THE JOURNAL OF PHYSICAL CHEMISTRY B

ARTICLE

pubs.acs.org/JPCB

Tunneling under Coherent Control by Sequences of Unitary Pulses

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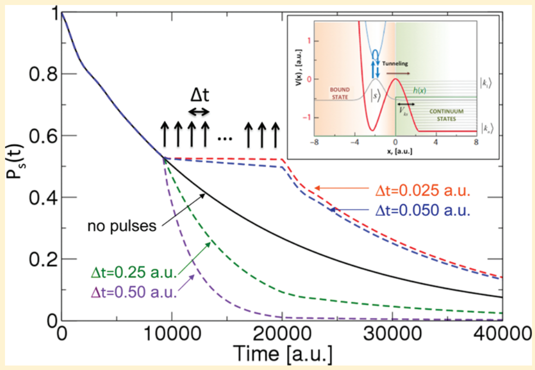
ABSTRACT: A general coherent control scenario to suppress or accelerate tunneling of quantum states decaying into a continuum is investigated. The method is based on deterministic, or stochastic, sequences of unitary pulses that affect the underlying interference phenomena responsible for quantum dynamics, without inducing decoherence, or collapsing the coherent evolution of the system. The influence of control sequences on the ensuing quantum dynamics is analyzed by using perturbation theory to first order in the control pulse fields and compared to dynamical decoupling protocols and to sequences of pulses that collapse the coherent evolution and induce quantum Zeno (QZE) or quantum anti-Zeno effects (AZE). The analysis reveals a subtle interplay between coherent and incoherent phenomena and demonstrates that dynamics analogous to the evolution due to QZE or AZE can be generated from stochastic sequences of unitary pulses when averaged over all possible realizations.



I. INTRODUCTION

Advancing our understanding of coherent control techniques to accelerate or suppress tunneling of quantum states decaying into a manifold of continuum states is a problem of great technological interest.[1−3] Tunneling is central to a wide range of molecular and electronic processes and often determines the lifetime of metastable states and the timescales of electron and proton transfer. During the past 30 years, several coherent control methods have been developed and optimized to manipulate a wide range of quantum processes[4−21] including tunneling dynamics.[6−8,13−19] This paper focuses on one of the most recently proposed methods,[15,22−24] based on sequences of unitary pulses that repetitively change the phases of interfering states responsible for quantum dynamics without inducing decoherence or collapsing the coherent evolution of the system. The method has been numerically demonstrated as applied to control superexchange electron tunneling dynamics in monolayers of adsorbate molecules functionalizing semiconductor surfaces when using either deterministic or stochastic sequences of unitary phase-kick pulses.[25−30] However, the underlying control mechanism induced by those sequences of unitary pulses has been difficult to elucidate from a cursory examination of the ensuing dynamics.

This paper reports a rigorous theoretical analysis of the origin of quantum control as resulting from the interplay between coherent and incoherent phenomena induced by deterministic or stochastic sequences. Control is analyzed by perturbation theory to first order in the pulse fields and compared with dynamical decoupling (DD) protocols[31−33] and sequences of pulses that periodically collapse the coherent evolution[34−37] and yield dynamics modulated by quantum Zeno (QZE) and quantum anti-Zeno (AZE) effects.[15,22,38] The reported results provide fundamental insights into the origin of suppression of quantum tunneling by sufficiently frequent perturbation pulses and acceleration induced by pulses separated by finite time intervals.

The analytic expressions reported for the description of short-time dynamics also provide understanding of the effect of randomization of pulse sequences and clarify how the ensuing dynamics depends on the average time period between perturbational phase-kick pulses when averaged of all possible realizations of control sequences. These results are particularly valuable, since stochastic pulse sequences have already been demonstrated to achieve control in condensed material systems[29,30] or predicted to outperform deterministic pulsed schemes in control of quantum coherences.[15,22] Considering that current laser technology can produce a wide range of pulses with ultrashort time resolution and extremely high-peak power, it is natural to expect that the quantum control techniques explored in this paper should raise significant experimental interest.[34,39]

The paper is organized as follows. Section II introduces the model system and the description of spontaneous decay due to tunneling into a continuum. Section III introduces coherent control based on equally time-spaced phase-kick pulses, as applied to the acceleration or suppression of tunneling into a continuum. Section IV analyzes a generalization of the method to sequences of randomly time-spaced pulses. Section V explores stochastic sequences, averaged over all possible realizations, as compared with DD protocols and quantum Zeno effects. Concluding remarks and future directions are presented in Section VI.

Special Issue: Shaul Mukamel Festschrift

Received: September 1, 2010
Revised: January 5, 2011

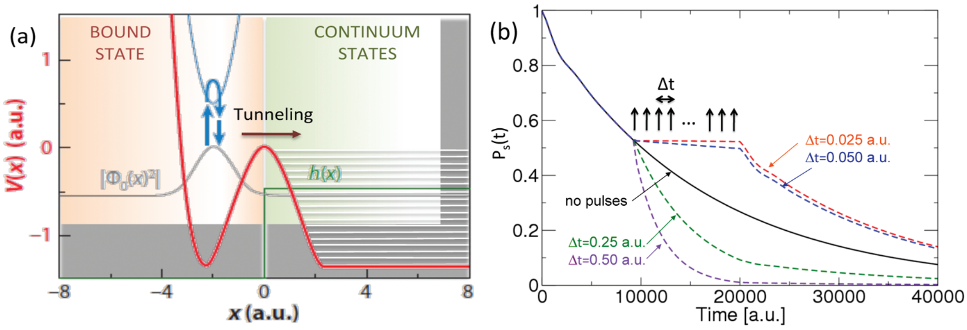


Figure 1. (a) Unstable quantum state $|s\rangle$ tunneling into a manifold of continuum states $|k_1\rangle|k_n\rangle$ with couplings V_{ks}. The initial metastable state $\langle x|s\rangle = \Phi_0(x)$ is defined as the ground state of an electron in the quadratic approximation to the quartic potential $V(x) = -\alpha x^2 + \beta x^4$ with $\alpha = 1/2$ and $\beta = 0.0461$. (b) Time-dependent population $P_s(t) = \langle\varphi_t|1 - h(\hat{x})|\varphi_t\rangle$ due to evolution on $V(x)$, with $V(x) = V(x_e)$ for $x < x_e = 2.33$ au, as modulated by instantaneous 2π pulses applied at time intervals Δt. Here, $h(x) = 1$ for $x < 0$ and $h(x) = 0$, otherwise.[22]

