PHYSICAL REVIEW A **91**, 042123 (2015)

Interaction-free evolution in the presence of time-dependent Hamiltonians

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(Received 23 February 2015; published 20 April 2015)

The generalization of the concept of interaction-free evolutions (IFE) [Napoli *et al.*, Phys. Rev. A **89**, 062104 (2014)] to the case of time-dependent Hamiltonians is discussed. It turns out that the time-dependent case allows for much richer structures of interaction-free states and interaction-free subspaces. The general condition for the occurrence of IFE is found and exploited to analyze specific situations. Several examples are presented, each one associated to a class of Hamiltonians with specific features.

DOI: 10.1103/PhysRevA.91.042123 PACS number(s): 03.65.Ta, 03.65.Aa, 42.50.Ct

I. INTRODUCTION

An interaction-free evolution (IFE) of a quantum system is an evolution which is not influenced by a certain part of the Hamiltonian, which is addressed as the interaction term [1]. In other words, the dynamics generated by the "unperturbed" Hamiltonian H_0 is essentially the same as the evolution generated by the total Hamiltonian, which is the sum of H_0 and the interaction term $H_{\rm I}$: $H = H_0 + H_{\rm I}$. This notion, which has been introduced in Ref. [1], is somehow related to the concept of decoherence-free subspaces (DFS) [2–6]. In spite of such connection, it should be stressed that the two concepts are still different in many aspects. Generally speaking, the notion of IFE can be relevant to composite systems with different dimensions (like a small system and its environment) or with similar dimensions (for example, two interacting qubits), but it can even concern different degrees of freedom of the same particle (for example, atomic and vibrational degrees of freedom of a trapped ion). One can even talk about IFE states in connection with the action of a classical field on a quantum system, for example, a spin under the action of a magnetic field.

Subradiance [7–13], in its original formulation, is surely a very famous phenomenon which can be thought of as an IFE involving a matter system (several atoms) and the vacuum electromagnetic field.

In this paper, we study the nontrivial extension of IFE states, which applies to those cases wherein the system is governed by a time-dependent Hamiltonian. The interest in this kind of problem is related to several aspects. On the one hand, generally speaking, the resolution of dynamical problems with time-dependent Hamiltonians is a tough job due to the highly nontrivial structure of the corresponding solution

$$U(t) = \mathcal{T} \exp\left(-i\int_0^t H(\tau)d\tau\right), \qquad (1)$$

where $\mathcal{T}$ denotes the chronological product. In general Eq. (1) is untractable and except for some lucky cases [14–16] it requires special assumptions, such as, for example, the adiabatic one [17], or suitable approximations, like in the perturbative treatment [18,19]. Therefore, even the partial resolution of a class of time-dependent problems in the presence of time-dependent Hamiltonians is of interest itself. Formula (1) simplifies if $H(t)$ defines a commutative family, i.e., $[H(t),H(t')] = 0$ for arbitrary t and t'. In this case the chronological product drops out and the entire evolution is controlled by the integral $\int_0^t H(\tau)d\tau$.

On the other hand, there could be important applications in the field of quantum control and in particular in the field of suppression of decoherence effects. Indeed, our analysis could pave the way to extensions of the concepts of subradiance and decoherence-free subspaces in the presence of time-dependent Hamiltonian of the system and even in the presence of time-dependent interaction between the system and its environment.

The paper is organized as follows. In the next section we introduce the problem and find out the general conditions that guarantee the interaction-free evolution. In Secs. III–V we provide several examples of IFE states belonging to different classes. In particular, after the simplest examples in Sec. III, we go on in Sec. IV by analyzing a case of IFE in the context of an adiabatic evolution, while in Sec. V we present some examples related to a more general class of IFE states. Finally, in Sec. VI, we give some conclusive remarks.

II. INTERACTION-FREE CONDITIONS

Let us recall the definition of interaction-free evolution (IFE): We say that a state $|\psi_0\rangle$ undergoes an IFE if it evolves as if the interaction term of the Hamiltonian (which can be time dependent) were absent. To better understand this definition, let us assume that our system is governed by a time-dependent Hamiltonian which can be split into two parts, one part that we call unperturbed and one part that we call the interaction term:

$$i\partial_t|\psi(t)\rangle = (H_0(t) + H_{\rm I}(t))|\psi(t)\rangle\,. \qquad (2)$$

The relevant evolution operator is denoted by $U(t)$, while $U_0(t)$ denotes the evolution operator associated to $H_0(t)$ only. This means

$$i\partial_t U(t) = [H_0(t) + H_{\rm I}(t)]U(t)\,, \qquad (3a)$$

$$i\partial_t U_0(t) = H_0(t)U_0(t)\,. \qquad (3b)$$

We are looking for those states $|\psi_0\rangle \in \mathcal{H}$ (the Hilert space of the system) for which the complete evolution is "essentially" equal to the unperturbed one:

$$U(t)|\psi_0\rangle = e^{iA(t)}U_0(t)|\psi_0\rangle\,, \qquad (4)$$

where $A(t)$ is a real function of time.

