Controllable Snail-Paced Light in Biological Bacteriorhodopsin Thin Film

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We observe that the group velocity of light is reduced to an extremely low value of 0.091 mm/s in a biological thin film of bacteriorhodopsin at room temperature. By exploiting unique features of a flexible photoisomerization process for coherent population oscillation, the velocity is all-optically controlled over an enormous span, from snail-paced to normal light speed, with no need of modifying the characteristics of the incident pulse. Because of the large quantum yield for the photoreaction in this biochemical system, the ultraslow light is observed even at low light levels of microwatts, indicating high energy efficiency.

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Ultraslow light is receiving a lot of attention in view of its interesting basic physics and potential impact on photonic technology [1,2]. The group velocity of light is reduced significantly in atomic vapor [3–6] by using electromagnetically induced transparency that permits light pulses to propagate through an opaque medium due to a quantum interference effect. Recently, two-wave coupling has been used to generate ultraslow pulse for which the energy is obtained from the diffraction of a strong permanent pump beam by a temporal grating with delayed buildup in a photorefractive crystal [7]. The technique of coherent population oscillation (CPO) [8–10] is also exploited to reduce the group velocity [11–13]. The beating between pump and probe leads to periodic modulation of the electronic-state population, creating a narrow spectral dip in the probe absorptive spectrum. The group velocity was reduced to 57.5 m/s propagating in a 7.25 cm long Ruby rod [11] and to 9600 m/s in quantum wells at a temperature of 10 k [13]. In these CPO studies, however, it is not feasible to achieve wide all-optical control of the light speed, with fixed incident pulse width and amplitude, due to the intrinsic property of electronic states characterized by fixed lifetime and transition rate. Although the group velocity may be varied by changing the incident pulse length or modulation frequency, it modifies the information carried by the input beam, which is a drawback for applications in optical data transport, computing, and storage.

In this Letter, we report interesting results on reducing the group velocity to an extreme value of only 0.091 mm/s using a photoisomerization system of biological bacteriorhodopsin (bR) at room temperature. More importantly, the light speed is controllable over a wide span from snail-paced to normal light speed with no need of changing any characteristics of the incident pulse. Unlike previous electromagnetically induced transparency and CPO experiments that were restricted to electronic states associated with atomic gases or crystals, in our system there are several chemically isomerized states (i.e., photoisomers) involved in the quantum effects for slow light. Figure 1(a) shows a schematic of the bR photocycle dynamics. The system is flexible and all-optically controllable since the process of photoisomerization has two different directions depending on the wavelengths, intensities, and polarizations of the incident light sources [14,15]. This offers a wide variety of possibilities for controlling photons. Because of the large quantum yield of the photoreaction, the ultraslow light can be observed even at microwatt levels, about five orders less than in crystals [11], indicating high energy efficiency. In addition, our study has potential applications to optical switching [14], specifically, all-optical switching with controllable delay using the same film. This could be particularly useful for an all-optical router, which directs information from one point to another in an all-optical network [2]. The realization of an all-optical router requires an optical buffer with all-optical switching schemes. Since a lot of information arrives at one point at the same time, the device needs to have the capability of wide all-optical control of the light speed, in order to delay and direct data according to their priority. Our results indicate that the potential for an optical buffer

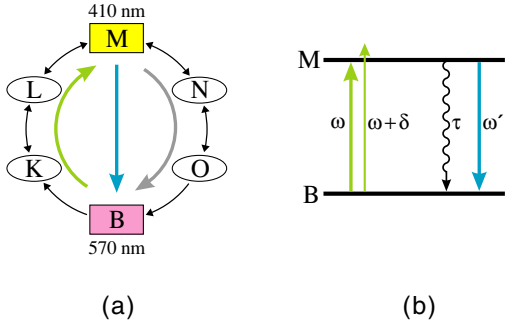

FIG. 1 (color online). Photoactive dynamics of bR molecule. (a) Schematic of bR photocycle. (b) Two-level model. The bR molecules can be excited from the B to M state by a pump beam with frequency ω and a probe beam with frequency $\omega + \delta$. The M state relaxes thermally back to the initial B state with lifetime τ or goes back directly when a blue beam with frequency ω' is incident.

and optical switch may be achieved simultaneously using one film.