II. TUNNELING INTO A CONTINUUM

We consider the system depicted in Figure 1, initially prepared in a bound metastable state $|s\rangle$ coupled to a continuum, as described by the following Hamiltonian:[39,40]

$$\hat{H} = \omega_s|s\rangle\langle s| + \sum_k \omega_k|k\rangle\langle k| + \sum_k (V_{ks}|k\rangle\langle s| + V_{sk}|s\rangle\langle k|) \tag{1}$$

where $|s\rangle$ and $|k\rangle$ are stationary eigenstates of $\hat{H}$, when $V_{ks} = 0$, with energies ω_s and ω_k, respectively. For simplicity, notation is kept in atomic units (with $\hbar = 1$). When $V_{ks} \neq 0$, state $|s\rangle$ is nonstationary. Therefore, a system initially prepared in state $|s\rangle$ spontaneously decays by tunneling into the continuum. In the absence of external perturbations, the time-evolution is described by the time-dependent wave function,

$$\Psi(t) = \alpha_s(t)\, e^{-i\omega_s t}|s\rangle + \sum_k \beta_k(t)\, e^{-i\omega_k t}|k\rangle \tag{2}$$

with $\alpha_s(0) = 1$, and $\beta_k(0) = 0$ for all $|k\rangle$. The equations of motion of the time-dependent expansion coefficients, introduced by eq 2, are obtained by solving the time-dependent Schrödinger equation, as follows:

$$\dot{\alpha}_s = -i\sum_k V_{sk} e^{i(\omega_s - \omega_k)t}\beta_k \tag{3}$$

$$\dot{\beta}_k = -iV_{ks} e^{i(\omega_k - \omega_s)t}\alpha_s \tag{4}$$

Integrating eq 4 from time t_b to time t yields

$$\beta_k(t) - \beta_k(t_b) = -i\int_{t_b}^{t} V_{ks} e^{i(\omega_k - \omega_s)t'}\, \alpha_s(t')\, dt' \tag{5}$$

and substituting eq 5 into eq 3 gives

$$\dot{\alpha}_s = -\sum_k \int_{t_b}^{t} |V_{ks}|^2 e^{i(\omega_k - \omega_s)(t' - t)}\, \alpha_s(t')\, dt' - i\sum_k V_{sk} e^{i(\omega_s - \omega_k)t}\, \beta_k(t_b) \tag{6}$$

Equation 6 can be solved exactly by using standard Laplace transform techniques;[34,41] however, for sufficiently short time intervals, one can approximate $\alpha_s(t') \approx \alpha_s(t_b)$ as shown in Appendix A and obtain the following solution of eq 6:[41,42]

$$\alpha_s(t) \approx \alpha_s(t_b)\left(1 - \sum_k |V_{ks}|^2 \int_{t_b}^{t} (t - t') e^{i\Omega_{ks}(t' - t_b)}\, dt'\right) - i\sum_k V_{sk} \int_{t_b}^{t} e^{i\Omega_{sk}t'}\, \beta_k(t_b)\, dt' \tag{7}$$

where we introduced $\Omega_{sk} = \omega_s - \omega_k$. Similarly, the expansion coefficients for states $|k\rangle$ are obtained from eq 5, as follows:

$$\beta_k(t) \approx \beta_k(t_b) - i\alpha_s(t_b)\, V_{ks} \int_{tb}^{t} e^{i\Omega_{sk}t'}\, dt' \tag{8}$$

It is important to note that the above approximation holds only in the limit of short time intervals for which $t - t_b < 2\pi/\Omega_{ks}$.

Equation 7 yields the standard expression for the spontaneous population decay of state $|s\rangle$ due to coupling to the manifold of continuum states $|k\rangle$, as follows:[15,22,43]

$$P_s(t) = |\alpha_s(t)|^2 = 1 - \sum_k \frac{|V_{ks}|^2}{\left(\frac{\Omega_{sk}}{2}\right)^2} \sin^2\left(\Omega_{sk}\frac{t}{2}\right) \tag{9}$$

and is valid up to second order in perturbation theory, since it neglects terms of $O(|V_{ks}|^3)$ and higher.

Sections III−V show that the spontaneous decay described by eq 9 can be suppressed or accelerated by perturbing the system with a train of unitary pulses (Figure 1) that change the *phase* of the wave function component along state $|s\rangle$ relative to the other terms in the coherent state expansion of eq 2. Section V also shows that eq 9 is recovered in the limit where the pulses have a low probability of inducing changes of phase.

III. PERIODIC PULSING

Consider the evolution of the system, introduced in SectionII, as perturbed by two consecutive instantaneous pulses, Q, spaced by a time interval Δt as follows:

1 Evolve the system for a short time period, Δt, using eqs 7 and 8.
2 Apply an instantaneous pulse, Q.
3 Continue the evolution, from $t = \Delta t$ to $t = 2\Delta t$, according to eqs 7 and 8.
4 Apply another pulse, Q.

Repeating steps 1−4 n times, evolves the system to time $t = 2n\Delta t$, yielding the expansion coefficients $\alpha_s(2n\Delta t)$ and $\beta_k(2n\Delta t)$ for states $|s\rangle$ and $|k\rangle$, respectively.

For simplicity, we consider the specific case of sequences of 2π pulses, for which each pulse, Q, changes the sign of the projection of the time-evolved wave function along the direction of $|s\rangle$ as follows:

$$\hat{Q}|\psi\rangle = |\psi\rangle - 2|s\rangle\langle s|\psi\rangle = \sum_k |k\rangle\langle k|\psi\rangle - |s\rangle\langle s|\psi\rangle \quad (10)$$

leaving unaffected the projection of $|\psi\rangle$ along the manifold of states $|k\rangle$ in the continuum. Therefore, 2π pulses can be represented as $Q_s = 1 - |s\rangle\langle s|$, yielding the following evolution for the expansion coefficients:

$$\begin{aligned}
\beta_k(\Delta t) &= \beta_k(0) - iV_{ks}\left(\frac{e^{i\Omega_{ks}\Delta t} - 1}{i\Omega_{ks}}\right)\alpha_s(0) \\
\alpha_s(\Delta t) &= \alpha_s(0)\left(1 - \int_0^{\Delta t}(\Delta t - t')\,K(t')\,dt'\right) \\
&\quad - i\sum_k V_{sk}\int_0^{\Delta t} e^{i\Omega_{sk}t}\,\beta_k(0)\,dt\alpha_s'(\Delta t) = -\alpha_s(\Delta t) \\
\beta_k(2\Delta t) &= \beta_k(\Delta t) - iV_{ks}\left(\frac{e^{i2\Omega_{ks}\Delta t} - e^{i\Omega_{ks}\Delta t}}{i\Omega_{ks}}\right)\alpha_s'(\Delta t) \\
\alpha_s'(2\Delta t) &= \alpha_s'(\Delta t)\left(1 - \int_{\Delta t}^{2\Delta t}(2\Delta t - t')\,K(t' - \Delta t)\,dt'\right) \\
&\quad - i\sum_k V_{sk}\int_{\Delta t}^{2\Delta t} e^{i\Omega_{sk}t}\,\beta_k(\Delta t)\,dt \\
\alpha_s(2\Delta t) &= -\alpha_s'(2\Delta t)
\end{aligned} \quad (11)$$

where $K(t) = \sum_k |V_{ks}|^2 e^{i\Omega_{sk}t}$. Collecting the expressions, introduced by eq 11, with $\alpha_s(0) = 1$ and $\beta_k(0) = 0$, we obtain

