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Phase control of photon-echo dynamics with overlapping pulse pairs

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We report on the dynamics of two-pulse photon echoes in a two-level system. We consider two different types of excitation pulse pairs: pulses with the same carrier envelope phases (CEP pulse pairs) and pulse pairs cut from the same carrier wave train (labeled as normal pulse pairs). We show that for CEP pulse pairs when the pulses overlap somewhat the photon-echo emission time is strongly sensitive to the relative phase (i.e., delay time) between the two pulses. We also show how the photon-echo emission time for such CEP pulse pairs depends on the dephasing time of the polarization ($T_2$) and the pulse width of the applied pulses. This phase sensitivity can be utilized to control the emission time of the photon echo which may prove useful in storing and retrieving light signals from an atomic ensemble. Our results also inform the interpretation of photon-echo signals at short delay time when used to measure fast coherence decay rates.

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I. INTRODUCTION

Spin echoes, also known as Hahn echoes [1], were first detected by Erwin Hahn in 1950. These echoes originate from the rephasing of spin coherence after the application of a pair of radio frequency excitation pulses delayed by time $\tau_d$. A photon echo is similarly the rephasing of the optical polarization after optical excitation by a pulse pair. Echoes have important applications in many fields including spectroscopy [2,3], quantum information processing [4], studying magnons and phonons in single crystals [5], and measurement of spin-spin or polarization relaxation times ($T_2$) [6]. The photon echo is one potential candidate for quantum storage of light in an atomic ensemble and photon echoes are being investigated for applications in long-term optical quantum memories [7–9]. Conventional photon echoes have low signal retrieval efficiency [10] and the time of the echo appearance after the second pulse is fixed near to the interpulse delay. Techniques for on-demand retrieval of the photon echo would be beneficial [11,12] and techniques like controlled reversible inhomogeneous broadening (transverse as well as longitudinal) are being developed to increase the signal retrieval efficiency of such systems [13]. Even after 65 years, the interest in echo phenomena is still rising as further applications emerge; therefore it is imperative that we study and understand all aspects of photon-echo dynamics.

In this paper, we study the echo dynamics for two different kinds of pulse pairs. For pulse pairs that each have the same carrier envelope phase, denoted CEP pulse pairs, the electric field can be written as

$$E(t) = \sum_{j=1,2} E_j(t) = \sum_{j=1,2} \mathcal{E}_j e^{-(t-t_j)^2/2\delta^2} \cos[\omega(t-t_j)],$$

where $\mathcal{E}_j$ is the pulse peak amplitude, $t_j$ locates the pulse Gaussian envelope in time, $\omega$ is the driving laser frequency, and $\delta$ is the temporal pulse width. Typically $\mathcal{E}_1, \mathcal{E}_2$ and $\delta$ are chosen to yield $\pi/2$ and $\pi$ pulse areas respectively but this is not crucial to the photon echo phenomenon. For pulse pairs that are cut out from the same carrier wave train, denoted here as normal pulses, the electric field can be written as

$$E(t) = \sum_{j=1,2} E_j(t) = \sum_{j=1,2} \mathcal{E}_j e^{-(t-t_j)^2/2\delta^2} \cos[\omega(t-t_j)].$$

Figure 1 illustrates these two different kinds of pulse pairs. We see that the phasing of the envelope and the carrier wave of both pulses in a CEP pulse pair is identical, whereas for normal pulse pairs this is not the case. Most of the studies (and applications hitherto) of photon and spin echoes consider only nonoverlapping pulses. By exploring the regime where pulses partially overlap, we found that the photon-echo signal is very sensitive to the relative phase (i.e., interpulse delay, $\tau_d$) between the pulses when a CEP pulse pair is used. Unsurprisingly the absolute value of the carrier envelope phase has no influence on the echo dynamics other than defining the phase of the underlying polarization oscillations. Using numerical simulations and approximate analytic solutions to the optical Bloch equations we find that large shifts in the echo emission time are possible by controlling parameters such as the delay between the pulses, the inhomogeneous broadening of the two-level ensemble, the pulse widths of the applied pulses, and the polarization dephasing time.

Results from this study show that we can achieve control over the emission time of the echo signal while using the same pulse sequence ($\pi/2-\pi$) as the conventional echoes. This wasn’t possible before in a two-level system. Therefore, just by overlapping the pulses we can increase the storage time of the signal (limited by $T_2$) by a significant amount as shown in the numerical simulations in Sec. III.

