Self-Stimulated Pulse Echo Trains from Inhomogeneously Broadened Spin Ensembles

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We show experimentally and describe theoretically how a conventional magnetic resonance Hahn echo sequence can lead to a self-stimulated pulse echo train when an inhomogeneously broadened spin ensemble is coupled to a resonator. Effective strong coupling between the subsystems assures that the first Hahn echo can act as a refocusing pulse on the spins, leading to self-stimulated secondary echoes. Within the framework of mean field theory, we show that this process can continue multiple times leading to a train of echoes. We introduce an analytical model that explains the shape of the first echo and numerical results that account well for the experimentally observed shape and strength of the echo train and provides insights into the collective effects involved.

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Introduction.—Electron spin resonance (ESR) [1,2] and nuclear magnetic resonance (NMR) [3,4] are used in diverse branches of science, ranging from spectroscopic studies in biochemistry and materials science [5–8] to imaging of internal organs in medicine [9]. In NMR and ESR, an ensemble of spins is typically placed within a resonator, controlled by the application of resonant pulses, and measured via emission of signals into a resonator mode. As the spin ensembles are typically inhomogeneous, a common solution is to use control pulses which refocus inhomogeneous interactions, reversing the time evolution of different spin packets to produce a spin echo [10,11] or “Hahn echo” [12]. Aside from being the cornerstone of pulsed NMR and ESR techniques, spin echoes have also become an essential ingredient in quantum information science due to their applications in ensemble quantum memories with ESR and optical transitions [13–17], and in nanoscale quantum metrology [18,19], as well as being a building block for more complex dynamical decoupling sequences used to extend qubit coherence times [20].

The Hahn echo sequence consists of an initial $\pi/2$ pulse, a time interval $\tau$ and a $\pi$ pulse leading to the emission of a spin echo at time $2\tau$. The $\pi/2$ pulse excites the spins into a coherent superposition state, which starts precessing at the Larmor frequencies of the individual spins, leading to dephasing of the collective spin. At time $\tau$, a $\pi$ rotation of the spins is performed in the Bloch sphere, which is equivalent to a time reversal operation, since the phases of the excited and ground state amplitudes get interchanged, i.e., $|g\rangle + Be^{-i\Phi}|e\rangle \rightarrow Be^{-i\Phi}|g\rangle + A|e\rangle$, where $\Phi$ is the relative phase between the ground state $|g\rangle$ and the excited state $|e\rangle$. As $\Phi \propto \omega_0\tau$, where $\hbar\omega_0$ is the energy difference between $|e\rangle$ and $|g\rangle$, the subsequent time evolution leads to refocusing of the spins to recover the original coherent superposition state and produce a spin echo at time $2\tau$.

A range of recent ESR studies have begun to examine increasing the coupling between the spin ensemble and the resonator, for example, to improve spin sensitivity in spectroscopic applications [21,22], or to improve the efficiency of a quantum memories or transducers [13–17,23–30]. However, increasing the spin-resonator coupling also introduces the possibility for the emitted echo itself to act as a sufficient perturbing field to drive further evolution of the spins. If the light-matter coupling becomes stronger than the dissipative losses, even the simple and ubiquitous Hahn echo sequence can yield nontrivial spin dynamics which lead to the emission of not just to one, but multiple spin echoes. Such multiple echoes are illustrated in Fig. 1(a) which shows the experimentally observed emission from an inhomogeneously broadened spin ensemble (Nd ions in $Y_2SiO_3$) subject only to a Hahn echo sequence. (Further details on the experiment are provided in the Supplemental Material [31]). The data reveal the conventional echo signal at time $2\tau$, followed by additional echoes separated by $\tau$. A similar result was reported recently in Ref. [33], which showed that the secondary echoes were absent when the light-matter coupling was reduced. Indeed, secondary echoes were discerned in the very first microwave spin echo experiment in 1958 [34], where highly doped samples were used to compensate low detector sensitivity. In this Letter we examine the “echo train” phenomenon experimentally and theoretically, with the goal of developing a better understanding of the underlying mechanisms, necessary for the exploitation of strongly coupled resonators and spin ensembles.
This rephasing occurs for all values of the frequency of the spins and leads to the usual Hahn echo.