We use a polymer thin film doped with bR molecules in our experiments. The thickness (L) of the bR polymer film is about 100 μm with an optical density (OD) of 5 at 570 nm. Bacteriorhodopsin has a molecular structure related to the human visual pigment rhodopsin, and is a photoactive retinal protein existing in the purple membrane of *Halobacterium salinarum* responsible for energetic processes in the bacterium [16,17]. The bR molecule is very resistant to degradation by environmental perturbations of heat and light [18–20] because of its adaptation to the highly saline, aerobic, and hot environments where the halobacteria normally grow. In the bR photocycle, the initial B state has an absorption band at 570 nm, while the long-lived M state has one at 410 nm. Upon excitation of the initial state, the molecule goes through the K, L intermediate states with short lifetimes to the relatively long-lived M state, and relaxes back to the initial state through the N, O steps spontaneously. The M state can also revert to the initial state directly upon excitation with blue light. When using light sources with wavelengths close to B and M bands, as in our experiments, the most relevant states in the bR photocycle are the B and M isomers, because the populations of the remaining short-lived intermediate states can be neglected. Accordingly, we approximate the photoactive dynamics of bR by using a simple two-level model as shown in Fig. 1(b). The unique advantage of this system is that one can all-optically control the process of photoisomerization, including isomerization directions, rates, and lifetimes.

To create a narrow spectral hole in the bR film, we use the technique of coherent population oscillation. The 568 nm light from a Ar-Kr ion laser is modulated at a frequency f by using an electro-optic modulator with a sinusoidal amplitude. The resulting pulse trains contain the beating beams of the pump $Ee^{-i\omega t}$ and the probe $E_1 e^{-i(\omega+\delta)t}$, where $\delta = 2\pi f$ is the detuning of the probe frequency from the pump frequency ω. The beam diameter is about 2 mm on the bR film. The initial B state of bR absorbs 568 nm light transforming to the M state. The beating between the pump and probe causes the molecular population to coherently oscillate between the B and M states in the bR photocycle at the beat frequency δ. The coherent population oscillation induces a dipole moment at a frequency $\omega + \delta$ with amplitude proportional to the pump field E. The amplitude of this dipole moment depends on the detuning frequency as well. In the region of resonance modulations $\delta \leq 1/\tau$, this is much enhanced. The induced dipole moment interferes with the dipole driven directly by the probe field, leading to a reduction in the absorption of the probe with an extremely narrow frequency interval ($\sim 1/\tau$). This indicates that the refractive index increases rapidly over the same spectral region, facilitating the enlargement of the group index $n_g = n(\omega) + \omega(dn/d\delta)$ and resulting in the ultraslow group velocity $v_g = d\omega/dk = c/n_g$. For a two-level resonant electronic-state system, the coherent population oscillation with wave mixing can be described by using the optical Bloch equations [11,21].

In our system, besides the direction of $B \rightarrow M$ photoisomerization induced by the 568 nm light, the bR molecule can also undergo $M \rightarrow B$ isomerization in the opposite direction induced by a blue beam. Thus, the coherent population oscillation induced by the 568 nm beam beating is controllable by the blue beam. The total isomerization rate γ from the M to B state depends on the intensity I_m of blue light and the thermal lifetime τ of M state, i.e., $\gamma = (1 + AI_m)/\tau = (1 + I_M)/\tau$, where A is a constant determined by both the incident light field and the molecular characteristics of the two-isomer isomerization system [15]. We can thus give the group index due to the coherent population oscillation as

$$n_g = n(\omega) + \frac{\alpha_0 c\tau I_B(1 + I_M)}{2[(1 + I_B)^2(1 + I_M)^2 + (\delta\tau)^2](1 + I_B)} = n(\omega) + \Delta n(\delta), \quad (1)$$

where α_0 is the linear absorption coefficient, $I_B = I_b/I_s = \kappa^2\tau\tau' E^2$ is the normalized intensity for 568 nm light with $\kappa = 2|\mu|/\hbar$, here μ is the transition dipole moment, and τ' is the dipole dephasing time. For a very large group index with a thin film, the delay ΔT of the ultraslow light pulse can be described as