$$\begin{aligned}
\alpha_s(2\Delta t) &= \left\{\left(1 - \int_0^{\Delta t}(\Delta t - t')K(t')\,dt'\right) - i\sum_k \int_0^{\Delta t} V_{sk}e^{i\Omega_{sk}t'}\beta_k(0)dt'\right\}\times \\
&\quad \left(1 - \int_{\Delta t}^{2\Delta t}(2\Delta t - t')K(t' - \Delta t)\,dt'\right) + i\sum_k \int_{\Delta t}^{2\Delta t} V_{sk}e^{i\Omega_{sk}t'}\beta_k(\Delta t)dt' + \cdots, \\
&= 1 - \underbrace{\int_0^{\Delta t}(\Delta t - t')K(t')\,dt' - \int_{\Delta t}^{2\Delta t}(2\Delta t - t')K(t' - \Delta t)\,dt'}_{I} \\
&\quad \underbrace{-i\sum_k \int_0^{\Delta t} V_{sk}e^{i\Omega_{sk}t'}\beta_k(0)dt'}_{II} + \underbrace{i\sum_k \int_{\Delta t}^{2\Delta t} V_{sk}e^{i\Omega_{sk}t'}\beta_k(\Delta t)dt'}_{III}
\end{aligned} \quad (12)$$

where terms of $O(|V_{ks}|^3)$ have been neglected. Terms II and III have opposite signs and, therefore, introduce the couplings to the continuum with interference effects.

Repeating the process described above n times, we obtain $\alpha_s(2n\Delta t)$. The contributions to $\alpha_s(2n\Delta t)$ from term I are

$$\begin{aligned}
\text{Terms from } I &= \sum_k |V_{ks}|^2 \int_0^{\Delta t}(\Delta t - t')e^{i\Omega_{sk}t'}\,dt' + \ldots \\
&+ \sum_k |V_{ks}|^2 \int_{(j-1)\Delta t}^{j\Delta t}(j\Delta t - t')e^{i\Omega_{sk}(t' - [j-1]\Delta t)}\,dt' + \ldots \\
&+ \sum_k |V_{ks}|^2 \int_{(2n-1)\Delta t} 2n\Delta t\,(2n\Delta t - t')e^{i\Omega_{sk}(t' - [2n-1]\Delta t)}\,dt'
\end{aligned} \quad (13)$$

and changing the limits of integration in eq 13, we obtain

$$\text{Terms from } I = 2n\Delta t\sum_k |V_{ks}|^2\int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)e^{i\Omega_{sk}t'}\,dt' \quad (14)$$

Similarly, contributions from terms II and III are obtained, as follows:

$$\text{TermsfromIIandIII} = \sum_k \sum_{j=1}^{2n-1}(-1)^j\int_{j\Delta t}^{\{j+1\}\Delta t} V_{sk}e^{i\Omega_{sk}t'}\,\beta_k(j\Delta t)\,dt' \quad (15)$$

where the summation over j starts at $j = 1$ because $\beta_k(0) = 0$. Substituting 14 and 15 into 12, we obtain

$$\begin{aligned}
\alpha_s(2n\Delta t) &= 1 - 2n\Delta t\int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)K(t')\,dt' \\
&\quad - i\sum_k \sum_{j=1}^{2n-1}(-1)^j\int_{j\Delta t}^{\{j+1\}\Delta t} V_{sk}e^{i\Omega_{sk}t'}\,\beta_k(j\Delta t)\,dt' \\
&= 1 - 2n\Delta t\int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)K(t')\,dt' \\
&\quad - i\sum_k \sum_{j=1}^{2n-1}(-1)^j\,V_{sk}\left(\frac{e^{i\Omega_{sk}t'} - 1}{i\Omega_{sk}}\right)\beta_k(j\Delta t)
\end{aligned} \quad (16)$$

Appendix B shows that one can introduce the substitution,

$$\beta_k(j\Delta t) = \frac{V_{ks}}{\Omega_{ks}}\left[\frac{e^{\Omega_{ks}\Delta t} - 1}{e^{\Omega_{ks}\Delta t} + 1}\right]\{(-1)^j e^{i\Omega_{ks}j\Delta t} - 1\} \quad (17)$$

into eq 16 to obtain

$$\begin{aligned}
\alpha_s(2n\Delta t) &= 1 - 2n\Delta t\int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)K(t')\,dt' \\
&\quad - \sum_k e^{i\Omega_{sk}\Delta t}\frac{|V_{ks}|^2}{\Omega_{ks}^2}\frac{(e^{i\Omega_{ks}\Delta t} - 1)^2}{e^{i\Omega_{ks}\Delta t} + 1}\sum_{j=1}^{2n-1}\{1 - (-1)^j e^{i\Omega_{skj}\Delta t}\}
\end{aligned} \quad (18)$$

The summation over j in eq 18, as detailed in Appendix D, leads to the central result of this paper:

$$\begin{aligned}
\alpha_s(2n\Delta t) &= 1 - 2n\Delta t\underbrace{\sum_k \int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)|V_{ks}|^2 e^{i\Omega_{sk}t'}\,dt'}_{A} \\
&\quad - \sum_k \underbrace{\frac{|V_{ks}|^2}{\Omega_{ks}^2}\left(\frac{e^{i\Omega_{ks}\Delta t} - 1}{e^{i\Omega_{ks}\Delta t} + 1}\right)^2\left(e^{-i\Omega_{ks}2n\Delta t} - 1\right)}_{\mathbf{F}_k^1} \\
&\quad - 2n\sum_k \underbrace{\frac{|V_{ks}|^2}{\Omega_{ks}^2}e^{-i\Omega_{ks}\Delta t}\frac{(e^{-i\Omega_{ks}\Delta t} - 1)^2}{(e^{-i\Omega_{ks}\Delta t} + 1)}}_{\mathbf{F}_k^2}
\end{aligned} \quad (19)$$

Equation 19 is an important result, since it provides an explicit description of the state amplitude, $\alpha_s(2n\Delta t)$, as a function of the time interval Δt between phase-kick pulses, yielding fundamental insight into the origin of interference phenomena introduced by the various terms. In addition, eq 19 allows for calculations of the survival probability of state $|s\rangle$:

$$|\alpha_s 2n\Delta t|^2 = 1 - \underbrace{2n\Delta t \left\{ 2\mathrm{Re} \sum_k \int_0^{\Delta t} (1 - \frac{t'}{\Delta t}) |V_{ks}|^2 e^{i\Omega_{sk}t'}\, dt' \right\}}_{A} - \underbrace{2\mathrm{Re} \sum_k \mathbf{F}_k^1}_{B} - \underbrace{2\mathrm{Re} \sum_k \mathbf{F}_k^2}_{C} + \cdots \quad (20)$$

with

$$A = 2Re \sum_k |V_{ks}|^2 \int_0^{\Delta t} \left(1 - \frac{t'}{\Delta t}\right) e^{i\Omega_{sk}t'}\, dt' = \Delta t \sum_k |V_{ks}|^2 \frac{\sin^2\left(\frac{\Omega_{sk}\Delta t}{2}\right)}{\left(\frac{\Omega_{sk}\Delta t}{2}\right)^2}$$

$$B = 2Re \sum_k F_k^1 = \sum_k \frac{|V_{ks}|^2}{\left(\frac{\Omega_{sk}}{2}\right)^2} \tan^2\left(\Omega_{sk}\frac{\Delta t}{2}\right) \sin^2\left(\Omega_{sk}\frac{2n\Delta t}{2}\right)$$

$$C = 2Re \sum_k F_k^2 = -2n \sum_k |V_{ks}|^2 \frac{\sin^2 \Omega_{sk}\frac{\Delta t}{2}}{\left(\frac{\Omega_{sk}}{2}\right)^2} \quad (21)$$

