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Ultrafast optical switching with CdTe nanocrystals in a glass matrix

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This letter describes a principle demonstration of an ultrafast optical switch operating at 1 Tbit/s using CdTe-quantum-dots-doped glasses. Using a three-beam pump and probe experiment, we showed that thermal effects are responsible for a baseline in the pump and probe graphs and the nonexistence of carrier accumulation effects. After eliminating the thermal effects, we showed that, when two pump pulses are delayed by 1 ps, each pump pulse modulates the probe pulse independently, making this material highly promising for ultrafast all optical switching. © 2005 American Institute of Physics. [DOI: 10.1063/1.1905805]

In this last decade, attention has been devoted to the development of new materials that combine ultrafast response times and high optical nonlinearities for applications in ultrafast all-optical switching at Tbit/s data transmission rate for optical communication systems.1,2

Semiconductor nanocrystals are among the most investigated materials for these applications because their optical properties can be controlled by their size and because their large surface to volume ratio supplies a large amount of surface trap states to accelerate the recovery process. Yu et al.3 have shown nonlinear refraction index as high as $n_2 \sim 10^{-12}$ cm$^2$/W for CdTe quantum-dot doped glass in the transparent region and Cotter et al.4 and Tsuda and Cruz5 showed that $n_2$ should increase closer to the band gap in quantum dots, due to the ac Stark effect. In a previous paper we have shown that it is possible to control the average recombination time, through the Auger recombination process, in the same material, down to a minimum of 250 fs.6 These characteristics indicate that a large figure of merit can be expected in CdTe quantum dots. Table I shows the reported typical values for $n_2$, response time, $\tau$, and the figure of merit for different semiconductor quantum dots. CdTe quantum dots present one of the greatest figures of merit. In CdTe quantum dots multilayers fabricated by laser ablation technique, Tsunetomo et al.1 have shown optical switching operating at 250 Gbit/s. For optical switching applications an important figure of merit involves the real part of the nonlinear susceptibility, $\chi(3)$, which is proportional to $n_2$, the nonlinear refraction index, the linear absorption, $\alpha$, and the recombination time, $\tau$: $F = \text{Re}(\chi(3))/\alpha \tau$. The higher this figure of merit, the better the optical device.2

The experimental results described in this letter demonstrate that CdTe quantum dots doped glass can operate as an optical switch at rates as high as 1 Tbit/s. Through a combined femtosecond and continuous wave excitation experiment we demonstrate that no accumulation process occurs for times longer than 1 ps, and only thermal effect could jeopardize the device’s operation. After eliminating the thermal effects we observe a complete recovery with an average response time of 300 fs. In a previous letter we observed the presence of deep trap states in our sample.6 In this letter we find that the deep-trapped electrons are not relevant for the transient transmission signal so that the main effect responsible for the baseline observed in the transient transmission graphs is thermal.

The CdTe QD doped glass was grown by melting the glass components, SiO$_2$, Na$_2$O, B$_2$O$_3$, ZnO, and Al$_2$O$_3$, together with metallic Te and CdO dopants and a subsequent heat treatment of 640 °C for 77 h.6 Due to this long heat treatment, the average radius of the QDs was 7.2 nm and the first absorption peak is at 745 nm. We used a three beam pump and probe setup1 to observe response differences between the sample when it is excited by one pump pulse or two pump pulses. Translation stages for the probe and one pump pulse control the delays between the three pulses. The pulses were generated from a tunable femtosecond Ti:sapphire laser operating at 80 MHz pumped by an argon laser. The two pumps and the probe pulses were split from the Ti:sapphire laser output in the ratio $f_{\text{pump1}}: f_{\text{pump2}}: f_{\text{probe}} = 20:20:1$. The two pump and the probe pulses are polarized perpendicularly to each other to avoid coherent artifacts, and chopped at different frequencies, $f_{\text{pump}}$ and $f_{\text{probe}}$. The lock-in was synchronized at $f_{\text{pump}}$, $f_{\text{pump2}}$ and $f_{\text{probe}}$ to guarantee that the signal detected was generated only by a joint pump and probe effect. The three beams were focused on the same spot in the sample with a spot size of $\sim 10$ μm in radius at HW1/e$^2$M by a 5 cm focal length lens. The transmitted probe passes through a polarizer to block light scattered from the pump beams and collected with a pin detector connected to a lock-in amplifier. The temporal FWHM of the pulses was 80 fs with 8 nm spectral line width. All measurements were performed at room temperature.

