Supplementary Information
for "Understanding of collective atom phase control in modified photon echoes for a near perfect, storage time extended quantum memory" by

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In Fig. S1(a), the D and R_1 pulses are resonant to the transition $|1\rangle - |2\rangle$. The two counter-propagating C_1 and C_2 control pulses are resonant to the transition $|2\rangle - |3\rangle$. The optical Bloch and the Maxwell-Schrödinger equations are:

$$\frac{\partial}{\partial t}\sigma_{11}(z,t,\Delta) = i\varepsilon_l(z,t)\big(\sigma_{12}(z,t,\Delta) - \sigma_{21}(z,t,\Delta)\big)\,, \tag{S1}$$

$$\frac{\partial}{\partial t}\sigma_{22}(z,t,\Delta) = i\varepsilon_l(z,t)\big(\sigma_{21}(z,t,\Delta) - \sigma_{12}(z,t,\Delta)\big) + i\varepsilon_j\big(\sigma_{23}(z,t,\Delta) - \sigma_{32}(z,t,\Delta)\big)\,, \tag{S2}$$

$$\frac{\partial}{\partial t}\sigma_{33}(z,t,,\Delta) = i\varepsilon_j\big(\sigma_{32}(z,t,\Delta) - \sigma_{23}(z,t,\Delta)\big)\,, \tag{S3}$$

$$\frac{\partial}{\partial t}\sigma_{12}(z,t,\Delta) = i\Delta\sigma_{12}(z,t,\Delta) + i\varepsilon_l(z,t)\big(\sigma_{11}(z,t,\Delta) - \sigma_{22}(z,t,\Delta)\big) + i\varepsilon_j(z,t)\sigma_{13}(z,t,\Delta), \tag{S4}$$

$$\frac{\partial}{\partial t}\sigma_{32}(z,t,\Delta) = i\Delta\sigma_{32}(z,t,\Delta) + i\varepsilon_j(z,t)\big(\sigma_{33}(z,t,\Delta) - \sigma_{22}(z,t,\Delta)\big) + i\varepsilon_l(z,t)\sigma_{31}(z,t,\Delta), \tag{S5}$$

$$\frac{\partial}{\partial t}\sigma_{13}(z,t,\Delta) = i\varepsilon_j(z,t)\sigma_{12}(z,t,\Delta) + i\varepsilon_l\sigma_{23}(z,t,\Delta)\,, \tag{S6}$$

$$\frac{\partial}{\partial z}\varepsilon_{l(j)}(z,t) = \frac{i\alpha}{2\pi}\int_{-\infty}^{\infty}\sigma_{12}(z,t,\Delta)d\Delta, \tag{S7}$$

where, l= D or R_1 and j= C_1 or C_2. The $\varepsilon_{l(j)}$ is the optical field, α is the optical depth parameter and $\Delta = \omega_{12} - \omega_l = \omega_{23} - \omega_j$ is the detuning of the atom. For the controlled single rephasing photon echo scheme without R_2, the controlled coherence conversion is studied below as discussed in numerically in ref. [18] and experimentally in ref. [19]. The pulse sequence is shown in Fig. S1(b).

I. D-pulse

A weak D-pulse propagates through the medium along z-direction. We assume that D-pulse is much weaker than the π-pulse and neglect population change by D: $\sigma_{11}(z,t,\Delta) = 1$. The resultant Maxwell-Bloch equations by D are obtained by putting $\varepsilon_l = \varepsilon_{\mathrm{D}}$ and $\varepsilon_j = 0$ in equations (S4) and (S7)

$$\frac{\partial}{\partial t}\sigma_{12}(z,t,\Delta) = i\Delta\sigma_{12}(z,t,\Delta) + i\varepsilon_{\mathrm{D}}(z,t)\,, \tag{S8}$$

$$\frac{\partial}{\partial z}\varepsilon_{\mathrm{D}}(z,t) = \frac{i\alpha}{2\pi}\int_{-\infty}^{\infty}\sigma_{12}(z,t,\Delta)d\Delta. \tag{S9}$$

The equation (S8) is the first order linear differential equation. By applying integrating factor $e^{-i\Delta t}$, the solution is obtained as:

$$\sigma_{12}(z,t,\Delta) = \sigma_{12}(z,-\infty,\Delta) + i\int_{-\infty}^{t}\varepsilon_D(z,\acute{t})e^{i\Delta(t-\acute{t})}, \tag{S10}$$

By setting the initial atomic coherence zero, $\sigma_{12}(z,-\infty,\Delta) = 0$,

$$\sigma_{12}(z,t,\Delta) = i\int_{-\infty}^{t}\varepsilon_D(z,\acute{t})e^{i\Delta(t-\acute{t})}\,d\acute{t}. \tag{S11}$$

