Dichromatic light halting using double spin coherence gratings

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*New Journal of Physics* **13** (2011) 083012 (11pp)
Received 12 March 2011
Published 12 August 2011
Online at [http://www.njp.org/](http://www.njp.org/)
doi:10.1088/1367-2630/13/8/083012

**Abstract.** Light control by another light has drawn much attention in nonlinear quantum optics. Achieving all-optical control of the refractive index has been a key issue in all-optical information processing. Ultraslow light has been a good candidate for this purpose, where a giant phase shift can be achieved. The recent presentation of stationary light utilizing ultraslow light is an advanced example of such research. The stationary light functions as cavity quantum electrodynamics, where no high-$Q$-factor mirror pair is needed. In this paper, we report on two-color halted light pulses inside a solid medium, where the trapping time is comparable with that of ultraslow light but is much longer than quantum mapping storage time. The observed two-color halted light is achieved by means of double Raman optical field-excited spin coherence gratings, where slow light enhanced backward nondegenerate four-wave mixing processes play a major role.

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1. Introduction

Electromagnetically induced transparency (EIT) [1, 2] resulting in absorption cancellation due to destructive quantum interference in a three-level system has been used in various nonlinear quantum optics applications [3–10]. Because absorption is correlated with dispersion by the Kramers–Kronig relation [11], the group velocity of a traveling light pulse can be controlled by using another (coupling) light. Although the group velocity of probe light can be extremely slow in a single-EIT model [4–6], the interaction time for the cross-phase modulation must be determined by the group velocity of the coupling light, which is fast enough, where a large phase shift cannot be obtained. In conventional nonlinear optics such as cross-phase modulation, the use of intense light has been a critical limit. Thus, the large phase shift of weak signal light has potential applications not only in nonlinear optics, for example, electro-optic switches\(^2\), but also in quantum information processing, for example, entanglement generation [12] and quantum nondemolition measurement [13]. About a decade ago, a double-EIT scheme was proposed for a cross-phase modulation, where coupled ultraslow light pulses result in a significant phase shift [14]. However, the cross-phase modulation model based on the double-EIT scheme is somewhat complicated, where at least five energy levels and four coherent light frequencies are required. Recently, the double-EIT scheme has been efficiently modified for stationary light in a backward propagation scheme resulting in trapped light [15]. Since the first observation of optically induced Bragg grating-based stationary light [15], intensive studies have been carried out [16–18]. Lukin and co-workers proposed a large phase shift model using double standing wave gratings (Bragg gratings) in a four-level EIT system with a giant Kerr nonlinearity [19].

Nondegenerate four-wave mixing processes have been intensively studied for nonlinear quantum optics using EIT for enhanced four-wave mixing generation [3, 20, 21]. Because the nondegenerate four-wave mixing signal is proportional to pumping field as well as Raman coherence strength, ultraslow light has been adapted for ultraefficient wave mixing processes, where nondegenerate four-wave mixing signal is proportional to EIT-induced Raman coherence [22, 23]. Recent observations of slow light-based matched pulses in nondegenerate four-wave mixing processes demonstrate that a giant phase shift can be obtained by lengthening interaction time in a slow light regime, where frequency selection is an inherent property [24]. Here we demonstrate two-color stationary light in a counterpropagating nondegenerate four-wave mixing scheme. Unlike the monochromatic light trapping based on Bragg gratings (standing-wave grating) excited by standing light fields [15–19], the present dichromatic stationary light is based on counterpropagating spin coherence gratings excited by double Raman fields [25]. More specifically, the present paper is a stationary light version of [24] and [25], satisfying a phase conjugate scheme. On the other hand, the works [15–19] never satisfy phase conjugation unless all light beams are collinear. In rare-earth-doped solids, however, the selection rules are not satisfied due to crystal field interactions [26], and thus the collinear scheme is not appropriate for stationary light.

