Coherent spin control by electromagnetic vacuum fluctuations

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In coherent control, electromagnetic vacuum fluctuations usually cause coherence loss through irreversible spontaneous emission. However, since the dissipation via emission is essentially due to correlation of the fluctuations, when emission ends in a superposition of multiple final states, correlation between different pathways may build up if the “which-way” information is not fully resolved (i.e., the emission spectrum is broader than the transition energy range). Such correlation can be exploited for spin-flip control in a Λ-type three-level system, which manifests itself as an all-optical spin echo in nonlinear optics with two orders of optical fields saved as compared with stimulated Raman processes. This finding may open a new class of nonlinear spectroscopy with some perturbative orders of the optical field replaced by the vacuum fluctuation.

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Electromagnetic vacuum fluctuations are fundamental in many physical processes (spontaneous emission, light scattering, Casimir effect, lasing, etc)1,2 and in a wide variety of applications (quantum information processing, quantum metrology, laser cooling, photonic engineering, etc)3,4. Particularly in quantum coherence control, the vacuum fluctuations are usually undesirable3,5 since they cause spontaneous photon emission and in turn irreversible loss of coherence of the systems under control. However, there are still some surprising effects. For instance, it was predicted and observed that in the stimulated Raman process for spin coherence generation in a Λ-type three-level system, the irreversible spontaneous emission (SE) from the excited state, when its spectrum is wide enough to cover both emission pathways and the two pathways couple to the same photon modes, will generate Raman coherence between different spin states7,8.

In this Letter, we predict yet another striking effect of the vacuum fluctuations and show how it manifests itself in nonlinear optics. The irreversible SE in a Λ-type three-level system can cause a spin flip and hence recover the dephased spin coherence by spin echo9. Such spin-flip control by vacuum fluctuations, when implemented in the standard spin coherence pump-probe spectroscopy, can realize spin echo in nonlinear optics, with two orders of optical fields saved as compared with the conventional methods using stimulated Raman processes. This effect reveals that the vacuum field can indeed replace some orders of the optical field in nonlinear spectroscopy. By studying the nonlinear optical signals of spin coherence in a fluctuating random field (due to environmental noises)10, we will also show that the SE-assisted spin flip has the same effect as a usual coherent r-rotation control in restoring the spin coherence lost within the memory time of the environmental noises11.

To illustrate the basic idea of spin flip control by vacuum fluctuations, let us first examine the stimulated Raman processes. Such processes are the fundamental mechanisms of many physical phenomena such as electromagnetically induced transparency12, stimulated Raman adiabatic passage5, and optical control of spins in semiconductors13-19. As shown in Fig. 1 (a), we consider two spin states |±⟩ coupled to the same excited state by a short laser pulse. The spin is flipped when |+⟩ and |−⟩ are exchanged, via two state transfer processes in parallel, namely, the stimulated Raman processes from |±⟩ to |∓⟩ mediated by the excited state. Similar to the photon echo in four-wave mixing20, the signature of the spin flip will appear as spin echo in nonlinear optics via a perturbative procedure with four orders of the optical field involved in the spin flip.

Now if the stimulated photon emission from the excited state is replaced by the SE [see Fig. 1 (b)], the spin flip can be realized by Raman processes involving only two orders of the laser field. Similar to the stimulated Raman processes, it is essential that the SE spectrum is broader than the spin splitting and the two emission pathways couple to the same photon mode. Such requirements indicate the fundamental basis of the predicted effect: The SE (dissipation) is due to the correlation of the vacuum fluctuations21, so when there are several final states in a SE process, coherent correlation between different quantum pathways may be generated when the

FIG. 1: (Color online) (a) Stimulated Raman processes for a spin flip control in a Λ-type three-level system. (b) Raman processes for a spin flip control, with emission caused by vacuum fluctuations (dotted arrows) instead of a laser field as in (a).
“which-way” information is not fully resolved. Such correlation may lead to coherence generation by SE \[8\] and even coherent spin control when there is initial spin coherence.

