

## Restoring Coherence Lost to a Slow Interacting Mesoscopic Spin Bath

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For a two-state quantum object interacting with a slow mesoscopic interacting spin bath, we show that a many-body solution of the bath dynamics conditioned on the quantum-object state leads to an efficient control scheme to recover the lost quantum-object coherence through disentanglement. We demonstrate the theory with the realistic problem of one electron spin in a bath of many interacting nuclear spins in a semiconductor quantum dot. The spin language can be easily generalized to a quantum object in contact with a bath of interacting multilevel quantum units with the caveat that the bath is mesoscopic and its dynamics is slow compared with the quantum object.

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The coherent superposition of states of a quantum object is the wellspring of quantum properties and key to quantum technology. Decoherence of a quantum object results from the entanglement with an environment by coupled dynamics [1–3]. Amelioration of decoherence becomes important in any sustained quantum process. Different types of amelioration include dynamical decoupling [4–6], decoherence-free subspace [7], quantum error correction (for a review, see [8]), and feedback control [9].

We offer an alternate approach to the restoration of coherence based on the theory that control of the quantum object can direct the quantum evolution of the bath to disentangle the object from the bath. The operation resembles the spin-echo schemes [10] but it removes the pure decoherence due to bath interaction dynamics as well as the inhomogeneous broadening effect. The key is that the environment is effectively a mesoscopic system, i.e., the number of particles  $N$  is small enough for the time scale of the quantum-object decoherence to be much smaller than its energy relaxation time  $T_1$  while large enough for ergodicity, specifically for the Poincaré period to be effectively infinite as compared to  $T_1$ . Our theoretical demonstration of coherence restoration uses one electron spin in a semiconductor quantum dot of many ( $N \sim 10^6$ ) nuclear spins, which serves as a paradigmatic system of a two-level system in a bath of interacting spins for decoherence physics [11] and for spin-based quantum technology [12]. Electron spin decoherence due to the hyperfine interaction with the nuclear spins has been much studied [13–19]. Theories of the effect of interaction between nuclear spins on the electron decoherence have recently appeared [18,19]. Our theory of coherence recovery by disentanglement is based on the previous finding [19] that the mesoscopic bath of slow dynamics is well described by a simple pseudospin model for the particle pair interaction in the bath.

The model for the coupled spin-bath system is a localized electron of spin  $\frac{1}{2}$  coupled to a bath of finite  $N$  mutually interacting nuclei with spin  $j$  in a magnetic field. The isolation of the electron spin plus the mesoscopic bath,

in the relevant time scale, from the rest of the Universe arises out of their weak coupling with the outside. The initial state of the electron spin,  $|\varphi^s(0)\rangle = C_+|+\rangle + C_-|-\rangle$ , is prepared as a coherent superposition of the spin up and down states  $|\pm\rangle$  in an external magnetic field. The state of the total system of the spin plus bath at that instant forms an unentangled state,  $|\Psi(0)\rangle = |\varphi^s(0)\rangle \otimes |\mathcal{J}\rangle$ . It evolves over time  $t$  to the entangled state  $|\Psi(t)\rangle = C_+(t)|+\rangle \otimes |\mathcal{J}^+(t)\rangle + C_-(t)|-\rangle \otimes |\mathcal{J}^-(t)\rangle$ , where the bath states  $|\mathcal{J}^\pm(t)\rangle$  are different. The electron spin state is now given by the reduced density matrix by tracing over the bath states,  $\rho_{\sigma,\sigma'}^s(t) = C_{\sigma'}^*(t)C_\sigma(t)\langle\mathcal{J}^{\sigma'}(t)|\mathcal{J}^\sigma(t)\rangle$ ,  $\sigma, \sigma' = \pm$ . The environment-driven transfer between  $\rho_{+,+}^s$  and  $\rho_{-,-}^s$  is longitudinal relaxation. Either off-diagonal element gives a measure of the coherence of the spin. The longitudinal relaxation contributes to the decoherence. When this contribution is removed, the remaining decoherence is called pure dephasing. For applications in quantum technology, the longitudinal relaxation can be virtually suppressed by a choice of system and of the electron Zeeman splitting much larger than the dominant excitation energies in the bath and the spin-bath coupling strength [20]. In time scale  $\ll T_1$ , the reduced Hamiltonian of the whole system is in the form diagonal in the electron spin basis,  $\hat{H} = |+\rangle\langle+| \otimes \hat{H}^+ + |-\rangle\langle-| \otimes \hat{H}^-$ . The bath evolves under the Hamiltonians  $\hat{H}^\pm$  into separate states  $|\mathcal{J}^\pm(t)\rangle \equiv e^{-i\hat{H}^\pm t}|\mathcal{J}\rangle$  depending on the electron basis states  $|\pm\rangle$ . Pure dephasing is then measured by  $\mathcal{L}_{+,-}^s(t) = |\langle\mathcal{J}|e^{i\hat{H}^- t}e^{-i\hat{H}^+ t}|\mathcal{J}\rangle|$ . The electron spin coherence may be restored by exploiting the dependence of the bath dynamics on the electron spin states to make the bifurcated bath pathways intersect at a later time, i.e.,  $|\mathcal{J}^+(t)\rangle = |\mathcal{J}^-(t)\rangle$ , leading to disentanglement.