For a coherence conversion process such as that in EIT or Raman scattering, the ultimate coherence decay time is dependent on overall spin dephasing time determined by spin inhomogeneous broadening, which determines optical storage time (shown in figure 4). In a rare-earth-doped solid medium, the overall spin dephasing time is ultrashort, much less than the optical phase decay time. To overcome such a short storage time, the photon echo...
method has been adapted to the ultraslow light regime to lengthen it [6]. In the present paper, we experimentally demonstrate an ultralong dichromatic stationary light in a solid medium whose trapping time is nearly two orders of magnitude longer than the critical constraint given by the spin inhomogeneous broadening. Unlike the Bragg grating method using atom population modulation [15], the physics of the present observation of light halt is in the mutual interactions of counterpropagating moving (spin) coherence gratings. To support the validity of the observed stationary light, we perform various supporting experiments with detailed analyses in figures 4–7.

2. Experimental details

Figure 1(a) shows a partial energy-level diagram of a rare-earth Pr$^{3+}$ (0.05 at.%)-doped Y$_2$SiO$_5$ (Pr:YSO). The resonant probe pulse P experiences an absorption cancellation under the coupling light C owing to EIT [2, 3, 6]. The pumping light A, whose propagation direction is opposite to that of the coupling light C, is used to generate backward spin coherence moving gratings, resulting in the nondegenerate four-wave mixing signal D [25]. The initial population redistribution process for $\rho_{11} = 1$ is accomplished by repumping fields resonant to the excited state from other hyperfine ground states excluding state $|1\rangle$, where the third ground state is not shown [2, 3]. As a result, the 4 GHz optical inhomogeneous width is shaped for 300 kHz, which is determined by the laser jitter.

In figure 1, we chose the $^3\text{H}_4 \rightarrow ^1\text{D}_2$ optical transition in Pr:YSO for a resonance frequency of $\sim$606 nm. The angle between light beams P and C is $\sim$25 mrad, where the beams remain overlapped at overall 80% inside the sample of Pr:YSO whose length is 5 mm. All light beams are focused into the sample by the same lens. The focused beam diameter (exp(−1) in intensity) of the light beam P at the focal point is $\sim$200 μm and that for the others is $\sim$600 μm. In figure 2, the light powers of R, P, C, A and Y are 8, 0.9, 10, 20 and 20 mW, respectively. Relative frequency adjustment for R, P, C, A and Y is achieved by using acousto-optic modulators driven by radio-frequency synthesizers. The repetition rate of the light pulse train is 10 Hz and

\[ \text{Figure 1. Schematic diagram of trapped light using double spin coherence.} \]
\[ \text{(a) A partial energy level diagram of Pr$^{3+}$ doped Y$_2$SiO$_5$. (b) A pulse sequence for (a). (c) A light propagation scheme.} \]
\[ \delta = 20 \text{kHz. S represents the slow light of P. D is a diffracted signal by A, which propagates backwards if C is turned off.} \]
Figure 2. Observations of trapped light for figure 1. (a) Ultraslow light under the action of the forward coupling light C only. (b) Trapped light by adding a backward light A into (a). (c) Photon switching with Y. Red color: diffracted signal YD by Y. The boxes P, A and Y represent relative temporal positions. The control C covers all the light pulses shown in figure 1(b).

30 samples are averaged for the measurement. The ground levels $|1\rangle$ and $|2\rangle$ in figure 1 represent Kramer’s doublets $\pm 5/2$ and $\pm 3/2$ of $^3\text{H}_4$, respectively, whereas the excited level $|3\rangle$ is for $^1\text{D}_2 (\pm 3/2)$ of Pr : YSO. The Pr : YSO sample in a flow-type liquid helium cryostat is kept at a temperature of 5–6 K.

