Two-photon and two-photon-assisted slow light

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We show that light pulses propagating in two-photon absorbing systems may present time delays like slow light produced via coherent population oscillations in one-photon interactions. Two regimes are numerically studied for a simplified two-level system: (a) a light pulse at frequency \(\omega/2\) undergoes two-photon absorption (TPA) and is delayed by the absorbing system (two-photon slow light) and (b) a light pulse at frequency \(\omega\) is delayed in a system prepared by TPA of a light pulse at frequency \(\omega/2\) (two-photon-assisted slow light). The study carried out in solutions of dyes and dendrites shows significant delays, low distortion, and good transmission for easily reachable experimental conditions. The working principle can be applied to other media and can be used in telecommunications technology. © 2011 Optical Society of America

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The study of light propagation through matter has always raised a lot of interest, and nowadays much activity is being done on group-velocity control, with useful applications, for example, in telecommunications [1]. Among the methods used at room temperature stands the one based on coherent population oscillations (CPO) [2], which is based on affecting the distribution of population among the levels of a system undergoing one-photon absorption. As a consequence of a time-dependent ground state population, the system shows a time-dependent absorbance, and by the Kramers-Krönig relationship, a changing refractive index, which influences the propagation of light through the system. The first observation of this process was done in a ruby crystal [3]. Because of its relative simplicity, slow light by CPO can be observed even in undergraduate laboratories [4].

Our aim is to show that two-photon absorption of light pulses in matter can substantially affect the population distribution and modify their absorbance and refractive index, leading to time delays in the pulse propagation, in the manner of slow light. The large intensities which are nowadays reachable focusing laser sources, allow for observation of two-photon absorption transitions in many systems, as long as a powerful light source is available at the desired frequency (\(\omega/2\) for a system with a resonance at \(\omega\)). Organic molecules are among the most frequently studied systems within this context [5,6]. They have two-photon absorption (TPA) cross sections which are large enough to even produce population inversion, leading to laser action [7,8], and can be specifically designed to have giant one- and two-photon absorption cross sections, as is the case of the dendrites [6]. Organic systems are characteristic of low-cost, easy preparation in solution and they can be also embedded in polymer matrices, if a solid state is desired.

A typical four-level energy diagram for an organic molecule is depicted in Fig. 1(a). Gray areas represent a ground and an excited singlet electronic state, each comprising a number of vibrational levels. Photons with frequency \(\omega\) can be absorbed with an absorption cross section \(\sigma^{(1)}\), promoting a transition from the lowest vibrational level 1 in the first singlet to one of the upper vibrational states (level 2) of the second singlet. For intense light sources, photons of frequency \(\omega/2\) can also be absorbed, with a TPA cross section \(\sigma^{(2)}\). From level 2, the system relaxes vibrationally to level 3, decays with a lifetime \(\tau\) to a vibrationally excited level 4 in the lower singlet, and then relaxes, back to level 1. Since \(\tau_{nr}(\approx 10^{-12}\text{ s}) \ll \tau(\approx 10^{-8} \text{ to } 10^{-9}\text{ s})\), level 1 depopulates essentially at a rate governed by the product of the corresponding absorption cross section and the light intensity [see Fig. 1(a)], and it populates essentially at rate \(\tau\).

Let us first consider the situation of a single pulse of frequency \(\omega/2\), duration \(t_{p}\), and photon intensity \(I_{\omega/2}\) traversing a medium of length \(L\) and refractive index \(n_{r}(\omega/2)\) [Fig. 1(b)]. If only TPA is present, the set of equations describing the evolution of the density of population \(n_{1}\) in level 1 and the propagation of the pulse photon intensity \(I_{\omega/2}\) through the medium is given by

\[
\frac{dn_{1}(z,t)}{dt} = -\sigma^{(2)}n_{1}(z,t)I_{\omega/2}(z,t) + \frac{n - n_{1}(z,t)}{\tau},
\]

where \(n_{r}\) is the real part of the refractive index.

Fig. 1. (a) Energy levels diagram for an organic molecule, (b) propagation of a light pulse at \(\omega/2\) through a two-photon absorbing system, (c) propagation of a weak light pulse at \(\omega\) through a TPA prepared system.

