Engineering arbitrary pure and mixed quantum states

Alexander Pechen[*,†]

Steklov Mathematical Institute, Russian Academy of Sciences, Gubkina str. 8, Moscow 119991, Russia

Department of Chemical Physics, Weizmann Institute of Science, Rehovot 76100, Israel

(Received 12 September 2010; published 6 October 2011)

Controlled manipulation by atomic- and molecular-scale quantum systems has attracted a lot of research attention in recent years. A fundamental problem is to provide deterministic methods for controlled engineering of arbitrary quantum states. This work proposes a deterministic method for engineering arbitrary pure and mixed states of a wide class of quantum systems. The method exploits a special combination of incoherent and coherent controls (incoherent and coherent radiation) and has two properties which are specifically important for manipulating by quantum systems: it realizes the strongest possible degree of their state control, complete density matrix controllability, meaning the ability to steer arbitrary pure and mixed initial states into any desired pure or mixed final state, and it is all-to-one, such that each particular control transfers all initial system states into one target state.

DOI: 10.1103/PhysRevA.84.042106 PACS number(s): 03.65.Yz, 02.30.Yy, 32.80.Qk

I. INTRODUCTION

Controlled manipulation by atoms and molecules using external controls is an active field of modern research with applications ranging from selective creation of atomic or molecular excitations to the control of chemical reactions and to the design of nanoscale systems with desired properties. The external control may be either coherent (e.g., a tailored laser pulse [1]) or incoherent (e.g., a specially adjusted or engineered environment [2,3] or quantum measurements commonly used with and sometimes without feedback [4]).

A challenging topic in quantum control is to provide practical methods for engineering arbitrary quantum states [5]. The interest in this topic is driven by fundamental connections to quantum physics as well as by potential applications to quantum state measurement [5] and quantum computing with mixed states and nonunitary quantum gates [6]. Various recipes for engineering arbitrary quantum states of light were proposed [7,8]. For matter, engineering an arbitrary open system's quantum dynamics with coherent control, quantum measurements, and feedback was shown to be achievable [9]. However, the problem of deterministic engineering of arbitrary quantum states of matter has generally remained unsolved.

This paper proposes a deterministic method for engineering arbitrary pure and mixed density matrices for a wide class of quantum systems. The method uses a combination of incoherent control by engineering the state of the environment (for which we consider appropriately filtered incoherent radiation) on the time scale of several orders of magnitude of the characteristic system relaxation time τ_{rel} followed by fast (e.g., femtosecond) coherent laser control to produce arbitrary pure and mixed states for a wide class of quantum systems. The method is deterministic in the sense that it does not use real-time feedback and can be applied to an ensemble of systems without the need for an individual addressing of each system. It is important that the suggested scheme requires the ability to manipulate both Hamiltonian and non-Hamiltonian aspects of the dynamics; a fundamental result of Altafini shows that varying only the Hamiltonian is not sufficient to produce arbitrary states of a quantum system [10].

Engineered environments were suggested for improving quantum computation and quantum state engineering [11], making robust quantum memories [12], preparing many-body states and nonequilibrium quantum phases [13], and inducing multiparticle entanglement dynamics [14]. This paper exploits incoherent control by an engineered environment [2] in combination with coherent control for producing arbitrary pure and mixed density matrices. While incoherent processes were used in various circumstances, for example, in cold molecule research, to prepare a pure state needed for full control over the system's pure states [15], their use in the proposed method serves a more general goal of engineering arbitrary pure and mixed quantum states.

The method has two special properties. First, it implements complete density matrix controllability, the strongest possible degree of state control for quantum systems, meaning the ability to prepare in a controllable way any density matrix starting from any initial state. Second, the produced controls are all-to-one: any such control c_* transfers all pure and mixed initial states into the same final state and thus can be optimal simultaneously for all initial states [16]. This property has no analog for purely coherent control of closed quantum systems with unitary evolution, where different initial states in general require different optimal controls. While an abstract theoretical construction of all-to-one controls was provided [16], the problem of their physical realization has remained open. The suggested method provides a solution for such a physical realization.