Figures 1(b) and (c) show schematic diagrams of the pulse sequence and propagation directions, respectively. Under the coupling light C, the probe P evolves into ultraslow light S due to a narrow EIT transparency window [6]. Because light–matter interaction time increases in the ultraslow light regime, the spin coherence excitation must be enhanced [21–24]. Thus, the nondegenerate four-wave mixing process must be efficiently enhanced, satisfying the phase matching condition: $k_D = k_C - k_P + k_A$. As a potential application, a delayed all-optical switching/routing has been reported recently in an ultraslow light scheme [22]. In the present paper, we set a backward slow light scheme (see figure 1(c)) and demonstrate a novel phenomenon of ultralong halted (trapped) light, the so-called dichromatic stationary light. Unlike the work [15], whose light trapping is due to population gratings excited by standing lights, the counterpropagating lights C and A in figure 1 have different frequencies, resulting in no standing-wave gratings. Instead, the pair of resonant Raman optical fields, P and C, generates spin coherence-moving gratings through the medium along the path of the light propagation [3]. A backward light A applied to the ultraslow light S generates an enhanced four-wave mixing signal D, whose propagation direction is also backward due to the phase matching condition [22, 25]. Then light pulses A and D can create another spin coherence-moving grating, but in the opposite direction. The forward ultraslow light S and the backward four-wave mixing light D are coupled together and can be halted completely.
3. Results and discussions

Figure 2 shows the result of the halted light, where the trapping time is much longer than the expected value determined by the ultrashort spin dephasing time ($T_2^S$)$^*$. The measured overall spin dephasing time is the same as that calculated from the inverse of the spin inhomogeneous width $\Delta_S$: ($T_2^S$)$^* = 1/(\pi \Delta_S)$. In Pr : YSO for the transition between states $|1\rangle$ ($\pm 1/2; ^3H_4$) and $|2\rangle$ ($\pm 3/2; ^3H_4$), the spin inhomogeneous width $\Delta_S$ is $\sim 30$ kHz [26], and the spin dephasing time is measured at $\sim 10 \mu$s [6], whereas the measured spin phase relaxation time $T_2^S$ is ultralong ($T_2^S \sim 500 \mu$s) [27]. In figure 2(a), an ultraslow light S is shown as a reference when only the pump light C is turned on. When a balanced backward coupling light A is also turned on in figure 2(b), both ultraslow lights S and D become trapped inside the medium until the light A (or the light C) is turned off. As shown in figure 2(b), the trapped light (see red dotted circle) regenerates or accelerates in the medium when the light A is turned off. The amplitude reduction of the regenerated light via atom interactions must be mainly due to the spin phase decay [22]; this will be discussed in figure 3. The shortened pulse duration of D (regenerated light) is due to atom depopulation by A in state $|1\rangle$, resulting in an accelerated group velocity (discussed in figure 5) [28].

To prove the origin of the regenerated light (see red dotted circle in figure 2(b)), a slow-light-based photon-switching technique is used for nondegenerate four-wave mixing processes [22–25]. Here, the photon switching is a direct result of the coherence conversion process from slow light-coupled spin coherence to the four-wave mixing signal. For this, a test light Y is applied to the regenerated light, where the frequency of the test light Y is 1 MHz blue-detuned from the light C, and its propagation direction is forward. Unlike the regenerated light in figure 2(b), the Y-diffracted light (YD: red curve) in figure 2(c) satisfies the following: $k_{YD} = -k_C + k_P + k_Y$; $\omega_{YD} = -\omega_C + \omega_P + \omega_Y$ [22]. In figure 2(c), the angle between Y and C is the same as that between P and C at 25 mrad. As shown in figure 2(c), the halted light intensity

\[ \text{Figure 3.} \text{ Trapped light intensity versus pulse duration of A. Inset: overlapped regenerated light signals for various pulse durations of A. Solid line: best-fit curve of equation (1). The open squares are not shown in the inset.} \]
Figure 4. Storage time versus spin dephasing. (a–d) Trapped light with temporal delay $T$. (e) Regenerated light intensity versus delay $T$. (f–i) Stopped light with temporal delay $T$. (j) Retrieved signal versus delay $T$.

