Arbitrarily Accurate Pulse Sequences for Robust Dynamical Decoupling

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We introduce universally robust sequences for dynamical decoupling, which simultaneously compensate pulse imperfections and the detrimental effect of a dephasing environment to an arbitrary order, work with any pulse shape, and improve performance for any initial condition. Moreover, the number of pulses in a sequence grows only linearly with the order of error compensation. Our sequences outperform the state-of-the-art robust sequences for dynamical decoupling. Beyond the theoretical proposal, we also present convincing experimental data for dynamical decoupling of atomic coherences in a solid-state optical memory.

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Introduction.—Quantum technologies are increasingly important nowadays for a multitude of applications in sensing, processing, and communication of information. Nevertheless, protection of quantum systems from unwanted interactions with the environment remains a major challenge. Dynamical decoupling (DD) is a widely used approach that aims to do this by nullifying the average effect of the unwanted qubit-environment coupling through the application of appropriate sequences of pulses [1–4].

Most DD schemes focus on dephasing processes because they have maximum contribution to information loss in many systems, e.g., in nuclear magnetic resonance and quantum information [5,6]. Then, the major limitation to DD are pulse imperfections whose impact often exceeds the effect of the perturbations from the environment [6–8]. Some sequences, e.g., the widely used Carr-Purcell-Meiboom-Gill (CPMG) sequence, work efficiently for specific quantum states only [6,9]. Robust sequences for any state with limited error compensation have been demonstrated experimentally, e.g., XY4 (PDD), Knill DD (KDD) [6]. Composite pulses, designed for static errors, were also recently shown to be robust to time-dependent non-Markovian noise up to a noise frequency threshold [10]. A common feature of most robust DD sequences so far is pulse error compensation in one or two parameters only (flip angle error, detuning). High fidelity error compensation has been proposed, e.g., by nesting of sequences, but only at the price of a very fast growth in the number of pulses [6].

In this Letter, we describe a general theoretical procedure to derive universally robust (UR) DD sequences that compensate pulse imperfections in any experimental parameter (e.g., variations of pulse shapes or intensities) and the effect of a slowly changing environment to an *arbitrary order* in the permitted error. We note that the term universal is applied for pulse errors. The UR sequences work at high efficiency for any initial condition. The number of pulses for higher order error compensation grows only linearly with the order of the residual error. The concept works for arbitrary pulse shapes. Our only assumptions are identical pulses in a sequence and a correlation time of the environment that is longer than the sequence duration—in order to maintain appropriate phase relations between the pulses. In the following, we will describe our theoretical approach and present convincing data from a demonstration experiment with relevance to applications in quantum information technology, i.e., DD of atomic coherences for coherent optical data storage in a $Pr^{3+}:Y_2SiO_5$ crystal (termed Pr:YSO). As our numerical simulations and the experimental data show, the UR sequences outperform the best robust DD sequences available so far.

The system.—We consider a system, consisting of an ensemble of noninteracting two-state systems in a dephasing environment, and assume we have no control of the environment. Similarly to previous work on robust DD sequences [11], we use a semiclassical approximation, where the free evolution Hamiltonian of a qubit includes an effective time-dependent Hamiltonian due to the system-environment interaction [12,13]. This is the case, e.g., when the changes in the environment are slow, compared to the delay between the pulses in the DD sequences [11]. Such systems are encountered in many solid-state spin systems, e.g., doped solids, electron spins in diamond, electron spins in quantum dots, etc.

We denote a qubit transition frequency as $\omega_S^{(k)}(t) = \bar{\omega}_S + \Delta^{(k)} + \epsilon(t)$, where $\bar{\omega}_S$ is the center frequency of the ensemble, and $\Delta^{(k)}$ is the detuning of kth qubit due to a slowly changing qubit-environment interaction, e.g., inhomogeneous broadening; $\epsilon(t)$ is a stochastic term due to a fast qubit-environment interaction, which cannot be refocused by DD and its effect is simply an additional exponential decay of the coherence—we omit it further on. The assumption for constant $\Delta^{(k)}$ during a DD sequence becomes feasible by shortening the time between the pulses. This was the inspiration for the introduction of

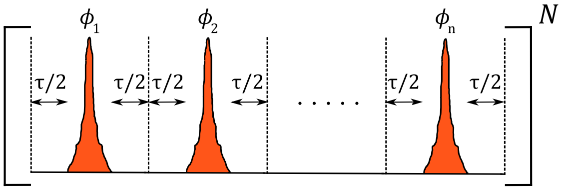

FIG. 1. Schematic description of a DD sequence with n equally separated phased pulses. A single cycle free evolution-pulse-free evolution lies within the dashed lines. The proper choice of the relative phases of the pulses compensates both pulse errors and dephasing due to the environment. The DD sequence is repeated N times during the storage time; the pulse shape can be arbitrary.