Note that terms A and C in eq 20 cancel each other, and term B determines the time-dependent survival probability, as follows:

$$|\alpha_s(2n\Delta t)|^2 = 1 - 2Re \sum_k F_k^1 = 1 - \sum_k \frac{|V_{ks}|^2}{\left(\frac{\Omega_{sk}}{2}\right)^2} \tan^2\left(\Omega_{sk}\frac{\Delta t}{2}\right) \sin^2\left(\Omega_{sk}\frac{2n\Delta t}{2}\right) \quad (22)$$

Equation 22 gives the survival probability, $P_s(t) = |\alpha_s(t)|^2$, as a function of the time interval Δt between pulses. Note that when the time interval between pulses is large, eq 22 is identical to eq 9 describing the spontaneous decay in the absence of pulses. However, due to the modulatory factor $\tan^2(\omega_s - \omega_k)\Delta t/2$ in eq 22, decay is suppressed when the time interval between pulses is sufficiently short, $\Delta t \to 0$ (with $t = 2n\Delta t$), and accelerated relative to spontaneous decay when $\tan^2(\Omega_{sk}\Delta t/2) = \tan^2((\omega_s - \omega_k)\Delta t/2) > 1$. Maximum acceleration is achieved when $\Delta t = \pi/(\omega_s - \omega_k)$. Equation 22 agrees with previous work,[23,24] including the study of decay into a continuum,[27,29,44] and the decay of coherences in a system of spin 1/2 qubits in contact with a bosonic bath when periodically pulsed by dynamical decoupling sequences.[29] The derivation presented in this section, however, is novel, since contrary to earlier studies, it is derived from eq 19, providing an explicit description of the evolution of the expansion coefficient, $\alpha_s(t)$, as a function of the time interval Δt between pulses.

IV. STOCHASTIC PULSING

This section analyzes stochastic sequences of 2π pulses, using the perturbational treatment introduced in Section III. Rather than pulsing the system deterministically, as in Section III, stochastic sequences pulse the system at time intervals Δt, but only with 50% probability.

To obtain the survival probability $P_s(t) = |\alpha_s(t)|^2$ at time $t = 2n\Delta t$, we analyze first the state of the system at time $t = 2\Delta t$, obtained by propagating the expansion coefficients for states $|s\rangle$ and $|k\rangle$, as follows:

$$\beta_k(\Delta t) = \beta_k(0) - iV_{ks}\left(\frac{e^{i\Omega_{sk}\Delta t} - 1}{i\Omega_{sk}}\right)\alpha_s(0)$$

$$\alpha_s(\Delta t) = \alpha_s(0)\left(1 - \int_0^{\Delta t} (\Delta t - t')\, K(t')\, dt'\right) - i\sum_k V_{sk} \int_0^{\Delta t} e^{i\Omega_{sk}t}\, \beta_k(0)\, dt$$

$$\alpha_s'(\Delta t) = \xi_1\, \alpha_s(\Delta t)$$

$$\beta_k(2\Delta t) = \beta_k(\Delta t) - iV_{ks}\left(\frac{e^{i\Omega_{sk}2\Delta t} - e^{i\Omega_{sk}\Delta t}}{i\Omega_{sk}}\right)\alpha_s'(\Delta t)$$

$$\alpha_s'(2\Delta t) = \alpha_s'(\Delta t)\left(1 - \int_{\Delta t}^{2\Delta t} (2\Delta t - t')\, K(t' - \Delta t)\, dt'\right) - i\sum_k V_{sk} \int_{\Delta t}^{2\Delta t} e^{i\Omega_{sk}t}\, \beta_k(\Delta t)\, dt$$

$$\alpha_s'(2\Delta t) = \xi_2\, \alpha_s'(2\Delta t) \quad (23)$$

where ξ_j are stochastic variables that take on values of ± 1 with equal probability and correspond to the system being perturbed (i.e., $\xi_j = -1$) by a 2π pulse (i.e., $Q = 1 - |s\rangle\langle s|$) at time $t_j = j\Delta t$, or not (i.e., $\xi_j = 1$).[46] The expansion coefficients for the continuum states are obtained as follows:

$$\beta_k(l\Delta t) = -iV_{ks}\left(\frac{e^{i(\omega_k - \omega_s)\Delta t} - 1}{i(\omega_k - \omega_s)}\right)\left(1 + \sum_{j=1}^{l-1} \xi_j e^{i(\omega_k - \omega_s)j\Delta t}\right) \quad (24)$$

and the time evolution of the initially populated state $|s\rangle$ is

$$\alpha_s(2n\Delta t) = \prod_{j=1}^{2n} \xi_j \underbrace{\left(1 - 2n\Delta t \int_0^{\Delta t} (1 - \frac{t'}{\Delta t})\mathrm{K}(t')\, dt'\right)}_{G} \underbrace{- \sum_k \sum_{l=1}^{2n-1} \prod_{j=l}^{2n} \xi_j \left(e^{-i\Omega_{ks}[l+1]\Delta t} - e^{-i\Omega_{ks}l\Delta t}\right) \frac{|V_{ks}|^2}{(\Omega_{ks})^2} (e^{i\Omega_{ks}\Delta t} - 1)\left(1 + \sum_{i=1}^{l-1} \xi_i e^{i\Omega_{ks}i\Delta t}\right)}_{F} \quad (25)$$

Note that in the limit when $\xi_j = 1$ (i.e., pulses with 0% efficiency), eq 25 yields

$$|\alpha(2n\Delta t)|^2 = 1 - \sum_k \frac{|V_{ks}|^2}{\left(\frac{\Omega_{sk}}{2}\right)^2} \sin^2\left(\Omega_{sk}\frac{2n\Delta t}{2}\right) \quad (26)$$

which is the expression for spontaneous decay, introduced by eq 9.[34] More generally, the survival probability of the system evolving under the effect of pulses with $\xi_j \neq 1$ is

$$|\alpha(2n\Delta t)|^2 = |G|^2 - 2Re(F^*G) + |F|^2$$

$$|G|^2 = 1 - 2Re\left(2n\Delta t \int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)K(t')\, dt'\right)$$

$$F^*G = \prod_{j=1}^{2n}\xi_j \sum_k \sum_{l=1}^{2n-1}\prod_{a=l}^{2n}\xi_a\left(\frac{e^{-i\Omega_{ks}[l+1]\Delta t} - e^{-i\Omega_{ks}l\Delta t}}{i\Omega_{sk}}\right)$$
$$\times\frac{|V_{ks}|^2}{(\Omega_{ks})^2}(e^{i\Omega_{ks}\Delta t} - 1)\left(1 + \sum_{b=1}^{l-1}\xi_b e^{i\Omega_{ks}b\Delta t}\right) \tag{27}$$

where $|F|^2$ is neglected, since it involves terms of $O(|V_{ks}|^4)$. Equation 27 shows that coherent control can be achieved with stochastic sequences of phase-kick pulses. Note that the population decay is suppressed (i.e., $|\alpha(2n\Delta t)|^2 \to 1$) when $\Delta t \to 0$. In addition, decay can be accelerated relative to the spontaneous behavior described by eq 9 for larger values of Δt.

