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Ultrafast decay of femtosecond laser-induced grating in silicon-quantum-dot-based optical waveguides

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Abstract

Femtosecond transient laser-induced grating (LIG) experiments were performed in planar optical waveguides made of luminescent silicon quantum dots. The LIG was created by interference of two pulses from the frequency-doubled output of a Ti–sapphire laser (400 nm, 400 fs, 1 kHz). The LIG exhibits an extremely fast decay in time (several picoseconds) that was found to decrease with decreasing grating period. The standard models based on lateral carrier diffusion cannot explain this observation (this procedure yields an unrealistically high diffusion constant of 420 $cm^2\ s^{-1}$). Instead, the results are explained by exciton diffusion and/or enhanced exciton radiative decay rate in a cavity represented by the periodically modulated planar waveguide (Purcell effect).

1. Introduction

Silicon quantum dots (silicon nanocrystals, Si-ncs) are efficient light emitters and have been employed to fabricate silicon-based light-emitting devices, such as light-emitting diodes. They have also been employed to fabricate active optical waveguide structures [1–3], in which complex spectral filtering of the self-guided photoluminescence (PL) has been observed [2, 3]. Many properties of such waveguides, which hold promise for exploitation in silicon photonics, remain to be explored and interpreted.

Transient laser-induced-grating (LIG) experiments are known to provide insight into fast non-equilibrium carrier recombination and diffusion in semiconductors. The LIG can decay in time due to both carrier recombination and lateral diffusion. When applied with picosecond laser pulses, a clear discrimination between both contributions can usually be obtained and the carrier ambipolar diffusion constant extracted [4,5]. Here we report on the application of ultrashort (femtosecond) laser pulses to investigate LIG decay in planar waveguides made of Si-ncs. We show that the observed grating efficiency decay is extremely fast, of the order of picoseconds, and we discuss possible reasons for such an unusual decay in a system composed of Si nanocrystals, which commonly exhibit much slower photocarrier recombination—in the microsecond range or even longer [6].

2. Experimental

Samples investigated in this study were prepared by implanting Si^+ ions into slabs of synthetic silica substrates (Infrasil)

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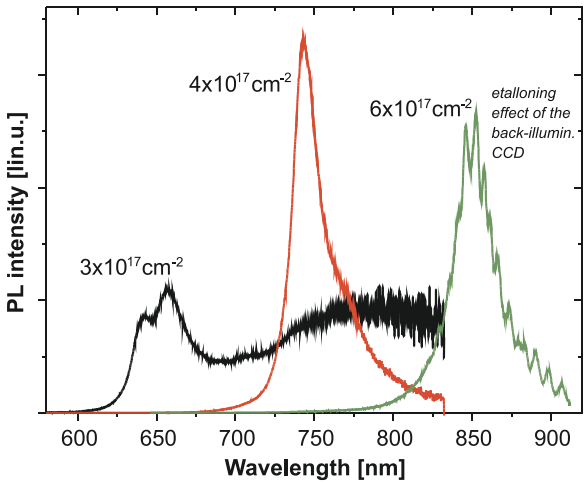

Figure 1. PL spectra taken from the edge of investigated samples ('waveguiding geometry'). Excitation with the 442 nm line of a cw HeCd laser, $T = 295$ K. Owing to insufficient resolving power the doublet structure of the peaks can be discerned in the sample 3×10^{17} cm^{-2} only.

with dimensions of about $8 \times 5 \times 1$ mm^3. The implantation energy was 400 keV and several implant fluences from 1 up to 6×10^{17} cm^{-2} were applied to different samples. A post-implant anneal at 1100 °C in N_2 ambient for 1 h caused precipitation of dispersed excess Si^+-ions into luminescent Si-ncs with a typical diameter of 4–5 nm. A further anneal for 1 h at 500 °C in forming gas (N_2/H_2) was employed to passivate non-radiative defects and enhance the Si-nc PL intensity [7]. Peak excess Si concentrations were up to 26 at% Si. To give an idea of how densely the Si-nc are packed, we suppose (i) the nanocrystal diameter to be 5 nm and (ii) only 50% of the implanted Si atoms have been accumulated to form the nanocrystals. For the excess Si concentration of 20 at% we get in this way the mean distance between nanocrystals of about 16 nm. The ensembles of Si-ncs with the nanocrystal concentration peaking at a mean depth of ~630 nm (the total thickness of the Si-nc layers being about 800 nm) represent planar waveguides buried in fused silica [3]. Room-temperature PL spectra of the three samples implanted to the highest fluences, as taken in the waveguiding geometry (i.e. from the edge of the samples), are shown in figure 1. The spectra consist of relatively narrow peaks whose position is systematically red shifted with increasing implant fluence. These peaks represent radiative modes of the waveguides [3] and are in fact TE/TM doublets; here, their doublet structure is clearly resolved in the 3×10^{17} cm^{-2} sample only.