In case of a perfect $\pi$ refocusing pulse, the state amplitudes $A_1$ and $B_1$ vanish, and the spins accumulate diverging phases in the time following this spin echo. Otherwise, all four terms in (2) contribute to the subsequent evolution of the system, and this is key for the production of further echoes. For large $N$, the Hahn echo pulse may be strong enough to significantly alter the spin states and thus populate a new superposition:

$$
(A_1|g\rangle + A_2|e\rangle) + e^{-i\omega_{\text{c}}\tau}(C_1|g\rangle + C_2|e\rangle) + e^{-2i\omega_{\text{c}}\tau}(B_1|g\rangle + B_2|e\rangle).
$$

As can be seen from Fig. 1(b), after a further time $\tau$, the underlined (and doubly underlined) terms come into phase and cause a second echo. The evolution and refocusing may occur also for terms with higher phase arguments, and the process can repeat and lead to multiple self-stimulated echoes.

To assess the validity of our qualitative discussion, we now proceed to investigate whether the spin echo train can be reproduced by a numerical treatment. We begin with a description of the model, followed by the numerical results, demonstrating clear evidence of the self-stimulated spin echo train. We then introduce a simplified analytical model that predicts various shapes of the echo pulse and provide insights into the collective effects involved.

Theoretical model.—We consider $N = 10^{10}$ Nd spins coupled to a resonator of linewidth $\kappa = 2\pi \times 150$ kHz and resonance frequency $\omega_{\text{c}}$. The spins are inhomogeneously broadened with transition frequencies $\omega_{\text{c}}^j$, following a Gaussian distribution with central frequency $2\pi \times 8$ GHz and FWHM of $\Gamma_{\text{inh}} = 2\pi \times 4$ MHz. An external coherent drive with frequency $\omega_{\text{c}}$ and amplitude $F$ is used to apply the two initial pulses. In the frame rotating with the pump frequency, the Hamiltonian ($\hbar = 1$) of the system can be written as

$$
H = \delta_c a^\dagger a + \sum_{j=1}^N \left[ \frac{\delta_{\text{c}}^j}{2} \sigma_z^j + g_j (a\sigma_-^j + a^\dagger\sigma_+^j) \right] + F(a + a^\dagger),
$$

where $\delta_{\text{c}} = (\omega_{\text{c}} - \omega_p)$ and $\delta_{\text{c}}^j = (\omega_{\text{c}}^j - \omega_p)$. $\sigma_z^j, \sigma_+^j, \sigma_-^j$ are the Pauli operators and $a, a^\dagger$ are the annihilation and creation operators of the cavity mode, obeying the usual commutation relation $[a, a^\dagger] = 1$. For numerical simplicity, we assume $g_j = g = 2\pi \times 8$ Hz. Despite this weak single spin-cavity coupling, the presence of $10^{10}$ spins significantly enhances the effective coupling. The exact dynamics of the system can be described by the master equation

$$
\dot{\rho} = -i[H, \rho] + \kappa D[a]\rho + \gamma \sum_{j=1}^N D[\sigma_-^j]\rho + \Gamma \sum_{j=1}^N D[\sigma_+^j]\rho.
$$
followed by a delay of variable duration pulse between \( t \) not crucial for the appearance of the echo as long as the \( \text{condition between the spin eigenstates and the exact intensity is finite pulse area causes some coherent transfer of population for } N_k \text{ the master equation for the full density matrix is impossible. Solving the master equation for the full density matrix is impossible, and we treat the model by discretizing the frequency distribution into } N_k = 10^3 \text{ frequency classes following a Gaussian distribution and employ mean field theory for the field and spin raising and lowering operators, assuming factorization of their products, i.e., } \langle AB \rangle \approx \langle A \rangle \langle B \rangle [35].