Figure 1 shows the transient transmission signal measured by pump and probe technique when the sample is excited by only one pump beam and by two pump beams. The second pump pulse, delayed by more than 1 ps causes a copy of the effect of the first one, which shows that its effect is not influenced by the first pump pulse. It also indicates that the difference between the peak and the baseline of the signal trace, $\beta$, does not depend on the number of pump pulses. However, in the same figure we also observe that the baseline increases from 0.06 mV for one pump pulse to 0.12 mV for two pump pulses, suggesting a baseline height proportional to the number of pulses, or the incident energy. The increasing in the baseline could be explained by an accumu-
Two pumps modulate the pump beam at 580 Hz. Highness is the same in both cases. The chopper modulates the pump beam for two pumps the baseline is higher than for only one, but the peak and 500 mW and with HW1/e2 M radius equal to 1 mm. The difficulties arising from thermal effects. Information traffic changes, which means that one can avoid difficulties arising from thermal effects.

Table 1. Nonlinear refractive index, $n_2$, response time, and figure of merit for different semiconductors quantum dots in doped glass.

<table>
<thead>
<tr>
<th>Quantum dots in doped glass</th>
<th>Linear absorption edge (nm)</th>
<th>$n_2$ ($\text{cm}^2/\text{W}$)</th>
<th>Response time (ps)</th>
<th>Figure of merit $\text{cm}^2/\text{W}$ $/\epsilon_0$ $a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CdTe</td>
<td>745</td>
<td>$-6 \times 10^{-13}$</td>
<td>0.3</td>
<td>&gt;9</td>
</tr>
<tr>
<td>CdSe</td>
<td>580</td>
<td>$-10^{-15}$</td>
<td>60</td>
<td>&gt;7.5 $\times 10^{-3}$</td>
</tr>
<tr>
<td>CdS, CdSe</td>
<td>790</td>
<td>$-10^{-14}$</td>
<td>46</td>
<td>&gt;10$^{-3}$</td>
</tr>
<tr>
<td>PbTe</td>
<td>1550</td>
<td>Not measured</td>
<td>100</td>
<td>...</td>
</tr>
<tr>
<td>PbS</td>
<td>1300</td>
<td>Not measured</td>
<td>4</td>
<td>...</td>
</tr>
<tr>
<td>Ge</td>
<td>780</td>
<td>$1.5 - 8 \times 10^{-12}$</td>
<td>65</td>
<td>~2.2</td>
</tr>
</tbody>
</table>

$^a$n$_2$ measured in the transparent region, in the absorptive region $n_2$ is higher (Refs. 4 and 5).

$^b$Figure of merit in units of $c\epsilon_0/\epsilon_0$, where $c$ is speed of light, $\epsilon_0$ is the electrical permittivity of the vacuum, and $a$ is the linear absorption.

$^c$See Refs. 3 and 6.

$^d$See Refs. 5 and 7.

$^e$See Refs. 8 and 9.

$^f$See Ref. 10.

$^g$See Ref. 11.

$^h$See Ref. 12.

The long-time accumulation of real carriers is an undesirable process for ultrafast all-optical switching because a long pulse sequence would degrade the nonlinearity and reduce the switching rate achievable. On the other hand, thermally induced baseline depends only on the average power of income signals, because thermal diffusion, in the ms time scale, is too slow for systems intended for Gbit/s rates but the average power will define the temperature of the device for a given operation. However, in this case, a simple feedback loop to control the device temperature can correct for fluctuations as the information traffic changes, which means that one can avoid difficulties arising from thermal effects.