To analyze the effect of averaging over all possible stochastic sequences, we consider independent random variables with $\langle\xi_j\rangle = 0$, for which,

$$\langle\xi_1\xi_2...\xi_n\rangle = \langle\xi_1\rangle\langle\xi_2\rangle\langle\xi_3\rangle...\langle\xi_n\rangle = 0 \tag{28}$$

Therefore, $\langle F^*G\rangle = 0$, and the average short-time population decay at $t = 2n\Delta t$ is

$$\langle|\alpha(2n\Delta t)|^2\rangle = |G|^2$$
$$|G|^2 = 1 - 2n\Delta t\left\{2Re\left(\int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)K(t')\, dt'\right)\right\}$$
$$= 1 - \gamma_{avg}2n\Delta t$$
$$\gamma_{avg} = 2Re\left(\int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)K(t')\, dt'\right)$$
$$= \Delta t\sum_k |V_{ks}|^2 \frac{\sin^2\left(\Omega_{ks}\frac{\Delta t}{2}\right)}{\left(\Omega_{ks}\frac{\Delta t}{2}\right)^2} \tag{29}$$

Interestingly, γ_{avg} is exactly the decay rate derived by Kofman and Kurizki in the context of QZE,[34−36] in which, contrary to unitary phase-kick pulses, the pulses *collapse* the coherent evolution, as due to a measurement, by projecting the time-evolved state into a state (e.g., $|s\rangle$). For comparison, Section V derives the QZE and AZE dynamics by using the perturbational treatment implemented in this section in conjunction with pulses that collapse the coherent evolution into state $|s\rangle$. The observed correspondence in the decay rates suggests that the dynamical effect of repetitive collapses is equivalent to the destructive interference induced by stochastic sequences of unitary pulses when averaged over all possible realizations.

V. QUANTUM ZENO AND ANTI-ZENO EFFECT

QZE and AZE occur when the coherent evolution is repetitively interrupted by collapsing the system onto state $|s\rangle$, as in a measurement process described by $\hat{P} = |s\rangle\langle s|$.[32] Sufficiently frequent pulses freeze decay dynamics (zeno effect),[31] whereas sequences with longer time intervals between pulses accelerate the decay (anti-zeno effect).[34] In their landmark work on the topic, Kofman and Kurizki elucidated the mechanism via which both of these effects set in, hinting at the relation between the density of states of the continuum and the time interval between measurements. We refer the reader to the original work of Kofman and Kurizki[34] for the relevant details of the processes. In this section, we compare the resulting dynamics to coherent control schemes, described in Sections III and IV.

We consider the Hamiltonian, introduced by eq 1, with $\hat{U}$ denoting the short-time evolution as described by eqs 7 and 8. $\hat{P} = |s\rangle\langle s|$. represents the measurement process, collapsing the system onto state $|s\rangle$ at time Δt, yielding a state with

$$\alpha_s(\Delta t) = \langle s|\hat{P}\hat{U}|\psi\rangle \tag{30}$$

and devoid of any population in states $|k\rangle$ (i.e., $\beta_k(0) = 0$). Now, if the time evolution proceeds in sufficiently small time steps of order Δt, then the population of states $|s\rangle$ will remain negligible for later times. Using eq 7 for computing the survival probability in state $|s\rangle$, we obtain

$$|\alpha_s(\Delta t)|^2 = |\alpha_s(0)|^2\left(1 - 2Re\left\{\sum_k |V_{ks}|^2\int_0^{\Delta t}(\Delta t - t')e^{i\Omega_{sk}t'}\, dt'\right\}\right) \tag{31}$$

Repeating the evolution and measurement steps, $2n$ times, we obtain the survival probability at time $t = 2n\Delta t$.

$$\left|\alpha_s(2n\Delta t)\right|^2 = \left|\alpha_s([2n-1]\Delta t)\right|^2\left(1 - 2Re\left\{\sum_k |V_{ks}|^2 \int_0^{\Delta t}(\Delta t - t')e^{i\Omega_{sk}t'}\, dt'\right\}\right) \tag{32}$$

Substituting eq 31 into eq 32 recursively, we obtain the survival probability at $t = 2n\Delta t$,

$$\left|\alpha_s(2n\Delta t)\right|^2 = \left|\alpha_s(0)\right|^2\left(1 - 2Re\left\{\sum_k |V_{ks}|^2 \int_0^{\Delta t}(\Delta t - t')e^{i\Omega_{sk}t'}\, dt'\right\}\right)^{2n}$$
$$\approx \left|\alpha_s(0)\right|^2\left(1 - 2Re\left\{\sum_k |V_{ks}|^2 2n\Delta t \int_0^{\Delta t}\left(1 - \frac{t'}{\Delta t}\right)e^{i\Omega_{sk}t'}\, dt'\right\}\right) \tag{33}$$

dx.doi.org/10.1021/jp108331x | *J. Phys. Chem. B* XXXX, XXX, 000–000

where we have neglected terms of $O(|V_{ks}|^3)$ and higher, as appropriate in the weak coupling limit. After the final integration, the above expression takes the form

$$|\alpha_s(2n\Delta t)|^2 = |\alpha_s(0)|^2 \left(1 - 2Re\left\{\sum_k |V_{ks}|^2 \int_0^{\Delta t}(\Delta t - t')e^{i\Omega_{sk}t'}dt'\right\}\right)^{2n}$$

$$\approx |\alpha_s(0)|^2 \left(1 - 2n\Delta t \underbrace{\sum_k |V_{ks}|^2 \Delta t \frac{\sin^2\left(\Omega_{ks}\frac{\Delta t}{2}\right)}{\left(\Omega_{ks}\frac{\Delta t}{2}\right)^2}}_{\gamma_{ZENO}}\right), \tag{34}$$

where the rate $\gamma_{\rm ZENO}$ is identical to term A in the expression of the survival probability for the system under the pulsed coherent evolution (see eq 29). Such a term A, therefore, leads to the effective emergence of QZE and AZE when terms B and C cancel.

VI. CONCLUSIONS

In this paper, we have shown that quantum tunneling can be suppressed or accelerated by using deterministic, or stochastic, sequences of unitary pulses that affect the underlying interference phenomena responsible for quantum dynamics, without inducing decoherence or collapsing the coherent evolution of the system. A rigorous theoretical analysis based on perturbation theory to first order in the control pulse fields showed that sufficiently frequent perturbation pulses suppress quantum tunneling whereas trains of pulses separated by finite time intervals accelerate tunneling relative to spontaneous decay. The reported expressions also provided understanding of the role of randomization and the emergence of dynamics analogous to the evolution due to QZE or AZE, generated by stochastic sequences of unitary pulses when averaged over all possible realizations. The comparison to DD protocols and to control schemes based on pulses that collapse the coherent evolution reveals a subtle interplay between coherent and incoherent phenomena when stochastic sequences of unitary pulses are averaged over all possible realizations. We emphasize, however, that the resulting coherent control induced by sequences of unitary pulses is due to *interference* effects, with destructive interference averaged over stochastic sequences yielding dynamics analogous to the behavior of the system in the presence of repetitive collapsing pulses.