is severely reduced due to depletion of spin coherence by $Y$. Thus, the regenerated light in figure 2(b) (red dotted circle) directly proves that the trapped light originates at the ultra-slow light $S$ as well as spin coherence $\rho_{12}$ [22]. More detailed discussions are given in figures 6 and 7. The first peak that appeared immediately after the light $A$ was turned on is coherence leakage due to both imperfect geometry of the spin gratings and partial reflection on each spin grating, as also shown in the Bragg grating-based trapped light [15].

The inset of figure 3 shows overlapped intensities of the regenerated light pulses for various pulse durations of the backward control field $A$ in figure 2(b) at a fixed delay $T$ of 3 $\mu$s. The best-fit curve for the data in figure 3 reveals a much longer decay time $\tau$ ($\tau = 588 \mu s$) in comparison with that of figure 4(e), which is nearly two orders of magnitude longer than the spin dephasing time ($T_S^*$):

$$I(t) = I_0 \exp\left(-2t/\tau\right).$$

This value $\tau$ in figure 3 is comparable with the value of spin phase decay time ($\sim 500 \mu s$) measured using a resonant three-pulse Raman echo technique [27]. This means that the trapping time of the present halted light mechanism is independent of the spin inhomogeneous broadening but dependent on slow light processes. Unlike conventional light storage methods limited by spin inhomogeneous decay time [6], the present technique shows a breakthrough in nonlinear optics using dichromatic ultralow light pulses. The present light trapping method is completely different from the quantum mapping process [6–9], as the quantum-mapping process cannot be used for the application of giant phase shift or enhanced nonlinearity. The present
method also reveals a novelty of light–matter interactions in an inhomogeneously broadened spin medium replacing the harsh constraint of ultrashort spin dephasing time by ultralong spin phase relaxation time. In addition to the enhanced nonlinear effect with longer interaction time, the use of wider bandwidth spin transitions for shorter probe pulses offers the benefit of increasing the value of the delay bandwidth product, where the delay bandwidth product has been a critical limitation in slow-light-based optical information processing [6–10].

In figures 4–7, we analyze the origin of the light trapping phenomenon observed in figure 2(b). First of all, we need to identify the same physics of spin coherence excited by EIT in figure 4. For this the regenerated light intensity (red dotted circle) is measured as a function of the elapse (delay) time of A from P and compared with that in conventional stopped light based on the quantum mapping process [6–9]. The left column of figures 4(a)–(e) is for the present halted (trapped) light. The right column of figures 4(f)–(j) is for the conventional stopped light [6–9]. As shown in figures 4(e) and (j), both show the same results, where both regenerated light intensities exponentially decrease as a function of the elapse time $T$. Here, in the right column, the optical coherence excited by P is fully transferred into spin coherence when C is turned off. This EIT-excited spin coherence, however, degrades according to overall spin phase relaxation accelerated by spin inhomogeneous broadening. In the left column, the magnitude of spin coherence read out by the light A is also decreased by the delay time $T$. Thus, the decision factor of the regenerated light intensity or storage time of the probe pulse without coupling light is due to the spin coherence degradation. For the best-fit curve, a Gaussian function is used because the transition $|1\rangle$–$|2\rangle$ is spin inhomogeneously broadened.

The measured decay time for figures 4(e) and (j) is 15 and 12.5 $\mu$s, respectively. These measured decay times indicate spin inhomogeneous width of 21 and 24 kHz, respectively. We believe that the discrepancy is due to line broadening resulting from the light–matter interactions in the left column. Thus, we have experimentally demonstrated that the trapped light in figure 2(b) has the same origin as the ultraslow light-based stopped light based on the quantum mapping process, where the converted spin coherence that rapidly decays according to the spin inhomogeneous broadening if no coupling light exists [6] plays a key role.