II. COHERENT AND INCOHERENT CONTROLS

The dynamics of a controlled n-level quantum system isolated from the environment is described by density matrix ρ_t, satisfying the equation

$$\dot{\rho}_t = -i\,[H_0 + u(t)V, \rho_t]\,, \qquad \rho_{t=0} = \rho_{\mathrm{i}}. \tag{1}$$

Here $H_0 = \sum \varepsilon_i |i\rangle\langle i|$ is the free system Hamiltonian (with eigenvalues $\varepsilon_1 < \varepsilon_2 < \cdots < \varepsilon_n$ and eigenvectors $|i\rangle$), and V is

[*]On leave from Department of Chemistry, Princeton University, Princeton, New Jersey 08544, USA.

[†]apechen@gmail.com

the interaction Hamiltonian describing coupling of the system to the control field $u(t)$ (e.g., a shaped laser pulse). The evolution is unitary, $\rho_t = U_t \rho_0 U_t^\dagger$, where the unitary evolution operator U_t satisfies the Schrödinger equation $\dot{U}_t = -i[H_0 + u(t)V]U_t$. The unitary nature of the evolution induced by the field $u(t)$ implies preservation of coherence in the system such that, for example, pure states will always remain pure; the corresponding control is called coherent.

If the system interacts with the environment, then its evolution in the absence of coherent control is described by the master equation for the reduced density matrix ρ_t [17,18]:

$$\dot{\rho}_t = -i\,[H_0 + H_{\rm eff}, \rho_t] + \mathcal{L}(\rho_t), \qquad \rho_{t=0} = \rho_{\rm i}, \tag{2}$$

where the superoperator $\mathcal{L}$ and the effective Hamiltonian $H_{\rm eff}$ describe the influence of the environment. The effective Hamiltonian $H_{\rm eff}$ represents spectral broadening of the system energy levels and typically commutes with H_0. For a Markovian environment, the superoperator $\mathcal{L}$ has the form $\mathcal{L}(\rho) = \sum_i (2L_i \rho L_i^\dagger - L_i^\dagger L_i \rho - \rho L_i^\dagger L_i)$ with some matrices L_i [17,19]. The explicit form of these matrices is determined by the state of the environment and by the details of the microscopic interaction between the system and the environment.

The matrices L_i are usually considered as fixed and having a deleterious effect on the ability to control the system: open quantum systems subject to the Markovian evolution (2) with constant $\mathcal{L}$ are uncontrollable [10]. However, the assumption of constant $\mathcal{L}$ is too restrictive since $\mathcal{L}$ can be manipulated by adjusting the state of the environment through its temperature, pressure, or, more generally, its distribution function. Control through adjusting the state of the environment in general does not preserve quantum coherence in the controlled system and for this reason is called incoherent [3].

This paper considers incoherent radiation as an example of a Markovian control environment, and the scheme is analyzed below for this case. Other environments, either Markovian or non-Markovian, can also be used as incoherent controls; however, the ability to use a particular environment for engineering arbitrary quantum states using the proposed scheme requires a separate analysis in each case. The state of the environment formed by incoherent photons is characterized by the distribution $n_{\mathbf{k}}$ of photons in momentum $\mathbf{k}$; in general, the distribution of photons in polarization α can also be exploited. For the purpose of this paper the directional dependence of the distribution function is not necessary, and its dependence only on the photon energy $\omega = |\mathbf{k}|$ is used; n_ω is assumed to be constant over the frequency range of significant absorption and emission for each system transition frequency $\omega_{ij} = \varepsilon_j - \varepsilon_i$. The distribution function n_ω can be experimentally manipulated, for example, by filtering.

