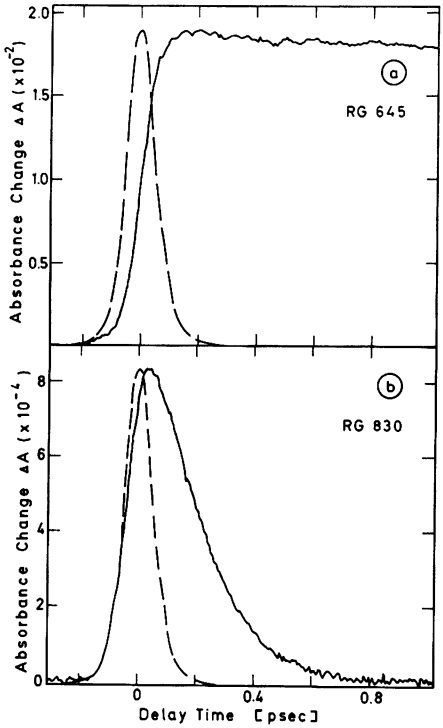

Fig.1 Femtosecond absorption recovery of semiconductor-doped glass filters RG 645 (Fig.1a) and RG 830 (Fig.1b).

Actually, ΔA decays with a long time constant of $\sim$ 100 ps (not shown in the figure). We attribute the 100 ps time constant to electron-hole recombination. This short recombination time is not unreasonable in the light of the large number of surface states in the semiconductor-doped glasses, reacting as efficient recombination centers.

The most interesting case is the semiconductor-doped glass RG 715 (Fig.2), where the initial carrier excess energy ΔE is $\sim$ 300 meV. Here, both the femtosecond decay process as well as the bandfilling can be traced. The three solid curves correspond to different excitation densities, where the lower curves are for two times (curve b) and four times (curve c) attenuated laser pulses, respectively. A semilog plot of the data reveals two time constants; 250 fs for the fast initial absorption recovery and 85 ps for the carrier recombination process. Significant is the superlinear decrease with carrier density of ΔA for times > 1 ps: ΔA decreases four times faster if the carrier density is reduced by a factor of 2.

The data can be explained assuming a carrier density dependent bandfilling process. In particular, we think of energy levels or traps below the conduction band, acting as a sink for the conduction band electrons. If the density of these levels is about the same as the carrier density ($\sim 10^{17}$ cm^{-3}) and the trapping time is several hundfred femtoseconds, the capture of the carriers into these levels becomes saturated, leading to the nonlinear dependence of ΔA on carrier density observed in Fig.2. The fast 250 fs relaxation process is a combination of cooling of the electron gas due to LO-phonon emission and trapping of charge carriers. From there, electron-hole recombination takes place via surface states in the middle of the bandgap with a time constant of 85-100 ps.

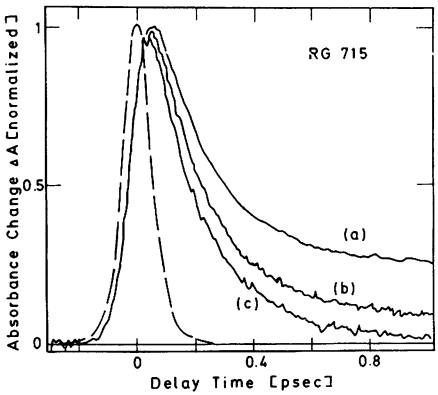

Fig.2 Absorption recovery of the semiconductor-doped glass RG 715

In conclusion, we point out that the absorption changes observed for the three semiconductor-doped glasses investigated here can be well explained within the framework of bulk semiconductor physics. The observed processes are cooling of a hot electron, electron trapping, and recombination. There is no indication of quantum size effects in the currently used glasses. We expect that excitonic effects will only be apparent for semiconductor-doped systems with smaller crystallites and with homogeneous size distribution.

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