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Entanglement in interference-based quantum control: the wave function is not enough

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We analyze the way entanglement affects features of interference-based quantum control. We show that quantum interferences vanishes in several cases in the process of extracting probabilities from wave functions. As an example, we discuss the loss of quantum interferences when tracing over *number* states of the radiation field. We also consider the way in which controllability is reduced when tracing over an entire manifold of states whose cumulative probability we wish to control. Finally, we show that it is impossible to control the relative populations of degenerate states (occurring, *e.g.*, in dissociation and chemical reactions) when the relevant transition amplitude is factorizeable, *i.e.*, when it can be written as a product of a purely classical field-dependent part and a purely material-dependent part. Differences between entanglement and non-factorizability of amplitudes are emphasized.

I. Introduction

It is well understood[1,2] that quantum interferences are responsible for active control over molecular processes, and that they are the single most important factor that distinguishes quantum control from classical control. For example, quantum interferences allow one to *completely* block off an undesired process,[3] thereby channelling the outcome to orthogonal subspaces with 100% probability.

It is often assumed that it is sufficient, in efforts to gain control, to attain final wave functions that are as close as possible to preset target states, a feat which is strongly dependent on quantum interferences.[4,5] In spite of this extremely useful strategy, achieving such a target wavefunction does not guarantee success in achieving desired product cross sections. This is because, in accord with quantum mechanics, predicting the probability of a given outcome entails squaring the wave function and summing (*tracing*) the result over all the events that contribute to the measured outcome. This tracing operation may well destroy interferences, thereby rendering the control-objective unattained, even though the final system wave function has been optimized to be in a very closely proximity of the target state.

In this paper we discuss several examples that show how tracing can nullify control. Of particular interest is the outcome of properly accounting for the *quantum* nature of the light source(s) used to navigate the system towards the target state. One finds that certain radiative states (such as number states) become entangled with a material system with which they interact. As a result, tracing over the final radiation states eliminates all the quantum interferences within the system, and control is lost.

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Another example where control is lost deals with situations, commonly encountered in *non-linear* optics, in which the relevant transition amplitudes are products of purely material ("response") entities, such as non-linear polarizabilities, and field dependent entities, such as some power of the radiation field. We show that under these circumstances, control over degenerate states (occurring for example in multi-channel photodissociation or branching chemical reactions, or indeed in any scattering process) cannot be attained.

II. Loss of control due to entanglement

A. The role of the radiative states

To illustrate the issues involved we focus on the 1-photon *vs.* 2-photon symmetry-breaking control scenario[6] and consider exciting a material system, initially in bound state $|E_g\rangle$, to the final dissociative state $|E, \mathbf{m}^-\rangle$, where E denotes the total energy and $\mathbf{m}$ all other quantum numbers ("channels"). For this example we use a radiation source composed of a two mode *number* state, $|N_\omega, N_{\omega/2}\rangle$, where $|N_\omega, N_{\omega/2}\rangle$ denotes $|0,\ldots, 0, N_\omega, 0,\ldots, 0, N_{\omega/2}, 0,\ldots\rangle$, a number state containing N_ω photons in the ω mode; $N_{\omega/2}$ photons in the $\omega/2$ mode; and zero photons in all the other modes. The initial state of the matter + radiation is then,

$$|\Psi^i\rangle = |N_\omega, N_{\omega/2}\rangle|E_g\rangle. \tag{1}$$

The final matter + radiation state is given (in second-order perturbation theory) as,

$$|\Psi^f\rangle = (2\pi i)^{\frac{1}{2}}\sum_{\mathrm{m}} |E, \mathbf{m}^-\rangle\Big\{\epsilon(\omega)\langle E, \mathbf{m}^- |\mathrm{d}|E_g\rangle|N_\omega - 1, N_{\omega/2}\rangle - \epsilon^2(\omega/2)$$

$$\times \langle E, \mathbf{m}^- |D|E_g\rangle|N_\omega, N_{\omega/2} - 2\rangle\Big\}, \tag{2}$$

where $\langle E, \mathbf{m}^- |\mathrm{d}|E_g\rangle$ is the one-photon transition-dipole matrix element, and $\langle E, \mathbf{m}^- |D|E_g\rangle$ is the two-photon matrix element, given as

$$\langle E, \mathbf{m}^- | D | E_g\rangle = \sum_j \langle E, \mathbf{m}^- | \mathrm{d} | E_j\rangle\langle E_j | \mathrm{d} | E_g\rangle\left[\hbar\omega/2 + E_g - i\epsilon - E_j\right]^{-1}. \tag{3}$$