The evolution of the system in the environment formed by incoherent radiation with distribution function n_ω is approximated by the master equation (2) which was derived from the exact dynamics in the weak coupling limit [20,21] and whose superoperator $\mathcal{L} = \mathcal{L}_{n_\omega}$ has the form

$$\mathcal{L}_{n_\omega}(\rho) = \sum_{i,j} [\gamma^+_{\omega_{ij}} + \gamma^-_{\omega_{ji}}](2\hat{\mu}_{ij}\rho\hat{\mu}_{ij}^\dagger - \hat{\mu}_{ij}^\dagger\hat{\mu}_{ij}\rho - \rho\hat{\mu}_{ij}^\dagger\hat{\mu}_{ij}), \tag{3}$$

where $\hat{\mu}_{ij} = \mu_{ij}|i\rangle\langle j|$, $\mu_{ij} = \langle i|\mu|j\rangle$, and μ is the dipole moment for coupling the system to radiation. The coefficients $\gamma^+_\omega = C_\omega[n_\omega + 1]$ and $\gamma^-_\omega = C_\omega n_\omega$ ($C_\omega > 0$ for $\omega > 0$ and $C_\omega = 0$ for $\omega \leqslant 0$) depend on the photon distribution n_ω and determine the transition rates between system energy levels with transition frequency ω. The Einstein A coefficients for spontaneous emission are defined by $A_{ij} = C_{\omega_{ji}}|\mu_{ij}|^2$. The radiative energy density per unit angular frequency interval is $\rho_\omega = \hbar\omega n_\omega(\omega^2/\pi^2c^3)$.

III. ENGINEERING ARBITRARY QUANTUM STATES

Let $\rho_{\rm i}$ be any (mixed or pure) initial state of the system and $\rho_{\rm f} = \sum p_i|\phi_i\rangle\langle\phi_i|$ be an arbitrary (mixed or pure) target (final) state. Without loss of generality we assume $p_1 \geqslant p_2 \geqslant \cdots \geqslant p_n$. The method can be used to prepare arbitrary nondegenerate states which form a dense set in the space of all density matrices, thus approximately realizing arbitrary pure and mixed quantum states. The system is assumed to be generic in the sense that all its transition frequencies are different (in particular, its spectrum is nondegenerate) and all off-diagonal matrix elements μ_{ij} of the dipole moment are nonzero. The goal is to find a combination of coherent and incoherent fields transforming all $\rho_{\rm i}$ into $\rho_{\rm f}$.

The scheme to steer $\rho_{\rm i}$ into $\rho_{\rm f}$ consists of two stages. In the first stage, the system evolves on the time scale of several orders of magnitude of the relaxation time, $t \approx a\tau_{\rm rel}$ (where a can be chosen in the range 2–10 depending on the required degree of accuracy) under the action of a suitable optimal incoherent control n^*_ω into the state $\tilde{\rho}_{\rm f} = \sum p_i|i\rangle\langle i|$ diagonal in the basis of H_0 and having the same spectrum as the final state $\rho_{\rm f}$. The state $\tilde{\rho}_{\rm f}$ has the same purity as $\rho_{\rm f}$; $\tilde{\rho}_{\rm f}$ is mixed if $\rho_{\rm f}$ is mixed, and $\tilde{\rho}_{\rm f}$ is pure if $\rho_{\rm f}$ is pure. In the second stage, the system evolves on the fast (e.g., femtosecond) time scale under the action of a suitable coherent laser control, which rotates the basis of H_0 to match the basis of $\rho_{\rm f}$.

