No-go theorem and optimization of dynamical decoupling against noise with soft cutoff

Zhen-Yu Wang and Ren-Bao Liu
Department of Physics and Center for Quantum Coherence, The Chinese University of Hong Kong, Shatin, N. T., Hong Kong, China

We study the performance of dynamical decoupling in suppressing decoherence caused by soft-cutoff Gaussian noise, using short-time expansion of the noise correlations and numerical optimization. For the noise with soft cutoff at high frequencies, there exists no dynamical decoupling scheme to eliminate the decoherence to arbitrary orders of the short time, regardless of the timing or pulse shaping of the control under the population conserving condition. We formulate the equations for optimizing pulse sequences that minimizes decoherence up to the highest possible order of the short time for the noise correlations with odd power terms in the short-time expansion. In particular, we show that the Carr-Purcell-Meiboom-Gill sequence is optimal in short-time limit for the noise correlations with a linear order term in the expansion.

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I. INTRODUCTION

Quantum information processing relies on the coherence of quantum systems. Unavoidable interactions between a quantum system and its environment (bath) introduce noise on the system and lead to errors (decoherence) of the quantum system. Various methods have been proposed to combat the decoherence, including decoherence-free subspaces, error-correction codes, and dynamical decoupling (DD). In particular, the DD scheme uses rapid unitary control pulses acting only on the systems to suppress the effects of the noise from the environments. DD has the advantages of suppressing decoherence without measurement, feedback, or redundant encoding. DD originated from the seminal spin echo experiment, in which the effect of a static random magnetic field (inhomogeneous broadening) is canceled. And more complex DD pulse sequences, such as the Carr-Purcell-Meiboom-Gill (CPMG) sequence, were designed to prolong the spin coherence time.

The early DD schemes only eliminate low-order errors, i.e., the errors of quantum evolutions up to some low order in the Magnus expansion. By unitary symmetrization procedure, DD cancels the first order (i.e., leading order) errors. To eliminate errors to the second order in short time, mirror-symmetric arrangement of two DD sequences can be used. The first explicit arbitrary $M$th order DD scheme, which suppresses errors to $O(T^{M+1})$ for short evolution time $T$, is the concatenated DD (CDD) proposed by Khodjasteh and Lidar. CDD sequences against pure dephasing were investigated for electron spin qubits in realistic solid-state systems with nuclear spins as baths. Experiments have tested the performance of CDD. CDD works for generic quantum systems coupled to a finite bath. However, since CDD uses recursively constructed pulse sequences to suppress decoherence, the number of pulses increases exponentially with the decoupling order. As pulse errors are inevitably introduced in each control pulse in experiments, finding efficient DD schemes with fewer control pulses is desirable. A remarkable advance is the Uhrig DD (UDD) scheme. UDD is optimal in the short-time limit in the sense that it suppresses the pure dephasing of a qubit coupled to a finite bath to the $M$th order using only $M$ qubit flips. The performance bounds for UDD against pure dephasing were established. Shaped pulses of finite amplitude can be incorporated into UDD. Many recent experimental studies demonstrated the performance of UDD.

It is important to find efficient schemes to suppress general decoherence (including pure dephasing and population relaxation). Yang and Liu extended UDD to the suppression of population relaxation. Inspired efficient ways to suppress the general decoherence of single qubits, including concatenation of UDD sequences (CUDD) and a much more efficient one called quadratic DD (QDD) discovered by West et al. Based on the proof in et al, Mukhtar et al generalized UDD to protect arbitrary multilevel systems with full prior knowledge of the initial states. One can actually preserve the coherence of arbitrary multi-qubit systems by protecting a mutually orthogonal operation set (MOOS). By nesting UDD sequences for protecting the elements in the MOOS, the nested UDD (NUDD) requires only a polynomially increasing number of pulses in the decoupling order. These universal DD schemes also work for analytically time-dependent baths.

The above-mentioned variations of UDD, however, rely on the finiteness of the baths, i.e., the existence of hard high-frequency cutoff in the noise spectra. Legitimate questions arise: For quantum systems coupled to an infinite quantum bath or affected by soft-cutoff noise, can any DD be designed to eliminate the decoherence to arbitrary orders of precision in the short-time limit? And if yes, how can such DD be designed? Such questions have been previously addressed in some specific noise models. Comparing the efficiency of various DD sequences in suppressing pure dephasing of a qubit due to classical noise, Cywiński et al observed that if the noise spectrum cutoff is not reached, CPMG sequences actually performs better than CDD and UDD sequences.

With the consideration of minimum pulse separations in phys-
ical systems, Viola et al observed that low-order DD sequences provide better performance than high-order DD when the rate of pulses is not faster than the correlation time of the noise \cite{48, 49}. It was confirmed by experiments that for $^{13}$C spin qubits in a $^1$H spin bath of which the high-frequency cutoff was not reached by the DD sequences, CPMG outperforms UDD \cite{50}. Also, Pasini and Uhrig derived the equations for minimizing decoherence for power-law spectra, and found that the numerically optimized sequences resemble CPMG \cite{51}. Chen and Liu proved that for telegraphlike noise the CPMG sequences are the most efficient scheme in protecting the qubit coherence in the short-time limit and the decoherence can be suppressed at most to the third order of short evolution time by DD \cite{52}. These results suggest that for noise with soft cutoff in the spectrum, there are certain constraints on the optimal order and decoupling scaling of DD. Ref. \cite{53} presented numerical optimization of bounded-strength DD for specific noise spectra. However, no conclusion has been drawn on the performance of DD with arbitrary timing and shaping for the general cases of soft-cutoff noise.

In this paper, we address the general question of the performance of DD against soft-cutoff noise based on the general modulation function induced by arbitrary DD with bounded-strength or pulsed control. We show that for the noise spectrum with a power-law asymptote at high frequencies, there exists no modulation function to eliminate the decoherence to an arbitrary order of the short time, regardless of the timing and shaping of the DD control under the population conserving condition. Although the decoherence can be suppressed to be arbitrarily small by DD with a sufficiently large number of pulses, the existence of the largest achievable decoupling order shows that DD against soft-cutoff noise does not have the order-by-order decoupling efficiency, which is possible for hard-cutoff noise. Since for soft-cutoff noise the decoherence cannot be eliminated at a certain order of short time, we derive a set of equations to minimize the leading-order term in the short-time expansion while eliminating the lower orders. These equations are numerically solved for optimal solutions. In particular, for noise correlations with a linear order term in time, we prove that the CPMG sequences are optimal. For other noise correlations with odd-order terms, the minimum pulse interval of the optimized sequences is larger than in UDD sequences. This feature is important in realistic experiments when there is a minimum pulse switching time \cite{49}.

This paper is organized as follows: In Sec. II we analyze the performance of DD against soft-cutoff noise, and we give the condition under which decoherence suppression to an arbitrary order of short-time scaling is impossible. The relation between the high-frequency cutoff and the short-time expansion of correlations is also discussed. In Sec. III we derive the equations for sequence optimization and obtain optimal DD for noise correlations with odd-power expansion terms. The conclusions are drawn in Sec. IV.