At temperature ( $\sim 10$  mK–1 K)  $\gg$  the nuclear Zeeman energy  $\omega_n$  ( $\sim$  mK)  $\gg$  nuclear spin interaction ( $\sim$  nK), the nuclear bath initially has no off-diagonal coherence and is described by  $\sum_{\mathcal{J}} P_{\mathcal{J}}|\mathcal{J}\rangle\langle\mathcal{J}|$  where  $|\mathcal{J}\rangle \equiv \bigotimes_n |j_n\rangle$ ,  $j_n$  is the quantum number for  $\hat{J}_n^z$ , the component of the  $n$ th nuclear spin along  $z$  (the magnetic field direction), and  $P_{\mathcal{J}}$  gives

thermal distribution. The essence of electron decoherence is contained in the consideration of each pure bath state  $|\mathcal{J}\rangle$  and later the ensemble average over  $\mathcal{J}$  is included. The transverse interaction  $\hat{J}_n^+ \hat{J}_m^-$  between two nuclear spins creates the pair-flip excitation,  $|j_n\rangle|j_m\rangle \rightarrow |j_n + 1\rangle|j_m - 1\rangle$ . We sort out all such elementary excitations from the “vacuum” state  $|\mathcal{J}\rangle$  and denote each by the flip-process of a pseudospin  $\frac{1}{2}$  indexed by  $k$ :  $|\uparrow_k\rangle \rightarrow |\downarrow_k\rangle$ , characterized by the energy cost  $\pm E_k + D_k$  and the transition matrix element  $\pm A_k + B_k$  depending on the electron  $|\pm\rangle$  state.  $\pm E_k$  ( $\sim \mathcal{A}/N$ ) is from the longitudinal interaction of the form  $\hat{S}^z \hat{J}_n^z$  between the electron spin  $\hat{S}^z$  and each of the two nuclear spins [21].  $A_k$  ( $\sim \mathcal{A}^2/N^2\Omega$ ,  $\Omega$  being the electron Zeeman energy) is the extrinsic nuclear interaction [19], i.e., the effective interaction mediated by hyperfine coupling with the single electron [see Fig. 1(a)]. Its dependence on the number of particles in the bath signifies its mesoscopic nature.  $B_k$  ( $\sim b$ ) is due to the transverse part of the intrinsic nuclear interaction (referring to nuclear interactions that exist in the semiconductor matrix, e.g., dipolar). The extrinsic interaction  $A_k$  couples any two spins in the mesoscopic bath, as opposed to the finite-range intrinsic interaction  $B_k$ .  $D_k$  ( $\sim b$ ) is due to the longitudinal part of the intrinsic nuclear interaction.