By inserting the ansatz $|\psi(t)\rangle = e^{iA(t)}U_0(t)|\psi_0\rangle$ into the Schrödinger equation (2) and exploiting Eqs. (3a) and (3b), we get that the following condition must be satisfied:

$$[H_\mathrm{I}(t) - \dot{A}(t)]U_0(t)|\psi_0\rangle = 0\,, \tag{5}$$

which means that, at every instant, the unperturbed evolution operator $U_0(t)$ maps the initial state $|\psi_0\rangle$ into an instantaneous eigenstate of the interaction term:

$$H_\mathrm{I}(t)U_0(t)|\psi_0\rangle = a(t)\,U_0(t)|\psi_0\rangle\,, \tag{6}$$

with $a(t) = \dot{A}(t)$. This condition is clearly necessary and sufficient, since the chain of implications that brings from Eqs. (4) to (6) can be followed backward, from Eqs. (6) to (4).

It is worth mentioning that all the states satisfying Eq. (6) with the same $a(t)$ form a subspace, that we will address as an IFE subspace. In fact, every state belonging to such a subspace evolves as if the interaction were not present. On the contrary, if one considers the superposition of two IFE states belonging to different IFE subspaces, a phase difference between such states will be accumulated (due to the different values of the eigenvalue $a(t)$), and then the evolution will be effectively different from the one obtained in the absence of interaction.

Let us observe that by applying $U_0^\dagger(t)$ to the both sides of Eq. (6) one gets

$$\tilde{H}_\mathrm{I}(t)|\psi_0\rangle = a(t)|\psi_0\rangle\,, \tag{7}$$

where $\tilde{H}_\mathrm{I}(t) = U_0^\dagger(t)H_\mathrm{I}(t)U_0(t)$ is the interaction term in the interaction picture. This means that the initial state $|\psi_0\rangle$ is supposed to be an eigenstate of $\tilde{H}_\mathrm{I}(t)$ for all t. It should be stressed that $|\psi_0\rangle$ being an eigenvector of $\tilde{H}_\mathrm{I}(t)$ does not need to be an eigenvector of $H_\mathrm{I}(t)$, which is clear from Eq. (6). Note, however, that if $|\psi_0\rangle$ satisfies

$$H_\mathrm{I}(t)|\psi_0\rangle = a(t)\,|\psi_0\rangle\,, \tag{8}$$

and

$$[H_\mathrm{I}(t) - a(t)\mathbb{I}]H_0(t_1)H_0(t_2)\ldots H_0(t_n)|\psi_0\rangle = 0\,, \tag{9}$$

for $n = 1,2,\ldots$, then Eq. (6) is surely satisfied (cf. Appendix A). It should be stressed that Eqs. (8) and (9) are only sufficient but not necessary conditions for $|\psi_0\rangle$ to be an IFE state. The condition in Eq. (7) [as well as that in Eq. (6)] is both necessary and sufficient for $|\psi_0\rangle$ to be IFE state.

Interestingly, in the time-independent case they reduce to

$$[H_\mathrm{I} - a\,\mathbb{I}]H_0^n|\psi_0\rangle = 0\,, \tag{10}$$

for $n = 1,2,\ldots,N-1$, where $N = \dim\mathcal{H}$. It was proved [1] that these conditions are both necessary and sufficient. It is therefore clear that the time-dependent case is much more complicated and rich, showing that $|\psi_0\rangle$ needs not to be eigenvector of $H_\mathrm{I}(t)$ for $t \neq 0$, but $U_0(t)|\psi_0\rangle$ must belong to an eigenspace of the interaction Hamiltonian $H_\mathrm{I}(t)$ at any time.

