

Optimally driving multiphoton transitions in the perturbative single-mode regime

Frieder Lindel^{1,2,*}, Stefan Yoshi Buhmann³, Andreas Buchleitner^{2,4,‡} and Edoardo G. Carnio^{2,4,§,||}

¹*Departamento de Física Teórica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, E-28049 Madrid, Spain*

²*Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Straße 3, D-79104 Freiburg, Germany*

³*Institut für Physik, Universität Kassel, Heinrich-Plett-Straße 40, 34132 Kassel, Germany*

⁴*EUCOR Center for Quantum Science and Quantum Computing, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Straße 3, D-79104 Freiburg, Germany*



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The rate of m -photon transitions in matter, induced by an incident light field, depends on the field's m th-order coherence function. Consequently, the coherence properties of the light field may be shaped to increase the rate of multiphoton transitions. Here, we determine the optimal state of a weak fixed-intensity, narrow-band incident light field, with a restricted maximal photon number, that optimally drives m -photon transitions in the case of a short-lived atomic multilevel system. We show that, in this case, no quantum properties of the light field need to be exploited, but that classical mixtures of coherent states are optimal.

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I. INTRODUCTION

The control of quantum systems lies at the heart of many quantum technologies. Standard control is achieved by using classical light fields, whose amplitude, polarization, frequencies, and relative phases are used as control knobs [1]. Improving the efficiency of such control protocols is crucial to avoid, e.g., decoherence during a targeted unitary evolution, or other side effects such as heating or photodamage in the sample.

To enhance the efficiencies of different control protocols, it has been suggested to harness the quantum properties of the control field [2]. A paradigmatic example is given by multiphoton absorption processes in the weak light-matter coupling regime. Here, only the lowest nonvanishing order of perturbation theory in the light-matter coupling has to be considered [3]. It was found theoretically [4–7] and experimentally [8,9] that, in this regime, light with strong number fluctuations, such as squeezed or thermal states of light, can drive multiphoton absorption processes in free space more efficiently than quasiclassical coherent states with the same intensity. If multimode control fields are employed, two frequency-entangled photons can induce two-photon transitions more

efficiently than any classically correlated photon pair [10–13]. The implications of these enhanced control protocols have been discussed for a range of different applications, such as spectroscopy [2,14,15], quantum imaging [16], or quantum sensing [17,18]. More recently, the potential of quantum states of light in case the control field is strongly coupled to the target system, as happens, e.g., in a cavity, [19,20] or in strongly driven systems [21,22], have been analyzed.

In our present contribution, we consider the following question: Which is the *optimal* initial state of a single-mode field to drive an m -photon absorption process in the weak (perturbative) light-matter coupling regime? By fixing the average number of photons in the (quantized) field, and optimizing over a finite-dimensional part of its Hilbert space, we find that the optimal initial field states can be very well approximated by classical mixtures of coherent states. This implies that no quantum feature is needed to induce maximal enhancement of single-mode-driven m -photon absorption rates in the perturbative regime, as realized in, e.g., [8,9], but classical mixtures of quasiclassical coherent states are sufficient. The origin of the enhancement is traced back to the nonlinear scaling of the m -photon transition rate with the number of photons in the light field.

II. MULTIPHOTON ABSORPTION PROCESSES

We consider a single atom with dipole operator $\hat{d}$ that is driven by a narrow-band field $\hat{E}(t)$ with central frequency ω_0 and bandwidth $\Delta\omega \ll \omega_0$. The light-matter interaction Hamiltonian is given by

$$\hat{H}_I = \hat{d}(t)[\hat{E}(t) + \hat{E}^\dagger(t)], \quad (1)$$

where $\hat{E}(t)$ denotes the positive frequency part of the field propagating in a single direction with cross section A , which is given in terms of creation and annihilation operators $\hat{a}^{(\dagger)}(\omega)$

*Contact author: flindel@phys.ethz.ch

†Present address: Institute for Theoretical Physics, ETH Zürich, Zürich 8093, Switzerland.

‡Contact author: andreas.buchleitner@physik.uni-freiburg.de

§Contact author: e.carnio@neqxt.org

||Present address: neQxt, Hansaring 12, D-50670, Cologne, Germany.