Photon-echo experiments which attempt to measure dephasing times which are close to the available pulse durations will naturally make some measurements with partially overlapping excitation pulses. Our results show that, depending on the relative phase difference between the pulses, strongly shifting (in time) photon echoes will arise. This may be misinterpreted as jitter or noise when, as we show, it is in fact a consequence of interference.

We next distinguish our results from previous published results where the phase difference between pairs of...
excitation pulses have proven useful in nonlinear spectroscopy. Reference [2] shows how the real and imaginary parts of the nonlinear response function can be measured independently using nonoverlapping phase-locked pulses by changing the phase between the two pulses to be in-phase and in-quadrature. Using heterodyne-detected phase-locked femtosecond stimulated photon-echo and phase-locked pump-probe techniques [14,15] it has been shown that the relative phase between two initial pulses and the relative phase between the last two pulses are both important individually rather than only their difference. Here again the second and third pulses never overlap in time. A shift in echo maximum with delay between the pulses is plotted and a quantum beatlike pattern is shown on the scale of 1 ps with a pulse width of 14 fs, i.e., much longer than the frequency and therefore not an interference phenomenon. A deformation of pulses when overlapped in the time domain is not considered here, which is the distinguishing feature in the present paper. The paper of Yano and Shinojima [16] considers a comparison between CEP and normal pulses for photon echoes and coherent population control using a perturbative approach which neglects any pulse overlap. Their results predict a shift in echo temporal position with respect to pulse widths when the dephasing and decay times differ, but this shift does not arise from the changing overlap between the pulses (as this is neglected from the start).

In coherent photon-echo simulations, there are five independent energy and time quantities which come into play: (i) the interpulse delay, (ii) the pulse widths, (iii) the inhomogeneous linewidth, (iv) the detuning of the excitation from the center of the inhomogeneous line, and (v) the dephasing time. In this large phase space there is no natural scaling, so we choose to present simulations for physical parameters corresponding to the $1s-2p_x$ transitions of phosphorus-doped silicon (Si:P) which has potential for quantum information processing [17,18]. The behavior we report however is generic and can be easily applied with appropriate scaling to other materials and systems.

FIG. 1. Illustration of a CEP pulse pair (top panel) and a normal pulse pair (bottom panel). The Gaussian envelope (dotted line) highlights the difference between CEP and normal pulses.
To get the full picture of how the echo signal is changing with respect to the delay between the pulses we solved the Bloch equations numerically for many different delays and plotted the echo signal as a heat map as shown in Fig. 3. This shows that the echo peak position and to a lesser extent the echo strength are very sensitive to the relative phase between the pulses.

When pulses do not overlap in the time domain we do not observe this phase sensitivity of the echo signal emission time whether the pulses used are CEP or normal. So the observed delay sensitivity is only seen for partially overlapping CEP pulses. This indicates that the origin of the phase sensitivity lies in the interference between the electric fields of the pulses and not between the polarization oscillation induced by the first pulse interfering with the second pulse. The result of such interference is seen in the lower part of Fig. 2; the constructive interference case (red, light shading) resembles a single pulse with an amplitude modulation while the nodal structure in the destructive interference case (blue, dark shading) produces two slightly shorter but sequential pulses. We use this observation to construct an analytical model of the system in Sec. IV.

This phase sensitivity has implications for the use of photon-echo techniques to measurements of dephasing ($T_2$) times. If the dephasing times are similar to the pulse length, care must be taken to either use normal pulse pairs or to account for the phase difference between the pulses in the analysis.

B. Influence of pulse overlap area

We next investigate how the area of overlap between the pulses quantitatively affects the photon-echo emission time and its dependence on the relative phase between the two pulses. This overlap area can be changed by either (i) changing the pulse width keeping the delay between the applied pulses constant or (ii) changing the delay between the applied pulses while keeping the pulse width constant.

Figure 4 shows the effect of changing the pulse width while keeping all other parameters, including the pulse areas, fixed. For destructive interference, around $\tau_d = 33.4 \text{ ps}$, as we increase the pulse width the shift in the echo peak position in time increases by over 15 ps. Also interesting are the qualitative changes that increasing the pulse width brings midway between constructive and destructive interference conditions around a delay of $\tau_d = 33.43 \text{ ps}$; the echo peak position is not even monotonic with the pulse width. For
constructive interference the shifts seen are rather small, indicating an insensitivity to pulse width in this regime.

