Relationship between phase-conjugation efficiency and grating response time in semiconductor-doped glasses

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It is shown experimentally that deviations from the square dependence of the phase-conjugate reflectivity in CdS_xSe_{1-x}-doped glasses on the reduced pump power can be explained by the dependence of the relaxation time of the induced optical nonlinearity on intensity. The same argument explains the diversity of several previously published results.

Nonlinear-optical properties of semiconductor-doped glasses have been extensively investigated by four-wave mixing[1–7] and optical interferometry.[8,9] Reported experimental data seem to be inconsistent and sometimes contradictory. For example, the published values for $\chi^{(3)}$ of CdS_xSe_{1-x}-doped glasses vary by 4 orders of magnitude: 10^{-11} esu,[3–5] 10^{-10} esu,[6] 10^{-9}–10^{-8} esu,[1,2] and 10^{-7} esu.[7] A similar spread exists for values of the response time of the phase-conjugate process. Roussignol *et al.*,[10] using single 28-psec pulses, measured a change of the decay time from 0.75 to 1.25 nsec with increasing intensity from 2.5 to 30 MW/cm^2. Similar results are presented by Nuttermann *et al.*[11] but over an intensity range of 0.1–6 MW/cm^2. Cotter,[12] using much shorter pulses, $\tau_p < 0.5$ psec, also noticed a strong dependence of the phase-conjugation decay time on intensity: from 10 psec to 10 nsec for a 2-order-of-magnitude intensity variation. Remillard and Steel,[7] using a low-power cw dye laser, observed a constant grating decay time of 72 μsec over an intensity range of 0.5–5 W/cm^2. The most controversial aspect among the published papers[1,4,11,13] is the variation of the observed dependence of the phase-conjugate reflectivity on the pump intensity. Some research groups[5–7,13,14] measured the slope of a log–log plot of the phase-conjugation reflectivity versus the reduced pump beam intensity to be 2, as expected for a third-order nonlinear process. However, for the same glass, other research groups reported a slope of less than 1 (below the saturation region).[1,4,11] From a phenomenological point of view this could mean that the phase conjugation in semiconductor-doped glasses is not a purely third-order nonlinear process. In Ref. 13 it is suggested that photochemical changes in so-called "dark spots" are the cause for the decreasing slopes. These dark spots are observed after the samples are irradiated with picosecond pulses having an energy higher than 0.5 mJ/cm^2. Jain and Lind[1] suggested that the destructive contribution of higher-order nonlinearities is one of the reasons for a slope of less than 2.

In this paper we report experimental evidence for a relationship between the induced grating decay time and slope of the log–log plot of the phase-conjugate reflectivity versus reduced pump intensity in CdS_xSe_{1-x}-doped glasses. The intensity dependence of the phase-conjugation efficiency is less than quadratic when the laser pulse duration is close to, or longer than, the induced grating decay time τ_{gr}, and τ_{gr} decreases with increasing intensity. Application of this explanation to previously published results confirms this hypothesis.

Our measurements were performed in the low-power region, using a cw mode-locked Nd:YAG laser (average power 1 W at 532 nm, 70-psec pulse width at a 82-MHz repetition rate). The experimental setup for phase conjugation by degenerate four-wave mixing using a retroreflection scheme was described previously.[15] All beams had parallel polarization and a radius of 1 mm in the glass sample. With a chopper located in the beam waist of a telescope consisting of two equal lenses ($f = 20$ mm), the probe beam was given a rectangular time profile consisting of pulses having a rise time of less than 500 nsec. The grating decay time was measured by observing the profile of the conjugate pulses on a fast-memory oscilloscope. The semiconductor glasses used in these experiments were Corning filter 3-68 and Schott filter OG530, both $CdS_{0.9}Se_{0.1}$-doped glasses. The thickness was 3 mm, and the transmission at 532 nm was 25% and 50%, respectively.