Figure 5(a) shows typical slow light experiments. In figure 5(b), the propagation direction of A (see figure 1(c)) is slightly tilted to break the trapping condition in figure 2(b).
demonstrated, the group velocity of the probe P is inversely proportional to the atom density in state $|1\rangle$, where the atom density can be affected by the control lights C and A [28]. In figure 5(b), the backward coupling light A applied to the same ground state together with P depletes more atoms, resulting in acceleration of the slow light S. The group velocity of the probe P depends on both atom density in state $|1\rangle$ and the control Rabi frequency. As the Rabi frequency of the forward coupling C increases, the EIT transparency window widens. According to the Kramers–Kronig relation, a widened transparency window induces a less stiff dispersion slope. Thus, the preexisting slow light should experience acceleration as the coupling C power increases as shown in figure 5(a). When the backward coupling Rabi frequency of A increases, the ultraslow light S also accelerates due to decreasing atom density in state $|1\rangle$: see figure 5(b). The frequency of A is nearly resonant with transition $|1\rangle$–$|3\rangle$. In both cases, as the group velocity increases, the pulse duration of the slow light also becomes narrower as discussed in figure 2(b).

Now we test the acceleration of the trapped light in figure 2(b) with various shapes of the counterpropagating light A. Figure 6(a) shows ultraslow light for a reference without the counterpropagating control A. The group velocity of the P must be accelerated when the control A is applied as demonstrated in figure 5(b) via atom depletion in state $|1\rangle$. When the pulse A is applied but switched off abruptly as shown in figure 6(b), the regenerated light intensity is maximized as discussed. If the pulse A is switched off slowly, the spin coherence readout

**Figure 6.** (a) Ultraslow light as a reference without the counterpropagating control A. (b) Abrupt switch-off of the backward control A (red). (c, d) Smooth switch-off of the backward control A (red).
process becomes inefficient due to an adiabatic-like process as shown in figures 6(c) and (d). Thus, figure 6 demonstrates that the efficiency of the regenerated light in figure 2(b) depends on the spin coherence readout process determined by the pulse edge shape of the backward control A, and proves that the regenerated light originates at the spin coherence excitation via the ultraslow light S-based trapped light.

Figure 7 presents the on-demand control of the trapped light shown in figure 2(b). Figure 7(a) is a reference without A. In figures 7(b)–(e), the switching-on time of the backward coupling A varies for a fixed switching-off time. As a result, figure 7 shows that the regenerated light intensity is proportional to the area of A or the amount of ultraslow light S covered by A. This leads us to conclude that the regenerated light intensity is proportional to the captured ultraslow light S by the backward coupling A. Here, the trapping time of light in the medium becomes lengthened up to the medium’s spin phase relaxation time as discussed in figure 3, which is 50 times longer than the spin dephasing time in the quantum mapping process as discussed in figure 4.

4. Conclusion

In summary, we have reported ultralong dichromatic stationary light in a solid medium by using electromagnetically induced transparency-based nondegenerate four-wave mixing processes, where the light trapping mechanism is based on double coherence spin excitations. The observed trapping time of the halted light is nearly two orders of magnitude longer than the critical constraint given by spin inhomogeneous width as in the quantum mapping process. This observation opens a door to potential applications of light–matter interactions in ultralow power.

Figure 7. (a–e) From top to bottom, the switching-on time of the backward coupling A (red) is varied to control the amount of trapped light. The greater the area of ultraslow light covered by A, the greater the intensity.
nonlinear optics as well as in increased delay bandwidth product using a solid medium. Such nonlinear optics may also apply for the manipulations of giant phase shift of a weak probe light. Frequency selection and phase conjugate of the regenerated light are additional advantages for all-optical information processing.

Acknowledgment

This work was supported by the Creative Research Initiative program (grant no. 2010-0000690) of the Korean Ministry of Education, Science and Technology via the National Research Foundation.

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