The first stage exploits incoherent control with any n^*_ω such that $n^*_{\omega_{ij}} = p_j/(p_i - p_j)$ to prepare the system in the state $\tilde{\rho}_{\rm f}$. Coherent control is switched off [$u(t) = 0$] during this stage, and the system dynamics is described by the master equation (2), which for off-diagonal matrix elements $\rho_{ln} = \langle l|\rho|n\rangle$ of the density matrix takes the form

$$\dot{\rho}_{ln} = -(i\alpha_{ln} + \Gamma_{ln})\rho_{ln}, \qquad \Gamma_{ln} = \sum_j (W_{jl} + W_{jn}) \geqslant 0,$$

where $\alpha_{ln} = \langle l|H_0 + H_{\rm eff}|l\rangle - \langle n|H_0 + H_{\rm eff}|n\rangle$ and $W_{ij} = 2(\gamma^+_{ji} + \gamma^-_{ij})|\mu_{ij}|^2$. Off-diagonal elements decay exponentially for $\Gamma_{ln} > 0$, $\rho_{ln} \sim \exp(-\Gamma_{ln}t)$. Diagonal elements satisfy the Pauli master equation

$$\dot{\rho}_{nn} = \sum_j (W_{nj}\rho_{jj} - W_{jn}\rho_{nn}).$$

For generic systems, all $n_{\omega_{ij}}$ can be independently adjusted, and the detailed balance condition $W_{nj}\rho_{jj} = W_{jn}\rho_{nn}$ implies $n^*_{\omega_{ij}} = p_j/(p_i - p_j)$. The master equation with an incoherent distribution n_ω such that $n^*_{\omega_{ij}} = p_j/(p_i - p_j)$ has $\tilde{\rho}_{\rm f}$ as the stationary state and exponentially quickly drives the system to $\tilde{\rho}_{\rm f}$ [22]. The case when some $p_i = p_j \neq 0$ formally requires infinite density at the corresponding transition (e.g., infinite temperature of the environment is required for creating the equally populated state $\tilde{\rho}_{\rm f} = \frac{1}{n}\mathbb{I}$), but for practical applications a reasonable degree of accuracy allowing for a finite $n^*_{\omega_{ij}}$ is always sufficient. The first stage, when necessary, can

be divided into two parts, diagonalization of the density matrix with any n_ω producing positive and sufficiently large Γ_{ln} followed by the evolution driven by any n^*_ω such that $n^*_{\omega_{ij}} = p_j/(p_i - p_j)$ to produce $\tilde{\rho}_\mathrm{f}$.

The second stage implements coherent dynamics with unitary evolution transforming the basis $\{|i\rangle\}$ into $\{|\phi_i\rangle\}$ and hence steering $\tilde{\rho}_\mathrm{f}$ into ρ_f. For successful realization of this stage we assume that any unitary evolution operator of the system can be produced with available coherent controls when $\mathcal{L} = 0$ on a time scale sufficiently shorter than τ_rel, i.e., that the system is unitary controllable when decoherence effects are negligible. Incoherent control is switched off during this stage by setting $n_\omega = 0$, and the dynamics is well approximated by the unitary evolution (1).

Necessary and sufficient conditions for unitary controllability were obtained by Ramakrishna *et al.* [23]. Sufficient conditions for complete controllability of n-level quantum systems subject to a single control pulse that addresses multiple allowed transitions concurrently were established [24]. Ramakrishna *et al.* showed that a necessary and sufficient condition for unitary controllability of a quantum system with Hamiltonian $H = H_0 + u(t)V$ is that the Lie algebra generated by the operators H_0 and V has the dimension n^2. We assume that the system satisfies this condition when $\mathcal{L} = 0$ and that any U can be produced on a time scale sufficiently shorter than the decoherence time scale, e.g., by using time-optimal control [25]. Under these assumptions a coherent field $u^*(t)$ exists that produces a unitary operator U transforming the basis $\{|i\rangle\}$ into $\{|\phi_i\rangle\}$ and therefore steering $\tilde{\rho}_\mathrm{f}$ into ρ_f. This field implements the second stage of the scheme to finish evolving ρ_i into ρ_f. The second stage can be implemented also on the long time scale using dynamical decoupling [26] to effectively decouple the system from the environment.