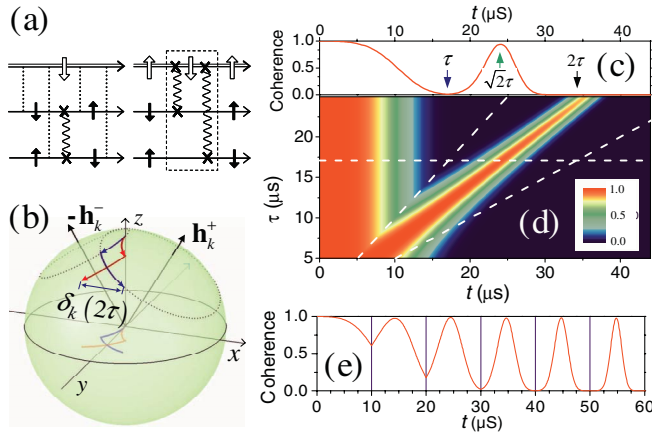


FIG. 1 (color). (a) Diagrams for the nuclear spin pair-flip by intrinsic (left) and extrinsic (right) interaction, where the single (double) horizontal lines stand for the nuclear (electron) spin propagators and the wavy (dotted) vertical lines for the transverse (longitudinal) spin interactions. The processes in the dashed box yield the extrinsic nuclear interaction. (b) Evolution of the pseudospin vector for the electron spin state  $|+\rangle$  (red) and  $|-\rangle$  (blue), under the control of a single pulse. (c) Electron spin coherence  $\mathcal{L}_{+,-}^s(t)$  under the control of a single flip pulse applied at  $\tau = 17 \mu\text{s}$  (indicated with the blue arrow). (d) Contour plot of the electron spin coherence  $\mathcal{L}_{+,-}^s$  under single pulse control as a function of time  $t$  and the pulse delay time  $\tau$  (indicated by the left tilted line). Right tilted line indicates conventional echo time  $2\tau$ . The horizontal line is the cut from the curve in (c). (e) Electron spin coherence with a sequence of  $\pi$  pulses (indicated by purple vertical lines) at intervals of  $\tau = 10 \mu\text{s}$ .

In the nuclear bath with the descending order of parameters,  $\Omega \gg \omega_n \gg \mathcal{A}/N \gg b$ , the bath dynamics is slow and the density of pair-flip excitations created from the vacuum state  $|\mathcal{J}\rangle$  is much less than unity in time scale of interest [19,22]. The excitations are almost always spatially separated, leading to the pair-correlation approximation [18,19] which treats pair flips as independent of each other. The bath, depending on the electron  $|\pm\rangle$  state, is then driven by the effective Hamiltonian derived from the first-principles interactions [19],

$$\hat{H}^\pm = \sum_k \hat{\mathcal{H}}_k^\pm \equiv \sum_k \mathbf{h}_k^\pm \cdot \hat{\sigma}_k/2, \quad (1)$$

where  $\hat{\sigma}_k$  is the Pauli matrix for pseudospin  $k$  driven by a pseudomagnetic field  $\mathbf{h}_k^\pm \equiv (2B_k \pm 2A_k, 0, D_k \pm E_k)$ .

From the justification that correlations of more than two spins are negligible [19,22], we derive the restrictions which the decoherence time scale places on the size of the bath  $N$ , given by  $N^2 b^2 \mathcal{A}^{-2} \ll 1 \ll \min(\sqrt{N}, N^4 b^2 \Omega^4 \mathcal{A}^{-6})$ , to be established below. The upper bound for  $N$  distinguishes the bath from a macroscopic system. It comes from the dominance of the pair correlation in the interaction dynamics of the bath spins over the correlations of more than two particles due to the intrinsic interaction. The lower bound,  $N^4 b^2 \Omega^4 \mathcal{A}^{-6} \gg 1$ , is by a similar consideration but due to the extrinsic interaction of the bath spins. The lower bound  $\sqrt{N} \gg 1$  simply signifies the necessary statistics for decoherence. In the case of the electron spin in a GaAs quantum dot, the theory is well justified for  $10^8 \geq N \geq 10^4$  which covers quantum dots of all practical sizes.

The theory of the interacting nuclear spin dynamics dominated by the pair excitation in the form of the pseudospin evolution leads to a simple physical picture of coherence decay and restoration. The initial unpolarized bath state  $|\mathcal{J}\rangle = \otimes_n |j_n\rangle$  can be replaced by the pseudospin product state  $\otimes_k |\uparrow_k\rangle$ . Each pseudospin, representing a nuclear spin states pair, initially points along the pseudospin  $+z$  axis and then precesses about the pseudomagnetic field  $\mathbf{h}_k^\pm$ ,  $|\psi_k^\pm\rangle \equiv e^{-(i/2)\mathbf{h}_k^\pm \cdot \hat{\sigma}_k t} |\uparrow_k\rangle$ , depending on the electron  $|\pm\rangle$  state. Thus, the electron spin coherence is measured by the divergence of the pseudospin paths,  $\mathcal{L}_{+,-}^s(t) = \prod_k |\langle \psi_k^- | \psi_k^+ \rangle| \cong e^{-\sum_k \delta_k^2/2}$ .  $\delta_k \equiv \sqrt{1 - |\langle \psi_k^- | \psi_k^+ \rangle|^2}$  is the geometric distance between the two conjugate pseudospin paths on Bloch sphere.