On the basis of Eq. (7) we can distinguish between two possible situations where the interaction picture interaction term is either time dependent or not. Nevertheless, in order to be effective, such a classification should explore in detail also a sort of " gray zone" which corresponds to all those cases where the Hamiltonian has a trivial time dependence, like, for example, $\tilde{H}_\mathrm{I}(t) = f(t)\tilde{H}_\mathrm{I}(0)$ (we will provide several examples of this kind). Though we will not go through such a *taxonomic* approach, in the examples given in the following sections we will always comment on the specific relevant properties of $\tilde{H}_\mathrm{I}(t)$.

III. SINGLE SYSTEMS SUBJECTED TO EXTERNAL FIELDS

As a class of time-dependent Hamiltonians that allow then occurrence of interaction-free evolutions we will consider the cases of magnetic moments immersed in suitable magnetic fields.

A. Spin-1/2 particle

Let us consider a spin-1/2 particle immersed in a time-dependent magnetic field. The corresponding Hamiltonian is expressible as follows:

$$H(t) = -\mu\mathbf{B}(t)\cdot\mathbf{S}\,, \tag{11}$$

where $\mathbf{S} = (\sigma_x,\sigma_y,\sigma_z)$. Then we take the z contribution, for the moment assumed to be time independent, as the unperturbed Hamiltonian, and the rest as the interaction term ($\hbar = 1$):

$$H_0(t) = \frac{\Omega}{2}\sigma_z, \tag{12a}$$

$$H_\mathrm{I}(t) = \alpha(t)[\cos(\Omega t + \phi)\sigma_x + \sin(\Omega t + \phi)\sigma_y]. \tag{12b}$$

We introduce the notation $\sigma_\theta = \cos\theta\sigma_x + \sin\theta\sigma_y$. The corresponding eigenvectors of σ_θ read

$$|\pm\rangle_\theta = \frac{1}{\sqrt{2}}(e^{-i\theta/2}|+\rangle \pm e^{i\theta/2}|-\rangle)\,, \tag{13}$$

where $|\pm\rangle$ are the eigenstates of σ_z.

Now, suppose that the initial state $|\psi_0\rangle$ is an eigenstate of the operator σ_ϕ: $|\psi_0\rangle = |\pm\rangle_\phi$. It is easy to show that the evolution operator associated to the unperturbed Hamiltonian, which is nothing but a rotation along the z axis, maps such an initial state into an instantaneous eigenstate of $H_\mathrm{I}(t)$:

$$U_0(t)|\pm\rangle_\phi = |\pm\rangle_{\Omega t+\phi} = \frac{1}{\sqrt{2}}(e^{-i(\Omega t+\phi)/2}|+\rangle \pm e^{i(\Omega t+\phi)/2}|-\rangle). \tag{14}$$

In such a case the total evolution is essentially given by the unperturbed evolution, up to a phase factor:

$$|\pm\rangle_\phi \to U(t)|\pm\rangle_\phi = e^{\mp iA(t)}U_0(t)|\pm\rangle_\phi = e^{\mp iA(t)}|\pm\rangle_{\Omega t+\phi}, \tag{15}$$

with $A(t) = \int_0^t \alpha(s)ds$. Of course in each subspace a different phase due to H_I is accumulated.

It is worth noting that we are beyond the trivial case where H_0 and H_I commute. In fact, they do not commute at all, but the operator $U_0(t)$ maps eigenstates of $H_\mathrm{I}(0)$ into eigenstates of $H_\mathrm{I}(t)$.

This results are still valid if we generalize the Hamiltonian model:

$$H(t) = H_0(t) + H_\mathrm{I}(t) = \frac{\Omega(t)}{2}\sigma_z + \alpha(t)[\cos(\Phi(t))\sigma_x + \alpha(t)\sin(\Phi(t))\sigma_y], \tag{16}$$

with

$$\Phi(t) = \int_0^t \Omega(s)ds + \phi\,. \tag{17}$$

There is a clear physical interpretation in terms of classical counterpart of such behaviors. We have a magnetic moment $\mathbf{m}$ on the xy plane which is rotating under the action of a magnetic field along z. Now we add another magnetic field of the xy plane, say $\mathbf{B}_\perp(t)$, which is always parallel to the magnetic moment. At any instant of time, the component $\mathbf{B}_\perp$ does not act on the spin, since the relevant torque is vanishing ($\tau = \mathbf{m} \times \mathbf{B}_\perp = 0$), and then the presence of $\mathbf{B}_\perp$ does not affect the motion of the spin.

It should be clear that if $H_\mathrm{I} = \alpha[\cos\phi\sigma_x + \sin\phi\sigma_y]$ does not depend on time, then there is no interaction-free state corresponding to $H_0 = \frac{1}{2}\Omega(t)\sigma_z$. This shows in a clear way the difference between time-independent and time-dependent cases.