Now we present the more detailed analysis with a model system of electron spins in quantum dots, a paradigmatic system in research of quantum optics, quantum computing, and mesoscopic physics. In a GaAs fluctuation quantum dot \[8\], for example, a normal incident light with circular polarization \(\sigma_+\) (or \(\sigma_-\)) couples only to the optical transition between the electron spin state \(|↑\rangle\) (or \(|↓\rangle\)) to the negatively charged exciton state, i.e., the trion state \(|t\rangle\) (or \(|\bar{t}\rangle\)), with the spin basis quantized along the growth direction [see Fig. 2(a)]. Under a transverse magnetic field [in the Voigt geometry, see Fig. 2(b)], the electron spin is split into two states \(|±\rangle\) quantized along the external field direction, but the trion states remain nearly degenerate due to the large energy mismatch between the heavy hole and the light hole and hence can still be quantized along the growth direction \[8\]. Without loss of generality, we set the pump, control, and probe pulses all \(\sigma_+\) polarized. Then, only the trion state \(|t\rangle\) will be excited and thus the system is modeled by a \(Λ\)-type three-level system consisting of \(|±\)\rangle and \(|t\rangle\). The all-optical spin echo is based on a standard pump-probe setup [see Fig. 2(b)] and the basic optical processes are illustrated in Fig. 2(c).

The initial step is optical pumping of spin coherence. A short \(\sigma_+\) pulse, with a bandwidth greater than the spin splitting, excites population from the spin state \(|↑\rangle\) to the trion state \(|t\rangle\), leaving the spins polarized along \(-z\)-direction and initiated to precess about the external field. The SE will bring the trion population back to the spin state \(|↑\rangle\), which tends to cancel the spin coherence generated by the stimulated Raman processes. As the SE takes a finite time during which the spins precess, the spin coherence will be only partially canceled and phase delayed \[8\]. The second-order optical processes for the spin coherence generation are described by [see Fig. 2(c)]

\[
\begin{align}
\rho_{++}^{(0)} &\rightarrow \rho_{++}^{(1)} &\rightarrow \rho_{++}^{(2)}, \\
\rho_{++}^{(0)} &\rightarrow \rho_{++}^{(1)} &\rightarrow \rho_{tt}^{(2)},
\end{align}
\]

where \(\rho_{ij}^{(n)}\) is the density matrix element between \(|i\rangle\) and \(|j\rangle\) in the \(n\)th order of the optical field, and \(E_k\) is the optical field of the \(k\)th pulse. The spin coherence after the pump, quantified as the off-diagonal matrix element, is \[8\]

\[
\rho_{tt}^{(2)}(t) \propto E_1 E_1^* \frac{\omega}{\omega + i \Gamma} e^{-i\omega t - i T_2/2},
\]

where \(\omega\) is the Zeeman splitting, \(\Gamma\) is the SE rate, and \(T_2\) is the spin decoherence time. In the presence of inhomogeneous broadening (a probability distribution of \(\omega\) assumed as \(e^{-(\omega-\omega_0)^2/(2\sigma^2)}\)), the ensemble-averaged spin coherence is

\[
\langle \rho_{tt}^{(2)}(t) \rangle \propto E_1 E_1^* \frac{\omega}{\omega + i \Gamma} e^{-i\omega_0 t - i \sigma^2 t^2/2}.
\]

As usually \(\sigma \gg 1/T_2\), the spin polarization decay is dominated by the inhomogeneous broadening effect. To resolve the “true” spin decoherence, spin echo may be invoked.