$$\Delta T = \Delta n(\delta) L/c. \quad (2)$$

The first reaction of bR in the photocycle is a light-induced photoisomerization of the active chromophore retinal, which is located in the middle of bR molecule. This occurs with a large quantum yield of 65% and is completed within 200 fs [22]. In addition, bR has little fluorescence during the photocycle, which further reduces the energy consumption for the coherent population oscillation. The B state can be pumped to the M state at light levels as low as 5 μW at 568 nm. In our experiments of slow light, the signal pulse emerging from bR film is compared with a reference pulse picked off before the film using a beam splitter. The optical path through the bR film has the same length as the reference path. The signal and reference pulses are incident on two identical fast detectors and monitored simultaneously using a digital oscilloscope. Figure 2(a) shows the temporal evolution of the signal and reference pulses with a 568 nm pump intensity of 0.05 mW, in the absence of the blue beam. The signal pulse is clearly delayed by the bR film relative to the reference pulse. No distortion of pulse shape is observed in the delayed signal pulse. The fractional delay in our study is also better than early CPO experiments, and we believe it can be optimized further by tailoring the bR characteristics or using other organic or biological isomerization systems. The magnitude of the time delay depends

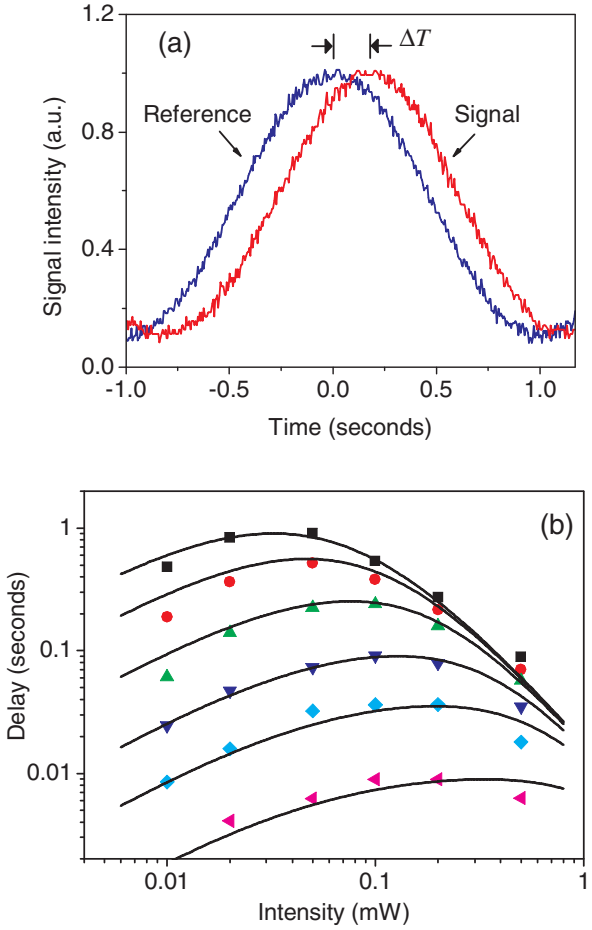

FIG. 2 (color online). Experimental results of ultraslow pulse delay. (a) Temporal comparison between the signal and reference pulses. ΔT is the delay of signal peak position relative to reference pulse. The input pump intensity is about 0.05 mW. (b) Dependence of the time delay on the pump intensity with several detuning frequencies: (■) 0.1, (●) 0.2, (▲) 0.4, (▼) 0.8, (◆) 1.4, and (◀) 3 Hz. The solid lines represent the theoretical best fit to the experimental data.

on the pump intensity and the detuning frequency as shown in Fig. 2(b). With a fixed pump intensity, the delay decreases when the detuning frequency is increased. For the same detuning frequency, there is an optimal pump intensity which produces a maximum time delay by satisfying

$$(2I_B - 1)(1 + I_B)^2 = (\tau\delta)^2, \tag{3}$$

which is derived from Eq. (1). With decreasing detuning frequency, the maximum time delay increases with its position shifting to low pump intensity. The pump intensity that causes the maximum delay is always larger than half the saturation intensity (I_s), since $I_B \geq 1/2$. For our bR film with OD $= \log_{10}(e^{\alpha_0 L})$, we obtained $\alpha_0 L = 11.5$. The theoretical curves in Fig. 2(b) are obtained from Eq. (2) with $I_s = 0.05$ mW and $\tau = 1.43$ s, which gave the best fit to the data. These values agree well with our measurements of the M-state lifetime and saturation absorption and are also in the range of other results for bR materials [23]. Our experiments show that the group velocity can be reduced to 0.091 mm/s obtained by using $\Delta T = 1.1$ s and $L = 100$ μm.