B. Spin-1 particle

Let us now consider a *toy model* involving spin-1 operators (cf. Appendix B). After introducing the following notation,

$$L_\phi = \cos\phi L_x + \sin\phi L_y\,, \tag{18}$$

we consider the following Hamiltonian:

$$H(t) = \Omega(t)L_z + \alpha(t)L^2_{\phi(t)}\,, \tag{19a}$$

with

$$\phi(t) = \int_0^t \Omega(s)ds + \phi(0)\,. \tag{19b}$$

As the initial condition we take the state

$$|\psi_0\rangle = c_-|-1\rangle_{\phi(0)} + c_+|+1\rangle_{\phi(0)}\,, \tag{20}$$

with

$$|\pm 1\rangle_\phi = \frac{e^{-i\phi}}{2}|+1\rangle \pm \frac{1}{\sqrt{2}}|0\rangle + \frac{e^{i\phi}}{2}|-1\rangle\,, \tag{21}$$

and $|-1\rangle$, $|0\rangle$, $|+1\rangle$ the eigenstates of L_z in the subspace with $l = 1$.

This is an example where the unperturbed Hamiltonian maps an eigenspace of the interaction Hamiltonian at the initial time to the corresponding eigenspace of the interaction Hamiltonian at time t. In fact, the operator $L^2_{\phi(t)}$ has a twofold degenerate subspace corresponding to the eigenvalue 1 and a singlet corresponding to zero. This means that the two states $|-1\rangle_{\phi(t)}$ and $|+1\rangle_{\phi(t)}$ do not "feel" the interaction Hamiltonian except for the (same) phase accumulated, which is $e^{-i\int_0^t \alpha(s)ds}$.

It deserves to be noted that the examples in this section are such that the relevant interaction Hamiltonian in the interaction picture provides a commutative family of operators, i.e., it has the following form $\tilde{H}_\mathrm{I}(t) = f(t)\tilde{H}_\mathrm{I}(0)$. In fact, for spin-1/2 we have

$$\tilde{H}_\mathrm{I}(t) = \alpha(t)\begin{pmatrix} 0 & e^{-i\phi} \\ e^{i\phi} & 0 \end{pmatrix}\,, \tag{22}$$

and hence it has time-independent eigenvectors $|\pm\rangle_\phi$ and time-dependent eigenvalues $\pm\alpha(t)$. For spin-1 one finds:

$$\tilde{H}_\mathrm{I}(t) = \alpha(t)\begin{pmatrix} 1 & 0 & e^{-2i\phi(0)} \\ 0 & 2 & 0 \\ e^{2i\phi(0)} & 0 & 1 \end{pmatrix} \tag{23}$$

which has a "static" doublet corresponding to the eigenvalue 1.

IV. ADIABATIC EVOLUTIONS

Also adiabatic evolutions can provide interesting examples of interaction-free evolutions, though approximated. Consider the Hamiltonian of the class used for stimulated Raman adiabatic passage (STIRAP) [20–23]. The unperturbed Hamiltonian in the basis $|1\rangle$, $|2\rangle$, $|3\rangle$ reads

$$H_0(t) = \begin{pmatrix} 0 & \Omega\sin\theta(t) & 0 \\ \Omega\sin\theta(t) & \Delta & \Omega\cos\theta(t) \\ 0 & \Omega\cos\theta(t) & 0 \end{pmatrix}. \tag{24}$$

The three instantaneous eigenvalues of H_0 are given by

$$\lambda = 0\,,\ \frac{\Delta \pm \sqrt{\Delta^2 + 4\Omega^2}}{2}\,. \tag{25}$$

The instantaneous eigenstate corresponding to the zero eigenvalue reads

$$|v(t)\rangle = \cos\theta(t)|1\rangle - \sin\theta(t)|3\rangle\,. \tag{26}$$

In the adiabatic limit, assuming $\theta(0) = 0$ and $\theta(\infty) = \pi/2$, one has that the state $|1\rangle$ is adiabatically mapped into $|3\rangle$. This is the essence of the counterintuitive STIRAP sequence.

Consider now the following additional interaction term:

$$H_\mathrm{I}(t) = \epsilon(t)\begin{pmatrix} \cos^2\theta(t) & 0 & -\sin\theta(t)\cos\theta(t) \\ 0 & 0 & 0 \\ -\sin\theta(t)\cos\theta(t) & 0 & \sin^2\theta(t) \end{pmatrix}. \tag{27}$$

It consists of a direct interaction between the states $|1\rangle$ and $|3\rangle$ and two shifts of the levels involved in such an interaction.