The field amplitude $\epsilon(\omega)$ is related to N_ω by:

$$\epsilon(\omega) = i\left(\frac{\hbar\omega N_\omega}{\varepsilon_0 V}\right)^{\frac{1}{2}}\exp(i\omega z/c - i\phi_\omega), \tag{4}$$

with z denoting the axis of propagation of the light beams, ε_0 the permittivity of the vacuum, and V the cavity volume.

The ϕ_ω phase in $\epsilon(\omega)$ is an added (controllable) phase that we can vary by, *e.g.*, modifying the optical path transversed by the field. As discussed in the context of the classical-field version of this scenario,[6] one controls the process by varying the $\phi_\omega - 2\phi_{\omega/2}$ phase difference and the $|\epsilon(\omega)/\epsilon(\omega/2)|$ amplitude ratio. Of interest is the probability $P_{\mathbf{m}}(E)$ of observing the dissociation products in a particular state $\mathbf{m}$ in the distant future, given by

$$P_{\mathrm{m}}(E) \equiv \left|\langle E, \mathbf{m}^- |\psi^f\rangle\right|^2 = 2\pi\Big|\epsilon(\omega)\langle E, \mathbf{m}^- |\mathrm{d}|E_g\rangle|N_\omega - 1, N_{\omega/2}\rangle$$
$$- \epsilon^2(\omega/2)\langle E, \mathbf{m}^- |D|E_g\rangle|N_\omega, N_{\omega/2} - 2\rangle\Big|^2. \tag{5}$$

Varying the phase difference allows controls of the interference term arising in this expression.

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The difference between the above formula and that obtained when using classical light[6] is that here the radiative states become entangled with the material states. A result of the orthogonality between the radiative states and the entanglement is that if we *do* measure either of the radiative states then we need to compute either

$$\left|\langle N_\omega - 1, N_{\omega/2}|\langle E, \mathbf{m}^-|\psi^f\rangle\right|^2 = 2\pi\left|\epsilon(\omega)\langle E, \mathbf{m}^-|\mathrm{d}|E_g\rangle\right|^2, \tag{6}$$

or

$$\left|\langle N_\omega, N_{\omega/2} - 2|\langle E, \mathbf{m}^-|\psi^f\rangle\right|^2 = 2\pi\left|\epsilon^2(\omega)\langle E, \mathbf{m}^-|D|E_g\rangle\right|^2. \tag{7}$$

Note that neither probability expressions contain any interference contribution. Furthermore, if we *do not* measure the radiative state explicitly then the observed result is obtained by summing (tracing over) these probabilities to obtain that

$$\begin{aligned}\overline{P}_{\mathbf{m}}(E) &\equiv \left|\langle N_\omega - 1, N_{\omega/2}|\langle E, \mathbf{m}^-|\Psi^f\rangle\right|^2 + \left|\langle N_\omega, N_{\omega/2} - 2|\right|\langle E, \mathbf{m}^-|\Psi^f\rangle\Big|^2 \\ &= |\epsilon(\omega)\langle E, \mathbf{m}^-|\mathrm{d}|E_g\rangle|^2 + |\epsilon^2(\omega/2)\langle E, \mathbf{m}^-|D|E_g\rangle|^2\end{aligned} \tag{8}$$

in which there are no interference terms either. We therefore conclude that coherent control cannot take place when the initial radiative states are *number states*. This property is not, as one might pre-suppose, the result of the indeterminacy of the electric field phase associated with number state. Rather, it is a result of the entanglement between the number states and matter states and the fact that the number states are orthogonal to one another.

Note that these results are not restricted to the one *vs.* two control scenario. As shown by Gong *et al.*,[7] similar conclusions hold for other coherent control scenarios as well.