II. NO-GO THEOREM ON DYNAMICAL DECOUPLING AGAINST NOISE WITH SOFT CUTOFF

We consider the pure-dephasing Hamiltonian for a single spin (qubit)

$$H = \frac{1}{2} \sigma_z [\omega_a + \beta(t)],$$  

(1)

where $\sigma_z = |+\rangle \langle +| - |\rangle \langle -|$ is the Pauli operator of the qubit, $\omega_a$ is the energy splitting of the qubit, and $\beta(t)$ describes random noise with average $\beta(\bar{t}) = 0$. Here the over bar denotes averaging over the noise realizations. We assume that the statistics of the noise fluctuations are Gaussian.

After a duration of free evolution time $T$, the noise induces between the two states $|\pm\rangle$ a random phase shift $\int_0^T \beta(t) dt$ that destroys the quantum coherence. We can suppress the decoherence by DD control on the qubit. There is only one noise source $\beta(t)$ in the model Eq. (1), and to suppress the decoherence we need to protect a MOOS which consists of a Pauli operator $\sigma_\chi$ (more generally $\sigma_x \cos \phi + \sigma_y \sin \phi$ with $\phi$ being real) \cite{27}. We will prove later that DD can suppress the decoherence (i.e., the protection of the MOOS $|\sigma_\chi\rangle$) only to a certain order of short evolution time for noise correlations that have odd-power expansion terms in time. We expect that the proof also applies to other quantum systems (e.g., multi-qubit systems) when the noise correlations have odd-power terms, since in those systems there are more noise sources and more system operators (e.g., a MOOS consisting of $L > 1$ Pauli operators) should be protected.

When we apply a sequence of instantaneous unitary operations $\sigma_\chi$ at the moments $T_1, T_2, \ldots, T_N$, the controlled evolution operator reads

$$U(T) = (\sigma_\chi)^N U(T_{N+1}, T_N) \sigma_\chi U(T_N, T_{N-1}) \cdots$$

$$\times \sigma_\chi U(T_2, T_1) \sigma_\chi U(T_1, T_0),$$

(2)

where $T_0 = 0, T_{N+1} = T$, and the free evolution operator

$$U(T_{j+1}, T_j) = e^{-i \int_{T_j}^{T_{j+1}} [\omega_a + \beta(t)] dt}.$$

(3)

Note that when $N$ is odd, we may apply an additional $\sigma_\chi$ pulse at the end of the sequence for the identity evolution. Using

$$\sigma_x U(T_{j+1}, T_j) \sigma_\chi = e^{-i \int_{T_j}^{T_{j+1}} [\omega_a - \beta(t)] dt},$$

(4)

we write the evolution operator as

$$U(T) = e^{-i \int_{T_j}^{T_{j+1}} \omega_a F_x(t/T) dt} e^{-i \int_{T_j}^{T_{j+1}} \beta(t) F_z(t/T) dt},$$

(5)

where we have defined the modulation function for instantaneous $\pi$-pulse sequences \cite{28, 47}

$$F_x(t/T) = \begin{cases} (-1)^j & \text{for } t \in (T_j, T_{j+1}) \\ 0 & \text{for } t > T, \text{ or } t \leq 0 \end{cases}.$$

(6)

The DD control is parametrized by the relative pulse locations $T_j/T$. At the moment $T$, the off-diagonal density matrix element of an ensemble is

$$\rho_{11}(T) = \rho_{11}(0) e^{-i \int_0^T \omega_a F_x(t/T) T dt} e^{-i \int_0^T \beta(t) F_z(t/T) T dt}.$$

(7)
The coherence is characterized by the ensemble-averaged phase factor
\[
W(T) = e^{-i\chi(T)},
\]
where the phase correlation for instantaneous pulses
\[
\chi_s(T) = \frac{1}{2} \int_0^T dt_1 \int_0^T dt_2 \beta(t_1)\beta(t_2)F_{\pi\pi}(\frac{t_1}{T})F_{\pi\pi}(\frac{t_2}{T}).
\]
can be written as the overlap between the noise spectrum and a filter function determined by the Fourier transform of the modulation function \[47\].

For Gaussian noise, the ensemble-averaged phase factor \(W(T)\) is determined by the two-point correlation function \(\beta(t)\beta(t)\) and \(W(T)\) becomes \[54\] \[55\]
\[
W(T) = e^{-i\chi(T)},
\]
where
\[
\chi(T) = \frac{1}{2} \int_0^T dt_1 \int_0^T dt_2 \beta(t_1)\beta(t_2)F_{\pi\pi}(\frac{t_1}{T})F_{\pi\pi}(\frac{t_2}{T}).
\]

Under DD control, the qubit is flipped at different moments, and the random field \(\beta(t)\) is modulated by the modulation function \(F_\pi(t/T)\). For multilevel systems, the modulation functions resulting from instantaneous \(\pi\)-pulse sequences may have values not restricted to \([-1, 1]\) for \(t \in (0, T]\) (see Appendix \[4\]). In Ref. \[56\], it is shown that for DD composed of specially designed finite-duration pulses, the effective modulation functions can take values from \([-1, 1, 0]\) alternatively. We may also encounter effective modulation functions which are triangle wave functions during the time of system evolution \[57\]. For a more general analysis, we assume that the control conserves the populations and the phase modulation function \(F_\pi(t/T)\) has a general form as
\[
F\left(\frac{t}{T}\right) = \begin{cases} 
\text{a bounded function} & \text{for } t \in (0, T] \\
0 & \text{otherwise}
\end{cases},
\]
which has a finite number of discontinuities. The more general phase correlation considered in this paper reads
\[
\chi(T) = \frac{1}{2} \int_0^T dt_1 \int_0^T dt_2 \beta(t_1)\beta(t_2)F_{\pi\pi}(\frac{t_1}{T})F_{\pi\pi}(\frac{t_2}{T}).
\]

We assume the noise is stationary, i.e., of time translation symmetry, \(\beta(t_1)\beta(t_2) = \beta(t_1 - t_2)\beta(0)\). Another symmetry is
\[
\beta(t)\beta(0) = \beta(0)\beta(t). \text{ These symmetries indicate that the noise correlation is an even function of time, i.e.,}
\]
\[
\beta(t)\beta(0) = C_{\text{corr}}(t) = C_{\text{corr}}(-t).
\]

The noise correlation can be transformed from the noise power spectrum \(S(\omega)\) as
\[
\beta(t)\beta(0) = \int_{-\infty}^{\infty} d\omega S(\omega) e^{-i\omega t}.
\]

Note that both \(\beta(t)\beta(0)\) and \(S(\omega)\) are real even functions.

The general filter function is defined as the Fourier transform of the general modulation function,
\[
\tilde{f}(\omega) = \frac{1}{T} \int_{-\infty}^{\infty} F\left(\frac{t}{T}\right) e^{i\omega t} dt,
\]

which has the power expansion
\[
\tilde{f}(\omega) = \sum_{m=0}^{\infty} \frac{(i\omega T)^m}{m!} \lambda_m,
\]

where \(s_j\) are the discontinuous points and \(|I_j| = \int_{s_j}^{s_{j+1}} |F(s)| ds\) is finite. Therefore we have
\[
|\tilde{f}(u)| \leq a_j/u,
\]

where the coefficient \(a_j = \sum_j |F(s_j)| + |F(s_{j+1})| + I_j\) is bounded.