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where $n$ is the total density of absorbing centers and is valid if $t_p \gg \tau_{nr}$. If a Gaussian pulse $I_{in}^{ω/2}(0, t)$ with an FWHM given by $t_p$ enters the medium and affects the populations $n_1(z, t)$ in such a way that the pulse at the exit $I_{out}^{ω/2}(L, t)$ is delayed with respect to a reference pulse $I_{ref}^{ω/2}(L, t)$ which propagated at velocity $c/n_\gamma(ω/2)$, we will identify this phenomenon as two-photon slow light.

Equations (1) and (2) were integrated in the lab coordinate system. For each time $t$, the spatial distributions of population $n_1(z, t)$ and the photon intensity $I_{in}^{ω/2}(z, t)$ were obtained in a $L = 1\, \text{cm}$ cell divided in $N = 400$ segments, using $dz = L/N$ and $dt = n_r/c \, dz$ ($c$ is light velocity in vacuum; $n_r = n_r(ω/2) = 1.34$). As a pump source we used an Nd:YAG laser (1064 nm) with Gaussian-shaped pulses ($t_p = 100\, \text{ps}$, centered at $4 \times t_p$) and having $2.8\, \text{mJ}$ maximum output energy per pulse. High input intensities can be reached by focusing on the sample with a $100\, \mu\text{m}$ beam size. Typical values for lifetimes and two-photon absorption cross sections of dyes and dendrites at 1064 nm are $[3-7]$ as follows: dyes, $\tau = 3 \times 10^{-8}\, \text{s}$, $σ^{(2)} = 1.8 \times 10^{-56}\, \text{m}^4\, \text{s}$; dendrites, $\tau = 1 \times 10^{-10}\, \text{s}$, $σ^{(2)} = 1 \times 10^{-55}\, \text{m}^4\, \text{s}$. Figure 2 shows the time delay $\Delta t$ between the reference and output pulses and the fractional delay $(\Delta t/t_p)$ for both systems, as a function of the intensity $I_{in}^{ω/2} \times h\, ω/2$ and at three different concentrations [molar concentrations 0.01, 0.005, 0.001, corresponding to $n = 6 \times 10^{25}$ (●); $3 \times 10^{25}$ (•); $6 \times 10^{25}$ $\text{m}^{-3}$ (▲)]. Insets contain some temporal profiles. It is clearly observable that (1) there exists a saturation intensity for which the delay is maximum, (2) the saturation intensity varies with concentration, (3) larger delays are obtained for higher concentrations, (4) for dendrites (larger TPA cross section and smaller lifetime) larger delays and lower saturation intensities are obtained. This behavior resembles slow-light behavior by CPO in one-photon absorbing systems. The delays are large and amount up to almost 0.3 fractional delay for high concentrations. Another important feature of slow-light systems is the maximum bandwidth achievable by the system. As in one-photon CPO-based slow light, this bandwidth is determined by the minimum time scale in which the population can respond to the intensity variations, being this time determined by the population decay time of the excited state. Also Eqs. (1) and (2) are similar to those used of one-photon CPO-based slow light, where a rate-equation analysis has proven to fully explain the spatial variation of the phase if the dipole dephasing time is much shorter than the ground state recovery time, as is the case under study $[4, 9]$.

The transmission was estimated by $\max(I_{out}^{ω/2}(L, t))/I_{in}^{ω/2}(0, t)$ and the distortion $D$ of the pulses was characterized by the quantity $[10]$ $D = \left[\int_{\infty}^{0} I'(t + Δt) - I'(t)\, dt\right]/\left[\int_{\infty}^{0} I'(t)\, dt\right]^{1/2}$, where $I'(t)$ and $I(t)$ are the normalized intensity shapes of the output $I_{out}^{ω/2}(L, t)$ and reference $I_{ref}^{ω/2}(L, t)$ pulses, respectively, and $Δt$ is the delay time. Figure 2 shows the transmission and distortion in both systems for the three concentrations.

Hence, we can conclude that a CPO slow-light behavior with large time delays, low pulse distortion without pulse peak splitting, and good transmission can be encountered in two-photon absorbing systems in experimental conditions that can be reached with commercial laser systems, and could even be the only way to produce slow light in large band-gap systems.