The observed phase-conjugate reflectivity R versus the reduced pump intensity, $I_R = (I_{p,1}I_{p,2})^{1/2}$, for the OG530 glass is shown in Fig. 1(a). The upper curve was measured in a fresh, unexposed spot, while the lower curve was measured in a long-time-irradiated spot (sometimes called a modified spot[13,16]). Detailed results of the experiments on the creation and properties of modified spots in this type of glasses will be published elsewhere. The phase-conjugation efficiency is less in a modified spot than in a new spot, and the slope of the log–log plot of R versus I_R is decreasing from 1.6 to 0.6 with increasing pump intensity. In an unmodified spot we observed an almost quadratic reflectivity dependence on pump intensity (slope 1.9). The slopes in the Corning glass CS 3-68 [Fig. 1(b)] were less than 2 for both types of spot: 1.2 for a modified spot and 1.1 for an unmodified spot of the sample. Special care was taken to minimize the exposure time during the experiments in unmodified spots. For this reason we used an electromechanical shutter. The total irradiation time during one complete experiment did not exceed 4 sec, whereas the time for creating a

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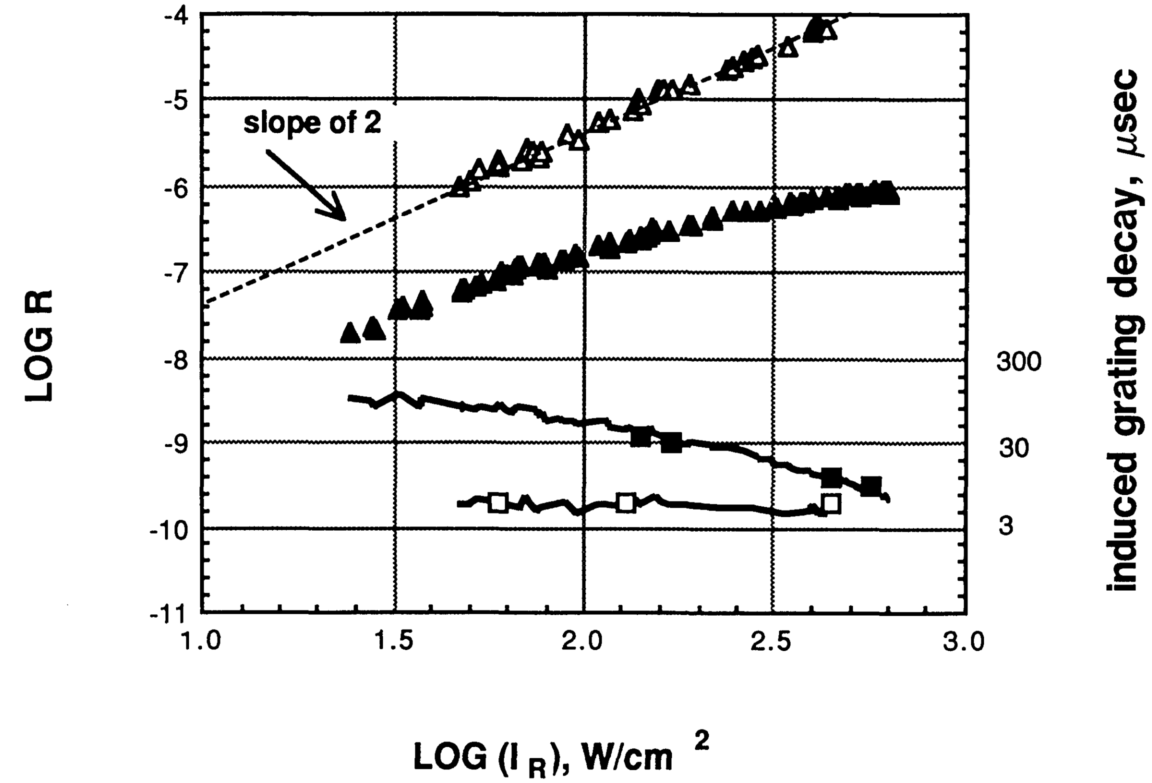
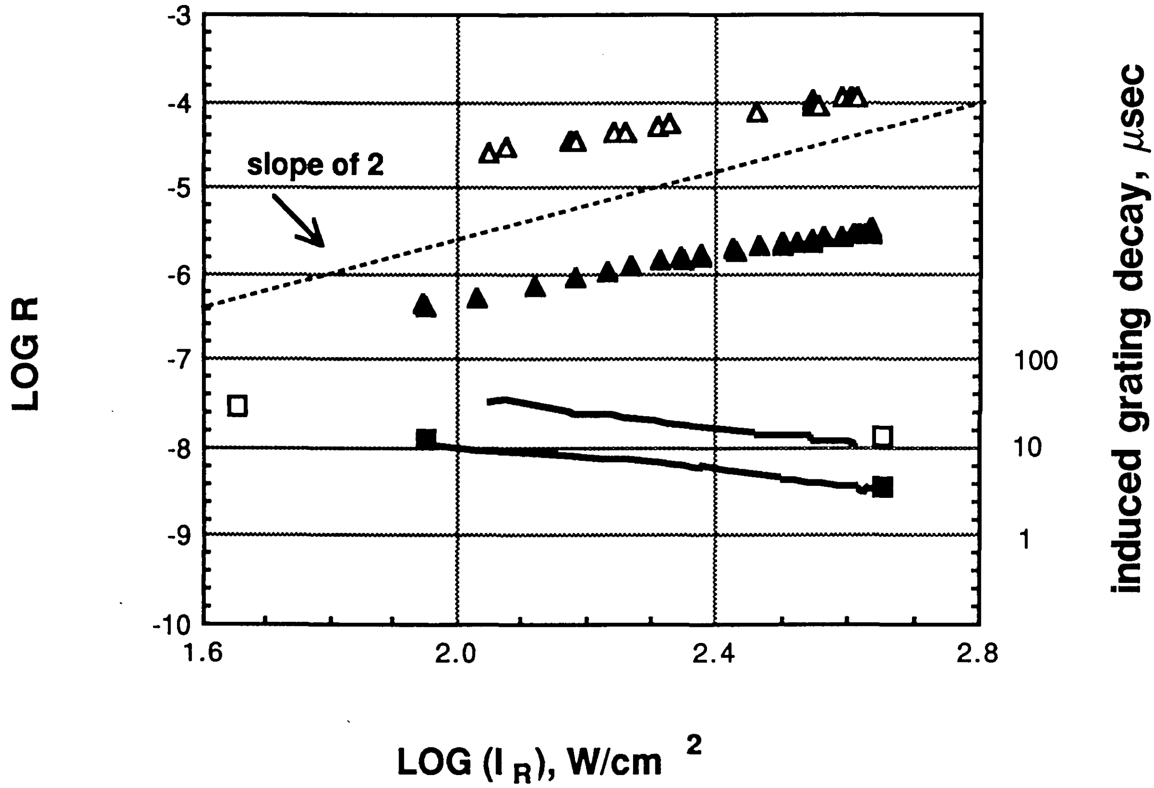


(a)



(b)

Fig. 1. Phase-conjugate reflectivity (▲, △) and induced grating decay time (■, □) versus the reduced pump intensity for (a) OG530 and (b) CS 3-68 filters. The open symbols are for unmodified spots, and the filled symbols are for modified spots. The solid curves represent the decay-time dependence on intensity calculated using our experimental reflectivity data and expression (2). The dashed curves represent the slope of 2.

modified spot was more than 2 min at the maximum pump intensity. The measured decay times of the induced grating are shown in the same figures. In the modified spots we find a decrease of the decay times with increasing pump intensity. The same behavior was observed for an unmodified spot of CS 3-68 glass. In an unmodified spot of OG530 glass, however, the phase-conjugation decay time was intensity independent and equal to 5 μsec. The correlation between the intensity dependence of the decay time and the slopes of the log–log plot of R versus I_R is obvious. Only in an unmodified spot in OG530, for which the grating decay time is essentially intensity independent, a slope of 2 was observed.

