Slowdown of nanosecond light pulses with photorefractive two-wave mixing

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In the demonstrations of slow light in photorefractive (PR) materials, the delays are only reported for long pulse durations (ms to s) that are in discrepancy with typical ns-pulse length in data processing applications. Here we demonstrate that the slowdown of light in a PR crystal can be also achieved in the nanosecond regime at room temperature. We experimentally show that the use of a pulsed laser with a high peak intensity may indeed reduce the $Sn_2P_2S_6$ crystal response time to a scale of ns and achieve a two-wave mixing (TWM) gain of 7.9 cm⁻¹. We show the slowdown of light pulses whose duration ranges from 10 to 100 ns with a fractional delay up to 0.3. These results are similar to those obtained in optical fibers, except that the slow light with PR TWM is achieved through smaller propagation lengths and at wide wavelength ranges. This could reveal potential applications of PR slow light systems and have a significant impact for future optical communications systems.

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Since the last decade, the deceleration of light pulses in high-dispersion nonlinear materials such as optical fibers [1,2] and photonic [3] and photorefractive [4–8] crystals, has attracted great interest for its potential applications, including controllable optical delay lines and buffers and quantum memories [9,10]. Recently, it has been studied in a wide range of photorefractive (PR) crystals such as $BaTiO_3$ [11] and $Sn_2P_2S_6$ (SPS) [4] using different nonlinear interaction mechanisms such as two-wave mixing (TWM) [4,11], four-wave mixing [12], and beam fanning [5]. All these processes can increase the dispersion in the PR material, hence allowing the amplification and slowing down of the output pulse. For example, using TWM, group velocities down to 1 cm/s are measured in different PR crystals [4,11] at room temperature.

So far, studies of PR nonlinear interactions leading to slow light have been limited to the so-called continuous regime. TWM is performed with a cw beam laser, which is split into two coherent beams, i.e., a continuous pump and a probe signal modulated by an electro-optic modulator [4]. In the cw regime, the slowdown of the light pulse can be achieved at visible [4,11] or infrared wavelengths [5] and not necessarily with high-intensity lasers. However, the major challenge that PR slow-light technologies are facing is that the group velocity is only reduced for light pulse duration equal to or superior to the material response time [4,5]. Indeed, the difficulty of decelerating very short pulses results from the fact that the PR crystal response time in the cw regime at visible wavelength is quite long, typically more than 1 ms for SPS (1 s for BaTiO₃), hence limiting to use of the PR effect for short time applications.

The key to slowdown short light pulses in a PR crystal is the use of a high pulse intensity laser that will be able to reduce the response time of the material to the μ s or ns timescale. Therefore, other applications can be foreseen for these systems as they present several advantages compared to other approaches that perform well at the microsecond and nanosecond timescale [1,13]. The nonlinearity in some PR crystals such as SPS extends over a wide range of wavelengths (from 633 to 1550 nm), including near IR range which contrasts with the optical fibers.

In this Letter, we present TWM experiments in a SPS with short pulses at the ns timescale. A maximum PR gain value of 7.9 cm^{-1} is measured at room temperature. This value is equivalent to those reported in other PR crystals in this regime [14-16]. As for the CW regime, we show that the PR gain value measured at a nanosecond timescale depends on the input beam intensities, the polarization, and the period of the grating recorded by the coupling of the pump and the probe pulses. In a second part, we show that the TWM process in the nanosecond regime can reduce the PR material response time τ to a magnitude of the same order as the laser pulse durations, in our case the ns timescale. Also, it is possible to slow down shorter light pulses and reduce the output pulse distortions. Delays that vary from 1.4 to 25 ns are measured for pulse duration between 10 and 100 ns. The slow light is achieved for pulses of the same duration as used in optical fibers [17] but over a shorter propagation length. Using the stimulated Brillouin scattering process in a km-long optical fiber, they observe a delay of 25 ns and a fractional delay of 0.4 for a 63-ns pulse duration.

