Optical nutation and stimulated photon echoes (SPEs) are common optical coherent transient phenomena that result from the coherent interaction between optical radiation fields and an inhomogeneously broadened absorbing medium. Optical nutation describes the modulation of transmission of a constant field interacting with the medium when the field’s duration is longer than a Rabi oscillation period. The SPE usually results from the interaction involving a sequence of three brief input pulses. Based on these two phenomena, well-established spectroscopic tools have been developed to study gaseous atomic and molecular systems and rare-earth-doped crystals.1–3

Optical nutation is used to measure the Rabi frequency of the optical field directly. The SPE is used to measure the coherence time, T2, and the excited-state lifetime, T1. With these tools, a variety of microdynamic processes of atoms, molecules, and solids, including molecular collisions, atom–atom and atom–lattice interactions, and excited-state relaxation, have been studied.

In this Letter we consider the combined effect of optical nutation and SPE, which results from two brief programming pulses, separated in time by τ21, followed by a long probe pulse with a duration of several Rabi periods and much longer than τ21. The temporal behavior of the output field, including optical nutation on the transmission and the SPEs, is investigated theoretically and experimentally. The commonly used angled (noncollinear) beam configuration was employed for spatial separation of the temporally overlapped probe and echo fields. Theoretical modeling of this angled configuration required the development of a formulation of the Maxwell–Bloch equations for an angled beam that could be numerically integrated. In this Letter we present a theoretical analysis of the optical nutation and the long probe pulse–induced SPEs for an angled beam configuration. We report what is, to the best of our knowledge, the first observation of the Rabi oscillation effects on SPEs and the existence of SPE responses in the noncausal direction, in which a typical (brief-pulse) SPE does not propagate because of spatial phase matching and the causality conditions.4,5 This study will help to clarify coherent saturation effects in optical coherent transient applications for optical storage and processing.6,7 It may also provide a novel background-free spectroscopic tool that can measure Rabi frequency, T2 and T1 simultaneously.

In general, optical coherent transient processes can be described by a set of Maxwell–Bloch equations.3 We first consider the configuration in which the input and output fields are collinear. When a resonant monochromatic plane-wave field starting at \( t = 0 \) interacts with an inhomogeneously broadened two-level atomic system, the coherent feedback from the atoms’ polarization to the input field gives rise to the nutation on the transmitted field. The oscillatory polarization has components that are in phase and in quadrature with the input field, respectively3:

\[
\begin{align*}
u(\Delta, t) &= \exp(-t/T_2)\omega_0(\Delta) \\
& \times \{1 - \cos[(\Omega_0^2 + \Delta^2)^{1/2}t]/(\Omega_0^2 + \Delta^2)\}, \\
\end{align*}
\]

where the on-resonant Rabi frequency, \( \Omega_0 = \mu E/h \), is determined by the electric dipole moment, \( \mu \), of the transition between the two energy levels and the input electric field, \( E \), under the single-dipole assumption; \( \Delta \) is the detuning from the resonance; and \( \omega_0(\Delta) \) represents the atomic population inversion between the two levels at time \( t = 0 \), which consists of inhomogeneous broadening and spectral structure created by the two programming pulses. For simplicity, we assume that the inhomogeneous bandwidth and the spectral range of the grating are much broader than \( \Omega_0 \) and the population inversion can be expressed as \( \omega_0(\Delta) = \omega_0 + \omega_0 \cos(\Delta \tau_{21}) \), where the two terms on the right-hand side represent a dc absorption background and a population spectral grating, respectively. The spectral grating period is determined by \( \tau_{21} \), the temporal separation between the two programming pulses. After integration over the detuning, the in-phase term that is an odd function of \( \Delta \) integrates to zero, and the collective polarization, \( p(t) = p_{\text{na}}(t) + p_{\text{echo}}(t) \), consists of two terms,

\[
p_{\text{na}}(t) \propto \exp(-t/T_2)\Omega_0 J_0(\Omega_0 t), \quad p_{\text{echo}}(t) \propto \begin{cases} 
\exp(-t/T_2)\Omega_0 J_0(\Omega_0 (t - \tau_{21})^2/2) & t > \tau_{21} \\
0 & t < \tau_{21}
\end{cases},
\]

where \( J_0(x) \) represents the 0th order of the first kind of Bessel function. Both terms show damped oscillations and have similar behavior when \( t \gg \tau_{21} \). Expression (1) is the term of the macroscopic polarization...
that emits the field simultaneous with the probe field known as optical nutation. Expression (2) represents the polarization that results from the spectral grating and does not start until \( \tau_{21} \) after the leading edge of the probe pulse. In this simplified case the fields induced by the two terms of the polarization can be identified as contributing to the optical nutation [Eq. (1)] and the delayed SPE [Eq. (2)]. However, it is impossible to study them separately in a collinear geometry experimentally, since all the induced and the transmitted fields overlap temporally and spatially.