Our theoretical procedure showed how to analyze coherent control techniques on the basis of sequences of unitary pulses, QZE, AZE, and DD techniques on an equal mathematical footing. The calculations essentially unify the treatments due to Kofman and Kurizki[34] and Agarwal et al.[23,24,29,44] and in the process go beyond their treatments to reveal the inherent intricacies of dynamics, showing that the decay pattern for deterministic decoupling is, in essence, universal rather than restricted to a particular system (e.g., a system of spin 1/2 qubits).[27] This assertion is supported by the analysis of a common system, tunneling to a continuum, as affected by the various control techniques. Our theoretical analysis has shown that common terms affect the evolution as modulated by both coherent and incoherent control schemes, with the manifestation of QZE emerging from averaging out some of the contributing terms. The emergence of such behavior upon random pulsing is due to the stochastic phase that washes out the coherent interference effects and brings forward the otherwise suppressed incoherent effects.

Considering the simplicity of sequences based on phase-kick pulses, the similarity to pulsed NMR techniques, and the fact that other pulse sequences have already been demonstrated to achieve control in condensed material systems, we anticipate that the control techniques analyzed in this paper should raise significant experimental interest.

APPENDIX A

This section derives the short-time approximation, introduced by eq 7. For a sufficiently short time-interval,$(t - t_b)$, we assume $\alpha_s(t') \approx \alpha_s(t_b)$ in eq 6,

$$\begin{aligned}\dot{\alpha}_s(t) \approx & -\alpha_s(t_b)\int_{t_b}^{t}\sum_k |V_{ks}|^2 e^{i\Omega_{sk}(t-t')}\,dt' - i\sum_k V_{sk}e^{i\Omega_{sk}t}\,\beta_k(t_b)\\ &= \alpha_s(t_b)\sum_k |V_{ks}|^2\frac{1-e^{i\Omega_{sk}(t-t_b)}}{i\Omega_{sk}}\\ &- i\sum_k V_{sk}e^{i\Omega_{sk}t}\,\beta_k(t_b)\end{aligned} \tag{A1}$$

where we use the definition $\Omega_{sk} = \omega_s - \omega_k$. Now, integrating eq A1A1 by parts, we obtain

$$\begin{aligned}\alpha_s(t) - \alpha_s(t_b) &= \alpha_s(t_b)\sum_k |V_{ks}|^2 t'\frac{1-e^{i\Omega_{sk}(t\prime - t_b)}}{i\Omega_{sk}}\Bigg|_{t_b}^{t}\\ &+ \int_{tb}^{t} t' e^{i\Omega_{sk}(t'-t_b)}\,dt' - i\sum_k V_{sk}\int_{t_b}^{t} dt' e^{i\Omega_{sk}t'}\beta_k(t_b)\\ &= -\alpha_s(t_b)\sum_k |V_{ks}|^2 t\int_{t_b}^{t} e^{i\Omega_{sk}(t-t_b)}\,dt'\\ &+ \int_{t_b}^{t} t' e^{i\Omega_{sk}(t'-t_b)}\,dt' - i\sum_k V_{sk}\int_{t_b}^{t} dt'\, e^{i\Omega_{sk}t'}\beta_k(t_b)\\ &= -\alpha_s(t_b)\sum_k |V_{ks}|^2\int_{t_b}^{t}(t-t')e^{i\Omega_{sk}(t'-t_b)}\,dt'\\ &- i\sum_k V_{sk}\int_{t_b}^{t} dt' e^{i\Omega_{sk}t'}\beta_k(t_b)\end{aligned} \tag{A2}$$

APPENDIX B

Using eq 5 and the scheme defined in eq 11, the evolution of the continuum states in steps Δt is obtained as follows:

$$\begin{aligned}\beta_k(\Delta t) &= \beta_k(0) - iV_{ks}\left(\frac{e^{i\Omega_{ks}\Delta t}-1}{i(\Omega_{ks})}\right)\alpha_s(0)\\ \beta_k(2\Delta t) &= \beta_k(0) - iV_{ks}\left(\frac{e^{i\Omega_{ks}\Delta t}-1}{i(\Omega_{ks})}\right)\alpha_s(0)\\ &- iV_{ks}\left(\frac{e^{i\Omega_{ks}2\Delta t}-e^{i(\Omega_{ks})\Delta t}}{i\Omega_{ks}}\right)\alpha_s'(\Delta t)\\ \beta_k(2n\Delta t) &= \beta_k(0) - iV_{ks}\left(\frac{e^{i(\Omega_{ks})\Delta t}-1}{i\Omega_{ks}}\right)\alpha_s(0)\\ &- iV_{ks}\left(\frac{e^{i\Omega_{ks}2\Delta t}-e^{i(\Omega_{ks})\Delta t}}{i\Omega_{ks}}\right)\alpha_s'(\Delta t)\\ &- iV_{ks}\left(\frac{e^{i\Omega_{ks}3\Delta t}-e^{i(\Omega_{ks})2\Delta t}}{i\Omega_{ks}}\right)\alpha_s(2\Delta t)\\ &+ iV_{ks}\left(\frac{e^{i\Omega_{ks}4\Delta t}-e^{i\Omega_{ks}3\Delta t}}{i\Omega_{ks}}\right)\alpha_s'(3\Delta t)\\ &\ldots iV_{ks}\left(\frac{e^{i\Omega_{ks}(2n-1)\Delta t}-e^{i\Omega_{ks}(2n-2)\Delta t}}{i\Omega_{ks}}\right)\alpha_s([2n-2]\Delta t)\end{aligned} \tag{B1}$$

where $\alpha_s''(l\Delta t') = -\alpha_s(l\Delta t)$ accounts for the phase flip due to the action of a 2π pulse. To obtain an expression of $\beta_k(2n\Delta t)$ of $O(|V_{ks}|^2)$ we keep only the zero-th order term in the expansions of $\alpha_s(l\Delta t)$ in powers of V_{ks} and we obtain the compact expressions for the continuum state amplitudes, as follows:

$$\beta_k(\Delta t) = \beta_k(0) - iV_{ks}\left(\frac{e^{i\Omega_{ks}\Delta t} - 1}{i\Omega_{ks}}\right)\alpha_s(0)$$

$$\beta_k(2\Delta t) = \beta_k(0) - iV_{ks}\left(\frac{e^{i\Omega_{ks}\Delta t} - 1}{i\Omega_{ks}}\right)\alpha_s(0) - iV_{ks}\left(\frac{e^{i\Omega_{ks}2\Delta t} - e^{i\Omega_{ks}\Delta t}}{i\Omega_{ks}}\right)\alpha_s'(0)$$