The state $|v(t)\rangle$ is an instantaneous eigenstate of the interaction term, corresponding to the eigenvalue $\epsilon(t)$. Therefore, in the adiabatic limit associated to the change of $H_0(t)$, the state $|v(0)\rangle$ is mapped into $|v(t)\rangle$, which does not feel $H_\mathrm{I}(t)$, except for the accumulation of a dynamical phase.

Of course, in this case the result is only approximated, since the adiabatic evolution is only an approximation of the complete evolution induced by $H_0(t)$.

Similar to the examples given in the previous section, even in this example that we have provided for adiabatic evolutions, the eigenstates of $\tilde{H}_\mathrm{I}(t)$ do not change. Indeed, since $v|(t)$ is common instantaneous eigenstate of $H_0(t)$ and $H_\mathrm{I}(t)$, then it turns out that $|v(0)$ is eigenstate of $\tilde{H}_\mathrm{I}(t)$ at every time, corresponding to the eigenvalue $\epsilon(t)$, and the remaining subspace is the kernel of $\tilde{H}_\mathrm{I}(t)$, and then $\tilde{H}_\mathrm{I}(t) = \epsilon(t)/\epsilon(0)\tilde{H}_\mathrm{I}(0)$.

V. ESSENTIAL TIME DEPENDENCE OF $\tilde{H}_\mathrm{I}$

Since all the examples given in the previous sections are related to those cases where $\tilde{H}_\mathrm{I}(t)$ has a trivial time dependence, in this section we provide some examples of *real* time-dependent $\tilde{H}_{\rm I}(t)$ which have some time-independent eigenstates.

A. The multiphoton nonlinear JC model

The Hamiltonian

$$H(t) = \omega\hat{n} + \frac{\Omega}{2}\sigma_z + \gamma[e^{-i(\Omega-k\omega-\Delta)t} f(\hat{n})\hat{a}^k\sigma_+ + \text{H.c.}] \tag{28}$$

can be obtained, for example, in the physical scenario of trapped ions subjected to a laser slightly off-resonant to the kth red sideband ($\omega_L = \Omega - k\omega - \Delta$), out of the Lamb-Dicke limit (which implies the presence of the "coefficient" $f(\hat{n})$) and in the rotating wave approximation [24].

Taking

$$H_0 = \omega\hat{n} + \frac{\Omega}{2}\sigma_z\,, \tag{29a}$$

$$H_{\rm I}(t) = \gamma[e^{i(\Omega-k\omega-\Delta)t} f(\hat{n})\hat{a}^k\sigma_+ + \text{H.c.}]\,, \tag{29b}$$

one can easily prove that

$$\tilde{H}_{\rm I}(t) = \gamma[e^{-i\Delta t} f(\hat{n})\hat{a}^k\sigma_+ + \text{H.c.}]\,, \tag{30}$$

and that the multiplet $\{|0,g\rangle,\ |1,g\rangle,\ldots,|k-1,g\rangle\}$ (with $\sigma_z|g\rangle = -|g\rangle$) defines an eigenspace of $\tilde{H}_{\rm I}(t)$. Of course, it is not an eigenspace of H_0, which implies that, though it is interaction free, in this subspace there could be a nontrivial evolution due to the action of H_0.

Note that the interaction term in the interaction picture $\tilde{H}_{\rm I}(t)$ in this case is time dependent, though it has a time-independent eigenspace (its kernel).

B. Sum of multiphoton JC models

Also the following Hamiltonian can be obtained in trapped ions scenario:

$$H(t) = \omega\hat{n} + \frac{\Omega}{2}\sigma_z + [(\gamma_k(t)\hat{a}^k + \gamma_l(t)\hat{a}^l)\sigma_+ + \text{H.c.}]\,. \tag{31}$$

The time dependence of the coupling parameters γ's can be realized through a modulation of the amplitudes of the laser fields.

Let us assume that $k > l$. If $\gamma_l = 0$ then the kernel of the interaction Hamiltonian is generated by all the states $|m,g\rangle$ with $m = 0,1,\ldots,k-1$, while in the other case we have only the states with $m = 0,1,\ldots,l-1$. Therefore, in the case where $\gamma_l(t)$ changes and vanishes at some instants of time, the kernel of $H_{\rm I}$ changes, but some states always belong to it. Such states ($m \leqslant l-1$) and all their linear combinations undergo interaction-free evolution.