To separate the temporally overlapped optical nutation and SPE fields, we resort to the angled beam configuration illustrated in Fig. 1, where programming pulses 1 and 2 propagate along \( \mathbf{k}_+ = \mathbf{k}_x + \mathbf{k}_c \) and \( \mathbf{k}_- = \mathbf{k}_x - \mathbf{k}_c \), respectively. The atomic population is then modulated with a spatial–spectral grating as the population inversion becomes \( w_0(\Delta, x) = w_0 + g_0 \cos(\Delta \tau_{21} + 2\delta) \), the spatial phase, \( \delta = k_x x \). It is well known that a probe pulse shorter than \( \tau_{21} \) has to be in the \( \mathbf{k}_+ \) direction to generate a real echo, which should propagate only along \( \mathbf{k}_- \) because of phase matching and causality.\(^4,5\) The causal (\( \mathbf{k}_- \)) and noncausal (\( \mathbf{k}_+ \)) directions are determined by the timing and the directions of the programming pulses.

Instead of a brief pulse, we consider a quasi-continuous probe field, starting at \( t = 0 \), incident along \( \mathbf{k}_+ \). The polarization-induced fields in the two directions have the following forms, respectively:

\[
E_+(t) \propto \int_0^{2\pi} \int_{-\infty}^{\infty} u(\Delta, t, x) \, d\Delta \, d\delta,
\]

\[
E_-(t) \propto \int_0^{2\pi} \int_{-\infty}^{\infty} [u(\Delta, t, x) \cos 2\delta - \nu(\Delta, t, x) \sin 2\delta] \, d\Delta \, d\delta,
\]

where \( E_+(t) \) corresponds to the optical nutation propagating along with the transmitted probe field and \( E_-(t) \) is the SPE along the causal direction.

By bringing \( u(\Delta, t, x), u(\Delta, t, x), w_0(\Delta, x) \) into expressions (3) and (4), we have

\[
E_+(t) = C \Omega_0 w_0 \pi \exp(-t/T_2) J_0(\Omega_0 t),
\]

\[
E_-(t) = 0.5 C \Omega_0 g_0 \exp(-t/T_2)
\times \left\{ -\pi \exp(-\Omega_0 \tau_{21}) + \pi J_0[\Omega_0(t - \tau_{21})^{1/2}] 
- \int_{-\infty}^{\infty} \frac{\Delta}{\Omega_0^2 + \Delta^2} \cos[(\Omega_0^2 + \Delta^2)^{1/2} t] 
\times \sin \Delta \tau_{21} \, d\Delta \right\}.
\]

where \( C \) is a common constant. Optical nutation (5) for the angled beam geometry has exactly the same form as \( p_{\text{nu}}(t) \) for the collinear case. The echo’s field [Eq. (6)], however, consists of a dc term and two time-varying terms, one of which cannot be solved analytically. We plot the fields of Eqs. (5) and (6) in Fig. 2 for \( w_0 < 0, \Omega_0 = 0.8 \, \text{MHz}, \tau_{21} = 0.3 \, \mu\text{s}, \) and \( T_2 = \infty \). The transmitted field was normalized to \( C \Omega_0 w_0 \) and the echo to \( C \Omega_0 g_0 \). One can see that the echo starts at \( t = \tau_{21} \) and oscillates around the dc background does not exist in collinear propagation.