TABLE I. Phases of the symmetric universal rephasing (UR) DD sequences with n cycles (indicated by the number in the label), based on Eq. (3). Each phase is defined modulo 2π.

Sequence	Phases	$\Phi^{(n)}$
UR4	$(0,1,1,0)\pi$	π
UR6	$\pm(0,2,0,0,2,0)\pi/3$	$\pm 2\pi/3$
UR8	$\pm(0,1,3,2,2,3,1,0)\pi/2$	$\pm\pi/2$
UR10	$\pm(0,4,2,4,0,0,4,2,4,0)\pi/5$	$\pm 4\pi/5$
UR12	$\pm(0,1,3,0,4,3,3,4,0,3,1,0)\pi/3$	$\pm\pi/3$
UR14	$\pm(0,6,4,8,4,6,0,0,6,4,8,4,6,0)\pi/7$	$\pm 6\pi/7$
UR16	$\pm(0,1,3,6,2,7,5,4,4,5,7,2,6,3,1,0)\pi/4$	$\pm\pi/4$

the widely used CPMG sequence, following the seminal work of Hahn [9,14]. Previous experiments have demonstrated that these are reasonable assumptions for comparison of robust DD sequences [11].

The Hamiltonian of the system in a rotating frame at an angular frequency $\bar{\omega}_S$ is $\hat{H}_f(t) = \Delta^{(k)}\hat{S}_z$ ($\hat{S}_z = \hbar\sigma_z/2$, $\hbar = 1$) during free evolution and $\hat{H}(t) = \hat{H}_f(t) + \hat{H}_p(t)$ during a pulse. The latter depends on qubit frequency offset $\Delta^{(k)}$, the (time-dependent) detuning of the applied field $\Delta^{(p)}(t) \equiv \bar{\omega}_S - \omega^{(p)}(t)$ and the Rabi frequency $\Omega(t) = -\mathbf{d}\cdot\mathbf{E}(t)/\hbar$. We make *no assumptions* about $\Omega(t)$ and $\Delta^{(p)}(t)$, which may vary for the different qubits.

The dynamics of a qubit due to a pulse is described by a propagator $\mathbf{U}_{\text{pulse}}$, which connects the density matrices of the system at the initial and final times t_{i} and t_{f}: $\rho(t_{\text{f}}) = \mathbf{U}_{\text{pulse}}\rho(t_{\text{i}})\mathbf{U}_{\text{pulse}}^{\dagger}$ and can be parametrized [15] as

$$\mathbf{U}_{\text{pulse}}(\alpha,\beta,p) = \begin{bmatrix} \sqrt{1-p}e^{i\alpha} & \sqrt{p}e^{i\beta} \\ -\sqrt{p}e^{-i\beta} & \sqrt{1-p}e^{-i\alpha} \end{bmatrix}, \quad (1)$$

where p is the transition probability, induced by a pulse; α, β are (unknown) phases. A phase shift ϕ in the Rabi frequency, $\Omega(t) \to \Omega(t)e^{i\phi}$, is imprinted in $\mathbf{U}_{\text{pulse}}$ as $\beta \to \beta + \phi$ [15–17]. The phase ϕ is assumed the same for every qubit (unlike β), which is usually experimentally feasible.