$$\beta_k([2n-1]\Delta t) = \beta_k(0) - iV_{ks}\left(\frac{e^{i\Omega_{ks}\Delta t} - 1}{i\Omega_{ks}}\right)\alpha_s(0) - iV_{ks}\left(\frac{e^{i\Omega_{k}2\Delta t} - e^{i\Omega_{ks}\Delta t}}{i\Omega_{ks}}\right)\alpha_s'(0) - iV_{ks}\left(\frac{e^{i\Omega_{ks}3\Delta t} - e^{i\Omega_{ks}2\Delta t}}{i\Omega_{ks}}\right)\alpha_s(0) + iV_{ks}\left(\frac{e^{i\Omega_{ks}4\Delta t} - e^{i\Omega_{ks}3\Delta t}}{i\Omega_{ks}}\right)\alpha_s'(0) \ldots iV_{ks}\left(\frac{e^{i\Omega_{ks}(2n-1)\Delta t} - e^{i\Omega_{ks}(2n-2)\Delta t}}{i\Omega_{ks}}\right)\alpha_s(0)$$

$$\beta_k(l\Delta t) = \frac{V_{ks}}{\Omega_{ks}}\sum_{j=1}^{l}(-1)^j(e^{i*j*\Omega_{ks}*\Delta t} - e^{i*(j-1)*\Omega_{ks}*\Delta t}) = \frac{V_{ks}}{\Omega_{ks}}\frac{(e^{i\Omega_{ks}\Delta t} - 1)((-1)^l e^{i\Omega_{ks}l\Delta t} - 1)}{e^{i\Omega_{ks}\Delta t} + 1} \quad (\because \beta_k(0) = 0) \tag{B2}$$

Note that the continuum-state amplitude at any particular time step accounts for all the continuum state amplitudes at prior time steps. Moreover, contributions from even and odd time steps occur with alternating signs. This is a direct consequence of the phase flip of the system state as a result of the pulsing, which affects the above evolution equations in the form of $\alpha_s(0)$. An interesting analogy emerges if one interprets the sign change as a time reversal of continuum dynamics under successive pulse applications.[45] In the context of NMR, this amounts to spin echoes[29] initiated with the purpose of negating the continuum-induced decoherence in spin−spin correlations. In the event of no pulses, the above expression becomes a telescoping sum, which eventually leads to the spontaneous decay behavior.

APPENDIX C

This section compares the coherent control scenario based on random variables $\xi_n = \pm 1$, introduced in Section IV, to the dynamical decoupling scheme based on random variables, $\chi_n = \{(-1)^n, n \in N\}$, considered by Santos and Viola for manipulating coherence in spin 1/2 qubits.[29] We modify the scheme defined in eq 23 as follows:

$$\beta_k(\Delta t) = \beta_k(0) - iV_{ks}\lambda_0\left(\frac{e^{i\Omega_{ks}\Delta t} - 1}{i\Omega_{ks}}\right)\alpha_s(0)$$

$$\alpha_s(\Delta t) = \alpha_s(0)\left(1 - \int_0^{\Delta t}(\Delta t - t')\,K(t')\,dt'\right) - i\sum_k V_{sk}\int_0^{\Delta t} e^{i\Omega_{sk}t}\lambda_0\,\beta_k(0)\,dt \beta_k(2\Delta t) = \beta_k(\Delta t) - iV_{ks}\lambda_1\left(\frac{e^{i\Omega_{ks}2\Delta t} - e^{i\Omega_{ks}\Delta t}}{i\Omega_{ks}}\right)\alpha_s(\Delta t)\alpha_s(2\Delta t) = \alpha_s(\Delta t)\left(1 - \int_{\Delta t}^{2\Delta t}(2\Delta t - t')\,K(t' - \Delta t)\,dt'\right) - i\sum_k V_{sk}\int_{\Delta t}^{2\Delta t} e^{i\Omega_{sk}t}\lambda_1\,\beta_k(\Delta t)\,dt \tag{C1}$$

where $K(t) = \sum_k |V_{ks}|^2\, e^{i\Omega_{sk}t}$ and $\lambda_j = (-1)^j$ for a deterministic pulsing scheme. If we collect the expressions from eq C1, we obtain

$$\begin{aligned}\alpha_s(2\Delta t) &= \left\{\alpha_s(0)\left(1 - \int_0^{\Delta t}(\Delta t - t')\,K(t')\,dt'\right) - i\sum_k\int_0^{\Delta t} V_{sk}e^{i\Omega_{sk}t'}\lambda_0\,\beta_k(0)\,dt'\right\} \\ &\quad\times\left\{\left(1 - \int_{\Delta t}^{2\Delta t}(2\Delta t - t')\,K(t' - \Delta t)\,dt'\right)\right\} - i\sum_k\int_{\Delta t}^{2\Delta t} V_{sk}e^{i\Omega_{sk}t'}\lambda_1\,\beta_k(\Delta t)\,dt' \\ &= 1 - \int_0^{\Delta t}(\Delta t - t')\,K(t')\,dt' - \int_{\Delta t}^{2\Delta t}(2\Delta t - t')\,K(t' - \Delta t)\,dt' - i\sum_k\int_0^{\Delta t} V_{sk}e^{i\Omega_{sk}t'}\lambda_0\,\beta_k(0)\,dt' - i\sum_k\int_{\Delta t}^{2\Delta t} V_{sk}e^{i\Omega_{sk}t'}\lambda_1\,\beta_k(\Delta t)\,dt' \\ &= 1 - 2\int_0^{\Delta t}(\Delta t - t')\,K(t')\,dt' - i\sum_k\int_0^{\Delta t} V_{sk}e^{i\Omega_{sk}t'}\lambda_0\,\beta_k(0)\,dt' - i\sum_k\int_{\Delta t}^{2\Delta t} V_{sk}e^{i\Omega_{sk}t'}\lambda_1\,\beta_k(\Delta t)\,dt'\end{aligned} \tag{C2}$$

and

$$\beta_k(l\Delta t) = -iV_{ks}\left(\frac{e^{i\Omega_{ks}\Delta t} - 1}{i\Omega_{ks}}\right)\left(\sum_{j=0}^{l-1}\lambda_j e^{i\Omega_{ks}j\Delta t}\right) \tag{C3}$$

It can be verified that using the above definition for continuum states and substituting it back into eq C1, one obtains exactly the expression derived in eq 24. We see from eq C3 that the continuum state amplitude is a combination of terms that alternate in sign as in eq B2B2 of Appendix B. Going back to our analogy of pulse applications and spin echoes (see

Appendix B), the application of periodic 2π pulses is equivalent to the initiation of successive π phase shifts in the continuum state amplitude. This is accomplished in this case by allowing the variables to be $\lambda_j = (-1)^j$. Using these definitions, the expression for the survival amplitude becomes

$$\alpha(2n\Delta t) = \left(1 - 2n\Delta t \int_0^{\Delta t} \left(1 - \frac{t'}{\Delta t}\right) K(t')\, dt'\right) - i \sum_k \sum_{l=1}^{2n-1} \int_{l\Delta t}^{\{l+1\}\Delta t} V_{sk} e^{i\Omega_{sk} t'} \lambda_l \beta_k(l\Delta t)\, dt' \quad (C4)$$