In the LIG technique, two coherent laser pulses interfere in the sample, creating a periodic, spatially modulated pattern. Diffraction of a third, time-delayed pulse is used to monitor the decay of the grating with time. Frequency-doubled pulses ($\lambda_p = 400$ nm, $h\nu = 3.1$ eV) of ~400 fs duration, provided by a cw doubled Nd^{3+}–YAG laser-pumped Ti–sapphire laser, were split into two pump pulses of equal intensity. These were used to define an LIG in the waveguides parallel to the sample surface. The grating period Λ was varied by varying the angle θ between the writing pulses ($\Lambda = \lambda_p/2\sin(\theta/2)$). The pump spot diameter was about 300 μm and the pump

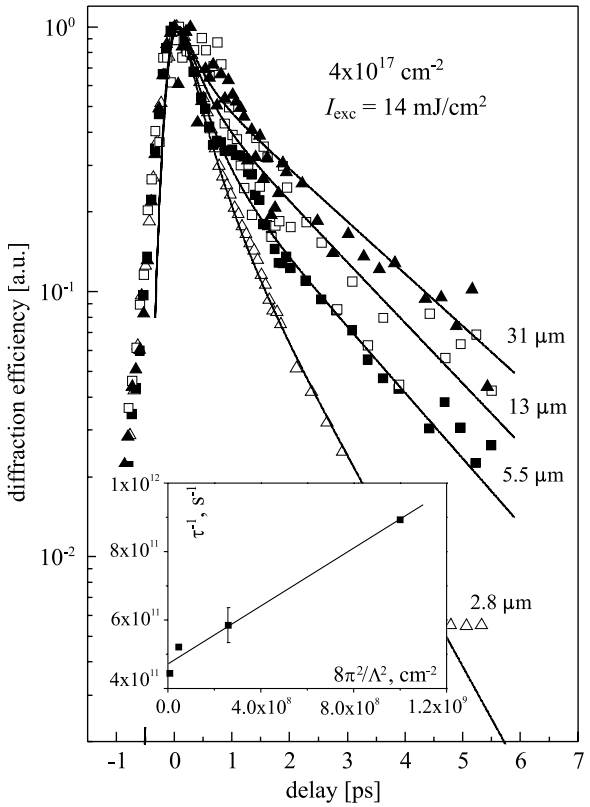

Figure 2. Normalized LIG decay curves for different grating periods Λ (next to the curves) in a waveguide implanted to a fluence of 4×10^{17} cm^{-2}. Pump energy density ~14 mJ cm^{-2}. The lines are guides to the eye. Inset represents 'standard' evaluation of the data (see text).

energy density ranged from ~2.8 mJ cm^{-2} to ~14 mJ cm^{-2} (peak power density $I_{exc} \sim 30$ GW cm^{-2}). A weaker, time-delayed test pulse of the fundamental frequency (800 nm) in the optically transparent region of the samples, with a diameter of ~100 μm, was aligned with the LIG and its diffracted intensity monitored using a conventional Si photodiode. All experiments were performed at room temperature.

3. Results and discussion

In what follows we focus our attention on the sample implanted to a fluence of 4×10^{17} cm^{-2}, in which the observed phenomena are most evident. The LIG dynamics in this sample are shown in figure 2. It can be seen that the grating decay time τ_g decreases with decreasing grating period Λ—an effect that is both distinct and interesting. In particular, our primary interest has been focused on the potential photocarrier lateral diffusion between nanocrystals because of the fundamental importance of this effect for electrical pumping of potential light-emitting devices based on Si-ncs. Such a lateral carrier diffusion, even if the Si-ncs are localized in an insulating matrix such as SiO_2, cannot be *a priori* excluded. It is true that in our samples the *mean* internanocrystal spacing (~16 nm) is quite large for an efficient charge transport. However, taking into account the non-uniform distribution of the nanocrystals and their tendency to form larger clumps with higher volume density, at many a