B. Entanglement in one photon dissociation

The above results, pertaining to the way entanglement eliminates interferences, are not restricted to quantum fields. Another example is ordinary multichannel photodissociation in the weak field domain. In this case we irradiate a molecule with weak cw light and the excited molecule breaks apart into a number of energetically accessible (open) internal fragment states **m** ("channels"). Such a process creates a superposition of all the open channels:

$$|\Psi^f\rangle \xrightarrow{t\to\infty} (2\pi i)^{1/2} \sum_{\mathbf{m}} |e_{\mathbf{m}}\rangle|k_{\mathbf{m}}\rangle\epsilon(\omega)\langle E, \mathbf{m}^-|\mathrm{d}|E_g\rangle, \tag{9}$$

where $e_{\mathbf{m}}$ is the internal energy of the photofragments and $k_{\mathbf{m}}$ is the momentum associated with their relative translational motion. The latter quantity is fixed by energy conservation

$$E = E_g + \hbar\omega = \hbar^2 k_{\mathbf{m}}^2/(2\mu) + e_{\mathbf{m}}. \tag{10}$$

Hence, a measurement of $e_{\mathbf{m}}$ fixes $k_{\mathbf{m}}$, and *vice versa*. Thus, energy conservation *entangles* the translational motion with the internal states of the fragments. A result of this entanglement is that although Ψ^f is a superposition of many dissociation channels, no interferences between these channels can be observed.[7,8] It is only when the bandwidth of the dissociating laser *exceeds* an internal energy difference $|e_{\mathbf{m}} - e_{\mathbf{n}}|$ that we can begin to see interferences between the $|k_{\mathbf{m}}\rangle$ and $|k_{\mathbf{n}}\rangle$ translational states.[8] This argument solidifies the well known prescription of scattering theory that the *total* cross section is the sum of the individual scattering amplitudes-squared, rather than the square of the sum of the scattering amplitudes.

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C. Regaining control *via* the use of coherent states

The situation is dramatically different than that in Sect. II A if the radiation field is comprised of a product of coherent states, $|\alpha_\omega\rangle$, defined as,

$$\hat{a}_\omega|\alpha_\omega\rangle = \alpha_\omega|\alpha_\omega\rangle. \tag{11}$$

In this case, the initial matter + radiation state associated with the one *vs.* two photons control scenario is

$$|\Psi^i\rangle = |\alpha_\omega, \alpha_{\omega/2}\rangle|E_g\rangle. \tag{12}$$

Because $|\alpha_\omega\rangle$ is an eigenstate of the annihilation operator, which is the only part of the quantum field operator that contributes in the rotating wave approximation, the absorption of one photon from an $|\alpha_\omega\rangle$ state results in the production of $\epsilon(\omega)|\alpha_\omega\rangle$, *i.e.* the initial coherent state multiplied by the field amplitude:

$$\epsilon(\omega) = i\left(\frac{\hbar\omega}{\epsilon_0 V}\right)^{\frac{1}{2}}\alpha_\omega \ \exp(i\omega z/c - i\phi_\omega). \tag{13}$$

Hence, the final matter + radiation state can be written as,

$$|\Psi^f\rangle = (2\pi i)^{\frac{1}{2}}\sum_{\mathbf{m}}|E,\mathbf{m}^-\rangle\left\{\epsilon(\omega)\langle E,\mathbf{m}^-|\mathrm{d}|E_g\rangle - \epsilon^2(\omega/2)\langle E,\mathbf{m}^-|D|E_g\rangle\right\}|\alpha_\omega,\alpha_{\omega/2}\rangle. \tag{14}$$

The resultant wave function $|\Psi^f\rangle$ is seen to be completely unentangled. That is, contrary to the number state situation discussed above, the coherent field states do not get entangled with the material system. Hence the interference terms do not vanish when we trace-over the radiative states. In other words, the measurement of the final radiation states (or lack thereof) collapses this final state to the material superposition of eqn (14), allowing for quantum control. Hence, coherent control with coherent states is seen to be essentially identical to the classical-field scenario, in keeping with the fact that coherent states are known to be the closest quantum states to classical electromagnetic fields.[9]

III. Reduced interference in the control of a *manifold* of states

Another example where the control objective cannot be expressed as that of attaining a particular final *wave function* is the case analyzed early on[3] in which one attempts to achieve control over a *manifold* of states. In this case as well we need to trace over states, but here it is not done with respect to unobserved states of the bath or radiation field. Rather the trace arises as the sum over all final states of interest. As a result, the control objective in this case is to maximize or minimize a *cumulative probability* given as the sum over many states:

$$P_q(E) = \sum_{\mathbf{m}} P_{q,\mathbf{m}}(E), \tag{15}$$

where q denotes the multi dimensional manifold to which we wish to navigate the system.