It is obvious that the group velocity can be varied by changing the modulation frequency. However, this modifies the information carried by the input beam. An important feature for the bR film is that the group velocity is controllable by applying a blue beam. In this experiment, keeping modulation frequency and intensity of the 568 nm beam constant, a 442 nm blue beam from a He-Cd laser, used as the control beam, illuminates the film in the same region overlapping the 568 nm beam. As shown in Fig. 3(a), the delay of the signal pulse is varied by turning on the blue control beam with different intensities. This effect on the delay does not rely on modifying any characteristics of the incident 568 nm pulse, such as the pulse width. With an increase of the blue light intensity, the delay time decreases gradually to zero as shown in Fig. 3(b). We fit the data by using Eq. (2). The previous values of the parameters $\alpha_0 L = 11.5$ and $I_s = 0.05$ mW are used here as constants. The best fit is found by using $A = 1.18$ mW^{-1} and $\tau = 0.28$ s. The lifetime is smaller than the value obtained without the blue beam, indicating that the blue beam increases the rate of thermal isomerization process in addition to creating the $M \rightarrow B$ photoisomerization process. The reduction of thermal lifetime may originate from the change of the local environment in the bR film induced by the blue beam. The control of the light speed is reversible; that is, the delay increases with decreasing intensity of blue light. The results indicate that we are able to control the group velocity from the order of 10^{-4} m/s to normal light speed. At high blue intensity, the initial coherent population oscillation induced by the 568 nm beam can be quenched and thus the delay disappears. The blue beam causes the molecules to switch back to the initial B state.

We considered the reversible control only in terms of decreasing the delay by increasing the blue beam intensity. It is quite possible to increase the delay as well by increasing the blue beam intensity. Since the linewidth of the spectral hole is related to the lifetime of M state, i.e., $\Delta_{HWHM} \propto 1/2\pi\gamma$, with the blue beam turned on, the resonance linewidth becomes wider due to the increase in the M-state decay rate. If the tuning is in the region of off resonance (or near resonance), the coherent population oscillation process can be ignited again (or enhanced) by turning on the blue beam, resulting in the control of increasing the delay.

With a faster isomerization rate from the M to B state, due to irradiation of the blue light, shorter pulses can be used in our ultraslow experiments. The time associated with thermal relaxation in bR can also be widely altered by different means, such as varying the temperature, pH, or by genetic mutation [24,25]. Thus, it is potentially possible

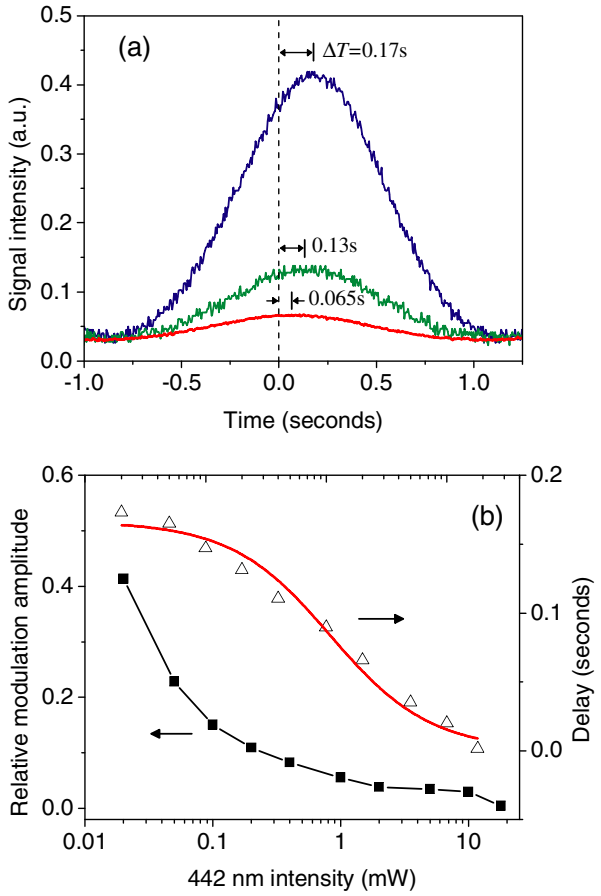

FIG. 3 (color online). Control of the delay time of the signal pulse. (a) Temporal evolution of signal pulses controlled by 442 nm beam. From top to bottom, the curves represent 568 nm signal pulses obtained with different 442 nm intensities of 0.02, 0.2, and 2 mW, respectively. Dashed line is the peak position of the reference pulse (not shown). (b) Dependence of the delay and relative modulation amplitude of the 568 nm pulse on the 442 nm intensity. The upper solid line represents the theoretical best fit to the experimental data of delay. The relative modulation amplitude (peak to valley difference) implying the strength of the coherent population oscillation also decreases with increasing the 442 nm light intensity.

to obtain various time responses using bR films for different applications in photonic devices. Alternately, the response time may also be improved by using other isomerized states involved in the bR photocycle.