For a sequence of \(N\) instantaneous \(\pi\) pulses, \(\lambda_m\) reads
\[
\lambda_m = \sum_{j=0}^{N} (-1)^j \left( \frac{T_{j+1}}{T} \right)^{m+1} \left( 1 - \frac{T_{j+1}}{T} \right)^{m+1}.
\]

Using Eqs. \[14\] and \[15b\], Eq. \[12a\] can be written as the overlap of the noise spectrum and filter function
\[
\chi(T) = T\int_{-\infty}^{\infty} \frac{d\omega}{\pi} S(\omega)|\tilde{f}(\omega T)|^2.
\]

It should be stressed that in Eq. \[20\] the filter function \(\tilde{f}(\omega T)\) is general and not limited to the case of instantaneous pulse sequences.
A. Scaling of decoupling orders

We separate the noise spectrum into two parts by a frequency $\Omega$. As the noise spectrum $S(\omega)$ in Eq. (20) induces decoherence linearly,

$$\chi(T) = \chi_{[0,\Omega]}(T) + \chi_{[\Omega,\infty]}(T),$$  \hspace{1cm} (21)

where

$$\chi_{[0,\Omega]}(T) = T^2 \int_0^\Omega \frac{d\omega}{\pi} S(\omega)(\tilde{f}(\omega T))^2,$$  \hspace{1cm} (22a)

$$\chi_{[\Omega,\infty]}(T) = T^2 \int_\Omega^\infty \frac{d\omega}{\pi} S(\omega)(\tilde{f}(\omega T))^2.$$  \hspace{1cm} (22b)

$\chi_{[0,\Omega]}(T)$ and $\chi_{[\Omega,\infty]}(T)$ account for the effects of the low- and high-frequency noise, respectively. Both $\chi_{[0,\Omega]}(T)$ and $\chi_{[\Omega,\infty]}(T)$ cause decoherence as $S(\omega) \gtrless 0$.

1. Effects of low-frequency noise

For the spectrum $S(\omega) = O(1/\omega^p)$ with $P < 1$ when $\omega \to 0$, Eq. (14) gives the noise correlation of the noise with the frequencies $\omega < \Omega$.

$$\beta(t)\beta(0)_{[0,\Omega]} = \frac{1}{\Omega} \int_0^\Omega \frac{d\omega}{\pi} S(\omega) \cos(\omega t) = \sum_{m=0}^\infty C_{2m} \Omega^{2m+1} t^{2m},$$  \hspace{1cm} (23a)

Here the coefficients

$$C_{2m} = (-1)^m \frac{1}{\pi} \int_0^1 \frac{du}{u} S(u\Omega) \frac{u^{2m}}{(2m)!},$$  \hspace{1cm} (24)

which depend on the noise spectrum $S(\omega)$ and $\Omega$, converge at low frequencies $\omega \to 0$.

Eq. (12b) gives

$$\chi_{[0,\Omega]}(T) = \sum_{m=0}^\infty C_{2m} \phi_{2m} \Omega^{2m+1} T^{2m+2},$$  \hspace{1cm} (25)

where the decoherence functions

$$\phi_k = \Re \int_0^1 ds_1 \int_0^{s_1} ds_2 (s_1 - s_2)^k F^*(s_1) F(s_2),$$  \hspace{1cm} (26)

are modified by the modulation function of DD. The even-order decoherence functions $\phi_{2k}$ control the effects of the low-frequency noise.

Therefore if the modulation function $F(t/T)$ is designed to make

$$\phi_{2m} = 0,$$  \hspace{1cm} (27)

the decoherence from low-frequency noise is eliminated to $\chi_{[0,\Omega]}(T) = O(T^{2M+2})$ (the prefactor of the scaling depends on the noise spectrum and $\Omega$). Note that $e^{-\int_0^T \omega_0 F(t/T) dt} = 1$ in Eq. (7) when $\phi_0 = 0$.

In Appendix B, we simplify the even-order $\phi_{2m}$ as

$$\phi_{2m} = \frac{(2m)!}{2} \sum_{r=0}^m (-1)^r \frac{\lambda_{2m-2r}}{r! (2m - r)!}.$$  \hspace{1cm} (28)

From Eq. (28), we find that the following two sets of equations are equivalent

$$\phi_{2m} = 0 \Rightarrow \chi_{[\Omega,\infty]}(T) = \frac{2 M - 1}{2 M + 1} \chi_{[\Omega,\infty]}(T) \equiv O(T^{2M+2}).$$  \hspace{1cm} (29)

For instantaneous $\pi$-pulse sequences, the optimal solution of the equation set $\chi_{[\Omega,\infty]}(T) = \chi_{[\Omega,\infty]}(T)$ is

$$T_{\pi}^{\text{UDD}} = T \sin^2 \left( \frac{\pi j}{2N + 2} \right),$$  \hspace{1cm} (30)

which is the timing of UDD sequences [28]. The conditions Eqs. (29) and (16b) are more general than the one that leads to UDD and may lead to more general designs of optimal DD.

For the power-law spectrum $S(\omega) \approx a/\omega^p$ with $P \geq 1$ at low frequencies, $\chi_{[0,\Omega]}(T) = T^2 \int_0^\Omega \frac{d\omega}{\pi} S(\omega)(\tilde{f}(\omega T))^2$ in general diverges. For the modulation function $F_{\text{opt}}(t/T)$, it was shown that the divergence of the integral can be eliminated by high-order DD sequences [51]. For the general modulation function $F(t/T)$ under the conditions $\chi_{[\Omega,\infty]}(T) \approx O(\Omega^{2M+1})$, we have

$$\frac{df(u)}{dt} \approx \frac{\pi j}{2N + 2},$$  \hspace{1cm} (32)

where $\Omega_\infty \gg 1/T$ and $\Omega \leq 1/T$. We assume that the decay of the noise at the frequencies $\omega \leq \Omega_\infty$ is not slower than $a/\omega^p$ and the contribution is negligible. We set $\Omega_\infty = \infty$. The high-frequency contribution Eq. (22b) reads

$$\chi_{[\Omega,\infty]}(T) \approx \chi_{[\Omega,\infty]}(T) \equiv T^{\beta + 1} \int_\Omega^\infty \frac{du}{\pi} a u^p [f(u)]^2.$$  \hspace{1cm} (32)

Since $f(u) = O(1/u)$ when $u \to \infty$ [Eq. (18)], $\chi_{[\Omega,\infty]}(T)$ converges when $P > -1$.

Using $[f(0)]^2 = |\alpha|^2$, we get $\lim_{T \to 0} \chi_{[\Omega,\infty]}(T) = 0$ even though the integral $\int_0^\Omega \frac{du}{\pi} a u^p [f(u)]^2$ may diverge when $T \to 0$. We have shown in Sec. I A 1 that under the conditions $\chi_{[\Omega,\infty]}(T) \approx O(T^{2M+2})$.