Consider first the low-power experiments with a cw mode-locked laser. As the observed grating decay times are much longer than the time between two mode-locked pulses (12 nsec), we can consider the chopped pulses as having a peak power equal to the peak power of the mode-locked pulses and an effective coherence time equal to its time duration determined by the chopper aperture and rotation speed.

At low excitation intensities the nonlinearity is due primarily to long-lived deep-trap levels, as known from photoconductivity measurement on single-crystal CdS_xSe_{1-x} samples[17] and cw phase-conjugation measurements by nearly degenerate four-wave mixing.[7] In this case the nonlinear process can be described by a three-level system, the deep-trap level being the long-lived third level. The shape of the phase-conjugated signal is well described by this model and will be reported in detail elsewhere. Since the length of the chopper pulses is much longer than the decay times, the phase-conjugate reflectivity reaches a quasi-steady-state level R, which can be described by a steady-state third-order susceptibility:

$$\chi^{(3)} = c\alpha\tau_{\mathrm{trap}}|\chi_{\mathrm{trap}}|/16h\omega, \tag{1}$$

where α is the low-intensity absorption coefficient, τ_{trap} being the lifetime of the sensitized traps and χ_{trap} their associated linear optical susceptibility. Since R is proportional to $(\chi^{(3)})^2 I_{p,1} I_{p,2}$, any dependence of τ_{trap} on the intensity will cause a deviation of the square dependence of R on the reduced pump power $I_R = (I_{p,1} I_{p,2})^{1/2}$. From a log–log plot of R versus I_R one can easily derive the intensity dependence of $\tau_{\mathrm{trap}}(I_R)$. According to

$$\log[\tau_{\mathrm{trap}}(I_R)] = \log(R)/2 - \log(I_R) + \mathrm{const.}, \tag{2}$$

we calculated curves proportional to τ_{trap} from the experimental dependence of R on I_r. These calculated curves and the experimentally observed values of τ_{trap} are shown together in Fig. 1, where the agreement between the experimental data and calculated curves is excellent. An intensity-independent value of τ_{trap} implies a slope of 2 for R, as is the case for an unmodified spot in OG530. This is also confirmed by previously published data.[7] These authors measured a constant τ_{trap} of 72 μsec by bandwidth measurements of nearly degenerate four-wave mixing phase conjugation and observed a slope of 2 in a log–log plot of R versus I.

Since these results clearly explain the origin of the deviation of the slope of a log(R) versus log(I) plot from the expected value of 2, we attempted to explain similar effects observed for phase conjugation in CdS_xSe_{1-x}-doped glasses at high pump intensities ($I_R > 10$ kW/cm^2). Although at these intensity levels the main mechanism for the optical nonlinear susceptibility is due not to traps but to band-gap related phenomena such as band filling and electron–hole plasma formation,[18,19] the finite decay time of the induced gratings plays a similar role in determining the magnitude of the phase-conjugate reflectivity. It is obvious that when the relaxation time of the induced optical nonlinearity is finite and shorter than the coherence time of the pump pulses, the coupling strength between the different waves is increased owing to a buildup of the induced gratings. It is important to notice that it is the (first-order) coherence time that is of importance here and not the actual width of the pulse. A quasi-steady state is reached during the pulse, the magnitude of which is proportional to the relaxation time τ_{gr} of the nonlinearity, described by Jain[18] through a steady-state third-order nonlinear susceptibility:

$$\chi^{(3)} = -\frac{\eta\alpha nce^2\tau_{\mathrm{gr}}}{8m_{\mathrm{eh}}{}^*h\omega^3}\left[\frac{\omega_g{}^2}{(\omega_g{}^2-\omega^2)}\right], \tag{3}$$

where η is the quantum efficiency for electron–hole pair creation, α is the absorption coefficient, n is the refractive index, ω_g is the band-gap frequency, and $m_{\mathrm{eh}}{}^*$ is the reduced carrier mass. Expression (3) describes the nonlinear response by free-carrier generation and is one of the main mechanisms contributing to the nonlinear-optical properties of semiconductor-doped glasses at high pump power. In this case, an intensity dependence of τ_{gr} will obviously result in an intensity-dependent value of $\chi^{(3)}$.

On the other hand, when the pulse coherence time τ_{coh} is much smaller than τ_{gr}, one probes the actual, instantaneous value of the third-order nonlinear susceptibility, and the effective $\chi^{(3)}$ is proportional to the coherence time of the pump pulses:

$$\chi^{(3)} = -\frac{\eta\alpha nce^2\tau_{\mathrm{coh}}}{16m_{\mathrm{eh}}{}^*h\omega^3}\left[\frac{\omega_g{}^2}{(\omega_g{}^2-\omega^2)}\right]. \tag{4}$$

In this case $\chi^{(3)}$ is independent of τ_{gr}, and one expects a square dependence of R on I_R as long as no depletion or saturation effects occur.

A compilation of experimental slopes and decay times for semiconductor-doped glasses, published by various research groups, is shown in Fig. 2. The intensity range of the slope measurement and the coherence time of the applied laser pulses are mapped on an intensity–time plane I–τ. Also indicated in Fig. 2 are the decay times of the phase-conjugation reflectivity versus the intensity at which they have been determined. The low-power experiments are located in the upper left-hand quadrant (low power, long decay times). Higher intensities are accompanied by shorter decay times in the nanosecond to picosecond region (lower right-hand quadrant).

Slope measurements performed using laser beams having a coherence time less than the decay time of the induced grating are located in the region below the decay-time plot. Slopes obtained with pulses having a coherence time longer than the decay time are located in the upper region of the I–τ plane. Values of the slope are indicated near the intensity regions over which they have been determined. The border between the two regions, made up by the τ_{gr}-versus-intensity curve, is not well defined because the data represented by Δ in Fig. 2 have been determined by using different CdS_xSe_{1-x}