As an example of this situation, consider bichromatic control attained by irradiating a system with two cw fields given as:

$$\varepsilon(t) = 2\sum_{i=1,2}\hat{\epsilon}_i|\epsilon(\omega_i)|\cos[\omega_i t - \phi(\omega_i)] = 2\sum_{i=1,2}\hat{\epsilon}_i Re[\epsilon(\omega_i)\exp(-i\omega_i t)]. \tag{16}$$

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Here the system is initially in a superposition of two bound states,

$$\chi = a_1|E_1\rangle + a_2|E_2\rangle. \tag{17}$$

Tuning the frequencies ω_1 and ω_2 such that $\omega_2 - \omega_1 = (E_1 - E_2)/\hbar$, we have that $P_q(E)$, the cumulative probability at energy $E = E_1 + \hbar\omega_1 = E_2 + \hbar\omega_2$, is given as

$$P_q(E) = 2\pi\left\{|a_1|^2|\epsilon(\omega_1)|^2 \mathrm{d}_q(22) + 2Re\left[a_1 a_2{}^* \epsilon(\omega_1)\epsilon^*(\omega_2)\mathrm{d}_q(12)\right]\right\}, \tag{18}$$

where

$$\mathrm{d}_q(ij) \equiv \sum_{\mathbf{m}} \langle E_i \,|\, \mathrm{d} \,|E, q, \mathbf{m}^-\rangle\langle E, q, \mathbf{m}^- \,|\, \mathrm{d} \,|E_j\rangle, \quad i,j = 1,2. \tag{19}$$

Expressing each complex dipole amplitude as its absolute value times a phase factor,

$$\langle E_i|\mathrm{d}|E, q, \mathbf{m}^-\rangle \equiv |\langle E_i|\mathrm{d}|E, q, \mathbf{m}^-\rangle|\exp(i\alpha^{(1)}_{q,\mathbf{m}}), \tag{20}$$

exposes the $\mathrm{d}_q(12)$ interference term as

$$\mathrm{d}_q(12) = \sum_{\mathbf{m}} |\langle E_1 \,|\, \mathrm{d} \,|E, q, \mathbf{m}^-\rangle\langle E, q, \mathbf{m}^- \,|\, \mathrm{d} \,|E_2\rangle| \, \exp(i\alpha^{(1)}_{q,\mathbf{m}} - i\alpha^{(2)}_{q,\mathbf{m}}). \tag{21}$$

As more and more $\mathbf{m}$ terms are added to the sum we expect the oscillatory $\exp(i\alpha^{(1)}_{q,\mathbf{m}} - i\alpha^{(2)}_{q,\mathbf{m}})$ terms to average out, thereby reducing the magnitude of the interference term. In the classical limit the size of the q manifold, and hence the number of $\mathbf{m}$ terms, becomes very large. In that limit, if the $\alpha^{(i)}_{q,\mathbf{m}}$ phases vary in an essentially random fashion, the interference term vanishes. Thus, the desire to control the population of an *entire* manifold, q, can result in a highly reduced interference term and the reduction of our ability to control the process, even when optimization procedures are nonetheless capable of attaining a single target wave function.

IV. Non-factorizability as a prerequisite for degenerate state control

Although, as discussed above, entanglement between the radiation field states and matter states can result in loss of control, the inability to factorize the wavefunction into a product of matter and radiation terms is sometimes necessary for control.