The conditions Eqs. (29) and (16b) are more general than the one that leads to UDD and may lead to more general designs of optimal DD.
Therefore when $M > (P - 1)/2$, in Eq. (32) we extend the bounds of integration to $(0, \infty)$,

$$\chi_{\Omega, \omega}(T) = C_\omega^{\text{soft}}T^{P+1} - O(T^{2M+1-P}) = O(T^{P+1}), \quad (33)$$

where $C_\omega^{\text{soft}} = \int_{-\infty}^{\infty} \frac{da}{\pi} |f(a)|^2$ is bounded when $\lambda_m = 0 = M-1$ with $2M + 1 + P > 0$ and $P > -1$. The scaling $\chi_{\Omega, \omega}(T) = O(T^{P+1})$ is the largest order of decoupling that can be achieved for the noise with soft cutoff. We have the following theorem.

**Theorem 1.** For the noise spectrum with a power-law asymptote $\alpha/\omega^P$ with $P > -1$ at high frequencies, the largest achievable decoupling order of DD with a general modulation function given by Eq. (11) is $O(T^{P+1})$ in short-time limit $T \to 0$.

This theorem holds for arbitrary non-zero modulation function $F(t/T)$, and it shows that one cannot suppress the decoherence to arbitrary order when the noise has a soft cutoff in the spectrum.

For example, the $1/f$ noise and the Lorentz spectrum $\alpha/(\Omega^2 + \omega^2)$ correspond to the cases of $P = 1$ and $P = 2$, respectively. After eliminating the effect of low-frequency noise by high-order DD which satisfy $\lambda_0 = 0$, we achieve the largest decoupling order $\chi_{\Omega, \omega}(T) = O(T^2)$ and $\chi_{\Omega, \omega, \omega}(T) = O(T^3)$.

Note that Theorem 1 applies to the order of short-time scaling and it does not mean that DD can not protect the coherence to arbitrarily high precision. The decoherence can be suppressed further by reducing the prefactor $C_\omega^{\text{soft}}$, i.e., the the overlap of the filter function $|\tilde{f}(\alpha, \omega/T)|^2$ and noise spectrum $S(\omega)$. We will discuss the optimization of pulse sequences based by minimizing the overlap in Sec. III.

The result in Ref. [32] that DD can suppress decoherence at most to the third order of short evolution time for telegraphlike noise is general for noise with arbitrary statistics. In deriving Theorem 1, we have made the assumption that the statistics of the noise are Gaussian and the decoherence is determined by the two-point noise correlation. It would be interesting to generalize the theorem to non-Gaussian noise.

The perturbative regime $T \lesssim 1/\Omega$ is limited by the technology of experiments. For power-law noise (e.g., $1/f$ noise), $\Omega \to 0$ and the conclusion applies to arbitrary duration of $T$ when $M > (P - 1)/2$.

**B. Noise correlation expansion and high-frequency cutoff**

The correlation in the short-time limit, which is due to high-frequency noise, can be written as

$$C_{\Omega, P}(t) = \Re \int_{\Omega}^{\infty} \frac{d\omega}{\pi} \frac{\alpha}{\omega^P} e^{-i\omega t} dz.$$  

(34)

As $C_{\Omega, P}(t)$ is an even function, we just calculate the integral for the case of $t > 0$. For $P > 1$ and $t > 0$, we have

$$C_{\Omega, P}(t) = \Re \left[ \int_{c_\Omega} + \int_{c_1} + \int_{c_\infty} \right] \frac{\alpha}{\omega^P} e^{\omega t} dz,$$

where the paths $c_\Omega$, $c_1$, and $c_\infty$ are shown in Fig. 1.

Since the maximum of $1/\omega^P \to 0$ as $|z| \to \infty$ in the upper half-plane, the contribution $\int_{c_\infty} \frac{\alpha}{\omega^P} e^{\omega t} dz = 0$. The contribution

$$\frac{\Re}{\pi} \int_{c_1} \frac{\alpha}{\omega^P} e^{\omega t} dz = \frac{\Re}{\pi} \int_{\Omega}^{\infty} \frac{\alpha}{\Omega^P} e^{\Omega t} e^{i\Omega t} d\theta$$

vanishes for even $P$. And

$$\frac{\Re}{\pi} \int_{c_{1,\infty}} \frac{\alpha}{\omega^P} e^{\omega t} dz = \frac{\Re}{\pi} \int_{\Omega}^{\infty} \frac{\alpha}{\Omega^P} e^{\Omega t} e^{i\Omega t} d\theta = f_{c_1}^{(1)} + f_{c_1}^{(2)},$$

(36)

where

$$f_{c_1}^{(1)} = \frac{1}{\pi} \Re \left[ \sum_{r=0}^{\infty} \frac{\alpha}{\Omega^r} \left( \frac{\alpha}{\Omega^r} \right)^P (P-1)! \right],$$

(37a)

and

$$f_{c_1}^{(2)} = \frac{1}{\pi} \Re \left[ \sum_{r=0}^{\infty} \frac{\alpha}{\Omega^r} \left( \frac{\alpha}{\Omega^r} \right)^P (P-1)! \right].$$

(37b)

For even $P$, $f_{c_1}^{(1)}$ is an expansion of even powers of $t$

$$f_{c_1}^{(1)} = \sum_{k=0}^{\infty} C_{2k} t^{2k}, C_{2k} = \frac{\alpha^{(1)^k+1}}{\pi(2k)!} (P+2k) \neq 0,$$

(38)

and there is only one odd-order expansion term, which is

$$f_{c_1}^{(2)} = \frac{1}{2} \frac{\alpha^{(1)^k}}{P-1}.$$

(39)

The existence of an odd-order term means that the correlation function is non-analytical, which indicates that the noise source cannot be a finite quantum bath. The noise with non-analytical correlation functions must come from the fluctuations of an infinite bath, since otherwise the unitary evolution of a finite quantum system will always lead to analytical correlation functions. For example, the noise correlation $\beta(t)\beta(0) = e^{-|t|/\eta}$ has odd-order terms in the time expansion, and the noise has a Lorentz spectrum, which has a power-law decrease at high frequencies. This kind of noise can be caused by Markovian (or instantaneous) collisions in the bath [61].

As an example, we consider the following noise spectrum

$$S^2_{2k}(\omega) \equiv \frac{\alpha}{\Omega^2k + \omega^{2k}},$$

(40)

for $K \in \{1, 2, \ldots\}$. $S^2_{2k}(\omega) \approx \alpha/\omega^{2k}$ when $\omega \gg \Omega_c$. For example, the measured ambient noise for ions in a Penning trap...
has an approximate $1/\omega^3$ spectrum at high frequencies and a flat dependence at low frequencies [35], which approximately corresponds to the noise spectrum Eq. (40) with $K = 2$. The corresponding correlation function of Eq. (40) is obtained by the inverse transform

$$\bar{\beta}(t)\bar{\beta}(0)_{2K} = \int_0^\infty \frac{d\omega}{\pi} \frac{\alpha e^{-i\omega t}}{\Omega^2 + \omega^{2K}}.$$  \hspace{1cm} (41)

Using the residue theorem, we have

$$\bar{\beta}(t)\bar{\beta}(0)_{2K} = \frac{i\alpha}{2K} \sum_{m=0}^K \frac{1}{\Omega - \omega^{2K}} = \frac{i\alpha}{2K} \sum_{m=0}^K \exp[-i\frac{\pi}{2}(2m+1)|\Omega|] \omega^{2K} - \frac{[(-1)^m + 1]}{\omega^{m+1}|\Omega|}.$$  \hspace{1cm} (42)

Expanding the right-hand side in powers of $t$, we get

$$\bar{\beta}(t)\bar{\beta}(0)_{2K} = \sum_{m=0}^\infty \frac{\exp[-i\frac{\pi}{2}(m+1)|\Omega|]}{m!} \frac{1}{\omega^{m+1}|\Omega|}.$$  \hspace{1cm} (43)

In Eq. (43), the leading odd-order term of the time expansion is $\frac{i\alpha}{2K} \exp(-i\frac{\pi}{2}K|\Omega|)\omega^{2K-1}$, as predicted by Eq. (39).