place the separation between *surfaces* of neighbouring Si-ncs may be considerably smaller, several tenths of nanometer only. Then, carrier tunnelling can occur (an observation of picosecond carrier tunnelling between Si-ncs has already been reported [8]). Moreover, the remaining excess Si atoms as well as the defects in the SiO_2 matrix could increase conductivity between the individual Si-ncs. As a result, a percolation conducting path in the Si-ncs/SiO_2 system can be established. To extract information about such carrier diffusion, however, is not easy.

It is almost certain that within the duration of the excitation pulse, *nonlinear* recombination processes (both radiative and non-radiative) occur in the excited Si-ncs, since, estimating the absorption cross section of Si-ncs at 400 nm for the samples with 20% excess Si concentration to be $\sigma \approx 2 \times 10^{-16}\,\mathrm{cm}^2$ [9, 10] and considering the pump energy density of $\sim$10 mJ cm^{-2}, we get the initial mean number of electron–hole pairs per nanocrystal as high as 3–5 (nevertheless, we were checking carefully that the pump power density was always below the threshold for irreversible sample changes, i.e. below the threshold for writing down a 'permanent grating' [11]). These nonlinear recombination processes in principle can be described by nonlinear terms in the relevant kinetic equations dealing with the time evolution of the LIG carrier population [4, 12] and could manifest themselves through a non-exponential decay of the LIG diffraction efficiency in time. However, the very formulation of such kinetic equations would be very difficult if not impossible, in view of the fact that the physics of highly excited Si-ncs has not been well understood till now. Instead, we apply here a simplified treatment of the experimental data, which is based on the following approach.

In order to estimate the involvement of carrier diffusion we have used the linear parts (in log scale) of the LIG decay curves at long delays only where the number of e–h pairs per Si-ncs can be supposed to be already low and the LIG decay should be driven mainly by relatively slow carrier lateral diffusion and recombination. The decay is then expected to be a single-exponential with a decay time τ_g; this is indeed the case (see figure 2). In this way we obtain $\tau_g(\Lambda = 2.8\,\mu\mathrm{m}) \approx 1.12$ ps, $\tau_g(\Lambda = 5.5\,\mu\mathrm{m}) \approx 1.71$ ps, $\tau_g(\Lambda = 13\,\mu\mathrm{m}) \approx 1.92$ ps and $\tau_g(\Lambda = 31\,\mu\mathrm{m}) \approx 2.25$ ps. Standard evaluation then consists of plotting $(\tau_g)^{-1}$ against $8\pi^2/\Lambda^2$, the slope of which yields the diffusion constant of carriers D [4]. Such a plot is shown in the inset of figure 2 and yields a value of D about 420 cm^2 s^{-1}, which is unrealistically high, bearing in mind that D in bulk Si is of the order of 10 cm^2 s^{-1} [13]. Carrier diffusion thus seems not to be involved in our ultrafast LIG decay. One has therefore to suggest another interpretation of the experimental data, in particular, of the peculiar variation of τ_g with Λ.

To assist in this regard, additional pump–probe experiments were undertaken. These used basically the same experimental set-up as above, only one of the writing beams was blocked and the energy density of the remaining one increased. The results, displayed in figure 3, revealed that the 'slow components' (delay $\geqslant$1.5 ps) of pump/probe and of LIG dynamics with large grating period Λ coincide. The decay