The key step in the all-optical spin echo is the control of spins. To illustrate the idea, let us start with the rotation of spins by the optical AC Stark shift which has been demonstrated in quantum dots \[14\][15][17][19]. Consider a \(\sigma_+\) pulse detuned well below the trion resonance. The virtual transitions between \(|t\rangle\) and the spin state \(|↑\rangle\) will induce an AC Stark energy shift of the spin state, which in turn will induce a rotation of the spin about the \(z\)-axis, with an angle \(\theta \propto |E_2|^2 + O(|E_2|^4)\). If \(\theta = \pi\), the spins are flipped. In reality, it is non-trivial to realize an exact \(\pi\)-rotation or even a large-angle rotation \[17\][19]. The idea of using nonlinear optical response to realize spin echo comes from the perturbative expansion of a small rotation

\[
\exp(i\theta S_z) = 1 + i\theta S_z + O(\theta^2).
\]

Thus, an infinitesimal rotation contains the rotation generator \(S_z\), i.e., the spin operator along the \(z\)-axis, which exchanges the states \(|+\rangle\) and \(|-\rangle\).

From Eq. (4), it is tempting to conclude that two orders of the control field can flip the spin coherence. A closer examination, however, reveals that we need actually four orders of the control field. To see the problem, let us consider a general spin state \(|\psi\rangle = C_+|+\rangle + C_-|-\rangle\). A small rotation about the \(z\)-axis transforms it into

\[
e^{i\theta \hat{S}_z} |\psi\rangle = \left(C_+ + \frac{i\theta}{2} C_-\right)|+\rangle + \left(C_- + \frac{i\theta}{2} C_+\right)|-\rangle + O(\theta^2).
\]

Before the pulse applied at \(t = \tau\), the spin coherence is \(\rho_{+-}(-\tau) = C_+ C_-^* \propto \exp(-i\omega \tau)\). For spin echo, we wish to pick up the spin-flipped term \(\rho_{+-}(\tau + 0)\) after the control pulse. Such a term in the leading order of \(\theta\) is \(\theta C_+ C_-^*/4\). Thus at least four orders of the control field are needed. This problem can also
be understood from the picture of stimulated Raman processes shown in Fig. 1 (a) or from the excitation pathways of the control process (see formula below). Starting from the spin coherence generated by the pump pulse, $\rho^{(2)}_{+-}$, the excitation by two orders of the control pulse follows the pathways

$$
\rho^{(2)}_+ \rightarrow E_z^{(2)} \rightarrow \rho^{(4)}_+ \rightarrow \rho^{(4)}_+ \rightarrow \rho^{(4)}_+ \rightarrow \rho^{(4)}_+ \rightarrow \rho^{(4)}_{++} \rightarrow \rho^{(4)}_{+-} \rightarrow \rho^{(4)}_{++} \rightarrow \rho^{(4)}_{+-}, \text{ or } \rho^{(4)}_{++}, \quad (6a)
$$

$$
\rho^{(2)}_- \rightarrow E_z^{(2)} \rightarrow \rho^{(4)}_- \rightarrow \rho^{(4)}_- \rightarrow \rho^{(4)}_- \rightarrow \rho^{(4)}_- \rightarrow \rho^{(4)}_{--} \rightarrow \rho^{(4)}_{++} \rightarrow \rho^{(4)}_{+-} \rightarrow \rho^{(4)}_{++} \rightarrow \rho^{(4)}_{+-}, \quad (6b)
$$

none of which results in a spin-flipped term $\rho^{(4)}_{+-}$. Note that the excitation pathways are independent of the detuning of light, and thus the problem discussed above is not limited to the spin rotation by the AC Stark effect of virtual excitation but applies also to real excitation.

We note that in Eq. (9) the trion population is also obtained if the excitation is in resonance with the trion. As discussed earlier for the optical pump of spin coherence, the SE will bring the trion population to the spin population $\rho^{(4)}_{+-}$. Thus with the SE included, the spin-flipped coherence is obtained through the quantum pathway [see Fig. 2(c)]

$$
\rho^{(2)}_{+-} \rightarrow E_z^{(2)} \rightarrow \rho^{(4)}_{+-} \rightarrow \rho^{(4)}_{+-} = \frac{1}{2} \left( \rho^{(4)}_{++} + \rho^{(4)}_{+-} + \rho^{(4)}_{-+} + \rho^{(4)}_{--} \right). \quad (7)
$$

Indeed, one can regard the SE as the contribution of two orders of the vacuum field to the nonlinear optical response, which is consistent with the observation that at least four orders of control field is needed to flip the spin coherence. Similar to the stimulated Raman processes, we also need the bandwidth of the SE to be comparable to or greater than the spin splitting (i.e., $\Gamma \gtrsim \omega$).