For simplicity, let us consider the noise correlations that have the power expansion

$$\bar{\beta}(t)\bar{\beta}(0) \equiv C_{\text{corr}}(t) = \sum_{k=0} C_k |t|^k,$$  \hspace{1cm} (44)

where the expansion coefficients

$$C_k \equiv \frac{1}{k!} \frac{d^k C_{\text{corr}}(t)}{dt^k} \bigg|_{t=0}.$$  \hspace{1cm} (45)

are finite real numbers with

$$C_{2k-1} = 0, \text{ for } k < K,$$  \hspace{1cm} (46a)

$$C_{2k-1} \neq 0.$$  \hspace{1cm} (46b)

The leading odd-order term in the short-time expansion of $C_{\text{corr}}(t)$ is $C_{2k-1}|t|^{2K-1}$. An example is $C_{\text{corr}}(t) = e^{-|t|^3}$ with $C_1 = 0$ and $C_3 = -1$. We assume that the noise correlation decreases smoothly at long correlation times, that is,

$$\lim_{t \to \infty} \frac{d^k}{dt^k} C_{\text{corr}}(t) = 0, \text{ for } k = 0, 1, \ldots, \hspace{1cm} (47)$$

and

$$I_4(\omega) \equiv \int_0^\infty e^{-i\omega t} dt.$$  \hspace{1cm} (48)

vanishes at large $\omega$ for $L = 0, 1, \ldots$ [22]. For the noise correlations that decay in the correlation time smoothly without fast oscillation, $I_4(\omega)$ vanishes at large frequency $\omega$. For example, the noise correlation $e^{-|t|^3}$ has $I_4(\omega) = i(-1)^3/(\omega + i) \to 0$ when $\omega \to \infty$.

We consider the high frequency behavior of the noise spectrum, $S_{2K}(\omega) = \int_0^\infty C_{\text{corr}}(t)e^{i\omega t} dt$. Integration by parts $L \geq (2K + 1)$ times gives

$$S_{2K}(\omega) = 2\Re \sum_{m=0}^L \frac{(-1)^m}{(m+1)!} (\frac{I_4(\omega)}{\omega^{m+1}} + \frac{I_L(\omega)}{(-i\omega)^L}),$$  \hspace{1cm} (49)

where we have used Eqs. (45) and (47).

Using Eqs. (46) and (48), we obtain for large $\omega$,

$$S_{2K}(\omega) \approx 2 \frac{(2K - 1)!}{(i\omega)^2} C_{2K-1} + O\left(\frac{1}{\omega^{2K+1}}\right).$$  \hspace{1cm} (50)

which is a power-law decrease at high frequencies.

When the noise correlation expansion contains only even-order terms, from Eq. (49) we have the noise spectrum $S_{\text{even}}(\omega) = 2\Re I_4(\omega)/(i\omega)^2$ for an arbitrarily large $L$. From the assumption $\lim_{\omega \to \infty} I_4(\omega) = 0$, we have $\lim_{\omega \to \infty} S_{\text{even}}(\omega)(i\omega)^2 = 0$ for an arbitrarily large $L$ and therefore the noise spectrum has a hard high-frequency cutoff. One example is the correlation function $e^{-t^2}$, which has the noise spectrum of exponential form $\sim \exp(-\frac{\omega^2}{4})$, and obviously the UDD sequence can suppress the noise effect order by order. The large $\omega$ behavior of other correlation functions of the form $\exp(-\sum_{j=1}^{p} \alpha_j t^{2j})$ can be calculated by the saddle point integration method, which gives a result of an exponential decrease at high frequencies (i.e., hard cutoff). For example, when $\omega$ is very large, $\int_0^\infty e^{i\omega t} e^{-|t|^3} dt \approx \frac{1}{2} \frac{1}{\sqrt{\pi}} e^{-\frac{\omega^2}{4}}$, and $a(\omega) = 12(\frac{\omega}{4})^\frac{5}{2} e^{-2\pi/3}$.

III. SEQUENCE OPTIMIZATION IN SHORT-TIME LIMIT

In this section, we optimize DD for the noise correlations that have the power expansion given by Eq. (44), $\bar{\beta}(t)\bar{\beta}(0) \equiv \sum_{k=0} C_k |t|^k$.

The performance of DD in the short-time limit is directly derived from the time-domain expansion. The time-domain expansion of noise correlations has the advantage to avoid the divergence of the decoherence $\chi(T)$. Using the expansion Eq. (44), we write

$$\chi(T) = \sum_{k=0} C_k \phi_k T^{k+2},$$  \hspace{1cm} (51)

where the decoherence functions $\phi_k$ is given by Eq. (26). It seems that we can find DD schemes to suppress the decoherence to an arbitrary order $\chi(T) = O(T^{M+2})$ by solving the equations $\phi_k = 0$ with $C_k \neq 0$ for $k < M$. However, we have shown in Sec. 11A2 by Theorem 1 that for soft-cutoff noise, there is a largest decoupling order.

The even-order functions $\phi_{2m}$ are given by Eq. (28). Using
Eq. (B10), we have the odd-order functions \( \phi_{2M-1} \),
\[
\phi_{2M-1} = \frac{(2M-1)!}{2} \sum_{r=0}^{2M-1} (-1)^r \frac{\lambda_r}{r! (2M-1-r)!} + (2M-1)! \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{|\tilde{f}(\omega)|^2}{(\omega_0)^{2M}} 
- \sum_{r=0}^{2M-1} \sum_{n=0}^{2M-1-r} (-1)^r \frac{\lambda_n}{(\omega_0)^r \Gamma(n+1) n!}.
\] (52)
The condition Eq. (29) gives
\[
\phi_{2M-1} = (2M-1)! \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{|\tilde{f}(\omega)|^2}{(\omega_0)^{2M}}.
\] (53)
Notice in the summation \( 2M-1-r < M \). We obtain
\[
\phi_{2M-1} = (-1)^M (2M-1)! \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{|\tilde{f}(\omega)|^2}{\omega^{2M}}.
\] (54)
In Eq. (54), the integrand \( |\tilde{f}(\omega)|^2/\omega^{2M} \geq 0 \) and it cannot vanish for all \( \omega \) from \( -\infty \) to \( \infty \). Thus we have the following theorem.

**Theorem 2.** There is no non-zero modulation function \( F(t/T) \) to eliminate the errors so that the equations \( \phi_{2m} = 0 \) \( m=0 \) and \( \phi_{2M-1} = 0 \) satisfy simultaneously.