DD sequences traditionally consist of time-separated pulses [9,14]. We consider DD with equal pulse separation (see Fig. 1), which was shown to be preferable for most types of environment [18]. The propagator for a single cycle, defined as free evolution—(phased) pulsed excitation—free evolution, is $\mathbf{U}(\phi) = \mathbf{U}_{\text{pulse}}(\alpha+\delta, \beta+\phi, p)$, where $\delta \equiv -\Delta^{(k)}\tau/2$ accounts for the effect of the environment during free evolution. The parameters α, β, δ, p may vary for the different qubits and are affected by many factors, e.g., field inhomogeneities, effect of the environment. The propagator of a DD sequence of n free evolution-pulse-free evolution cycles, where the kth pulse is phase shifted by ϕ_k (see Fig. 1), takes the form $\mathbf{U}^{(n)} = \mathbf{U}(\phi_n)\ldots\mathbf{U}(\phi_2)\mathbf{U}(\phi_1)$, where $\phi_1,\ldots,\phi_{\text{n}}$ are free control parameters. The DD sequence can be repeated N times for decoupling during the whole storage time.

Derivation of the UR DD sequences.—Our goal is to preserve an arbitrary qubit state, which can be achieved (up to a phase shift) if a DD sequence has an even number of (phased) pulses, and each performs complete population inversion, i.e., $p = 1$ (see Supplemental Material at [19]). Thus, we define our target propagator as $\mathbf{U}_0 = \mathbf{U}^{(n)}(p=1)$. We choose the phases $\phi_1,\ldots,\phi_{\text{n}}$, so that systematic errors in a pulse cycle are compensated by the other cycles in a DD sequence, similarly to the technique of composite pulses [22]. The DD sequence performance is characterized with the fidelity [6]

$$F = \frac{1}{2}|\text{Tr}(\mathbf{U}_0^{\dagger}\mathbf{U}^{(n)})| \equiv 1 - \varepsilon_{\text{n}}, \quad (2)$$

where ε_{n} is the fidelity error of a DD sequence of n cycles.

In order to minimize ε_{n}, we perform a Taylor expansion with respect to the transition probability p at $p = 1$ (ideal π pulse) and use the control parameters ϕ_k to nullify the series coefficients for every α, δ, and β up to the largest possible order of p. The phase ϕ_1 has a physical meaning only with respect to the (unknown) phase of the initial coherence, so we take $\phi_1 = 0$ without loss of generality. In the case of two cycles, e.g., the well-known CPMG sequence, the fidelity error is $\varepsilon_2 = 2(1-p)\cos^2(\alpha+\delta-\phi_2/2)$. Thus, error compensation is not possible by a proper choice of ϕ_2, except for a particular $\alpha+\delta$ or for certain initial states [6]. However, error compensation for an arbitrary initial state becomes possible with four or a higher even number of cycles.

We derive a general formula for the phases of a UR sequence of n pulses (see also Supplemental Material at [19])

$$\phi_k^{(n)} = \frac{(k-1)(k-2)}{2}\Phi^{(n)} + (k-1)\phi_2, \quad (3a)$$

$$\Phi^{(4m)} = \pm\frac{\pi}{m}, \qquad \Phi^{(4m+2)} = \pm\frac{2m\pi}{2m+1}. \quad (3b)$$