Results.—We apply a strong classical square pulse of amplitude \( F = 50 \text{ GHz} \) from \( t_1 = 0.20 \mu s \) to \( t_2 = 0.42 \mu s \), followed by a delay of variable duration \( \tau \) and a second pulse between \( t_3 = t_2 + \tau \) and \( t_4 = t_3 + 0.43 \mu s \). Any finite pulse area causes some coherent transfer of population between the spin eigenstates and the exact intensity is not crucial for the appearance of the echo as long as the pulses excite the spins by a significant amount.

In Fig. 2(a) we present the dynamics of the spin ensemble for \( \tau = 60 \mu s \), \( \Gamma = 2\pi \times 0.5 \text{ kHz} \). The top panel shows the square of the cavity field amplitude, i.e., the intracavity photon number, \( \langle |a|^2 \rangle = \langle \sigma_z \rangle \). The first two peaks, which extend beyond the border of the figure, correspond to the two external driving pulses, and we observe that the Hahn echo sequence generates multiple echoes separated by \( \tau = 60 \mu s \). To understand this better, we plot \( \langle |a|^2 \rangle \) for single spins in different frequency classes close to the resonance in the center panel of Fig. 2(a), where \( \Delta = \delta_i - \delta_c \). For simplicity, we consider \( \omega_c = \omega_p \) and hence \( \Delta = \delta_i \). The refocusing of the spins is characterized by \( \langle \sigma_x \rangle \) and \( \langle \sigma_y \rangle \) converging to the same finite value for a range of detunings at \( 2\pi \) and with reduced strength at later multiples of \( \tau \). The lower panel of Fig. 2(a) shows the dynamics of the \( z \) component of the spin vector for individual spins in few selected frequency classes. The blue boxes highlight the region where the individual spins get a boost due to the strong mean field, appearing in the cavity when the collective spin refocuses.

In Fig. 2(b) we analyze the shape of the echo signal by averaging multiple trajectories (shown in grey) with randomly sampled Gaussian distributed frequency classes for \( \tau = 45 \mu s \) and \( \Gamma = 2\pi \times 0.5 \text{ kHz} \). A consistent plot of \( \langle |a|^2 \rangle \) demands many closely spaced frequency classes \( (N_k \gg 10^3) \), which is a challenging task even with a mean field approximation. Averaging multiple realizations results in a more consistent data and \( \langle |a|^2 \rangle \) averaged over 45 realizations is shown with the bold curve. The rate \( \Gamma \), which takes into account dephasing due to different mechanisms such as mutual interactions of the spins, spectral diffusion due to crystal deformation and phonon induced energy shifts, leads to vanishing of \( \langle \sigma_x \rangle \) and \( \langle \sigma_y \rangle \) and breakdown of refocusing at large \( t \). The separation between the echoes increases with increasing \( t \), as evident from Figs. 2(a)–2(c).
and the phase of the output cavity field due to the echoes at \( t = 2\tau, 3\tau, \ldots \) depends on the phase of the refocusing pulse [31].

Figure 2(c) shows how an increase of the spin dephasing rate \( \Gamma \) causes a flattening of the shape of the echoes for \( \tau = 30 \, \mu s \). It also explains the symmetric shape of the echo observed in the experiments with the dephasing rate \( \Gamma = 2\pi \times 2.5 \, kHz \) [Fig. 1(a)] and in Ref. [31].

Following Ref. [33], in Fig. 2(d), we plot the emitted photon number in each echo \( A_{\text{echo}} = \int_{\text{echo}} |d|^2 dt \) as a function of its order of emission from both experiments and numerical calculations with \( \tau = 30 \mu s \) and \( \Gamma = 2\pi \times 2.5 \, kHz \).