To appreciate the difference[10] between "non-factorizable" and "entangled", consider eqn (14). This wavefunction is an *unentangled product* of a matter state $|E, \mathbf{m}^-\rangle$ and a state of the radiation field $|\alpha_\omega, \alpha_{\omega/2}\rangle$. However, the field and matter dependence is given by

$$\left\{\epsilon(\omega)\langle E, \mathbf{m}^-|\mathrm{d}|\rangle E_g - \epsilon^2(\omega/2)\langle E, \mathbf{m}^-|\mathrm{D}|E_g\rangle\right\}, \tag{22}$$

which is a *non-factorizable* product of matter and field components. It is this last feature that will be shown necessary for control in the cases discussed below. Specifically, we now show that in one such case, the control of the branching ratios between *degenerate* states, a situation common to all multichannel dissociation and scattering phenomena in general, non-factorizability is essential for interference-based control.[11]

This statement is an extension of the well known result [12] that it is impossible to control the branching ratio between two dissociation channels, and indeed all processes involving degenerate final states, using weak field one-photon transitions. It is also an extension of the (possibly less well known) result [1,2] that it is impossible to control the branching ratio between degenerate final states even in the strong field domain, if the process involves the net absorption of one photon from a *single* initial ("precursor") state.

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The prevailing opinion[13,14] was, in fact, that the above conclusions do not hold in the (non-linear) N-photon absorption case. This perspective was supported by a series of seminal experiments[15–19] that showed that the use of coherent control techniques in the short pulse realization of N-photon processes, whether performed *via* intermediate resonances or not, allows one to control the transitions to *non-degenerate* states.

Here we show that the situation concerning the control over *degenerate* final states is different: If the relevant amplitudes can be factorized into radiative and material parts, no control over branching ratios of degenerate states can be attained, even in the non-linear N-photon absorption case. That is, the mere existence of two or more interfering pathways to the same final states,[1,2] though *necessary* for quantum control, is not *sufficient*. Rather, in addition to this necessary condition, one must also satisfy certain conditions on the radiative states, as discussed above, and the non-factorizability condition, to be discussed below.

In order to see why non-factorizability is necessary, we utilize N-order perturbation theory to write the N-photon transition amplitude between the initial state $|E_g\rangle$ and a final state $|E_f\rangle$, where the former is subject to the action of a broad-band pulse of light as,

$$b_{E_f}^{(N)} = \int \cdot\cdot \int (\Pi_{j=1}^{N-1} d\omega_j)\epsilon(\omega_N) \times \sum_{i_1,\ldots,i_{N-1}} \langle E_f|\mathrm{d}|E_{i_{N-1}}\rangle \Pi_{k=1}^{N-1} \frac{\epsilon(\omega_k)\langle E_{i_k}|\mathrm{d}|E_{i_{k-1}}\rangle}{\sum_{j=1}^{k}\hbar\omega_j - (E_{i_k} + \Delta_{i_k} - i\Gamma_{i_k}/2 - E_g)}, \quad (23)$$

where $\omega_N = \omega_{f,g} - \sum_{j=1}^{N-1}\omega_j$, $i_0 = g$, with each i_k, $k = 1,\ldots, N-1$, ranges over all the intermediate states. In the absence of intermediate resonances, *e.g.* when $|\omega_{f,g}| \ll |E_i - E_g|/\hbar$ for all $i \neq f$ or g, we can replace the term $\sum_{j=1}^{k}\hbar\omega_j - (E_{i_k} + \Delta_{i_k} - i\Gamma_{i_k}/2 - E_g)$ in the denominator with a constant term $\hbar\omega\bar{\omega} - (E_{i_k} + \Delta_{i_k} - i\Gamma_{ik}/2 - E_g)$, and write that

$$b_{E_f}^{(N)} = \langle E_f|M^{(N)}|E_g\rangle \int \ldots \int (\Pi_{j=1}^{N-1} d\omega_j)\Pi_{k=1}^{N}\epsilon(\omega_k) \quad (24)$$

where

$$\langle E_f|M^{(N)}|E_g\rangle \equiv \sum_{i_1,\ldots,i_{N-1}} \langle E_f|\mathrm{d}|E_{i_{N-1}}\rangle \Pi_{k=1}^{N-1} \frac{\langle E_{i_k}|d|E_{i_{k-1}}\rangle}{\hbar\overline{\omega} - (E_{i_k} + \Delta_{i_k} - i\Gamma_{i_k}/2 - E_g)}.$$