Figure 1 shows the experimental setup, which uses a Sbdoped SPS crystal with dimensions $8.1 \times 8 \times 8.1 \text{ mm}^3$ along *x*, *y*, and *z* axes. The axes are defined in the same manner as in other works [5,6,18,19]. This sample ensures a strong PR gain

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FIG. 1. Experimental setup of slow light with TWM using a pulsed laser at 1064 nm; *P* is the polarizer, *A* is the attenuator, *BS* are the separator, $M_{1,2}$ are the mirrors, $D_{1,2}$ are the detectors, and *L* is the lens. P_s is the spontaneous optical axis of the SPS crystal.

and has been used for light pulse group velocity manipulations [19] at the visible wavelength (633 nm). It is characterized by a response time of the order of 10 ms and a low absorption coefficient $\alpha < 1 \text{ cm}^{-1}$ at $\lambda > 700 \text{ nm}$ [18]. The laser used in the experiment is a Quantel double YAG-Nd laser series that can generate light pulses from 7 to 100 ns with a repetition rate of 20 Hz and near field beam diameter of 2.6 mm (for a pulse duration of 7 ns). It can operate at 1064 nm with a maximum energy of 17 mJ. The laser beam is split into two arms corresponding to the pump and the probe pulses with different input powers ($P_s = 0.1P_p$). The two light pulses with the same widths are injected through the z face of the PR SPS crystal and propagate in the XZ plane. The intensity profile of the input and output pulses are measured with two amplified NewFocus Model 1611 IR 1-GHz low noise photoreceiver and analyzed by a digital oscilloscope of 4 GHz bandwidth.

The first analysis of the pulse intensity profiles measured by the photodetectors shows that the transmitted signal pulse gets amplified at the output of the sample when the signal pulse is overlapped with the pump pulse. It is worth mentioning that the basic theory of the TWM effect in a PR crystal has been studied in detail in several works [4,20,21]. To determine if this amplification is due to the photorefractive TWM between the pump and the signal pulses, we measure the values of the gain Γ under different parameters: the incidence angle θ (or, equivalently, the grating spatial period $\Lambda = \lambda/2\sin\theta$) and the polarization of the input pulse. In both cases, we measure the intensity of the signal at the input and output of the SPS crystal in the presence of the pump pulse and calculate the resulting amplification related to the gain $\Gamma = (1/d) \ln(I_{s \text{ with pump}}/I_{s \text{ without pump}})$, where d is the sample thickness.

In the following, the gain values achieved with a pulsed laser are analyzed and compared to those obtained using a cw laser at 1064 nm in the same sample. First, we measure Γ versus the ratio between the pump and signal pulse intensities for horizontal (*x*), vertical (*y*), and azimuthal (*x*-*y*) or 45° polarization. The results obtained with pulse intensity $I_L = 2.8$ W/cm² and duration of 10 ns [Fig. 2(a)] are compared to those achieved with a cw laser with $I_L = 1.5$ W/cm²



FIG. 2. Photorefractive gain in SPS doped Sb as a function of the ratio between the pump and signal beams for horizontal (parallel to the spontaneous polarization of the crystal), vertical, and 45° azimuthal polarization. The incident angle between the two pulses is $\theta = 45^{\circ}$. (a),(b) The results achieved with a pulsed laser and cw laser respectively at $\lambda = 1064$ nm. (c) The photorefractive gain of the Sb-doped SPS crystal versus the spatial period Λ for a pulsed and cw (c) laser at $\lambda = 1064$ nm with intensities of 2.8 and 1.5 W/cm² respectively. In both cases, the results are obtained for the case of the horizontal polarization.

[Fig. 2(b)]. By analyzing the curves of Figs. 2(a) and 2(b), we can note that in both the cw and the pulsed regime, the gain increases with the ratio of input intensities. From Figs. 2(a) and 2(b), we note that Γ reaches higher values in the case of horizontal polarization (parallel to the axis of the crystal spontaneous polarization) because this geometry corresponds to the largest component of the electro-optic tensor r_{111} [6,18]. On the other hand, the gain achieved with TWM at the nanosecond regime is larger than the one measured with the continuous pump. This result can be explained by the increase of the photoinduced charge mobility in the SPS crystal caused by the high input intensity of the pulsed laser. Or, in this case, the TWM is produced by other physical processes besides the diffusion (continuous case) such as the two-photon absorption process [22].