Figure 5 shows the photon-echo peak position in time versus delay between the pulses for a fixed (intensity) pulse duration of 17 ps (FHWM). We show scans from destructive interference through to constructive interference for three different delays around (a) \( \tau_d = 40.1 \) ps, (b) \( \tau_d = 33.4 \) ps, and (c) \( \tau_d = 25.1 \) ps. The change in overlap area of the two pulses is negligible within one scan but appreciable between the three subplots. We can see that here also the qualitative nature of the echo signal changes as we increase the delay; for shorter delays constructive interference echoes are emitted at times later than those for mixed quadrature excitation. Overall, for all interference conditions, longer pulse excitation tends to bring the emission back towards the \( t = 2 \tau_d \) line.

We conclude from these simulations that it is the specific combination of pulse width and interpulse delay that determines the qualitative behavior of echo, not simply the area of overlap between the pulses. We have also run simulations where by changing both the pulse duration and the interpulse delay we hold the pulse overlap constant and find no universal dependence on the overlap area.

IV. ANALYTICAL FORMULAS FOR PHOTON-ECHO SIGNALS

In this section we describe three special cases where we can derive an analytical formula for the shifts in the position of photon echo in the CEP case. The electric field of the pulses are assumed to be top hat in time and have the same amplitude. The second pulse is twice as long as the first and thus has the required area for a \( \pi \) pulse. The three cases are (i) when the applied pulses do not overlap, (ii) when the applied pulses interfere destructively, and (iii) when the applied pulses interfere constructively.

A. Nonoverlapping pulses

For comparison in the case where pulses do not overlap in time there is an existing result \[19\],

\[
P(t_d) = -2P_0 e^{-\sigma^2(t_{d1} - t_{d2} - 1/\Omega_1)^2/2} \sin(\omega t_d),
\]

where \( P(t_d) \) is the polarization after the application of the second pulse, \( P_0 \) is a constant, and \( \Omega_1 \) is the Rabi frequency of the initial \( \pi/2 \) pulse. \( t_{d1} \) is the time interval between the two pulses and \( t_{d2} \) is the time coordinate relative to the end of the second pulse. The echo appears when these two are almost equal, with a small correction due to the finite pulse width.

B. CEP pulses: Destructive interference

In this case the total electric field can be written as a piecewise constant field of varying amplitude [see Fig. 6(a)]. When the pulses overlap the electric field is zero so instead
with amplitude of the electric field to double at those times. In effect this allows an interval of free polarization oscillation and rephasing. For provides a region of zero field when the pulses are overlapping which destructively and (b) constructively. The destructive interference gives rise to a modified echo which is the origin of the phase control of the photon-echo dynamics we report here. In each section of the pulse sequence, we can write the solution in the control of the photon-echo dynamics we report here. In each gives rise to a modified echo which is the origin of the phase position of echo depends on the area of overlap \((t_1, t_2)\) as the pulse width, we can have control over the emission time by controlling the area of overlap between the pulses as well as the pulse width, we can have control over the emission time of the echo, which might be important in storing light using atomic ensembles.

\[ P(t_4) = P_0 \left[ e^{-\frac{A(t_4-t_1)}{\Omega_1}} - e^{-\frac{A(t_4-t_2)}{\Omega_1}} \right] \sin(\omega t_4), \]

with
\[
C = (1 + \sin 2A)/\Omega_1,
\]
\[
D = (\sin A \sec^2 A - \sec^2 A - \tan A)/\Omega_1,
\]
\[
E = (\cot A + \csc A)/\Omega_1,
\]

\(A\) is the area of overlap between the pulses (between 0 and \(\pi/2\)), \(\omega\) is the applied laser frequency, and \(t_{21} = t_2 - t_1\) and \(t_{13} = t_4 - t_3\) are the times as shown in Fig. 6. As area \(A\) is always less than \(\pi/2\), \(C\) and \(E\) are always positive, while \(D\) is always negative. Superposition of the first four terms in the formula defines the echo profile and the arrival time, while the last term doesn’t actually contribute to the echo signal after time \(t_3\) because \(E\) is always positive, which implies that the peak of the Gaussian lies before \(t_3\) and hence does not affect the echo that comes at times later than \(t_3\). The shift in the position of echo depends on the area of overlap \((A)\) between the pulses and the Rabi frequency of the applied pulses. For \(A = 0\) this result reduces to Eq. (6) as it must.