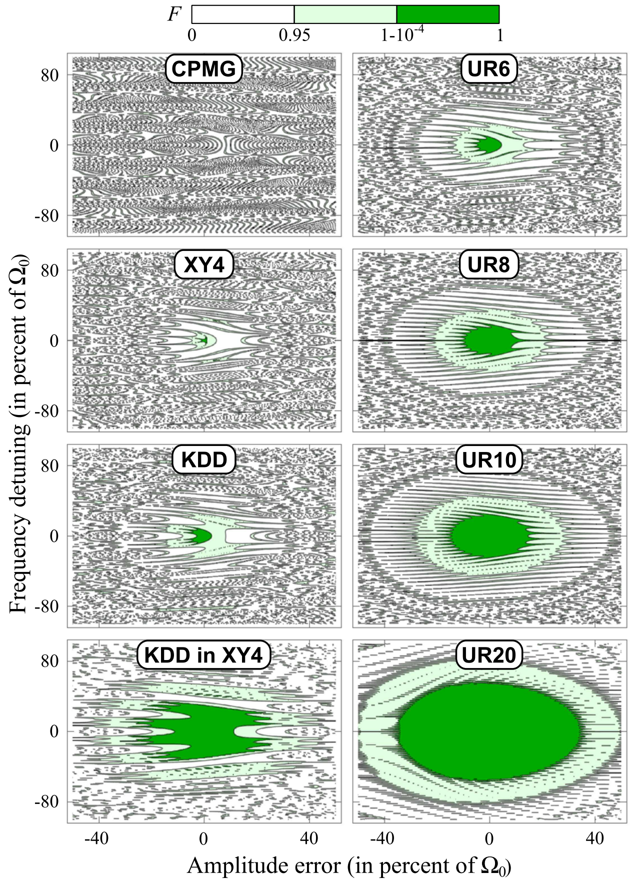

FIG. 2. Numerically simulated fidelity F vs detuning and amplitude errors for DD sequences from Table I and [6] for a total of 120 cycles. The DD pulses are rectangular with a duration of $T = \pi/\Omega_0$ and time separation of $\tau = 4T$.

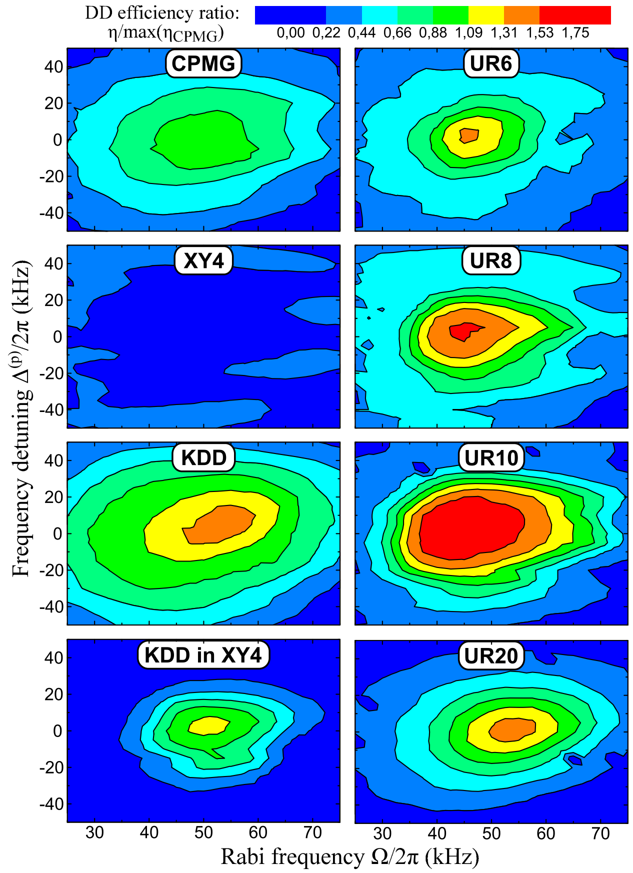

FIG. 3. Experimentally measured ratio of light storage efficiency for DD sequences from Table I and [6], and the maximum efficiency of the CPMG sequence. A total of 120 rectangular pulses with duration $T = 10\ \mu$s and time separation $\tau = 40\ \mu$s were applied; storage time is 6 ms. The performance of longer DD sequences is expectedly reduced by decoherence as the sequence duration approaches $T_2 = 500\ \mu$s.

The addition of an arbitrary phase $\tilde{\phi}$ to all phases does not affect the overall performance, while ϕ_2 can be chosen at will to perform an arbitrarily accurate phase gate $\exp(i\chi\hat{S}_z)$, $\chi = n(\phi_2 - \tilde{\phi})/2$ without additional pulses. We note that for $n = 4$, $\phi_2 = \pi/2$, we obtain the well-known XY4 sequence [6]. The simplest symmetric UR DD sequences with a target $\mathbf{U}_0 = (-1)^{n/2}\mathbf{I}$ are given in Table I. It is notable that the order of error compensation increases *linearly* with the number of cycles n:

$$\varepsilon_n = 2(1-p)^{n/2}\sin^2\left[\frac{n}{2}(\alpha + \delta - \pi/2 - \phi_2/2)\right]. \quad (4)$$

This is the central result of this Letter. Arbitrarily accurate error compensation is achievable even for small single pulse transition probability for any pulse shape, e.g., also for chirped pulses [23]; the linear rise in the number of pulses for higher order error compensation is superior to traditional techniques, e.g., nesting of sequences [6]; the analytic formula for UR DD allows for fine-tuning to the specific pulse errors and environment.