There exist other methods to prepare arbitrary quantum states. An example is the method of engineering arbitrary Kraus maps proposed by Lloyd and Viola [9], which, in particular, can be used to produce arbitrary quantum states. Another option is to cool an ensemble of systems to a pure state $|\phi\rangle$ and then apply randomly and independently to each system a control producing a unitary operator U_i transforming $|\phi\rangle$ into $|\phi_i\rangle$. Implementing each U_i with probability p_i will effectively produce the systems in the state $\rho_\mathrm{f} = \sum p_i |\phi_i\rangle\langle\phi_i|$. Both these methods require independent addressing of individual systems in the ensemble that in general is hard to realize. The proposed method is free of this shortcoming and can be applied to an ensemble without the need to independently address each system. This advantage should significantly simplify practical realization when individual addressing of quantum systems is hard to realize. We remark that the first control stage requires individual addressing of each transition frequency. If the system is nongeneric, in particular, if it is degenerate, then the method may work if the degenerate states can be selectively addressed by using polarization of the incoherent radiation. For degenerate systems (harmonic oscillator) another scheme was proposed [5].

Determining the correct form of the master equation for systems with time-dependent Hamiltonians in general is a nontrivial problem [27,28]. However, the proposed scheme does not use the master equation with time-dependent Hamiltonian $u(t)V$, and therefore the problem of choosing the correct form of the master equation with time-dependent Hamiltonian is not relevant for this work. The scheme in its first step exploits incoherent control when the coherent control is switched off, and therefore the system Hamiltonian is time independent. In this case the system dynamics under Markovian approximation is governed by a master equation of the form (2). The second step exploits fast coherent control on a short time scale when incoherent control is switched off and the decoherence effects are negligible. In this case $\mathcal{L} \approx 0$ and the dynamics is well approximated by (1).

If the quantum system is sufficiently simple and all its relevant parameters are known, then methods of optimal control theory (OCT) can be used to find optimal fields n^*_ω and $u^*(t)$, as shown in Fig. 1(a). If the system is complex and/or some of its relevant parameters are unknown, then various adaptive learning algorithms can be used to implement the proposed scheme for engineering arbitrary quantum states [Fig. 1(b)]. In this case, the creation of a desired diagonal mixed state $\tilde{\rho}_\mathrm{f}$ during the first stage of the control scheme can be realized with learning algorithms [2]. The second stage can be implemented using learning algorithms for coherent control [29]. Learning algorithms were shown to be efficient for both cases, and therefore they will be efficient when used in a successive combination as required in the proposed scheme.

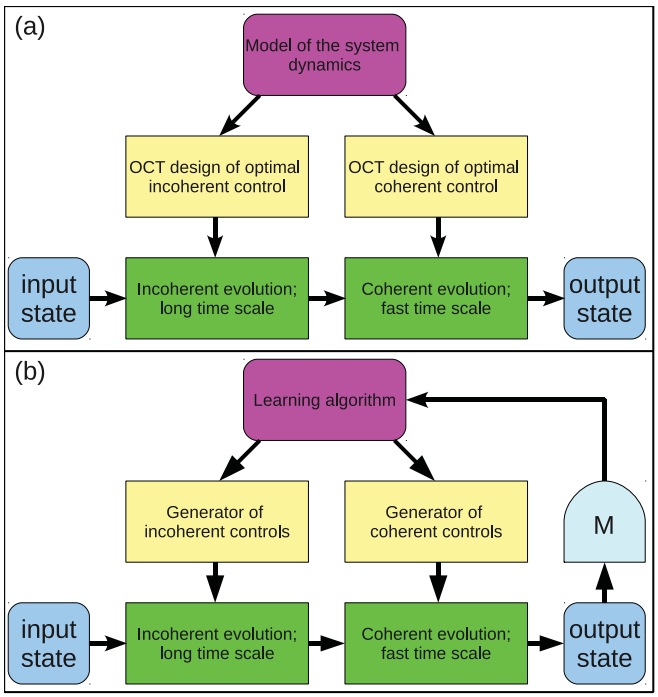

FIG. 1. (Color online) Implementing complete density matrix controllability with (a) OCT and (b) learning control. In (a), methods of OCT and a theoretical model of the system dynamics are used to find optimal incoherent and coherent controls n^*_k and $u^*(t)$. In (b), the objective $J = \|\rho_T - \rho_\mathrm{f}\|$, where ρ_T is the output state, is measured (block with the letter M) at the end of each iteration, and the result is used to design a new control, ideally with better performance. The cycle is repeated until a satisfactory output is obtained.