Using the fact that the Fourier transform of a product of functions is the convolution integral of their respective Fourier transforms,

$$\int_{-\infty}^{\infty} dt\varepsilon'(t)\varepsilon''(t)\exp(-i\omega t) = 2\pi \int_{-\infty}^{\infty} d\omega'\epsilon'(\omega')\epsilon''(\omega - \omega'), \quad (25)$$

we obtain, by repeated application of eqn (25), that $b^{(N)}{}_{E_f}$ in eqn (24) can be written as,

$$b_{E_f}^{(N)} = \frac{\langle E_f|M^{(N)}|E_g\rangle}{(2\pi)^{N-1}} \int dt\varepsilon^N(t)\exp(-i\omega_{f,g}t). \quad (26)$$

It follows from eqn (26) that it is possible to control the strength of a general N-photon transition between *non-degenerate* states, by essentially using variants of the pulse modulations of the $N = 2$ case.[20] However, it also follows from eqn (26) that if the final state is energetically *degenerate* it is not possible to use N-photon processes to optically control the *relative* populations of its components in the absence of

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intermediate resonances. In the continuum case this means that such N-photon processes cannot be used to control *branching ratios* to different scattering channels of the same total energy.

This extension of the *one-photon* result of ref. 12 follows because in the absence of intermediate resonances the transition amplitude $b^{(N)}{}_{E_f}$ factorizes into a product of a material part $\langle E_f|M^{(N)}|E_g\rangle$ and a radiative part $\int dt\varepsilon^N(t)\exp(-i\omega_{f,g}t)$. Since the only information regarding the final state appearing in the radiative part is its energy (contained in the $\omega_{f,\,g}$ factor), there is no way that the shape of the pulse can affect the populations of different states with the *same* energy.

We can extend this result to the strong field domain where N-order perturbation theory is not valid by including the time dependent initial coefficient in our expression. In particular, in a multi-channel photodissociation process, in the absence of an intermediate resonance, we can write the non-perturbative amplitude at time t for observing channel $\mathbf{n}$ in the far future as,

$$b^{(N)}_{E,\mathbf{n}}(t) = \frac{\langle E,\mathbf{n}^-|M^{(N)}|E_g\rangle}{(2\pi)^{N-1}}\int_{-\infty}^{t} dt'\varepsilon^N(t')\exp(-i\omega_{E,g}t')b_{E_g}(t'), \tag{27}$$

where $b_{E_g}(t)$ is the amplitude of the initial state, and

$$\langle E,\mathbf{n}^-|M^{(N)}|E_g\rangle = \sum_{i_1,\ldots,i_{N-1}}\langle E,\mathbf{n}^-|\mathrm{d}|E_{i_{N-1}}\rangle\Pi_{k=1}^{N-1}\frac{\langle E_{i_k}|\mathrm{d}|E_{i_{k-1}}\rangle}{\hbar\bar{\omega}-(E_{i_k}+\Delta_{i_k}-i\Gamma_{i_k}/2-E_g)}, \tag{28}$$

where $|E_{i_0}\rangle \equiv |E_g\rangle$. We see that the branching ratio between different final channels is given as

$$R_{\mathbf{n},\mathbf{m}}(t) \equiv \left|\frac{b^{(N)}_{E,\mathbf{n}}(t)}{b^{(N)}_{E,\mathbf{m}}(t)}\right|^2 = \left|\frac{\langle E,\mathbf{n}^-|M^{(N)}|E_g\rangle}{\langle E,\mathbf{m}^-|M^{(N)}|E_g\rangle}\right|^2, \tag{29}$$

and the pulse attributes, as well as the detailed evolution history of the initial state coefficient $b_{E_g}(t')$, have completely disappeared! Thus it is not possible, by manipulating the laser field(s), to control the branching ratio into different dissociation channels in a fixed-N multi-multi-photon process which starts with only one precursor state.