Equation (7) describes well the observed behaviors for destructive interference. Increasing the pulse duration for a fixed delay, or decreasing the delay time for a fixed pulse duration, increases the pulse overlap area \(A\). As \(A\) increases, \(C\) becomes larger and \(D\) more negative and both these trends lead to an increase in the shift of the echo emission time. Both these trends are seen in the full numerical simulations of Figs. 4 and 5.

The FWHM of each term is inversely related to the FWHM of the inhomogeneous broadening distribution, i.e., the broader the ensemble linewidth (larger \(\sigma\)), the narrower the Gaussians in each term of the formula. Therefore, the superposition between these four terms will change if we change the inhomogeneous broadening distribution. This implies that the echo position also depends on the ensemble linewidth. Now by controlling the area of overlap between the pulses as well as the pulse width, we can have control over the emission time of the echo, which might be important in storing light using atomic ensembles.

C. CEP pulses: Constructive interference

For constructive interference between the CEP pulses [see Fig. 6(b)], we find

\[ P(t_4) = -P_0 e^{-\frac{A(t_4-t_1)}{\Omega_1}} \sin(\omega t_4). \]

This result is independent of the overlap between the pulses, which is consistent with the rather weak shifts seen for constructive interference in Figs. 4 and 5. This formula is only valid when \(A \neq 0\) and therefore it is not equivalent to Eq. (6). Effectively within this analytic model there is no real echo emitted. This can be understood by noting that there is no free precession interval (the electric field in the interval \(t_{21}\) is not zero) between the two pulses. This means the macroscopic polarization does not have time to freely unphase; therefore we do not get a distinct photon-echo signal from the rephasing of oscillators after the excitation pulses are gone.

V. THE INFLUENCE OF DEPHASING

In Fig. 7 we show the effect of introducing a finite depolarization dephasing time, \(T_2 = 120\) ps, on the echo peak position in time. For destructive interference we find that the finite dephasing time shifts the curve to earlier times while for constructive interference we find essentially no effect. For shorter pulses (not shown) there is also a shift for constructive interference to earlier times too, but a full exploration of this phase space is beyond the scope of this paper.

Photon-echo decay with a nonoverlapping (\(\pi/2, \pi\)) pulse sequence is the conventional technique to measure the polarization relaxation times \(T_2\). The echo signal strength is measured for a range of interpulse delays, and an exponential decay in strength (with the rate of decay depending on \(T_2\)) is observed. Such measurements are most easily done when the dephasing time is considerably longer than the pulse duration, allowing for time discrimination over a decade or more in signal decay. Our results on the strong phase sensitivity of the echo arrival time inform such measurements in two ways.
First at early times when the pulses overlap to some degree, unless care is taken over the precise interpulse phase at each delay selected, there will appear in the data some scatter reflecting the spread of excitation phases used. Without an appreciation of the source of this scatter it may be wrongly interpreted as due to some other source of noise. Second, often it is desirable to push the techniques to measure dephasing times which approach the pulse duration and hence the phase sensitivity reported here would come into play. To explore if new information could be gained with knowledge of the sensitivity reported here would come into play. To explore

FIG. 7. Echo peak position in time vs delay between the pulses for different dephasing times. The solid line is the infinite dephasing time, and the dashed line is the dephasing time $T_2 = 120$ ps. The green dotted line indicates the time $\tau_d$ after the second pulse peak. Other parameters are as in Fig. 2.

VI. CONCLUSIONS

To conclude, we have shown numerically and analytically that when carrier-envelope-phase pulses overlap we see a strong phase dependence of the echo position (in time). Shifts of over 10 ps have been seen. The qualitative nature of this phase (delay) dependence changes depending on the pulse overlap area, the pulse width of the applied pulses, the dephasing time, and the inhomogeneous linewidth of the system. In the case of well-separated carrier-envelope-phase pulses, the echo peak position (in time) is independent of the phase difference between the applied pulses. For all normal pulses, the echo peak position (in time) is insensitive to wavelength-scale changes of the delay between the applied pulses. In all cases, there is an inherent small shift in the echo peak position in time due to the finite pulse width of the applied pulses. Our results inform both the interpretation and design of ultrafast polarization relaxation measurements using photon-echo techniques as they guide the interpretation of the measured signals when the pulses overlap. Making photon-echo measurements in the destructive interference condition should allow the resolution of dephasing times shorter than those in the constructive interference configuration.

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