Figure 2 demonstrates the theoretical fidelity of several DD sequences against frequency detuning and Rabi frequency errors for a single qubit. The applied rectangular pulses differ only in their phases, and each sequence is repeated to ensure a total of 120 pulses (e.g., UR10 is repeated $N = 12$ times). The parameter range corresponds to the experimental Fig. 3. The simulations show that the fidelity of the CPMG sequence is very sensitive to pulse errors, while the robustness of the UR sequences increases quickly with the sequence order. It is remarkable that the fidelity error ε_n for UR20 stays below the 10^{-4} quantum information benchmark even with amplitude errors and frequency offset of nearly 40% of the Rabi frequency. We note that the ultrahigh fidelity range expands even more with shorter pulse separation and higher order sequences. Finally, UR20 is more robust than the current state-of-the-art sequence for pulse error compensation KDD in XY4 (also of 20 pulses) [6]. This is not surprising since the fidelity error $\varepsilon_{20} \sim (1-p)^6$ for KDD in XY4 is larger than $\varepsilon_{20} \sim (1-p)^{10}$ for UR20 ($p \to 1$).

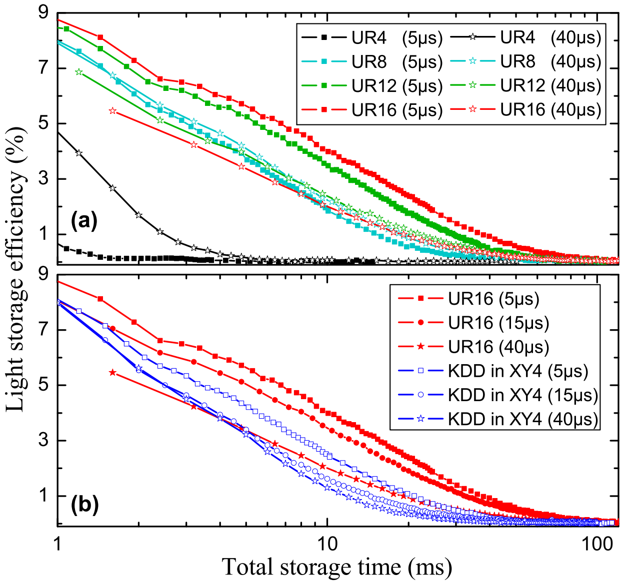

FIG. 4. Experimentally measured efficiency of stored light for several DD sequences, defined in Table I and [6] for different pulse separation. DD is performed with rectangular rf pulses with a frequency of 10.2 MHz, duration of 10 μs and a Rabi frequency $\Omega_0 \approx 2\pi$ 50 kHz, optimized for a maximum efficiency with the CPMG sequence for a storage time of 100 μs. Note the logarithmic scale on the time axis.

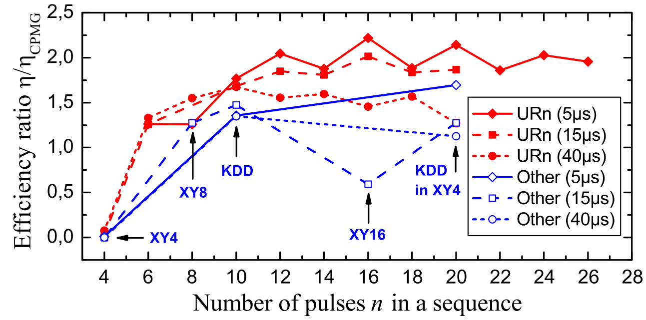

FIG. 5. Experimentally measured ratio of light storage efficiency for DD sequences from Table I and [6]. Experimental parameters are identical to Fig. 4, storage time is (approximately) 6 ms.

Experimental demonstration.—We experimentally verified the performance of the UR sequences for DD of atomic coherences for optical data storage. In the experiment, we generate a coherence on a radio frequency (rf) transition between two inhomogeneously broadened hyperfine levels of the Pr:YSO crystal. The coherence is prepared and read-out by electromagnetically induced transparency (EIT) [21]. EIT in a doped solid was already applied to drive an optical memory with long storage times [24] or high storage efficiency [25]. The concept and experimental setup for (single-pass) EIT light storage are described in Supplemental Material at [19] and [25].