Here, the coherence is positive (absorptive), so does the echo. It should be noted that $\sigma_{12}(z,t,\Delta) = -\rho_{12}(z,t,\Delta)$, where $\rho_{12}(z,t,\Delta)$ is the density matrix element. Taking the Fourier transform of equation (S11) and substituting into the Fourier version of (S9), we obtain:

$$\frac{\partial}{\partial z}\varepsilon_{\mathrm{D}}(z,t) = -\frac{\alpha}{2\pi}\int_{-\infty}^{\infty}\varepsilon_{\mathrm{D}}(z,\omega)\left[\frac{1}{i(\omega-\Delta)}+\pi(\omega-\Delta)\right]d\Delta = -\frac{\alpha}{2}\varepsilon_{\mathrm{D}}(z,\omega). \tag{S12}$$

The solution of equation (S12) in time domain is:

$$\varepsilon_{\mathrm{D}}(z,t) = e^{-\frac{\alpha z}{2}}\varepsilon_{\mathrm{D}}(0,t). \tag{S13}$$

The D-pulse exponentially decays as it propagates through the medium.

II. R_1-pulse

To retrieve D, we apply a π R_1-pulse in delay time T after the D-pulse. By the R_1-pulse the atoms are excited ($\sigma_{22}(z,t,\Delta) = 1$), and the corresponding equations of motion are:

$$\frac{\partial}{\partial t}\sigma_{12}(z,t,\Delta) = i\Delta\sigma_{12}(z,t,\Delta) - i\varepsilon_{\mathrm{R}_1}(z,t)\,, \tag{S14}$$

$$\frac{\partial}{\partial z}\varepsilon_{\mathrm{R}_1}(z,t) = \frac{i\alpha}{2\pi}\int_{-\infty}^{\infty}\sigma_{12}(z,t,\Delta)d\Delta. \tag{S15}$$

The equations (S14) and (S15) are obtained by substituting $\varepsilon_l = \varepsilon_{\mathrm{R}_1}$ and $\varepsilon_j = 0$ into equations (S4) and (S7). The solution of equation (S14) yields:

$$\sigma_{12}(z,t,\Delta) = e^{i\Delta(t-t_{\mathrm{R}_1})}\sigma_{12}(z,t_{\mathrm{R}_1},\Delta) - i\int_{t_{\mathrm{R}_1}}^{t}\varepsilon_{\mathrm{R}_1}(z,\acute{t})e^{i\Delta(t-\acute{t})}\,d\acute{t}. \tag{S16}$$

The R_1-pulse results in a phase conjugate of the D-excited coherence, so the coherence at $t = t_{\mathrm{R}_1}$ is equal to the conjugate of equation (S11):

$$\sigma_{12}(z,t,\Delta) = -\,i\,e^{-i\Delta(2t_{\mathrm{R}_1}-t)}\int_{-\infty}^{\infty}\varepsilon_{\mathrm{D}}^{\dagger}(z,\acute{t})e^{i\Delta\acute{t}}\,d\acute{t} - i\int_{t_{\mathrm{R}_1}}^{t}\varepsilon_{\mathrm{R}_1}(z,\acute{t})e^{i\Delta(t-\acute{t})}\,d\acute{t}. \tag{S17}$$

The negative sign in equation (S17) shows the emissive coherence of the photon echo. With some mathematical calculations, we obtain the following equation in a frequency domain:

$$\sigma_{12}(z,\omega,\Delta) = -\,i\,e^{-2i\Delta t_{\mathrm{R}_1}}2\pi\delta(\omega-\Delta)\int_{-\infty}^{\infty}\varepsilon_{\mathrm{D}}^{\dagger}(z,\acute{t})e^{i\Delta\acute{t}}d\acute{t} - i\varepsilon_{\mathrm{R}_1}(z,\omega)\left[\frac{1}{i(\omega-\Delta)}+\pi\delta(\omega-\Delta)\right]. \tag{S18}$$

By taking the Fourier transform of equation (S15) and substituting equation (S18), we get:

$$\varepsilon_{\mathrm{R}_1}(z,t) = \varepsilon_{\mathrm{R}_1}(0,t)e^{\frac{\alpha z}{2}} + 2\sinh\left(\frac{\alpha z}{2}\right)\varepsilon_{\mathrm{D}}^{\dagger}(0,2t_{\mathrm{R}_1}-t). \tag{S19}$$

The echo is emitted at $t = 2t_{\mathrm{R}_1} - t_{\mathrm{D}}$. The efficiency of the echo is $4\sinh^2\left(\frac{\alpha z}{2}\right)$, which is greater than unity for a large optical depth due to the stimulated emission in the inverted medium [1,2].