In addition to the input intensity and the polarization, we show that the PR effect depends on the spatial period of the grating index. Figure 2(c) shows Γ values as a function of the spatial period Λ . The results demonstrate that the increase of the PR gain is observed for small values of the spatial period, typical for charge transport by diffusion [23]. For $\Lambda = 1.4 \ \mu$ m, maximum gains Γ up to 4.3 and 3.2 cm⁻¹ are respectively measured in the case of the pulsed light and the case of the cw beams. The results of Fig. 2 confirm that the amplification of the pulse changes with light polarization, input intensity, and spatial period. It can then be concluded that the gain results from the PR TWM process between the



FIG. 3. Temporal envelopes of the normalized input (black line) and output (red line) pulses as a function of the time for $I_L =$ 2.8 W/cm². (a) Time delay $\Delta \tau = 0$ ns for $t_0 = 8$ ns, (b) $\Delta \tau = 1.6$ ns for $t_0 = 14$ ns, (c) $\Delta \tau = 4$ ns for $t_0 = 28$ ns, (d) $\Delta \tau = 7.2$ ns for $t_0 = 34$ ns, (e) $\Delta \tau = 10$ ns for $t_0 = 42$ ns, and (f) $\Delta \tau = 17.4$ ns for $t_0 = 70$ ns.

pump and signal pulses. In the following, we show that as the TWM at pulsed (nanosecond) regime permits the light pulse amplification at the output of the Sb-doped SPS crystal, it appears that this method can also reduce the response time τ of the crystal. Indeed, by varying the input intensity I_0 between 2.5 and 4 W/cm², τ (inversely proportional to I_0) can vary between 32 and 30 ns. Therefore, it permits the slowdown of ns light pulses at room temperature. The analysis of the pulse profiles at the input and output of the crystal shows that the output pulse amplifies and its maximum is shifted compared to the input one. Figure 3 shows the temporal envelopes of input (black line) and output (red line) pulses for different input pulse widths $t_0 = 8$, 14, 28, 34, 42, 70 ns for (a)–(f), respectively. No delay is measured for an input pulse of 8 ns. However, if the input pulse duration increases, we observe that the maximum of the output pulse is shifted compared to the input one and delays $\Delta \tau$ of 1.6, 4, 7.2, 10, and 17.4 ns are respectively measured for (b)-(f). On the other hand, in all cases, we note that the output pulse widening is smaller than the one observed for the slow light using a cw red laser [4]. A 10-ms pulse widens by twice its initial width at the cw regime [4]; while a 20-ns pulse widens only by a factor of 1.05 of its initial duration at the nanosecond regime. This means that the TWM in the nanosecond regime can reduce the distortion factor of the ns output pulse when compared to the cw regime. Indeed, at the nanosecond regime, the delay and the output pulse profile do not only depend on the PR gain and pulse



FIG. 4. Temporal envelopes of the normalized input (black line) and output (red line) pulses as a function of the time for input pulse duration $t_0 = 32$ ns. (a) For $\Gamma = 5.9$ cm⁻¹, the delay is of 9.2 ns. (b) For $\Gamma = 7.9$ cm⁻¹, the delay is of 12.4 ns.

duration of the input signal but also on the duration of the pump pulse. Also, we note that when the input pulses (signal and pump) have the same duration, the output pulse does not distort even if we increase the gain.

We also analyze the dependency of the delay and of the fractional delay, which is the ratio between the delay and the full output width at half maximum as a function of the PR gain and the width of the input pulses. First, we adjust the duration of the input pulse for example to $t_0 = 32$ ns. For the laser pulse intensities of 3.4 and 4.7 W/cm², we measure respectively a photorefractive gain of $\Gamma = 5.9$, 7.9 cm⁻¹. Figure 4 shows the output pulse intensities corresponding to the measured gains. As shown in Fig. 4, the delay values vary with the photorefractive gain. Delays of $\Delta \tau = 9.2$, 12.4 ns are measured respectively, for $\Gamma = 5.9$, 7.9 cm⁻¹. In Fig. 5, we plot the variation of delay and fractional delay of 32 ns input pulse as a function of the PR gain. We note that the delay time increases with the Γ and its values vary from 9.2 to 12.4 ns. The resulting fractional delay increases also with Γ . A maximum fractional delay of 0.3 is measured for $\Gamma = 7.9 \text{ cm}^{-1}$.