For example, for the noise with the correlation function \( e^{-|t/t_1|} \), one cannot simultaneously eliminate the two leading decoherence terms \( C_{0}\phi_0 \) and \( C_{1}\phi_1 \), and the error induced by the noise is at least \( O(T_2) \). The result is consistent with Theorem 1 since the noise correlation \( e^{-|t/t_1|} \) implies a spectrum with a soft cutoff at high frequencies \( S(\omega) \sim \frac{1}{\omega^{2M}} \).

### A. Sequence optimization

In this paper, we optimize the DD performance in the short-time limit. As indicated in Eq. (54), a smaller \( \tilde{f}(\omega T) \) at low frequencies will give a smaller \( \phi_{2M-1} \). Here we focus on DD with ideal instantaneous \( \pi \) pulses. We use more pulses to construct a more efficient modulation function \( F(t/T) = F_\pi(t/T) \) to minimize \( \phi_{2M-1} \), with the conditions \( \phi_{2m} = 0 \) \( m=0 \) [Eq. (29)]. Using the method of Lagrange multipliers, we need to solve a set of nonlinear equations as
\[
\nabla_{y,T} G_M = 0,
\] (55a)
\[
G_M \equiv \sum_{j=0}^{M-1} y_j^2 \lambda_j^{(\pi)} + \phi_{2M-1},
\] (55b)
\[
\nabla_{y,T} \equiv \left( \frac{\partial}{\partial y_1}, \ldots, \frac{\partial}{\partial y_N}, \frac{\partial}{\partial y_0}, \ldots, \frac{\partial}{\partial y_{M-1}} \right).
\] (55c)
The introduced variables \( y_j \) are the Lagrange multipliers. Note that the sequence optimization in Ref. [51] also used the constraints \( \{\lambda_j^{(\pi)} = 0\} \), but the constraints were used there to guarantee the convergence of the calculation of \( \lambda(T) \). Here, the constraints eliminate the lowest orders of errors \( (\phi_{2m} = 0 \) \( m=0 \)) [see Eq. (29)] in short-time limit. In particular, the decoherence from inhomogeneous broadening is eliminated when \( \phi_0 = 0 \).

We calculate Eq. (56) by separating the domain of integration according to the value of \( F_\pi(t/T) \). For \( k \geq 0 \), we obtain
\[
\phi_k = \frac{-1}{T^{k+2}(k+1)(k+2)} \left[ \sum_{j=2}^{N} \left( T_j - T_k \right)^{k+2} (-1)^{j+k+2} \right]
+ \left( T_{N+1} - T_0 \right)^{k+2} \left( -1 \right)^{N+1} + 2 \sum_{j=1}^{N} \left( T_j - T_0 \right)^{k+2} (-1)^j
+ 2 \sum_{j=1}^{N} (T_{N+1} - T_k)^{k+2} (-1)^{N+1+j} \right].
\] (56)
Then we have
\[
\frac{\partial \phi_{2M-1}}{\partial (T_k/T)} = \frac{-(-1)^k}{T^{2M} M} \left[ 2 \sum_{j=k+1}^{N} \left( T_j - T_k \right)^{2M} (-1)^j - \left( T_k - T_0 \right)^{2M} + (T_{N+1} - T_k)^{2M} (-1)^{N+1} - 2 \sum_{j=1}^{k-1} \left( T_j - T_k \right)^{2M} (-1)^j \right].
\] (57)
For the special case of \( M = 1 \), \( G_1 = y_0 \lambda_0^{(\pi)} + \phi_1 \), we find that the CPMG sequences are solutions to Eq. (55). The timing of an \( N \)-pulse CPMG sequence reads
\[
T_j^{CPMG} = \frac{2 j - 1}{2N} T, \text{ for } j = 1, \ldots, N.
\] (58)
The CPMG sequences obviously satisfy the constraint \( \lambda_0^{(\pi)} = 0 \) [see Eq. (19)], which is the so-called echo condition that eliminates the effect of static inhomogeneous broadening. Eqs. (57) and (19) give
\[
\frac{\partial \phi_1}{\partial (T_k/T) \mid_{CPMG}} = \frac{-(-1)^{k+1}}{4 N^2} \left[ 1 + (-1)^N \right],
\] (59a)
\[
\frac{\partial \lambda_0^{(\pi)}}{\partial (T_k/T) \mid_{CPMG}} = 2 (-1)^{k+1} y_0.
\] (59b)
Thus the CPMG sequences also satisfy Eq. (55) with \( y_0 = -\frac{1}{2N} \left[ 1 + (-1)^N \right] \), so they are at least the locally optimal pulse sequences. It has been proved that CPMG sequences are the most efficient pulse sequences in protecting the qubit coherence against telegraph-like noise in the short-time limit [52]. With numerical evidence, we conjecture that it is also true that the CPMG sequences are globally optimal in the short-time limit when the time expansion of the noise correlation function has the two leading terms \( C_0 \) and \( C_1 \).

For other cases of minimizing \( \phi_{2M-1} \) with the condition \( \lambda_0^{(\pi)} = 0 \) \( m=0 \), one can see that the short-time optimized DD (ODD) coincides with UDD for pulse number \( N \leq M \). And as \( N \) increases, the ODD sequences gradually approach the
FIG. 2: (Color online). Comparison of the ODD, UDD and CPMG sequences for different pulse number \(N\). Squares (blue), triangles (green), and circles (red) correspond to UDD, CPMG, and ODD. The ODD sequences are optimized to minimize \(\phi_1\) under the constraints \(\phi_0 = \phi_2 = 0\).

CPMG sequences. For example, ODD for the noise correlation

\[
\hat{\beta}(t)\hat{\beta}(0) = C_0 + C_2 t^2 + C_3 |t|^3 + O(t^4),
\]

is shown in Fig. 2 in comparison with UDD and CPMG. The ODD sequences resemble the CPMG sequences when \(N\) is large.

We show in Fig. 3(a) the performance of three DD schemes against the noise described by Eq. (60). A comparison is also shown in Fig. 3(b) by considering a hard high-frequency cut-off \(\omega_c = 40\). In Fig. 3(a), we can see that ODD sequences give better performance than UDD and CPMG sequences. These ODD sequences are optimal for a wide range of noise which has the noise correlation given by Eq. (60). When we introduce a high-frequency cutoff in the noise spectrum, as the case in Fig. 3(b), the ODD is the best initially when the number of pulses \(N \leq \omega_c T/2 \approx 10\), and the UDD sequences become better and suppress the decoherence order by order when \(N\) is large and the hard cutoff is reached. In Fig. 3(b), for large \(N\) UDD is better than ODD, since the ODD sequences are optimized for soft-cutoff noise rather than hard-cutoff noise. In Fig. 3(a) the decreasing of \(\chi(T)\) is a linear decrease in the double-logarithm plot, but in Fig. 3(b) it is much faster. This confirms that DD is not so efficient against soft-cutoff noise.