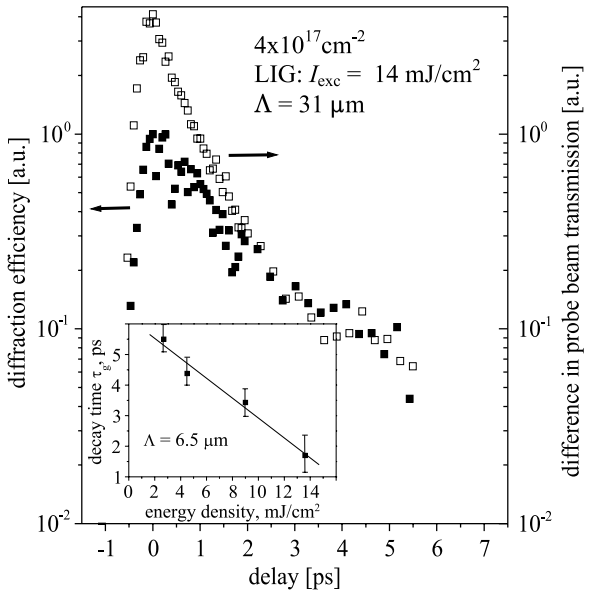

Figure 3. Comparison of the LIG dynamics ($\Lambda = 31\,\mu$m, full squares) with a pump-and-probe decay curve (open squares), measured on one and the same place of the sample. The 'slow' parts of both curves coincide for delays $\geqslant$1.5 ps. Inset shows decrease in τ_g with increasing pump energy density. At the initial stage of decay both the curves are not directly comparable because the energy density of the pump beam in the pump-and-probe experiment was chosen slightly higher ($\sim$21 mJ cm^{-2}) than that of the total pump in the case of LIG ($\sim$14 mJ cm^{-2}).

time τ_g was also studied as a function of pump intensity and it was found that increasing the pump intensity led to a reduction of τ_g (inset of figure 3).

The most obvious interpretation of these observations—fast non-radiative Auger recombination of carriers in Si-ncs—cannot readily account for the fact that the decay time depends on the grating period Λ. We therefore speculate about the following effects.

(i) Exciton (not free carrier) migration and tunnelling between neighbouring nanocrystals was proposed to be responsible for fast LIG decay in CdTe/ZnSe quantum dots under femtosecond pumping [14]. Both optical and acoustic phonons can participate in this exciton tunnelling at room temperature in order to compensate for the energy difference between different nanocrystals [15]. Due to the concentration gradient, the exciton diffusion tends to smooth the LIG and such a migration process should manifest itself predominantly at small Λ, in good agreement with our observation, see figure 2. Quite recent theoretical calculations confirm even the possibility of resonant (no-phonon) energy transfer between Si quantum dots [16] following the Förster mechanism. The energy transfer rate varies as $\sim 1/R^6$ where R is the interdot distance; for a very small R (of the order of nanocrystal radius) the transfer rate has been shown to increase even much faster. In this case submicrosecond transfer time can be possibly achieved.

(ii) Radiative recombination of excitons in the LIG is very probably involved in the LIG decay because intense PL is observed in our experiments under 400 nm femtosecond

pumping and the occurrence of a very fast (subpicosecond) component of Si-ncs luminescence has been evidenced recently by our group using an independent experimental set-up [17]. It is very interesting to note in this context that superradiance of an inhomogeneously broadened ensemble of semiconductor quantum dots has been experimentally demonstrated under 1.5 ps quasi-resonant excitation (i.e. excitation below the barrier bandgap energy and above the exciton ground state energy of the quantum dot) quite recently [18]. This effect of cooperative radiation leads to the enhancement of the exciton radiative recombination rate and thus to PL decay shortening. The basic requirement for the onset of the collective superradiant coupling, namely, a closely spaced ensemble of nanocrystals, has been met in our samples: we recall that the mean distance between (5 nm sized) Si-ncs in our samples with the excess Si concentration of 20 at% can be estimated to amount to ∼16 nm while Scheibner *et al* [18] observed the effect of superradiance in CdSe/ZnSe quantum dots up to the average interdot spacing of 150 nm. Nonetheless, even if we admit that a kind of superradiance occurs in our ensembles of Si-ncs, this effect alone can hardly explain the striking observation, namely, the LIG decay shortening with decreasing Λ. To this end we invoke the effect of the periodic modulation of the refractive index by the LIG, which defines a resonant optical structure that enhances the overlap between the Si-ncs emission frequencies and the available spectral density of photon modes. As a result, the spontaneous radiative lifetime of the nanocrystals can be modified (Purcell effect [19]). A variation of Λ can therefore affect the enhancement of the dipole-field coupling and cause changes in the radiative lifetime of the LIG as a function of Λ.