In such a coherent optical memory, it is crucial to reverse the effect of dephasing of atomic coherences during storage due to inhomogeneous broadening of the hyperfine levels ($T_{\rm deph} \approx 13$ μs). Additionally, stochastic magnetic interactions between the dopant ions and the host matrix lead to a decoherence time of $T_2 \approx 500$ μs. DD is ideally implemented with instantaneous resonant π pulses, which are not feasible in our experiment due to inhomogeneous broadening and the spatial inhomogeneity of the rf field. In order to permit a much broader operation bandwidth, we replace the identical pulses in the widely used CPMG sequence [9] with phased pulses. In all experiments, the optical "write" and "read" sequences were kept the same, while the DD sequences with the same pulse separation have identical duty cycle (total irradiation time divided by total time) for a fair comparison; therefore, the energy of the retrieved signal measures the DD efficiency (see also Supplemental Material at [19]).

In the first experiment (Fig. 3), we compare the efficiency of several DD sequences for a storage time of 6 ms, i.e., much longer than $T_2 \approx 500$ μs. Matching to the simulations in Fig. 2, we implement DD with 120 rectangular rf pulses. We vary the Rabi frequency and the detuning to obtain a 2D plot of the relative storage efficiency. The experimental results confirm the theoretical prediction that the efficiency increases with the UR order until the longer sequences are significantly affected by decoherence. We note that Figs. 2 and 3 are expected to differ as the former simulates only the DD fidelity for a single qubit in a constant environment. For example, the CPMG sequence has a higher storage efficiency than XY4 in the experiment because it works very well for some initial quantum states [6], i.e., some atoms in the ensemble. However, applying the CPMG sequence with pulse errors effectively projects the qubits on such states, thus making it unsuitable for quantum storage. The UR sequences significantly outperform the traditional sequences, e.g., the efficiency of UR10 is about 75% higher than the CPMG sequence. We also verified the superior performance of the UR sequences for a pulse separation $\tau = 15$ μs and with Gaussian pulses (see Supplemental Material at [19]).

In a second experiment, we compare DD efficiencies at different storage times. Figure 4(a) also confirms the theoretical prediction that the UR efficiency increases with the sequence order. The highest efficiency is achieved with UR16 for a pulse separation of $\tau = 5$ μs, while UR12 is better for 40 μs. This is explained by the trade-off between longer sequences that compensate pulse errors better and shorter sequences that suffer less from decoherence. The UR analytical formula and the fast improvement in error compensation allow for fine-tuning of the optimal sequence to the specific environment. Shorter (than 5 μs) pulse separation and even continuous UR sequences are theoretically possible and should provide even better performance. However, these were not possible in our experimental setup. Figure 4(b) compares the performance of the UR16 and the state-of-the-art KDD in XY4 sequence [6]. The experimental data show that UR16 performs remarkably better than KDD in XY4. This is expected from theory since UR16 has both higher order pulse error compensation and less pulses. The improvement is less for $\tau = 40$ μs as the duration of both sequences exceeds T_2.

Figure 5 summarizes the experimental performance of UR and other sequences [6] vs number of pulses n in a sequence. The data confirm the UR superior performance and the improvement of error compensation with n. The optimal sequence changes with pulse separation due to the trade-off between pulse errors and decoherence during a sequence, which affects its error self-compensatory mechanism [6]. The slight oscillation in UR efficiency is likely due to higher-order effects for the particular time separation. Finally, we note that we also verified the superior performance of the UR sequences in comparison to composite pulses with the same duty cycle, e.g., U5a and U5b [15].

Conclusion.—We theoretically developed and experimentally demonstrated universally robust DD sequences, which compensate systematic errors in any experimental parameter and the effect of a slowly changing dephasing environment to an arbitrary order for any pulse shape and initial condition. The UR sequences require a linear growth in the number of pulses for higher order error compensation, which is faster than traditional methods, e.g., nesting of sequences. The only assumptions made are those of a coherent evolution during the DD sequence (the correlation time of the environment is longer than the sequence duration) and identical phased pulses. We also experimentally confirmed the superior performance of our UR sequences for DD for coherent optical data storage in a Pr:YSO crystal.

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