To discriminate between the thermal and cumulative process we added a nonmodulated cw excitation to the pump and probe experiment and observed the behavior of the baseline. The cw excitation is a single-line argon laser at 514 nm and 500 mW and with HW1/e2 M radius equal to 1 mm. The cw excitation generated a residual carrier concentration in the sample. The carrier density excited by the cw laser is around $10^{21}$ cm$^{-3}$ s$^{-1}$, and each pulse excites a carrier density of $10^{14}$ cm$^{-3}$. If there are long lived states with lifetime of $\mu$s, this cw excitation should generate a residual carrier density of around $10^{15}$ cm$^{-3}$, in those states. This should cause the baseline to disappear or, at least, decrease, since this density is one order of magnitude higher than that generated by each pulse. However, if there are no long lived states, that excitation would generate a residual population of around only $10^8$ cm$^{-3}$ in the excited states and no changes in the baseline should be observed. Figure 2 shows no differences for the ultrafast transmission measured with and without the cw excitation, strongly suggesting that carrier accumulation effects are not important for the baseline. On the other hand, if the baseline is due to a modulated thermal effect due to the low frequency chopper modulation of the pump beams we expected a frequency response in the Hz–kHz range because thermal diffusion processes are slow. The modulated temperature change tends to disappear at high frequencies. The fact that CdTe quantum dot absorption is temperature dependent couples the transmission detected by the lock in amplifier to temperature modulations.

Our 1 cm in diameter, 600 $\mu$m of thickness sample can be modeled as a two-dimensional diffusion problem. The heat source comes from the focal spot of about 10 $\mu$m that can be considered as a Dirac Delta function in the space domain but time modulated with frequency $\omega=2\pi f_{\text{pump}}$. The heat diffusion equation, then, becomes

$$\nabla^2\Theta - \frac{1}{\alpha} \frac{\partial \Theta}{\partial t} = \frac{P_0}{K} \delta(r-r_0)e^{-iut}, \quad (1)$$

where $\Theta$ is the ac temperature component, $\alpha$ is the thermal diffusivity coefficient, $P_0$ is the average pump power, and $K$ is thermal conductivity. Solving this equation with the boundary conditions of no heat flux at the sample air interface (the diffusion through the sample is much more important than the heat loss to air), we obtain the ac temperature. The baseline, in the limit of small linear variations, is proportional to this temperature variation and is given by

$$\beta = c\Theta = k \frac{P_0}{\omega} \sin\omega t, \quad (2)$$

where $c$ and $k$ are constants.
Figure 3(a) shows that the baseline height is inversely proportional to chopper frequency, $B \propto \omega^{-1}$, as expected by the diffusion model result. Figure 3(b) shows that the baseline is proportional to the pump power for low modulation frequency (500 Hz), and that it is almost zero at high modulation frequency (3.1 kHz) and does not vary with the pump power. It is possible to extract the constant $k$ from the slope of the straight line of the plots in Fig. 3. We found that $k=1.58 \text{ V s/W}$ in Fig. 3(a) and $1.60 \text{ V s/W}$ in Fig. 3(b). The baseline behavior in both graphs and the proximity between the two values calculated for $k$ confirms the model results and suggests that the thermal effect is the most important effect that, if not eliminated, could cause problems in optical switching for telecommunications. Modulating the pump laser at high frequencies we can eliminate the baseline.

Finally, Fig. 4 shows another transient transmission signal, for one pump pulse and for two pump pulses, like in Fig. 1, but at a higher chopper frequency, 3.1 kHz. Confirming the previsions, we observe no baseline both for the case of one and two pump beams. We also observed again the non-influence of the second pump pulse in the probe modulated by the first pulse when the delay is larger than 1 ps. However, strong interference between the pump pulses is observed for time delays below 1 ps. It means that modulating the pump beams at higher frequencies we could eliminate the baseline in our experiment, and it confirms that the baseline is only due to the thermal effect and that there is no carrier accumulation effect, then the total recovery time is faster than 1 ps, and an optical device with such sample could operate up to 1 Tbit/s rates before pulses interactions starts to appear.

In conclusion, ultrafast nonlinear transient transmission was demonstrated using CdTe quantum dots doped glass with a three beam pump and probe measurement. There is no evidence of carrier accumulation and the thermal effects are the main reason for the baseline observed in the experiments. However, the ac thermal effects tend to disappear at high modulation frequency and shall not pose any problem for the optical device. Transient thermal effects when turning the system on and off or fluctuations in the average power can be a problem, but one which engineers have always dealt with. The high optical nonlinearity shown by Yu et al., the fast response time and the absence of long-lived carrier accumulation suggest that CdTe quantum dots doped glass is a promising material for Tbit/s all-optical switching devices.

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