Considering the spin precession during the SE, the spin coherence generated by the SE is [8]

$$
\rho^{\text{SE}(4)}_{+-}(t) = \frac{1}{2} \rho^{(4)}_{+-} \frac{i\omega t}{\omega^2 + \Gamma^2} e^{i\omega t/\tau} e^{i(\omega t - \tau)/\tau}, \quad (8)
$$

where the 4th order trion population is

$$
\rho^{(4)}_{+-} \approx E_z^{(2)} \rho^{(2)}_{+-} \frac{i\omega t}{\omega^2 + \Gamma^2} e^{i(\omega t - \tau)/\tau} \rho^{(2)}_{+-}(\tau). \quad (9)
$$

Thus we obtain the spin-flipped coherence term

$$
\rho^{(4)}_{+-}(t) \approx E_z^{(2)} \frac{i\omega t}{\omega^2 + \Gamma^2} e^{i(\omega t - \tau)/\tau} \rho^{(2)}_{+-}(\tau). \quad (10)
$$

With the spin coherence generated by the pump pulse given in Eq. (2), the ensemble average of the spin-flipped term is

$$
\left\langle \rho^{(4)}_{+-}(t) \right\rangle \approx \left| E_1 \right|^2 \left| E_2 \right|^2 \frac{i\omega t}{\omega^2 + \Gamma^2} e^{i(\omega t - \tau)/\tau} \rho^{(4)}_{+-}(\tau) \right \rangle \cdot (11)
$$

The spin echo is seen by noticing that the phase factor $e^{i\omega t}$ accumulated in $\rho^{(4)}_{+-}(\tau)$ is canceled in $\rho^{(4)}_{+-}(t)$ at $t = 2\tau$.

The differential transmission of a $\sigma^-$ pulse probes the population change of the spin state $\uparrow$ due to the pump and the control pulses. With two orders of the pump field and two orders of the control field carried by the spin coherence, the differential transmission is a $\chi^5$ optical response

$$
\Delta T^{(5)}(t) \approx \left( \rho^{(4)}_{+-} \right) = \left( \rho^{(4)}_{+-} + \rho^{(4)}_{+-} + 2\mathfrak{R}\rho^{(4)}_{+-} \right)/2 \right \rangle \cdot (12)
$$

where $C$ consists of all the background terms and the oscillation terms without spin flip. As the spin polarization processes about the external field with decoherence, the differential transmission signal will oscillate sinusoidally and decay. For an ensemble with inhomogeneous broadening $\sigma$, the signal at a long time ($t \gg \sigma^{-1}$) will present oscillation only near the echo time $t = 2\tau$. The decay of the echo signal as a function of $\tau$ reveals the “true” decoherence excluding the inhomogeneous broadening effect.

To check whether the spin-flip control by SE can suppress the decoherence in a “slow” bath the same way as a coherent $\pi$-rotation in spin echo [11], we simulate the decoherence by a spectral diffusion model in which the local magnetic field $\omega(t) = \omega + X(t)$ contains a dynamically fluctuating part $X(t)$ [10]. The accumulated random phase $\phi(t_2, t_1) \equiv \int_{t_1}^{t_2} X(t) dt$ causes the spin decoherence. For a Gaussian fluctuation to which Wick’s theorem applies, the spin-flipped coherence term in Eq. (11) becomes [10]

$$
\left\langle \rho^{(4)}_{+-}(t) \right\rangle \approx e^{i\omega_0(t - \tau)/\tau} e^{-\sigma^2(t - \tau)^2/2} \left\langle \left[ \phi(t_2, t_1) - \phi(t_1) \right]^2 \right\rangle/2. \quad (13)
$$

To be specific, we employ a noise correlation of the exponential form [11] $\langle X(t_2)X(t_1) \rangle = \langle X^2(0) \rangle \exp(-|t_2 - t_2|/\tau_c)$. The spin echo not only eliminates the static inhomogeneous broadening effect but also partially recovers the decoherence resulting from the dynamical fluctuation provided that the pulse delay time $\tau$ is comparable to or shorter than the noise correlation time $\tau_c$ [11].