Now we examine the consequences of the pseudospin echo. A fast  $\pi$  pulse applied at  $t = \tau$  to flip the electron spin [13] would cause the pseudospin evolution

$$|\psi_k^\pm(t)\rangle = e^{-(i/2)\mathbf{h}_k^\mp \cdot \hat{\sigma}_k(t-\tau)} e^{-(i/2)\mathbf{h}_k^\pm \cdot \hat{\sigma}_k \tau} |\uparrow\rangle. \quad (2)$$

To find out how to control the decoherence, we neglect for the time being the diagonal nuclear spin interaction  $D_k$ , which contributes to the same component of the pseudomagnetic field as  $E_k$  but much smaller. Because the pseudofields dominated by the extrinsic nuclear spin

interaction,  $\mathbf{h}_k^\pm \equiv \pm(2A_k, 0, E_k)$ , invert exactly into each other, disentanglement of the electron spin from the affected bath spin pairs follows at  $2\tau$  as in the classic spin echo to remove the inhomogeneous broadening effect. The pseudofields dominated by the intrinsic interaction,  $\mathbf{h}_k^\pm \equiv (2B_k, 0, \pm E_k)$ , do not exactly invert under the influence of the electron spin flip and the resultant pseudospin paths are illustrated in Fig. 1(b). Disentanglement from the affected nuclear pairs and, hence, recovery of the electron spin coherence occurs, by the rotation kinematics, at time  $\sqrt{2}\tau$ , distinct from the classic echo. Figure 1 gives the computed results for a GaAs dot with thickness  $d = 8.5$  nm in growth direction [001] and lateral Fock-Darwin radius  $r_0 = 25$  nm, on the large- $N$  side of the mesoscopic regime where the intrinsic nuclear interaction dominates [19]. The electron  $g$  factor is  $-0.44$  and  $\mathbf{B}_{\text{ext}} = 10$  T along the [110] direction. The initial bath state is randomly chosen from a thermal ensemble at temperature  $T = 1$  K. Figures 1(c) and 1(d) reveal the coherence recovery after a flip of the electron spin at a range of values for  $\tau$ , even after the coherence has apparently vanished. The restoration of the coherence is pronounced at  $\sqrt{2}\tau$  whereas no coherence peak is visible at the conventional echo time  $2\tau$ .

Furthermore, the coherence may be restored by a sequence of electron spin flips. For example, with a sequence of  $\pi$  pulses evenly spaced with interval  $\tau$ , the disentanglement from the bath will occur at  $\sqrt{n(n+1)}\tau$  between the  $n$ th and the  $(n+1)$ th pulses, as illustrated in Fig. 1(e). Consider the correction from the small term  $D_k$  in the pseudofields, the residue decoherence, at the disentanglement point  $\sqrt{n(n+1)}\tau$ , is measured by  $\delta_k^2 \sim (E_k B_k D_k \tau^3)^2$ . Compared with the free-induction decay [19] where  $\delta_k^2(\tau) \sim E_k^2 B_k^2 \tau^4$ , the decoherence is reduced by a factor of  $\sim D_k^2 \tau^2$  ( $\sim 10^{-4}$  for  $\tau \sim 10$   $\mu\text{s}$ ).