Next, we analyze the slow light performances as a function of the input pulse duration. The results of Fig. 6 are achieved for $\Gamma = 6 \text{ cm}^{-1}$.

In Fig. 6(a), it is seen that the delay increases with the input pulse duration and its maximum value of 25 ns (corresponding to group velocity $v_g = 400$ km/s) is achieved for pulse duration $t_0 = 100$ ns and laser intensity $I_L = 3.8$ W/cm². This output pulse group velocity is reduced by a factor of 1000 compared to the speed of light in the vacuum because the



FIG. 5. Performances of the slow light as a function of the photorefractive gain. Panels (a) and (b) are, respectively, time delay $\Delta \tau$ and fractional delay FD as a function of Γ .



FIG. 6. Performances of the slow light pulses according to the input pulse duration t_0 for input laser intensity $I_L = 3.8 W/cm^2$ and $\Gamma = 6 cm^{-1}$. (a) Delay curve, (b) output pulse durations and (c) fractional delay.

TWM at nanosecond timescale increases the dispersion of the material which permits decreasing v_g . The values of the delay are similar to those measured in the optical fiber for the same pulse widths [17]. It is worth noting that in this case, like for slow light in optical fibers, the group velocity is not reduced as much as in our earlier works in PR crystals [4,5] because the propagation time in 8 mm crystal is much longer than the pulse width. Figure 6(b) shows that the output pulse width increases with the input pulse duration. The output pulse widening is observed for pulse duration less than 40 ns and is smaller than the one for the CW TWM. Instead of widening, the pulse is compressed at the output of the crystal for $t_0 > 70$ ns. However, in all cases, the corresponding fractional delay remains smaller than the one achieved in [4]. A maximum fractional delay value FD = 0.3 is measured for pulse width $t_0 = 100$ ns and $I_L = 3.8$ W/cm².

In conclusion, we have reported TWM in the nanosecond regime in a PR SPS:Sb crystal. We have demonstrated a large gain value $\Gamma = 7.9 \text{ cm}^{-1}$ and a crystal response time close to a few ns for laser intensity of 4.7 W/cm². This method is used to demonstrate the slowdown of the light in a PR SPS sample with pulse durations which vary from 10 to 100 ns. A maximum delay value of 25 ns is measured for 100 ns pulse, corresponding to a group velocity $v_g = 400$ km/s and a fractional delay close to 0.3. The slow light performances achieved here are similar to those reported so far using nonlinearities in optical fibers, but with the additional advantage that PR nonlinearity extends over a wide range of wavelengths from visible to far-infrared [24]. By the end, the TWM in the nanosecond regime has distinctive features and advantages compared to the known TWM in the CW regime. It enables us to significantly reduce the response time of the crystal and permits the slowdown of nanosecond pulses. the achieved fractional delay is low compared to the one obtained in the cw regime [4]. The slow light performance for nanosecond pulses could be improved by increasing the PR TWM gain by means of a higher intensity of the input pulses, or by using a PR crystal with a response time longer than the signal pulse duration. In the future, further systematic studies on TWM in pulsed regime will be performed in different types of SPS crystals. In addition, the value of the pump pulse duration can have a significant effect on the delay and the output pulse distortion. Increasing the pump pulse duration compared to the signal pulse width may improve the value of the fractional delay.

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- Y. Okawachi, J. E. Sharping, C. Xu, and A. L. Gaeta, Large tunable optical delays via self-phase modulation and dispersion, Opt. Express 14, 12022 (2006).
- [2] L. Thévenaz, Slow and fast light in optical fibres, Nat. Photonics 2, 474 (2008).
- [3] T. Baba, Slow light in photonic crystals, Nat. Photonics 2, 465 (2008).
- [4] N. Bouldja, M. Sciamanna, and D. Wolfersberger, Improved slow light performances using photorefractive two-wave mixing, Opt. Lett. 44, 1496 (2019).
- [5] N. Bouldja, M. Sciamanna, and D. Wolfersberger, Slow light with photorefractive beam fanning, Opt. Express 28, 5860 (2020).