The experimental intensity data is not absolutely calibrated and has been displaced (in the \( y \) direction) by an arbitrary amount in the plot for better comparison with theory. An exponential fit, which is independent of the arbitrary displacement, reveals that the echoes decay faster than the spin dephasing rate \( \Gamma \) [see also Figs. S1(a)–S1(d)] while we observe an excellent agreement between theory \((2\pi \times 6.29 \, kHz)\) and experiment \((2\pi \times 6.84 \, kHz)\). We attribute this faster decay to the incomplete refocusing of the spins by the weakening pulses associated with the observed distortion and lengthening of the pulse shapes and the reduction in their amplitude. The inhomogeneous spin-cavity coupling may also affect the effective nonlinear dynamics.

We can understand the shape of the first Hahn echo observed in Fig. 2(b) by a simple analytical model. Rather than solving the complete spin dynamics analytically, we assume that a perfect \( \pi/2 \) and \( \pi \) pulse have been applied at \( t = -\tau \) and \( t = 0 \), respectively, to all the spins. This implies that right after \( t = 0 \), the spin excited states have acquired a phase \( \exp(i\Delta_j \tau) \) with respect to the spin ground states. Since the spin excited states evolve as \( \exp(-i\Delta_j \tau) \), they come in phase at \( t = \tau \). To model the resulting Hahn echo pulse shape, we employ the Holstein-Primakoff approximation [36] and treat all spins as harmonic oscillators prepared in a coherent state of complex amplitude \( \beta \exp(i\Delta_j \tau) \) at \( t = 0 \). Assuming \( \Gamma = 0 \), the mean field equations for the intracavity field operator and the spin lowering operator take the following form in the frequency domain

\[
-i\omega \langle \tilde{a}(\omega) \rangle = -\frac{\kappa}{2} \langle \tilde{a}(\omega) \rangle - i \sum_j g_j \langle \tilde{a}_j(\omega) \rangle,
\]

\[
-i\omega \langle \tilde{a}_j(\omega) \rangle = -(\gamma + i\Delta_j) \langle \tilde{a}_j(\omega) \rangle - ig_j \langle \tilde{a}(\omega) \rangle + \frac{\beta}{\sqrt{2\pi}} e^{i\beta \gamma t}.
\]

The above equations can be formally solved, which yields

\[
\langle \tilde{a}(\omega) \rangle = \frac{-\frac{\kappa}{2} \sum_j g_j e^{i\beta \gamma t} / (\gamma + i\Delta_j - i\omega)}{\frac{1}{2} - i\omega + \sum_j g_j^2 / (\gamma + i\Delta_j - i\omega)}.
\]
and has a more complex structure. Note that the mechanism leads to even later echoes which may all carry contributions from the refocusing by both the most recent and earlier echo pulses. Our numerical results reproduce most of the qualitative features observed in the experiments and show excellent quantitative agreement with the observed decay of the integrated echo pulse intensities.

Given the complexity of the model, it is difficult to analytically predict the shape of secondary echoes. Future efforts shall be devoted to understand the scaling laws that govern the gradual reduction of the self-stimulated echo amplitudes, and how these depend on the spin and cavity parameters. It is evident from the present study that in addition to the cavity linewidth and the spin dephasing rates [33], this decay will also depend nonlinearly on the intensity of the echoes, e.g., due to the incomplete refocusing by previous echoes, and the total number of spins effectively coupled to the resonator. Other factors such as the distribution of detunings and coupling strengths may offer varying contribution to the precise behavior of this decay. Although the secondary echoes studied here may constitute an unavoidable feature to be mitigated in quantum memory or transducer protocols involving spin ensembles, they may also offer opportunities in ESR spectroscopy. Aside from the potential to improve signal-to-noise by averaging multiple echoes in one train, these secondary echoes could provide an efficient route to obtaining information on loss rates in the system [33] or reveal spectroscopic information, for example in more complex spin systems with unresolved hyperfine couplings. The understanding and theoretical framework that we present here forms a basis to explore such opportunities in more detail.

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