IV. COMPLETE DENSITY MATRIX CONTROLLABILITY

Depending on a particular problem, various notions of controllability for quantum systems are used [10,16,23,30–32], including unitary controllability, pure state, density matrix and observable controllability [30], and complete density matrix controllability [16]. Unitary controllability means the ability to produce with available controls any unitary evolution operator U. Density matrix controllability means the ability to transfer one arbitrary density matrix into another with the same spectrum (i.e., kinematically equivalent density matrices); a particular case is pure state controllability, which is the ability to transfer one arbitrary pure state into another. Complete density matrix controllability means the ability to steer any initial (pure or mixed) density matrix ρ_{i} into any (pure or mixed) final density matrix ρ_{f}, irrespective of their relative spectra [16]. This notion is different from density matrix controllability [30], where only kinematically equivalent density matrices are required to be accessible, and is the strongest among all degrees of state controllability for quantum systems; complete density matrix controllability of a quantum system implies in particular its pure state and density matrix controllability. The suggested scheme allows for transferring one arbitrary density matrix into another, thereby approximately realizing complete density matrix controllability of quantum systems, the strongest possible degree of their state control.

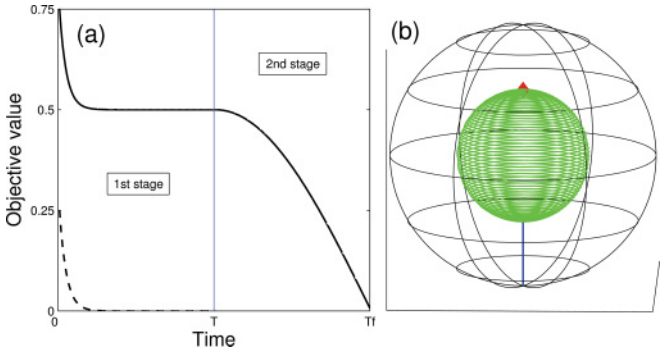

FIG. 2. (Color online) Application of the control scheme for engineering a desired mixed state of two Ca energy levels. (a) The behavior of the objective vs time during the two stages of the control. The time scale is not uniform; the duration of the first stage is $T = 50$ ns, and that of the second stage is $T_{\mathrm{f}} - T = 1310$ fs. The solid line is for $\|\rho_t - \rho_{\mathrm{f}}\|$, and the dotted line is for $\|\rho_t - \tilde{\rho}_{\mathrm{f}}\|$; the latter quantity almost completely vanishes during the first stage of the control. (b) The corresponding evolution of the Bloch vector. The initial system state corresponds to the south pole, the vertical blue line represents incoherent evolution during the first stage, and the green line shows coherent rotations during the second stage under the action of a resonant field of amplitude $E = 10^7$ V/m. The evolved state approaches the target state, indicated by the red marker at the top.

V. ALL-TO-ONE CONTROLS

An all-to-one control c_* is a control that steers all initial states into one final state. Such controls can be optimal simultaneously for all initial states [16], and their importance is motivated by the following. Let $J(c) \equiv J(\rho_T)$ be an arbitrary control objective determined by the system density matrix ρ_T evolving from the initial state ρ_0 to the final time T under the action of the control c (e.g., $J = \mathrm{Tr}\,[\rho_T O]$ for some Hermitian observable O or $J = \|\rho_T - \rho_{\mathrm{f}}\|$). In general, optimal controls (controls minimizing the objective) are different for different initial states. However, if c_* is an all-to-one control steering all states into a state ρ_{f} minimizing J, then c_* will optimize the objective for any initial system state. While an abstract theoretical construction of all-to-one controls was provided [16] in terms of special Kraus maps, whose definition is also provided below, their physical realizations has remained an open problem. The proposed control scheme provides a physical realization of all-to-one controls and the corresponding Kraus maps for any pure or mixed state ρ_{f}. The all-to-one property is achieved during the first stage, where n^*_ω produces the same density matrix $\tilde{\rho}_{\mathrm{f}}$ independently of the initial state.