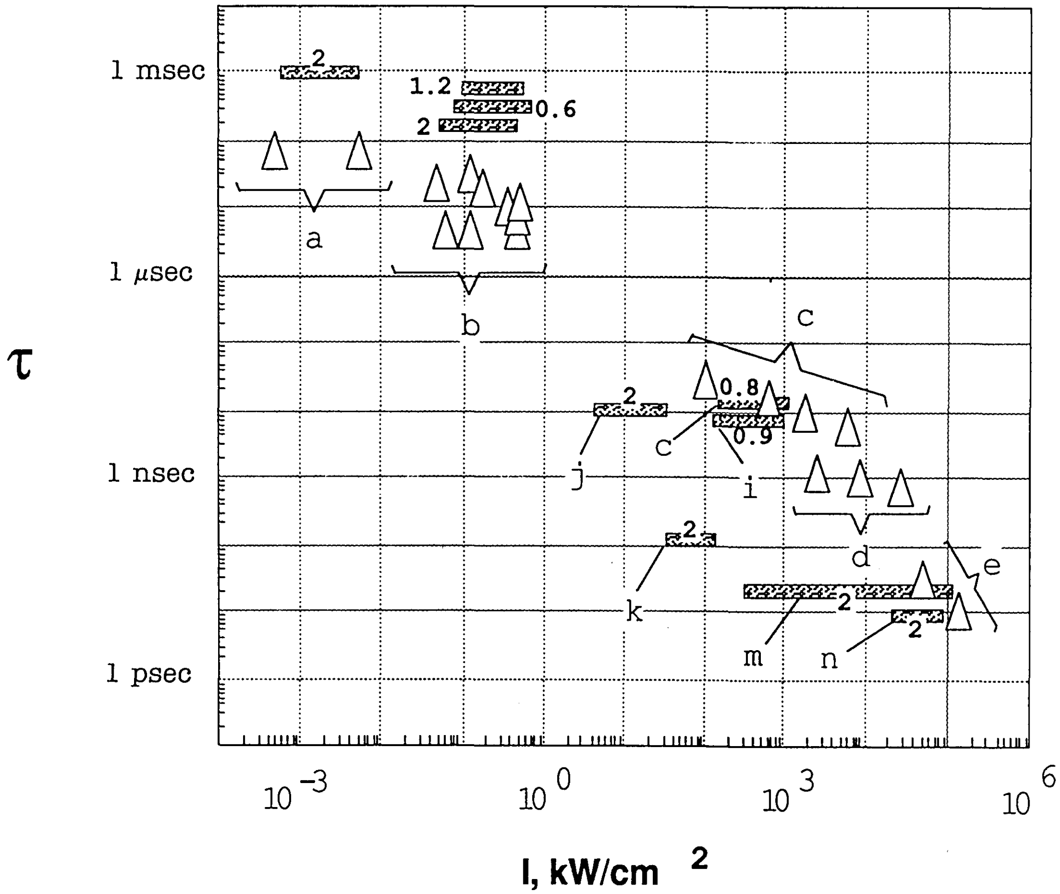


Fig. 2. Summary of reported experimental data for grating decay times (△), τ_{gr}, as a function of pump intensity in semiconductor-doped glasses. The shaded bars denote the intensity range of the pump used to measure the slope of the log–log plot of phase-conjugate reflectivity versus reduced pump intensity. The numbers beside the bars denote the observed slope. The position on the time axis corresponds to the duration τ_p or coherence time τ_{coh} of the laser pulses. The letters indicate the references: a, Ref. 7; b, this paper; c, Ref. 11; d, Ref. 10; e, Ref. 12; i, Ref. 1; j, Ref. 13; k, Ref. 6; m, Ref. 10; n, Ref. 14.

glasses, laser systems, and wavelengths. In the lower region, where τ_{coh} is smaller than the grating decay time τ_{gr}, all observed slopes are equal to 2. In the upper region, where τ_{coh} is larger than τ_{gr}, the slope is determined by the intensity dependence of τ_{gr}. When $d\tau_{gr}/dI$ is 0, the slope is 2, and when $d\tau_{gr}/dI$ is negative, i.e., τ_{gr} decreases with increasing pump intensity, the slope is less than 2. A quasi-steady state will be obtained only when τ_{coh} is at least two times shorter than τ_{gr}. This means that the transition region should be described in the same way.

In conclusion, experimental results are presented for the phase-conjugate reflectivity and its decay time in CdS_xSe_{1-x}-doped glasses. The deviation of the expected square-law dependence of the phase-conjugate reflectivity on the reduced pump intensity is explained by an intensity-dependent decay time of a long-lived deep-trap level.

The apparently contradictory results of slope measurements in previously reported experiments using high-power pulsed laser systems can be explained successfully by considering an effective value for $\chi^{(3)}$, depending on the coherence time of the laser pulses and the intensity dependence of the decay time of the induced gratings. When the laser pulse coherence time is longer than the relaxation time τ_{gr} of the nonlinear effect, the effective value of $\chi^{(3)}$ is proportional to τ_{coh}, whereas in the opposite case $\chi^{(3)}$ is independent of τ_{coh}. This argument is independent of the microscopic mechanism of the nonlinear-optical effect and is an addition to the well-known empirical rule that the nonlinear efficiency of systems is inversely proportional to the response time of the systems.

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