To assess the magnitude of the Purcell effect the following simplified model was employed: periodic modulation of the refractive index with a Gaussian envelope across the plane of the waveguide (see inset to figure 4)

$$n(x) = n_{\mathrm{F}} + \frac{\Delta n}{2}\left[1 + \cos\left(\frac{2\pi x}{\Lambda}\right)\exp\left(-\frac{x^2}{\sigma^2}\right)\right], \quad (1)$$

where Δn stands for the maximum of the induced change in the refractive index and σ is the (Gaussian) beam width. The propagation of light waves in this structure was then calculated using an effective refractive index [20] for the guided modes in the waveguide. Since the increase in the dipole moment implies a decrease in τ_{g}, figures 2 and 4 display qualitatively the same behaviour. For large values of Λ ($\geqslant 35\,\mu$m) the effect in figure 4 shows a tendency to disappear. This agrees with the observation in figure 3, namely, that decay curves for an LIG with $\Lambda = 31\,\mu$m and for a probe beam passing through a non-periodic excited spot ($\Lambda \to \infty$) are basically identical. Estimating the nonlinear susceptibility of Si-ncs (with the mean diameter of 5 nm) embedded in an SiO_2 matrix under femtosecond pumping to be [21] $\chi^{(3)} \approx 0.2 \times 10^{-9}$ (esu) $\approx 3 \times 10^{-18}$ ($\mathrm{m^2\,V^{-1}}$) we get a nonlinear refractive index $\gamma = 3\chi^{(3)}/4c\varepsilon_0 n_{\mathrm{F}}^2 \approx 3 \times 10^{-12}\,\mathrm{cm^2\,W^{-1}}$. The maximum

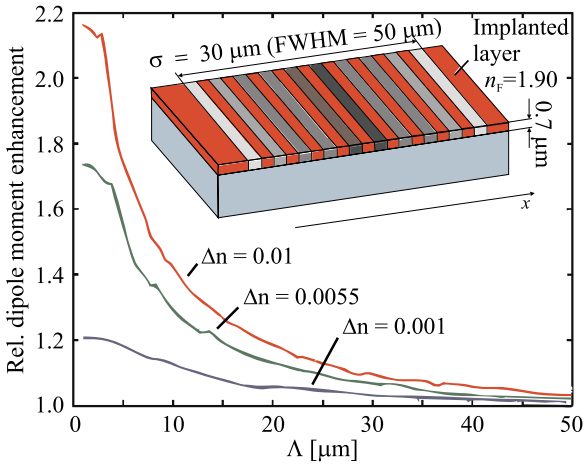

Figure 4. Calculated effective dipole moment enhancement for Si-ncs located at the maximum of the modulation profile as a function of Λ. The inset shows a scheme of the structure under consideration. For computational reasons, the Gaussian pump spot width was considered $\sigma = 30\,\mu$m only. The parameters Δn next to the curves are the amplitudes of the nonlinear refractive index modulation.

induced refractive index change $\Delta n = \gamma I_{\mathrm{exc}}$ thus amounts (for $I_{\mathrm{exc}} \sim 30\,\mathrm{GW\,cm^{-2}}$) to the orders of 0.01–0.1. This, referring to figure 4, can result in a substantial increase in the dipole moment magnitude. Since the first-order signal dynamics in a four-wave mixing process (as observed in our experiment) is driven by the speed of deformation of the cosine-like interference profile, i.e. predominantly by the maximum dipole enhancement (figure 4), the increase in the radiative recombination rate due to the Purcell effect could be quite significant.

(iii) An increase in the (linear) dipole coupling between the nanocrystals and the optical field can also lead to an increase in the nonlinear dipole interaction, i.e. an increase in the stimulated emission rate and the appearance of optical amplification. Here it is worth stressing the following facts. The ultrashort blue pump pulses (∼100 fs, ∼400 nm) from a frequency-doubled Ti–sapphire system have proved recently to be very efficient in producing room-temperature positive optical gain in ensembles of densely arranged Si-ncs. Experiments using the variable-stripe-length method [22, 23] yielded values of the gain coefficient as high as $25\,\mathrm{cm^{-1}}$.

4. Conclusions

In conclusion, we have performed transient LIG experiments with picosecond resolution in planar waveguides composed of luminescent Si-ncs. We explain the observed very fast LIG decay time and its variations with grating period by a model invoking exciton diffusion and/or enhancement of the radiative recombination rate in the periodic distribution of the electromagnetic field amplitude. An additional contribution to the LIG decay shortening can originate from possible involvement of stimulated emission in the waveguide.

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