III. C_1 and C_2 -pulses

The function of C_1–pulse is to convert the optical coherence $\sigma_{12}(z,t,\mathit{\Delta})$ into spin coherence $\sigma_{13}(z,t,\mathit{\Delta})$. The coherence at t_{C_1} is given by:

$$\sigma_{12}(z,t,\mathit{\Delta}) = -\, i\, e^{-i\Delta(2t_{R_1}-t_{C_1})} \int_{-\infty}^{\infty} \varepsilon_{D}^{\dagger}(z,\acute{t}) e^{i\Delta \acute{t}}\, d\acute{t}. \tag{S20}$$

Here, we consider the first term of equation (S17) related to the evolution of the coherences excited by the D-pulse. The optical coherence is transferred to spin coherence via relation; $\sigma_{12}(z,t,\mathit{\Delta}) = \cos\left(\frac{\pi}{2}\right)\sigma_{12}\left(z,t_{C_1},\mathit{\Delta}\right) = 0$ and $\sigma_{13}(z,t,\mathit{\Delta}) = e^{i\pi/2}\sigma_{12}\left(z,t_{C_1},\mathit{\Delta}\right)$ [17,18]. The π C_1 –pulse resulting additional $\pi/2$ phase shift, leading to the freezing of the optical coherence. The C_2 –pulse transfers back the spin coherence to optical coherence. By setting $\varepsilon_l = 0$ and $\sigma_{13}(z,t,,\mathit{\Delta}) = 0$ in equation (S4), we get:

$$\frac{\partial}{\partial t}\sigma_{12}(z,t,\mathit{\Delta}) = i\mathit{\Delta}\sigma_{12}(z,t,\mathit{\Delta}). \tag{S21}$$

The solution of (S21) is:

$$\sigma_{12}(z,t,\mathit{\Delta}) = \sigma_{12}\left(z,t_{C_2},\mathit{\Delta}\right)e^{i\Delta(t-t_{C_2})}, \tag{S22}$$

The π C_2 –pulse re-swaps the coherences with another $\pi/2$ phase shift.

$$\sigma_{12}\left(z,t_{C_2},\mathit{\Delta}\right) = e^{i\pi/2}\sigma_{13}(z,t,\mathit{\Delta}) = e^{i\pi}\sigma_{12}\left(z,t_{C_1},\mathit{\Delta}\right) = i\, e^{-i\Delta(2t_{R_1}-t_{C_1})}\int_{-\infty}^{\infty}\varepsilon_{D}^{\dagger}(z,\acute{t})e^{i\Delta\acute{t}}\, d\acute{t}. \tag{S23}$$

Substituting equation (S23) into (S22), we obtain:

$$\sigma_{12}(z,\mathrm{t},\mathit{\Delta}) = i\, e^{-i\Delta(t_{C_2}-t_{C_1}+2t_{R_1}-t)}\int_{-\infty}^{\infty}\varepsilon_{D}^{\dagger}(z,\acute{t})e^{i\Delta\acute{t}}\, d\acute{t}. \tag{S24}$$

Here the final atomic coherence is positive. Thus, the echo becomes absorptive and cannot be radiated from the medium [18]. If the area of the C_2 is 2π, it brings the atomic coherence halted ($\sigma_{12}(z,t) = 0$) again transferring it to the spin state $|3\rangle$. For 3π-C_2, the atoms are returned in the excited state $|2\rangle$, and $\sigma_{12}\left(z,t_{C_2}t_{C_2},\mathit{\Delta}\right) = -\sigma_{12}\left(z,t_{C_2}t_{C_2},\mathit{\Delta}\right)$ in equation (S24), where the echo becomes emissive. Therefore, π-3π control pulse sequence is valid for a single rephasing scheme as shown in ref. [18]. Thus, the controlled AFC echoes with the π-π control pulse sequence [8,9] results in an absorptive echo [3]. The observation of the controlled AFC is due to coherence leakage by spatial Gaussian distribution of laser light [20].

References

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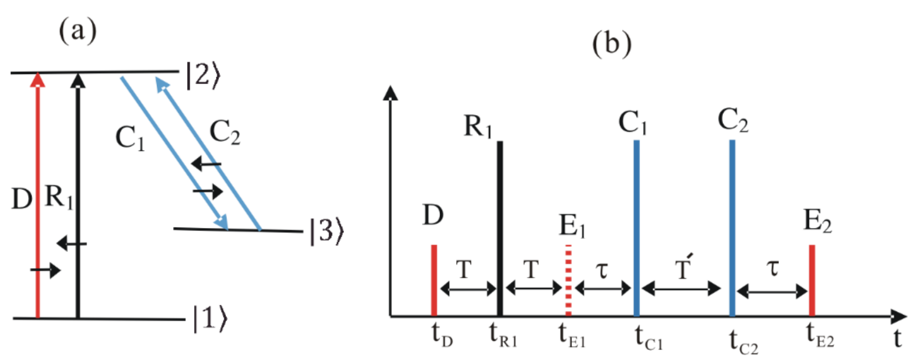

Fig. S1. (a) Schematic single rephasing echoes in a three-level system. (b) Pulse sequence for (a). tj is the arrival time of pulse j