![Fig. 3: (Color online) Real-time envelopes of the $\chi^5$ differential transmission of singly-charged quantum dots, calculated analytically (solid lines) or numerically (symbols), with the control pulse applied at (a) $\tau = 0.5 \mu$s or (b) $\tau = 1.5 \mu$s (signal amplified by 100). The insets show the oscillations in a few small time-windows. To model realistic conditions, the parameters are chosen as $\tau_c = 1 \mu$s, $T_2 \equiv \langle X^2(0) \rangle^{-1} \tau_c^{-1} = 0.1 \mu$s, $\omega_0 = 10 \mu$eV, and $\Gamma = 10 \mu$eV. The inhomogeneous broadening is set to zero.](image-url)
inset shows the real-time dependence of the differential transmission near the echo time for $$\tau$$ free-induction decay without the inhomogeneous broadening at $$t = 2\tau$$ (dotted line). Open square and circles are numerical results. The inset shows the real-time dependence of the differential transmission near the echo time for $$\tau = 0.5\,\mu s$$. The parameters are the same as in Fig. 3 except for the inhomogeneous broadening $$\sigma = 0.1\,\text{ns}^{-1}$$.

The partial recovery from the spin decoherence caused by dynamical fluctuations is seen in numerical simulation as shown in Fig. 3. To show the effect of dynamical fluctuation, the inhomogeneous broadening $$\sigma$$ is set to be zero. When the pulse delay time is shorter than the noise correlation time [Fig. 3 (a)], the coherence is recovered close to $$2\tau$$, the same as in spin echo for static inhomogeneous broadening which can actually be understood as spectral diffusion with infinite correlation time $$(\text{11})$$. For longer pulse delay times, the recovery is less perfect and the peak approaches $$0$$ with infinite correlation time $$(\text{11})$$, as evidenced in Fig. 3 (b). If the fluctuation is rapid ($$\tau_c \ll \tau$$), the random field correlation has a white noise form $$\langle X(t_1)X(t_2) \rangle = T_2^{-1}\delta(t_1 - t_2)$$, and the echo signal presents no coherence recovery (not shown).

When the inhomogeneous broadening is included, the signal for $$\tau \gg 1/\sigma$$ is visible only near the echo time $$2\tau$$, as shown in the inset of Fig. 4. The main part of Fig. 4 plots the echo signal as a function of the pulse delay time. When $$\tau \leq \tau_c$$, the spin decoherence due to the dynamical fluctuation is partially recovered, and the echo signal decays slower than the free-induction decay signal [$$\propto \mathcal{R} \langle e^{i\delta(2\tau,0)} \rangle$$] without the inhomogeneous broadening ($$\sigma = 0$$).

In conclusion, we have explored a striking effect of correlation between different quantum pathways of spontaneous emission in a $$\Lambda$$-type three-level system, namely, the coherent spin control by SE and its role in all-optical spin echo. It is shown that two orders of optical field can be replaced by the vacuum field in the nonlinear optical spectroscopy of spin coherence. It is conceivable that in more general multi-level systems, (higher order) correlations between multiple SE pathways could lead to a wealth of new physics to be unveiled.

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![Graphical representation of spin echo amplitude](image-url)

FIG. 4: (Color online) The spin echo amplitude (solid line), and the free-induction decay without the inhomogeneous broadening at $$t = 2\tau$$ (dotted line). Open square and circles are numerical results. The inset shows the real-time dependence of the differential transmission near the echo time for $$\tau = 0.5\,\mu s$$. The parameters are the same as in Fig. 3 except for the inhomogeneous broadening $$\sigma = 0.1\,\text{ns}^{-1}$$.

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