These two examples can be properly generalized considering, for example, $\Omega(t)$ instead of a time-independent Ω, in order to have a time-dependent H_0.

VI. DISCUSSION

In this paper we have generalized the concept of IFE to the case of time-dependent Hamiltonians. We have first of all provided necessary and sufficient conditions for such an occurrence. Then, we have presented several examples, related to different possible structures of the system under scrutiny. The very first examples (spin-1/2 and spin-1) analyze small quantum systems interacting with time-dependent classical fields. In particular, in the case of spin-1 we discuss the case where an IFE eigenspace is present (the doublet corresponding to angular momentum projections equal to -1 and $+1$). In the subsequent example we have considered IFE states in the presence of an adiabatic evolution, especially in the context of STIRAP. Finally, in Sec. V we have considered two cases of spin-boson interaction (for example the vibrational and electronic degrees of freedom of a trapped ion). In such a situation, we have two interacting subsystems, each one not feeling the interaction with the other, if the total system is prepared in suitable (IFE) states. Moreover, in one case, the IFE subspace has dimension varying in time.

On the basis of the analysis developed in Sec. II, we know that the more compact conditions to find out IFE subspaces is that IFE states are nothing but states which are eigenstates of the interaction-picture interaction Hamiltonian at every time instant, which really clarify the physical origin of the dynamical features of such states.

At this point, it is worth mentioning that the concept of IFE states (whether with time-independent or time-dependent Hamiltonian), when applied to a system interacting with its environment, has some connection with the concept of decoherence-free subspaces, as already pointed out in Ref. [1]. Nevertheless, reporting on a detailed analysis of the relation between IFE and DFS is beyond the scope of this paper and will be presented elsewhere.

ACKNOWLEDGMENTS

We thank the anonymous referee for valuable comments. D.C. was partially supported by the National Science Center Project No. DEC-2011/03/B/ST2/00136.

APPENDIX A

The evolution operator $U_0(t)$ associated to the unperturbed Hamiltonian $H_0(t)$ can be expanded as

$$\begin{aligned} U_0(t) &= \mathbb{I} - i\int_0^t H_0(t_1)dt_1 \\ &\quad + (-i)^2\int_0^t dt_1\int_0^{t_1} H_0(t_1)H_0(t_2)dt_2 + \cdots\,. \end{aligned} \tag{A1}$$

Thus,

$$\begin{aligned} &H_{\rm I}(t)U_0(t)|\psi_0\rangle \\ &\quad = H_{\rm I}(t)|\psi_0\rangle - i\int_0^t H_{\rm I}(t)H_0(t_1)dt_1|\psi_0\rangle \\ &\qquad + (-i)^2\int_0^t dt_1\int_0^{t_1} H_{\rm I}(t)H_0(t_1)H_0(t_2)dt_2|\psi_0\rangle + \cdots\,. \end{aligned} \tag{A2}$$

Starting from Eq. (A2) it is immediate to convince oneself that if $|\psi_0\rangle$ satisfies Eqs. (8) and (9) then

$$H_{\rm I}(t)U_0(t)|\psi_0\rangle = a(t)\,U_0(t)|\psi_0\rangle\,, \tag{A3}$$

that is, $|\psi_0\rangle$ is an IFE state.

APPENDIX B

The spin-1 operators are defined as follows:

$$L_x = \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \tag{B1a}$$

$$L_y = \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & -i & 0 \\ i & 0 & -i \\ 0 & i & 0 \end{pmatrix}, \tag{B1b}$$

$$L_z = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}. \tag{B1c}$$

The operator L_ϕ has eigenvalues $\{0,1,-1\}$ corresponding to the following eigenstates:

$$|0\rangle_\phi = \frac{e^{-i\phi}}{2}|+1\rangle - \frac{e^{i\phi}}{2}|-1\rangle, \tag{B2a}$$

$$|\pm1\rangle_\phi = \frac{e^{-i\phi}}{2}|+1\rangle \pm \frac{1}{\sqrt{2}}|0\rangle + \frac{e^{i\phi}}{2}|-1\rangle. \tag{B2b}$$

Its square,

$$L_\phi^2 = \frac{1}{2}\begin{pmatrix} 1 & 0 & e^{-2i\phi} \\ 0 & 2 & 0 \\ e^{2i\phi} & 0 & 1 \end{pmatrix}, \tag{B3}$$

has the same eigenstates and the following eigenvalues: 0 (singlet) and 1 (doublet).

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