Consequently, the resulting survival amplitude is

$$|\alpha(2n\Delta t)|^2 = |G|^2 + 2Re(F^*G) + |F|^2$$

$$|G|^2 = 1 - 2Re\left(2n\Delta t \int_0^{\Delta t} \left(1 - \frac{t'}{\Delta t}\right) K(t')\, dt'\right) = 1 - 2n\Delta t \sum_k \Delta t \frac{\sin^2 \Omega_{sk} \frac{\Delta t}{2}}{\left[\frac{\Omega_{sk}\Delta t}{2}\right]^2} |V_{ks}|^2$$

$$2Re(F^*G) = 2Re\left(\sum_k \sum_{l=1}^{2n-1} \sum_{m=0}^{l-1} \lambda_l \lambda_m \frac{|V_{ks}|^2}{(\Omega_{ks})^2} (e^{i\Omega_{ks}\Delta t} - 1)^2 e^{-i\Omega_{ks}(l-m+1)\Delta t}\right) = -2Re\left(\sum_k \sum_{l=1}^{2n-1} \sum_{m=0}^{l-1} \lambda_l \lambda_m \frac{|V_{ks}|^2}{(\Omega_{ks})^2} \left(2 \sin \Omega_{ks} \frac{\Delta t}{2}\right)^2 e^{-i\Omega_{ks}(l-m)\Delta t}\right)$$

$$= -2 \sum_k \sum_{l=1}^{2n-1} \sum_{m=0}^{l-1} \lambda_l \lambda_m \frac{|V_{ks}|^2}{(\Omega_{ks})^2} \left(2 \sin \Omega_{ks} \frac{\Delta t}{2}\right)^2 \cos[\Omega_{ks}(l-m)\Delta t] \quad (C5)$$

where $|F|^2$ has been neglected, since it is $O(|V_{ks}|^4)$. Assuming that $\lambda_j = (-1)^j$,

$$2Re(F^*G) = -2 \sum_k \sum_{l=1}^{2n-1} \sum_{m=0}^{l-1} (-1)^{l+m} \frac{|V_{ks}|^2}{(\Omega_{ks})^2} \left(2 \sin \Omega_{ks} \frac{\Delta t}{2}\right)^2 \cos(\Omega_{ks}(l-m)\Delta t)$$

$$= -\sum_k |V_{ks}|^2 \tan^2 \Omega_{ks} \frac{\Delta t}{2} \frac{\sin^2 \Omega_{ks} \frac{2n\Delta t}{2}}{\left(\frac{\Omega_{ks}}{2}\right)^2} + 2n \sum_k |V_{ks}|^2 \frac{\sin^2 \Omega_{ks} \frac{2n\Delta t}{2}}{\left(\frac{\Omega_{ks}}{2}\right)^2} \quad (C6)$$

Substituting eq C6 back into eq C5, one obtains exactly the result derived in eq 27, within the context of deterministic pulses.

When the variables are allowed to be stochastic, the expression for the survival probability, eq C5, resembles the one obtained for qubit coherence under similar conditions.[22]

APPENDIX D

Equation 18 gives

$$\alpha_s(2n\Delta t) = 1 - 2n\Delta t \int_0^{\Delta t} \left(1 - \frac{t'}{\Delta t}\right) K(t')\, dt' - \sum_k e^{i\Omega_{sk}\Delta t} \frac{|V_{ks}|^2}{{\Omega_{ks}}^2} \frac{(e^{i\Omega_{ks}\Delta t} - 1)^2}{e^{i\Omega_{ks}\Delta t} + 1} \sum_{j=1}^{2n-1} \{1 - (-1)^j e^{i\Omega_{skj}\Delta t}\} \quad (D1)$$

The summation over j is completed using $\sum_j^{2n-1} R^j = (R(1 - R^{2n-1}))/(1-R)$ with $R = -e^{i\Omega_{sk}\Delta t}$. This leads to

$$\alpha_s(2n\Delta t) = 1 - 2n\Delta t \int_0^{\Delta t} \left(1 - \frac{t'}{\Delta t}\right) K(t')\, dt' - \sum_k e^{i\Omega_{sk}\Delta t} \frac{|V_{ks}|^2}{{\Omega_{ks}}^2} \frac{(e^{i\Omega_{ks}\Delta t} - 1)^2}{e^{i\Omega_{ks}\Delta t} + 1} \left(2n - 1 + \frac{e^{i\Omega_{sk}2n\Delta t} + e^{i\Omega_{sk}\Delta t}}{e^{i\Omega_{sk}\Delta t} + 1}\right)$$

$$= 1 - 2n\Delta t \int_0^{\Delta t} \left(1 - \frac{t'}{\Delta t}\right) K(t')\, dt' - \sum_k e^{i\Omega_{sk}\Delta t} \frac{|V_{ks}|^2}{{\Omega_{ks}}^2} \frac{(e^{i\Omega_{ks}\Delta t} - 1)^2}{e^{i\Omega_{ks}\Delta t} + 1} \left(2n + \frac{e^{i\Omega_{sk}2n\Delta t} - 1}{e^{i\Omega_{sk}\Delta t} + 1}\right)$$

$$= 1 - 2n\Delta t \int_0^{\Delta t} \left(1 - \frac{t'}{\Delta t}\right) K(t')\, dt' - \sum_k e^{i\Omega_{sk}\Delta t} \frac{|V_{ks}|^2}{{\Omega_{ks}}^2} \frac{(e^{i\Omega_{ks}\Delta t} - 1)^2}{e^{i\Omega_{ks}\Delta t} + 1} 2n - \sum_k e^{i\Omega_{sk}\Delta t} \frac{|V_{ks}|^2}{{\Omega_{ks}}^2} \left(\frac{e^{i\Omega_{ks}\Delta t} - 1}{e^{i\Omega_{ks}\Delta t} + 1}\right)^2 (e^{i\Omega_{sk}2n\Delta t} - 1) \quad (D2)$$

Using the definition $k(t) = \Sigma_k |V_{ks}| e^{i\Omega_{sk}t}$ in eq D2, we retrieve eq .

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ACKNOWLEDGMENT

We are grateful to Prof. Shaul Mukamel for teaching us the beauty of quantum mechanics as applied to the description of light-matter interactions. We thank Prof. Lea F. Santos (Yeshiva University) for helpful comments on a preliminary version of this manuscript. V.S.B. acknowledges supercomputer time from NERSC and support from NSF grants CHE-0911520 and ECCS-1028066. Preliminary work on quantum dynamics for coherent control has been funded by the Division of Chemical Sciences, Geosciences, and Biosciences, Office of Basic Energy Sciences of the U.S., Department of Energy (DE-FG02-07ER15909).

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(46) A comparative analysis of the resulting stochastic sequence to a dynamical decoupling scheme based on random variables $x_n = \{(-1)^n, n \in N\}$, as previously considered by Santos and Viola for manipulating coherences in spin 1/2 qubits,[41,42] is discussed in Appendix C.

dx.doi.org/10.1021/jp108331x |*J. Phys. Chem. B* XXXX, XXX, 000–000