Ensemble average over the mixed bath states is necessary in two scenarios, namely, observation of decoherence of an ensemble of quantum objects and observation of a single quantum object repeated in a time sequence [13–16]. The coherence of the electron spin is now  $\rho_{+-}(t) = C_-^* C_+ \mathcal{L}_{+-}^s(t) \mathcal{L}_{+-}^0(t)$ , where  $\mathcal{L}_{+-}^0(t) \equiv \sum_{\mathcal{J}} P_{\mathcal{J}} e^{-i\phi_{\mathcal{J}}(t)}$  is the inhomogeneous broadening factor due to the probability distribution  $P_{\mathcal{J}}$  of the initial bath state  $|\mathcal{J}\rangle$  (different nuclear bath state may result in different Overhauser energy-splitting  $\mathcal{E}_{\mathcal{J}}$  of the electron) [19].  $\phi_{\mathcal{J}}(t) = \mathcal{E}_{\mathcal{J}}[\tau_1 - (\tau_2 - \tau_1) + \dots + (-1)^n(t - \tau_n)]$  under the control of a sequence of  $\pi$  pulses on electron spin at  $\tau_1, \tau_2, \dots$ , and  $\tau_n$ . The coherence factor  $\mathcal{L}_{+-}^s(t)$  is insensitive, up to a factor of  $1/\sqrt{N} \ll 1$ , to the selection of initial bath state  $|\mathcal{J}\rangle \equiv \otimes_n |j_n\rangle$  (verified by numerical evaluations), and is taken out of the summation [19].

Both the inhomogeneous broadening and the pure decoherence due to the extrinsic nuclear interaction are shown to be removed at the classic spin-echo time  $2\tau$  in contrast to the unusual recovery time of  $\sqrt{2}\tau$  in the case of intrinsic

nuclear interaction. We need a pulse sequence to produce a time where the decoherence from all three sources can be removed. A solution is a two-pulse control. Figure 2(a) shows that, after a second electron spin flip at  $3\tau$ , the two pseudospin paths corresponding to the electron  $|\pm\rangle$  states, driven by the intrinsic nuclear interaction, cross again at  $4\tau$ , coinciding with the secondary spin-echo time for the other two causes. This two-pulse sequence is well known as Carr-Purcell sequence in NMR spectroscopies [10]. The residual decoherence at  $t = 4\tau$  is  $\delta_k^2 \sim 16(E_k B_k - A_k D_k)^2 D_k^2 \tau^6$ . The restoration by two-pulse control of coherence in the presence of both pure and ensemble decoherence is demonstrated by the results of numerical evaluation in Fig. 2 for a smaller quantum dot ( $d = 2.8$  nm and  $r_0 = 15$  nm) with identical geometry and external field as that studied in Fig. 1. The nuclear bath is assumed initially in thermal equilibrium at  $T = 1$  K. Figures 2(b) and 2(c) show that the electron spin coherence is restored at  $4\tau$  by the second pulse even when the first spin echo at  $2\tau$  has completely vanished, illustrating the remarkable observation [23] that the absence of spin echo does not mean irreversible loss of coherence. To make the echo visible in the plot, we have artificially set the en-

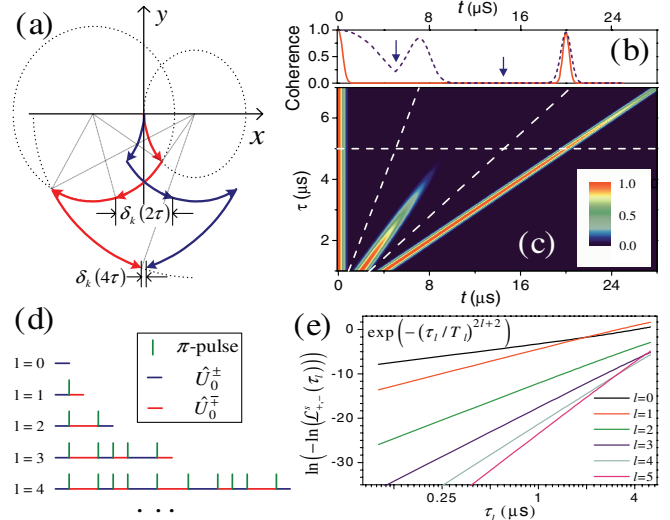


FIG. 2 (color). (a) Pseudospin paths (projected on  $x$ - $y$  plane) driven by intrinsic nuclear interaction with two flips of the electron spin at  $\tau$  and  $3\tau$ . (b) Evolution of the electron spin coherence under two-pulse control with  $\tau = 5$   $\mu\text{s}$ . The dashed purple line denotes the pure state dynamics part  $\mathcal{L}_{+-}^s$  and the solid red line includes the inhomogeneous broadening factor  $\mathcal{L}_{+-}^0$ . The blue arrows indicate the times of the electron spin flips. (c) Contour plot of the ensemble-averaged electron spin coherence under the two-pulse control. The tilted lines indicate the pulse times and the horizontal line is the cut for the curve in (b). (d) Concatenated pulse sequences. The  $(l+1)$ th order sequence is constructed by two subsequent  $l$ th order sequences, with a pulse inserted if  $l$  is even. (e) Dependence of echo magnitude on the echo delay time  $\tau_l$  under the control of the concatenated pulse sequences in ensemble measurement.