IV. SUMMARY AND CONCLUSIONS

We have studied the dynamical decoupling control of decoherence caused by Gaussian noise with soft cutoff in a general modulation formalism. We have proved Theorem 1 which shows that, for the soft cutoff with the power-law asymptote \(\alpha/\omega^P\) at high frequencies, DD can only suppress decoherence to \(O(T^{P+1})\), where \(P\) does not need to be an integer. When the short-time expansion of noise correlation has the \((2K + 1)\)th odd expansion term, DD can only suppress decoherence to \(O(T^{2K+1})\) (Theorem 2). The existence of odd-order terms in the short-time expansion corresponds to a soft high-frequency cutoff (\(\sim \alpha/\omega^{2K}\)) in the noise spectrum. For these noise spectra, we have derived the equations for pulse sequence optimization, which minimizes the leading odd-order decoherence function and eliminates even-order decoherence functions of lower orders. The ODD sequences obtained by this method coincide with the UDD sequences when the pulse number \(N\) is small, and they resemble CPMG sequences when \(N\) is large. For the special case that the short-time correlation function expansion has a linear term in time (i.e., a soft cutoff \(\sim \alpha/\omega^2\)), the ODD sequences are exactly the CPMG sequences.

Although we derived the theorems from a pure dephasing model, we expect that the results of the existence of the largest decoupling order in short-time limit can be extended to the general decoherence model (including both dephasing and relaxation) of quantum systems. It is desirable to study the DD in suppressing the general decoherence of quantum systems in the soft-cutoff noise in the future.

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Appendix A: Modulation Functions in Multiqubit Systems

Consider an \(L\)-qubit (or \(2^L\)-level) system suffering pure dephasing described by the Hamiltonian

\[
H = \sum_{m=0}^{2^L-1} |m\rangle\langle m| [\omega_m + \beta_m(t)],
\]
where \( \omega_m \) is the energy and \( \beta_m(t) \) is the fluctuation on the state \( |m\rangle \). Here \( m = (m_1 \cdots m_{2m}) \) is a binary code with \( m_l = 0 \) or \( 1 \) for the \( l \)th qubit.

The Pauli operator for the \( l \)th qubit is

\[
\sigma^{(l)}_x = \sum_{m=0}^{m=2^{-1}} \left( m + 2^{-\frac{1}{3}} \right) |m\rangle + H.c.,
\]

which exchanges two basis states \( |m\rangle \) and \( |m'\rangle \) if \( m \) and \( m' \) differ at and only at the \( l \)th bit.

After a sequence of \( \sigma^{(l)}_x \) pulses and a final pulse \( \sigma_{add} = \sigma^{(l)}_x \cdots \sigma^{(l)}_x \), the evolution operator is

\[
U(T) = \sigma_{add} U(T_{N+1}, T_N) \sigma^{(l)}_x U(T_{N}, T_{N-1}) \cdots \times \sigma^{(l)}_x U(T_2, T_1) \sigma^{(l)}_x U(T_1, T_0) \sigma^{(l)}_x,
\]

where \( T_0 = 0, T_{N+1} = T \), and \( \sigma^{(l)}_x \equiv I \). The free evolution operator

\[
U(T_{j+1}, T_j) = \exp \left[ -i \int_{T_j}^{T_{j+1}} \sum_{m=0}^{m=2^{-1}} \langle m | [\omega_m + \beta_m(t)] dt \right].
\]

We write the evolution operator as

\[
U(T) = e^{-i \sum_{m=0}^{m=2^{-1}} \int_{l=0}^{l=N} \sum_{i=0}^{i=l} \sum_{n=0}^{n=m} \eta |o_m + \beta_n(t)| dt},
\]

with \( \mathcal{E}_{m,j} \equiv \sigma_x^{(l)} \cdots \sigma_x^{(l)} |m\rangle \langle m| \sigma_x^{(l)} \cdots \sigma_x^{(l)} \). The phase factor between the states \( |p\rangle \) and \( |q\rangle \) changed during the evolution time \( T \)

\[
\varphi_{pq}(T) = \langle p | U(T) | q \rangle\langle q | U^\dagger (T) | p \rangle,
\]

where

\[
\langle p | U(T) | q \rangle = e^{-i \sum_{m=0}^{m=2^{-1}} \int_{l=0}^{l=N} \sum_{i=0}^{i=l} \sum_{n=0}^{n=m} \eta |o_m + \beta_n(t)| dt},
\]

Note that \( \sigma_x^{(l)} \cdots \sigma_x^{(l)} |p\rangle = |p \oplus [0 \cdots l]\rangle \) with \( p \oplus [0 \cdots l] \equiv p \) for \( j > 0 \) and \( p \oplus [0 \cdots l] \equiv p \) for \( j = 0 \) and \( p \oplus [0 \cdots l] \). Here \( \oplus \) denotes addition on binary digits, that is, \( p \oplus [0 \cdots l] \) is obtained by flipping the \( l_1 \ldots l_j \)th bits of \( p = (p_L \cdots p_{2p}) \) in binary code. We obtain

\[
\langle p | U(T) | q \rangle = e^{-i \sum_{m=0}^{m=2^{-1}} \int_{l=0}^{l=N} \sum_{i=0}^{i=l} \sum_{n=0}^{n=m} \eta |o_m + \beta_n(t)| dt},
\]

Therefore the coherence between the states \( |p\rangle \) and \( |q\rangle \) decreases by the average of the random phase

\[
e^{-i \sum_{m=0}^{m=2^{-1}} \int_{l=0}^{l=N} \sum_{i=0}^{i=l} \sum_{n=0}^{n=m} \eta |o_m + \beta_n(t)| dt}
\]

When each of the qubits feels the same noise, \( \beta_{m_l}(t) = (\sum_{l=1}^{L} m_l) \beta(t) \) for the \( r \)th and

\[
\varphi_{pq}(T) = e^{-i \int_{T_0}^{T} F_{pq}(t) dt} e^{-i \int_{T_0}^{T} F_{pq}(t) dt},
\]

where the modulation function is defined as

\[
F_{pq}(t) = (\sum_{l=1}^{L} \tilde{p}_k) - (\sum_{k=1}^{L} \tilde{q}_k),
\]

with \( \tilde{p} = p \oplus [0 \cdots l] \) and \( \tilde{q} = q \oplus [0 \cdots l] \) for \( t \in (T_j, T_{j+1}) \). For example, when \( L = 2 \), \( F_{pq}(t) \) in \([0, \pm 1, \pm 2]\).