Let us now consider the medium traversed by two pulses [Fig. 1(c)]. If an intense pulse $I_{in}^{ω/2}(0, t)$ at a certain frequency (here $ω/2$) enters the medium and affects the populations $n_1(z, t)$ in such a way that another weak pulse $I_{in}^{ω}(0, t)$ at another frequency (here $ω$) entering the medium exits as $I_{out}^{ω}(L, t)$, showing a delay with respect to a reference pulse $I_{ref}^{ω}(L, t)$ which propagated at velocity $c/n_\gamma(ω)$, we will identify this phenomenon as two-photon-assisted slow light. This situation is interesting for delaying weak pulses which are unable to produce slow light by themselves. The set of equations describing the evolution of the density of population $n_1$ in level 1 and the propagation of the photon intensities $I_{in}^{ω/2}$ and $I_{in}^{ω}$ through the medium is given by

\[
\frac{n_1(ω/2)}{c} \frac{dI_{out}^{ω/2}(z, t)}{dt} + \frac{dI_{out}^{ω/2}(z, t)}{dz} = -2σ^{(2)} n_1(z, t) I_{in}^{ω/2}(z, t),
\]

\[
\frac{n_1(ω)}{c} \frac{dI_{out}^{ω}(z, t)}{dt} + \frac{dI_{out}^{ω}(z, t)}{dz} = -σ n_1(z, t) I_{in}^{ω}(z, t).
\]

Fig. 2. Time and fractional delays between reference $I_{ref}^{ω/2}(L, t)$ and output $I_{out}^{ω/2}(L, t)$ pulses in (a) dye solution and (b) dendrite solution at three concentrations (see text). Inset, temporal profiles of a reference and output pulses.

Fig. 3. Distortion and transmission of calculated $I_{out}^{ω/2}(L, t)$ pulses in (a) dye solution and (b) dendrite solution at three concentrations (see text).
is self-affected during its propagation. 

\[ \frac{dn_1(z, t)}{dt} = -\sigma^{(1)} n_1(z, t) I_\omega(z, t) - \sigma^{(2)} n_1(z, t) I_{\omega/2}^2(z, t) + \frac{n - n_1(z, t)}{\tau}, \]

\[ \frac{n_r(\omega/2) dI_{\omega/2}(z, t)}{c dt} + \frac{dI_{\omega/2}(z, t)}{dz} = -2\sigma^{(2)} n_1(z, t) I_{\omega/2}^2(z, t), \]

\[ \frac{n_r(\omega) dI_\omega(z, t)}{c dt} + \frac{dI_\omega(z, t)}{dz} = -\sigma^{(1)} n_1(z, t) I_\omega(z, t), \]

and were integrated as before. Figure 4 shows the delay, distortion, and transmission of a weak pulse (532 nm, 1 nJ energy focused in 100 μm, Gaussian-shaped with \( t_p = 100 \) or 50 ps) propagating across a solution of dye \((n = 1 \times 10^{23} \text{ m}^{-3}, \sigma^{(1)} = 1 \times 10^{21} \text{ m}^{-2})\) in an \( L = 1 \) cm cell for different relative delays between pump and probe pulses. The medium was prepared with an Nd:YAG pulse (1064 nm, \( t_p = 100 \) ps, \( I_{\omega/2,h}^0/2 = 1.6 \times 10^{14} \text{ W m}^{-2} \)) with \( n_r(\omega/2) = 1.3, n_r(\omega) = 1.4. \) While the pump pulse has no distortion for these sets of parameters, significant delays can be achieved with very low distortion, no pulse peak splitting, and good transmission (see inset in Fig. 4) for the probe pulse. Larger delays, but also larger distortions, are obtained when pump and probe pulses have the same duration. The use of two different wavelengths has the added value of being easy to separate experimentally. This behavior is worth being studied in other systems such as a doped glass or a semiconductor with a resonance at the telecommunications wavelength 1.55 μm.

In conclusion, we have shown that TPA in a system with a resonance at frequency \( \omega \) can be employed to delay pulses in two different regimes: two-photon slow light (the pump pulse at \( \omega/2 \) is self-affected during its propagation) and two-photon-assisted slow light (the propagation of a weak pulse at \( \omega \) is affected by an intense pump pulse at \( \omega/2 \)). These results could motivate research in materials with large two-photon absorption cross sections at 1.55 μm, to be used in fiber telecommunications technology.

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