semble dephasing time  $T_2^*$  in  $\mathcal{L}_{+,-}^0$  to  $0.5 \mu\text{s}$ , about 100 times greater than its realistic value.

The power of concatenation of pulse sequences has been shown in the context of dynamical decoupling in quantum computation [4]. Similarly, the control of disentanglement of the bath states from the quantum object may be enhanced by concatenation. The pseudospin evolution with the two-pulse control of the quantum object can be constructed recursively from the free-induction evolution  $\hat{U}_0^\pm = e^{-i\mathbf{h}_k^\pm \cdot \hat{\sigma}_k \tau/2}$ , by the concatenation,  $\hat{U}_l^\pm = \hat{U}_{l-1}^\pm \hat{U}_{l-1}^\pm$ ,  $l = 1, 2$ . The process can be extended by iteration to any level  $\hat{U}_l^\pm = e^{-i\boldsymbol{\theta}_l^\pm \cdot \hat{\sigma}_k \tau/2}$  as shown in Fig. 2(d), where  $\boldsymbol{\theta}_l^\pm$  is the rotation vector along the axis of rotation through an angle  $\theta_l^\pm$ . Disentanglement occurs at  $\tau_l \equiv 2^l \tau$  coinciding with the classic spin echo. For small  $\theta_l^\pm$ , the recursion relation is  $\boldsymbol{\theta}_{l+1}^\pm = \boldsymbol{\theta}_l^\pm + \boldsymbol{\theta}_l^\mp + \boldsymbol{\theta}_l^\pm \times \boldsymbol{\theta}_l^\mp$ . At each iteration, the rotation vectors of the conjugate pseudospin states have their mean  $(\boldsymbol{\theta}_l^+ + \boldsymbol{\theta}_l^-)/2$  increased by a factor of 2 and their difference  $(\boldsymbol{\theta}_l^+ - \boldsymbol{\theta}_l^-)$  reduced by a factor of  $\theta_l^\pm \sim 2^l b\tau$  (deduced by induction from  $\theta_1^\pm \sim 2b\tau$ ). The decoherence is reduced by an order of  $b^2 \tau_l^2$  at  $\tau_l$  for each additional level of concatenation till saturation at the level  $l_0 \approx -\log_2(b\tau)$ . Hence, the coherence echo magnitude scales with the echo delay time according to  $\exp[-(\tau_l/T_1)^{2l+2}]$  as shown in Fig. 2(e). Our result shows the protection of electron spin coherence by pulse sequences with interpulse interval up to  $\sim 10 \mu\text{s}$ .

In conclusion, we note that our scheme of restoring the coherence depends on the pure decoherence being driven by the interaction in the spin bath and by the domination of the bath pair excitation in the slow bath dynamics. The pulse sequence design is borrowed from the dynamical decoupling schemes in NMR spectroscopies [10] and in quantum computation [4] but the disentanglement method aims directly at the bath dynamics. Our method seeks not to eliminate the object-bath interaction by dynamical averaging, but to disentangle by controlling the quantum object to maneuver the bath evolution. Thus, elimination of coupling between the quantum object and the bath is not a necessary condition for their disentanglement, as illustrated by coherence recovery at  $t = \sqrt{n(n+1)}\tau$  where effective object-bath interaction does not vanish even in the first order of hyperfine coupling  $\mathcal{A}/N$ . Direct observation of coherence echoes at such magic times is possible with the narrowing of inhomogeneous distribution by measurement projection [24]. The control of bath spins may well develop into a valuable addition to the collection of armaments of coherence preservation for quantum information processing.

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*Note added.*—Recently, a report appeared [25] that contains results similar to our Carr-Purcell control.

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