As discussed in ref. 1, 2, the remedy in this case is to interfere N photon processes with $L \neq N$ photon processes, both leading to the same final state $|E,\mathbf{n}^-\rangle$. In this case the transition amplitude is given in a field-matter *non-factorizable* form,

$$b_{E,\mathbf{n}}(t) \;=\; b^{(N)}_{E,\,\mathbf{n}}(t) \;+\; b^{(L)}_{E,\mathbf{n}}(\mathrm{t}) \;=$$

$$\left\{\frac{\langle E,\mathbf{n}^-|M^{(N)}|E_g\rangle}{(2\pi)^{N-1}}\int_{-\infty}^{t} dt'\varepsilon^N(t') + \frac{\langle E,\mathbf{n}^-|M^{(L)}|E_g\rangle}{(2\pi)^{L-1}}\int_{-\infty}^{t} dt'\varepsilon^L(t')\right\}e^{-i\omega_{E,g}t'}b_{E_g}(t'). \tag{30}$$

Alternatively we can start from two ($|E_1\rangle$ and $|E_2\rangle$) or more precursor states, in which case

$$b_{E,\mathbf{n}}(t) = \left(\frac{1}{2\pi}\right)^{N-1}\{\langle E,\mathbf{n}^-|M^{(N)}|E_1\rangle\int_{-\infty}^{t} dt'\varepsilon_1^N(t')e^{-i\omega_{E,1}t'}b_{E_1}(t') + \langle E,\mathbf{n}^-|M^{(N)}|E_2\rangle\int_{-\infty}^{t} dt'\varepsilon_2^N(t')e^{-i\omega_{E,2}t'}b_{E_2}(t')\}. \tag{31}$$

Controllability over degenerate states can also be attained if there exists an intermediate (M-photon) resonance, because in that case the photon variables and the

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material factors are also non-factorizable. Specifically, in the case of an intermediate resonance we have that

$$b_{E_f}^{(N)} = \left\langle E, \mathbf{n}^- | M'^{(N)} | E_g \right\rangle \int \cdot \cdot \int (\Pi_{j=1}^{N-1} d\omega_j) \Pi_{k=1}^{N} \epsilon(\omega_k) + \int \cdot \cdot \int (\Pi_{j=1}^{N-1} d\omega_j) \epsilon(\omega_N) \sum_{i_1,\ldots,i_{M-1},i_{M+1},\ldots,i_{N-1}} \langle E_f | \mathrm{d} | E_{i_{N-1}} \rangle$$

$$\frac{\epsilon(\omega_M) \langle E_{i_M} | \mathrm{d} | E_{i_{M-1}} \rangle}{\sum_{j=1}^{M} \hbar\omega_j - (E_{i_M} + \Delta_{i_M} - i\Gamma_{i_M}/2 - E_g)} \cdot \Pi_{k\neq M}^{N-1} \frac{\epsilon(\omega_k) \langle E_{i_k} | \mathrm{d} | E_{i_{k-1}} \rangle}{\sum_{j=1}^{k} \hbar\omega_j - (E_{i_k} + \Delta_{i_k} - i\Gamma_{i_k}/2 - E_g)}, \quad (32)$$

where

$$\langle E, \mathbf{n}^- | M'^{(N)} | E_g \rangle = \sum_{i_1,\ldots,i_{M-1},i_M \neq i'_M,i_{M+1},\ldots,i_{N-1}} \langle E, \mathbf{n}^- | \mathrm{d} | E_{i_{N-1}} \rangle \Pi_{k=1}^{N-1} \frac{\langle E_{i_k} | \mathrm{d} | E_{i_{k-1}} \rangle}{\hbar\overline{\omega} - (E_{i_k} + \Delta_{i_k} - i\Gamma_{i_k}/2 - E_g)}$$

V. Conclusions

In this paper we have shown that, in certain cases, attaining a particular target wave function is insufficient for control. For example, when entanglement is involved, the need to sum probabilities over all events that contribute to a given outcome, whether such events are measured explicitly or not, can destroy all interferences, making interference-based control impossible. Several examples were given: the vanishing of quantum interferences when material states are entangled with number states, and the reduction in controllability when control of an entire manifold of states is desired. An alternate example, where field-matter non-factorizability (as distinct from entanglement) is essential for exercising control over degenerate states was also discussed.

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