In conclusion, a group velocity as slow as 0.091 mm/s is obtained using bR films. The slow light is arbitrarily controllable by using a second beam which can significantly vary the coherent population oscillation process. In view of their large quantum yield, bR films are easily bleached even at low intensities, resulting in high transmission. We may get near 100% transparency in selected films. When the bR molecules are in ''working'' status with coherent population oscillation, the transmission of the film is much higher than its linear transmission. Bacteriorhodopsin materials are commercially available and environment friendly, offering several advantages such as stability and ease of preparation and of tailoring optical properties for practical applications in photonic technology [14,26,27]. It is also possible to design new systems and exploit new quantum effects by involving other isomerized states in the bR photocycle, as well as by using new organic or biological materials for quantum information applications.

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[1] M. D. Lukin and A. Imamoğlu, Nature (London) **413**, 273 (2001).
[2] J. T. Mok and B. J. Eggleton, Nature (London) **433**, 811 (2005).
[3] L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, Nature (London) **397**, 594 (1999).
[4] D. Budker, D. F. Kimball, S. M. Rochester, and V. V. Yashchuk, Phys. Rev. Lett. **83**, 1767 (1999).
[5] A. Kasapi, M. Jain, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **74**, 2447 (1995).
[6] M. M. Kash *et al.*, Phys. Rev. Lett. **82**, 5229 (1999).
[7] E. Podivilov, B. Sturman, A. Shumelyuk, and S. Odoulov, Phys. Rev. Lett. **91**, 083902 (2003).
[8] S. E. Schwarz and T. Y. Tan, Appl. Phys. Lett. **10**, 4 (1967).
[9] M. Sargent III, Phys. Rep. **43**, 223 (1978).
[10] A. D. Wilson-Gordon, Phys. Rev. A **48**, 4639 (1993).
[11] M. S. Bigelow, N. N. Lepeshkin, and R. W. Boyd, Phys. Rev. Lett. **90**, 113903 (2003).
[12] M. S. Bigelow, N. N. Lepeshkin, and R. W. Boyd, Science **301**, 200 (2003).
[13] P.-C. Ku *et al.*, Opt. Lett. **29**, 2291 (2004).
[14] P. Wu, D. V. G. L. N. Rao, B. R. Kimball, M. Nakashima, and B. S. DeCristofano, Appl. Phys. Lett. **81**, 3888 (2002).
[15] P. Wu *et al.*, Phys. Rev. B **57**, 3874 (1998).
[16] R. R. Birge and C.-F. Zhang, J. Chem. Phys. **92**, 7178 (1990).
[17] J. K. Lanyi, Biochim. Biophys. Acta **1183**, 241 (1993).
[18] Y. Shen, C. R. Safinya, K. S. Liang, A. F. Ruppert, and K. J. Rothschild, Nature (London) **366**, 48 (1993).
[19] D. Oesterhelt, C. Bräuchle, and N. Hampp, Q. Rev. Biophys. **24**, 425 (1991).
[20] R. R. Birge, Biochim. Biophys. Acta **1016**, 293 (1990).
[21] R. W. Boyd, *Nonlinear Optics* (Academic Press, Amsterdam, 2003), 2nd ed.
[22] J. Sasaki *et al.*, Science **269**, 73 (1995).
[23] F. J. Aranda *et al.*, Appl. Phys. Lett. **67**, 599 (1995).
[24] Z. Chen, A. Lewis, H. Takei, and I. Nebenzahl, Appl. Opt. **30**, 5188 (1991).
[25] R. Korenstein and B. Hess, Nature (London) **270**, 184 (1977).
[26] A. Seitz and N. Hampp, J. Phys. Chem. B **104**, 7183 (2000).
[27] C. Bräuchle, N. Hampp, and D. Oesterhelt, Adv. Mater. **3**, 420 (1991).