**Appendix B: Decoherence Functions \( \phi_{k} \)**

As \( F(t/T) = 0 \) for \( t \notin (0, T) \), we extend the bounds of integration for \( t \) to infinity and transform Eq. (26) to

\[
\phi_{k} = \mathcal{R} \frac{\partial^k}{\partial (\nu)^k} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \times \tilde{f}(\eta) f(\omega) e^{-i\omega_2t_2} e^{-i\omega t_1} e^{-i\nu t},
\]

where we set \( \eta \to 0 \) after differentiation. Integrations over \( t_2, t_1 \) and \( \omega \) give

\[
\phi_{k} = \mathcal{R} \frac{\partial^k}{\partial (\nu)^k} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \int_{-\infty}^{\infty} dt \tilde{f}(\eta) f(\omega) e^{-i\omega t},
\]

Using the formulas

\[
\frac{\partial^k}{\partial (\nu)^k} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} v(\nu) = \sum_{r=0}^{k} \frac{k!}{r!(k-r)!} \frac{\partial^{k-r} v(\nu)}{\partial (\nu)^{k-r}} \frac{\partial r}{\partial (\nu)^r},
\]

and

\[
\frac{\partial^k}{\partial (\nu)^k} \tilde{f}(\eta) = \lambda_\nu, \text{ for } r \geq 0,
\]

we have

\[
\phi_{k} = k! \mathcal{R} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \left( \frac{\tilde{f}(\omega)}{(-i\omega)^{k+1}} - \sum_{r=0}^{k} \frac{\tilde{f}(\omega)}{(-i\omega)^{k-r+1}} \frac{\lambda_\nu}{r!} \right).
\]

Changing the summation index, we obtain

\[
\phi_{k} = k! \mathcal{R} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \left( \frac{\tilde{f}(\omega)}{(-i\omega)^{k+1}} - \sum_{r=0}^{k} \frac{\tilde{f}(\omega)}{(-i\omega)^{k-r+1}} \frac{\lambda_\nu}{r!} \right).
\]

Note that the summation over \( r \) and the integration over frequency cannot be exchanged when the integration does not converge for each individual term. Using Eq. (16a), we have

\[
\phi_{k} = k! \mathcal{R} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \left( \frac{\tilde{f}(\omega)}{(-i\omega)^{k+1}} + \sum_{r=0}^{k} \frac{\tilde{f}(\omega)}{(-i\omega)^{k-r+1}} \frac{\lambda_\nu}{r!} \right).
\]

We simplify the last line by using Eq. (16b) and the equality

\[
\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \sum_{n=r}^{\infty} (\pm i\omega)^{r-n} \frac{1}{n!} = \frac{1}{r!} \frac{1}{2 (r-1)!}, \text{ for } r \geq 1, t \geq 0,
\]

which is proved in Appendix C. We obtain

\[
\phi_{k} = k! \mathcal{R} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \left( \frac{\tilde{f}(\omega)}{(-i\omega)^{k+1}} + \sum_{r=0}^{k} \frac{\tilde{f}(\omega)}{(-i\omega)^{k-r+1}} \frac{\lambda_\nu}{r!} \right).
\]

**Note:** The above derivation assumes a specific form of the modulation function \( F_{pq}(t) \), which may not hold in all cases. Further analysis or assumptions are required to generalize the results.
For even number $2m$, \( \frac{\partial f(\omega)}{\partial \omega} \) is an odd function and its integral vanishes. Eq. (B10) gives

\[
\phi_{2m} = (2m)! R \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \sum_{r=0}^{2m-1} \frac{(-1)^r}{(i\omega)^{2m-r-n+1}} \left( \sum_{n=0}^{\infty} \frac{\Lambda_n^r \Lambda_n}{r! n!} \right)
\]

\[
+ \frac{(2m)!}{2} \left( \sum_{r=0}^{2m} (-1)^r \frac{\Lambda_n^r}{r!} \frac{R_{2m-r}}{(2m-r)!} \right),
\]

(B11)

which is decomposed as (with the changes of summation order and indices)

\[
\phi_{2m} = (2m)! R \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \sum_{r=0}^{2m-1} \frac{(-1)^r/2}{(i\omega)^{2m-r-n+1}} \left( \sum_{n=0}^{\infty} \frac{\Lambda_n^r \Lambda_n}{r! n!} \right)
\]

\[
+ \frac{1}{2} \sum_{r=0}^{2m} \frac{(-1)^r/2}{(i\omega)^{2m-r-n+1}} \left( \sum_{n=0}^{\infty} \frac{\Lambda_n^r \Lambda_n}{r! n!} \right)
\]

\[
+ \frac{(2m)!}{2} \left( \sum_{r=0}^{2m} (-1)^r \frac{\Lambda_n^r}{r!} \frac{R_{2m-r}}{(2m-r)!} \right),
\]

(B12)

The integrals of the terms with odd-power of \( \omega \) vanish. And for even functions of \( \omega \), the sum \( n + r \) is an odd number, so \( (-1)^r + (-1)^r = 0 \). Thus the real part of the integral in Eq. (B12) vanishes. From the last line of Eq. (B12), we obtain Eq. (28), i.e.,

\[
\phi_{2m} = \frac{(2m)!}{2} \sum_{r=0}^{2m} (-1)^r \frac{\Lambda_n^r}{r!} \frac{R_{2m-r}}{(2m-r)!},
\]

(B13)

**Appendix C: Proof of Equation (B9)**

To prove Eq. (B9), we just need to prove

\[
\lim_{R \to \infty} \int_{-R}^{R} \frac{dx}{x} \sum_{n=r}^{\infty} \frac{e^{i\omega x}}{n!} = \frac{\pi}{(r-1)!}, \quad \text{for } r \geq 1,
\]

(C1)

where the bounds in the integral guarantee that the modulation function \( F(t) \) is a real function. Using

\[
\int_{-R}^{R} \frac{dx}{x} \sum_{n=r}^{\infty} \frac{e^{i\omega x}}{n!} = \sum_{n=1}^{\infty} \frac{R^n}{(n + r - 1)!n!} (e^{\omega R} + c.c.),
\]

(C2a)

\[
\frac{1}{(r-1)!} \int_{-R}^{R} \sin x x \, dx = \sum_{n=1}^{\infty} \frac{R^n}{n!n!(r-1)!} (e^{\omega R} + c.c.),
\]

(C2b)

and \( \lim_{R \to \infty} \int_{-R}^{R} \frac{\sin x}{x} \, dx = \pi \), we just need to prove

\[
\lim_{R \to \infty} \int_{-R}^{R} \frac{\sin x}{x} \, dx = \pi.
\]

(C3)

For \( r = 1 \), it obviously holds. For \( r \geq 2 \), we can show the difference

\[
\Delta = \sum_{n=1}^{\infty} \frac{R^n}{(n + r - 1)!} \left( 1 - \left( \frac{1}{R} \right)^{n - 1} \right) = O\left( \frac{1}{R} \right),
\]

(C4)

so \( \lim_{R \to \infty} \Delta = 0 \). By expanding the terms in the square brackets of Eq. (C4), we have

\[
\Delta = \sum_{n=1}^{\infty} \frac{R^n}{(n + r - 1)!} \left( \sum_{k=0}^{r-2} a_k R^k \right),
\]

(C5)

where \( a_k \) is a number independent of \( n \). We arrange the terms in the square brackets and get

\[
\Delta = \sum_{n=1}^{\infty} \frac{R^n}{(n + r - 1)!} \left( \sum_{k=0}^{r-2} b_k \sum_{j=1}^{k} [(n + r - j) + b_j] \right),
\]

(C6)

with \( b_j \) independent of \( n \). After some simplification it becomes for \( r \geq 2 \)

\[
\sum_{k=0}^{r-2} b_k \sum_{j=1}^{n} \left( e^{R} \sum_{n=0}^{\infty} \frac{a_k}{n!} R^n + c.c. \right) = O\left( \frac{1}{R} \right).
\]

(C7)

Hence \( \Delta = O\left( \frac{1}{R} \right) \), and Eq. (B9) is proved.

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