The density matrix representing the state of an n-level quantum system is a positive, unit-trace $n \times n$ matrix. We denote by $\mathcal{M}_n = \mathbb{C}^{n\times n}$ the set of all $n \times n$ complex matrices and by $\mathcal{D}_n := \{\rho \in \mathcal{M}_n \,|\, \rho = \rho^\dagger, \rho \geqslant 0, \mathrm{Tr}(\rho) = 1\}$ the set of all density matrices. A map $\Phi : \mathcal{M}_n \to \mathcal{M}_n$ is positive if $\Phi(\rho) \geqslant 0$ for any $\rho \geqslant 0$ in $\mathcal{M}_n$. A map $\Phi : \mathcal{M}_n \to \mathcal{M}_n$ is completely positive (CP) if for any $l \in \mathbb{N}$ the map $\Phi \otimes \mathbb{I}_l : \mathcal{M}_n \otimes \mathcal{M}_l \to \mathcal{M}_n \otimes \mathcal{M}_l$ is positive ($\mathbb{I}_l$ is the identity map in $\mathcal{M}_l$). A CP map is trace preserving if $\mathrm{Tr}[\Phi(\rho)] = \mathrm{Tr}(\rho)$ for any $\rho \in \mathcal{M}_n$. Completely positive trace-preserving maps are referred to as Kraus maps; they represent the reduced dynamics of quantum systems initially uncorrelated with the environment [18,33].

Any Kraus map Φ can be represented using the operator-sum representation (OSR) as $\Phi(\rho) = \sum_{i=1}^{l} K_i \rho K_i^\dagger$, where $\{K_i\}$ is a set of complex $n \times n$ matrices satisfying the condition $\sum_{i=1}^{l} K_i^\dagger K_i = \mathbb{I}_n$ to ensure trace preservation [18,33]. The OSR is not unique: any Kraus map Φ can be represented using infinitely many sets of Kraus operators.

The all-to-one Kraus map for a given final state $\rho_{\mathrm{f}} = \sum_i p_i |\phi_i\rangle\langle\phi_i|$ is defined as a Kraus map $\Phi_{\rho_{\mathrm{f}}}$ steering all initial states into ρ_{f}, i.e., such that $\Phi_{\rho_{\mathrm{f}}}(\rho) = \rho_{\mathrm{f}}$ for all $\rho \in \mathcal{D}_n$ [16]. If ρ_{f} is a density matrix maximizing a given objective J and c_* is a control producing the map $\Phi_{\rho_{\mathrm{f}}}$, then this control will be simultaneously optimal for all initial states; i.e., the same c_* will maximize the objective for any initial system state. An OSR for a universally optimal Kraus map $\Phi_{\rho_{\mathrm{f}}}$ can be

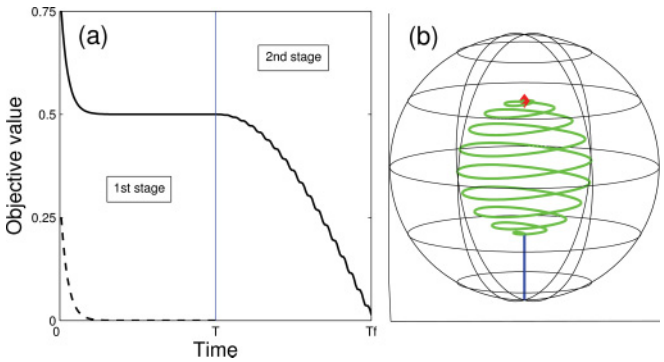

FIG. 3. (Color online) The same as Fig. 2, except that $E = 10^9$ V/m and $T_{\mathrm{f}} - T = 13.1$ fs.

constructed by using $K_{ij} = \sqrt{p_i}|\phi_i\rangle\langle\phi_j|$ as the Kraus operators. Indeed, for any ρ, $\sum_{i,j=1}^{n} K_{ij}\rho K_{ij}^{\dagger} = \rho_{\rm f}$.