- [6] A. Shumelyuk and S. Odoulov, Light pulse manipulation in sn₂p₂s₆, J. Opt 12, 104015 (2010).
- [7] G. Zhang, F. Bo, R. Dong, and J. Xu, Phase-Coupling-Induced Ultraslow Light Propagation in Solids at Room Temperature, Phys. Rev. Lett. 93, 133903 (2004).
- [8] S. Residori, U. Bortolozzo, and J.-P. Huignard, Slow and Fast Light in Liquid Crystal Light Valves, Phys. Rev. Lett. 100, 203603 (2008).
- [9] A. Walther, A. Amari, S. Kröll, and A. Kalachev, Experimental superradiance and slow-light effects for quantum memories, Phys. Rev. A 80, 012317 (2009).
- [10] B. Wu, J. F. Hulbert, E. J. Lunt, K. Hurd, A. R. Hawkins, and H. Schmidt, Slow light on a chip via atomic quantum state control, Nat. Photonics 4, 776 (2010).

- [11] E. Podivilov, B. Sturman, A. Shumelyuk, and S. Odoulov, Light Pulse Slowing Down Up to 0.025 cm/s by Photorefractive Two-Wave Coupling, Phys. Rev. Lett. 91, 083902 (2003).
- [12] K. Shcherbin, G. Gadret, H. R. Jauslin, A. Kamshilin, and P. Mathey, Slowing down of light pulses using backward-wave four-wave mixing with local response, J. Opt. Soc. Am. B 32, 2536 (2015).
- [13] L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, Light speed reduction to 17 metres per second in an ultracold atomic gas, Nature (London) **397**, 594 (1999).
- [14] W. She, Q. Wu, Q. Li, Z. Yu, and D. Mo, Picosecond light pulse shaping in photorefractive crystal by two-wave coupling, Opt. Commun. 101, 65 (1993).
- [15] N. Huot, J. Jonathan, G. Roosen, and D. Rytz, Two-wave mixing in photorefractive batio₃: Rh at 1.06 μm in the nanosecond regime, Opt. Lett. **22**, 976 (1997).
- [16] M. S. Petrovic, A. Suchocki, R. C. Powell, G. C. Valley, and G. Cantwell, Picosecond two-beam coupling and polarization rotation by scalar gratings in undoped cadmium telluride at 1.064 μ m, Phys. Rev. B **43**, 2228 (1991).
- [17] Y. Okawachi, M. S. Bigelow, J. E. Sharping, Z. Zhu, A. Schweinsberg, D. J. Gauthier, R. W. Boyd, and A. L. Gaeta, Tunable All-Optical Delays Via Brillouin Slow Light in an Optical Fiber, Phys. Rev. Lett. 94, 153902 (2005).

- [18] T. Bach, M. Jazbinšek, G. Montemezzani, P. Günter, A. A. Grabar, and Y. M. Vysochanskii, Tailoring of infrared photorefractive properties of sn₂p₂s₆ crystals by *te* and *sb* doping, J.Opt. Soc. Am. B 24, 1535 (2007).
- [19] A. Grabar, P. Mathey, and G. Gadret, Manipulation of fast light using photorefractive beam fanning, J. Opt. Soc. Am. B 31, 980 (2014).
- [20] A. Shumelyuk, S. Odoulov, D. Kip, and E. Krätzig, Electricfield enhancement of beam coupling in sn2p2s6, Appl. Phys. B 72, 707 (2001).
- [21] N. V. Kukhtarev, V. B. Markov, S. G. Odulov, M. S. Soskin, and V. L. Vinetskii, Holographic storage in electrooptic crystals.: I. steady state, *Landmark Papers On Photorefractive Nonlinear Optics* (World Scientific, Singapore, 1995), pp. 37–48.
- [22] G. C. Valley, J. Dubard, A. L. Smirl, and A. M. Glass, Picosecond photorefractive response of gaas: El2, inp: Fe, and cdte: V, Opt. Lett. 14, 961 (1989).
- [23] S. G. Odoulov, A. N. Shumelyuk, U. Hellwig, R. A. Rupp, and A. A. Grabar, Photorefractive beam coupling in tin hypothiodiphosphate in the near infrared, Opt. Lett. 21, 752 (1996).
- [24] R. Mosimann, Photorefractive effects in $Sn_2P_2S_6$ at near bandgap and telecommunication wavelengths, Ph.D. thesis, ETH Zurich, 2008.