Typically, a brief probe along \( \mathbf{k}_+ \) should not generate an echo. But a quasi-continuous probe field along \( \mathbf{k}_- \) gives a different result. Through a derivation similar to that presented above, the induced fields obtained are

\[
E_+(t) = 0.5 C \Omega_0 g_0 \exp(-t/T_2)
\times \left\{ -\pi \exp(-\Omega_0 \tau_{21}) + \pi J_0[\Omega_0(t - \tau_{21})^{1/2}] 
+ \int_{-\infty}^{\infty} \frac{\Delta}{\Omega_0^2 + \Delta^2} \cos[(\Omega_0^2 + \Delta^2)^{1/2} t] 
\times \sin \Delta \tau_{21} \, d\Delta \right\},
\]

\[
E_- (t) = C \Omega_0 w_0 \pi \exp(-t/T_2) J_0(\Omega_0 t).
\]

The optical nutation on the transmission, \( E_-(t) \), still has the same form as expression (5). However, the field in noncausal direction \( \mathbf{k}_- \) does not cancel as it would for the typical brief-pulse SPE. From Fig. 2 (bottom trace) one can see that Eq. (7) also represents an echo field with the same delay as the echo in the causal direction. The existence of this echo does not violate causality. The reason that an echo appears in the noncausal direction is that the probe pulse is longer than the delay \( \tau_{21} \). The front part of the probe pulse interacts with the grating and creates a coherence that does not generate a propagating field because of the

\[ \text{Fig. 1. (a) Angled beam configuration and (b) input timing showing the brief-pulse programming and quasi-continuous probe.} \]

\[ \text{Fig. 2. Calculated results of the induced fields from a quasi-continuous plane-wave probe in an optically thin spatial–spectral grating with } w_0 < 0, \Omega_0 = 0.8 \, \text{MHz}, \tau_{21} = 0.3 \, \mu\text{s}. \text{ Top trace, optical nutation on the normalized transmitted field. } E_+(t) \text{ and } E_-(t) \text{ are the normalized echo fields in the causal and noncausal directions, calculated from Eqs. (6) and (7), respectively.} \]
phase-matching condition. However, the probe field that is incident into the medium anytime $\tau_{21}$ later than the front part rephases this coherence into the propagating direction $\mathbf{k}_+$ and emits an echo. The echoes in both directions oscillate at the same Rabi frequency.

To verify the theory described above, we performed experiments in a 7-mm Tm:YAG (0.1-at.%) crystal with an absorption length of $\alpha L = 1.4$ and $T_2 \sim 10 \mu s$ (at 4.2 K). The resonant transition used was $^3H_4 \rightarrow ^3H_6$ at 793 nm.\(^8\) The population decay in approximately a millisecond has an insignificant effect on our experimental results. A cw Ti:sapphire laser was frequency locked to a spectral hole in another Tm:YAG crystal with $\alpha L = 4$.\(^9\) The frequency was stabilized to $\sim 10$ kHz over a time period of a signal shot experiment. The input beams were separated by an angle of $\sim 0.05$ rad and focused in the crystal to a spot of $\sim 75 \mu m$ (1/e waist). The three input pulses were created with two acousto-optic modulators, which also controlled the timing, direction, and power of the inputs. The duration of the two programming pulses was set to 100 ns, the first along $\mathbf{k}_+$ and the second along $\mathbf{k}_-$. The probe duration was 10 µs. The peak power of each programming pulse was $\sim 200$ mW while the probe was reduced to $\sim 28$ mW. The powers of the echo fields were measured in the causal ($I-$) and the noncausal directions ($I+$) at various delays. Two sets of results are shown in Fig. 3, one for $\tau_{21} = 150$ ns and one for $\tau_{21} = 300$ ns. From the experiment, we observed the following trends in the echoes in both directions: (1) The echoes are delayed by the time set by the programming pulses, (2) the echoes oscillate at the same Rabi frequency, and (3) the echoes consist of the same dc component, which decreases with delay. All these observations are consistent with the theory. From the optical nutation observed in the transmission [Fig. 3(c)], the Rabi frequency corresponding to the center of the Gaussian probe beam can be estimated to be $\sim 0.8$ MHz, assuming an optically thin medium.\(^{10}\) However, the local Rabi frequency in the medium varies with the probe intensity across the beam due to the beam’s Gaussian profile, as well as along the propagation due to the absorption. The echoes and the transmission shown in Fig. 3 are the collective effects of all these Rabi components. The damping of the oscillation on both the echoes and the transmission has two major causes. The first is the coherent decay on the time scale of $T_2 \sim 10 \mu s$. The second is that the oscillations at different Rabi frequencies average out after the first few periods. All these effects have to be considered in simulating the experimental results, this requires numerical solution of the angled beam Maxwell–Bloch equations.

In conclusion, we have analyzed the temporal behavior of echoes stimulated by probe pulses with time durations comparable to the medium’s coherence time and the Rabi oscillation period. Echo in the noncausal direction and the Rabi oscillation effects on the echoes were predicted and observed.

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References