All-to-one Kraus maps were constructed theoretically in [16]. The two-stage control scheme described in this paper provides an approximate physical realization of all-to-one Kraus maps $\Phi_{\rho_{\rm f}}$ for all $\rho_{\rm f}$ (therefore any all-to-one Kraus map can be produced using this scheme) for generic systems that are unitary controllable on the time scale which is small with respect to $\tau_{\rm rel}$.

VI. EXAMPLE: CALCIUM ATOM

The scheme is illustrated below with an example of a two-level atom whose relevant parameters are known and whose controlled dynamics are easily analytically understood and visualized. We consider calcium upper and lower levels $4\,^1P$ and $4\,^1S$ as two states $|1\rangle$ and $|0\rangle$ of the two-level system. For this system the transition frequency is $\omega_{21} = 4.5 \times 10^{15}$ rad/s, the radiative lifetime $t_{21} = 4.5$ ns, the Einstein coefficient $A_{21} = 1/t_{21} \approx 2.2 \times 10^8$ s^{-1}, and the dipole moment $\mu_{12} = 2.4 \times 10^{-29}$ C m [34]. The method works equally well for any initial and target states; we take for the sake of definiteness $\rho_0 = |0\rangle\langle 0|$ and $\rho_{\rm f} = \frac{1}{4}|0\rangle\langle 0| + \frac{3}{4}|1\rangle\langle 1|$.

The system is generic, all its relevant parameters are known, and we can analytically find optimal controls. The goal of the first (incoherent) stage is to prepare the mixture $\tilde{\rho}_{\rm f} = \frac{3}{4}|0\rangle\langle 0| + \frac{1}{4}|1\rangle\langle 1|$. This goal is realized by applying to the system incoherent radiation with a distribution satisfying $n_{\omega_{21}} = 1/2$ during time T of several magnitudes of decoherence time t_{21}; we choose $T = 50$ ns. The goal of the second (coherent) stage is to rotate the state produced at the end of the first stage to transform it into $\rho_{\rm f}$. This goal can be realized, for example, by applying a resonant π pulse $E(t) = E\cos(\omega_{12}t)$. Other methods of coherent control that produce the same unitary transformation can be used as well. The electric field amplitude that makes the Rabi frequency equal to the radiative decay rate is $E \approx 10^3$ V/m [34]. The proposed scheme requires the duration of the second stage to be significantly shorter than the decay time. This can be satisfied by choosing $E \gtrapprox 10^4$ V/m. We take resonant electromagnetic field $E(t) = E\cos(\omega_{12}t)$ of amplitude $E = 10^7$ V/m acting on the system during the time interval $T_{\rm f} - T = 1310$ fs. The Rabi frequency for a field of such amplitude is $\Omega_{\rm R} \approx 1/2320$ fs^{-1}; thus the field acts as a π pulse transforming the state $\tilde{\rho}_{\rm f}$ into $\rho_{\rm f}$. The results of the numerical simulation are shown in Fig. 2. The time interval 1310 fs is much less than t_{21}, and decoherence effects are negligible during the second stage. To better visualize the trajectory during the second stage we provide in Fig. 3 simulation results for $E = 10^9$ V/m and $T_{\rm f} - T = 13.1$ fs. The method can be applied to produce arbitrary pure or mixed target density matrices from any pure or mixed initial state, thereby implying the complete density matrix controllability of this system.

ACKNOWLEDGMENTS

This research was supported by the Marie Curie Action within the 7th European Community Framework Programme.

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