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via [23]

$$\hat{E}(t) = i \int_0^\infty d\omega \left(\frac{\hbar\omega}{4\pi c \epsilon_0 A} \right)^{1/2} \hat{a}(\omega) e^{-i\omega t}. \quad (2)$$

We assume that the atom is initially in the ground state $|G\rangle$, with energy ω_G , and there is a single atomic level $|F\rangle$, with energy ω_F , which is resonant with an m -photon transition such that $\omega_F = \omega_G + m\omega_0$. While other discrete atomic levels may be present, those do not matter for the discussion that follows [5], and we therefore do not specify them further. For $m = 2$, one finds that the probability $p_F(t)$ of finding the system in the state $|F\rangle$ at times $t \gg (\Delta\omega)^{-1}$ (i.e., after the driving field has faded out), is given, in lowest-order perturbation theory in $\hat{H}_I$, by [5]

$$p_F(t) = 2t g' \int dt' e^{2i\omega_F t' - \gamma_F |t'|} G^{(2)}(-t', -t', t', t'). \quad (3)$$

Here, g' is a coupling constant (which encodes the electronic structure of the atom), and we have defined $G^{(2)}(-t, -t, t, t) = \langle \hat{E}^\dagger(-t) \hat{E}^\dagger(-t) \hat{E}(t) \hat{E}(t) \rangle$. Moreover, $\langle \cdot \cdot \cdot \rangle$ indicates the expectation value with respect to the state of the field. If the lifetime of the target state $|F\rangle$ is much shorter than the coherence time of the field—or equivalently, if the linewidth γ_F of level $|F\rangle$ greatly exceeds the field bandwidth, $\gamma_F \gg \Delta\omega$ [5]—the atomic system in state $|F\rangle$ decays on timescales much shorter than the pulse coherence time. In this regime, p_F therefore only depends on $G^{(2)}(0, 0, 0, 0) \equiv G^{(2)}$, and Eq. (3) simplifies to [5,7]

$$p_F(t) \propto G^{(2)} t. \quad (4)$$

Here, $G^{(2)}$ is the equal-time, normally ordered, unnormalized second-order coherence function of the field,

$$G^{(2)} = \langle (\hat{a}^\dagger)^2 \hat{a}^2 \rangle, \quad (5)$$

where $\hat{a}$ is the annihilation operator of the field mode with frequency ω_0 . Similarly, one finds that the probability for an m -photon transition is proportional to the m th-order coherence function $G^{(m)}$ [4,24–26], defined by

$$G^{(m)} = \langle (\hat{a}^\dagger)^m \hat{a}^m \rangle. \quad (6)$$

All following results in this contribution are based on Eq. (4), and thus apply whenever this equation is valid. As we have just discussed, this is the case for a short-lived atomic multilevel system that is driven by a narrow-band field.

III. OPTIMAL CONTROL OF MULTIPHOTON ABSORPTION PROCESSES

To find the optimal field state that maximizes the m -photon transition probability, one has to maximize the normally ordered, m th-order coherence function $G^{(m)}$ of the field over all possible single-mode quantum field states. However, $G^{(m)}$ is, in general, not bounded from above. For example, for coherent states of light, one has $G^{(m)} \propto (n_{\text{av}})^m$ with the average number of photons $n_{\text{av}} = \langle \hat{a}^\dagger \hat{a} \rangle$. To find the optimally tailored quantum statistics that drive the m -photon transition with the highest efficiency, we therefore restrict our discussion to field states with fixed n_{av} . This constraint still does not bound $G^{(m)}$ from above in general. As shown in Ref. [27], such a bound exists if we restrict the infinite-dimensional Hilbert space of

the single-mode field to a subspace $\mathcal{H}_{N_{\text{max}}}$ spanned by all Fock states with at most a finite maximal number of photons N_{max} . The maximal value of $G^{(2)}$ and $G^{(m)}$ is then given by [27]

$$G^{(2)} \leq n_{\text{av}}(N_{\text{max}} - 1), \quad (7)$$

$$G^{(m)} \leq \frac{(N_{\text{max}} - 2)!}{(N_{\text{max}} - m)!} G^{(2)}, \quad (8)$$

and is attained by so-called coin states [27]

$$|\text{coin}\rangle = \frac{1}{\sqrt{N_{\text{max}}}} (\sqrt{N_{\text{max}} - n_{\text{av}}} |0\rangle + e^{i\phi} \sqrt{n_{\text{av}}} |N_{\text{max}}\rangle). \quad (9)$$

Note that the value of $G^{(2)}$ is independent of the relative phase ϕ between the states $|N_{\text{max}}\rangle$ and $|0\rangle$ in Eq. (9). These coin states are the states with the broadest photon number distribution around the average value n_{av} , which still belong to the Hilbert space with maximally N_{max} photons. Consequently, they exhibit the largest possible fluctuations of the average photon number, intuitively explaining their maximization of $G^{(2)}$, which is known to quantify photon number fluctuations [28].

Which is the increase in efficiency achieved when using coin states instead of coherent states $|\alpha\rangle$ with average photon number $|\alpha|^2 = n_{\text{av}}$? To answer this question, we consider the ratio

$$\frac{G_{\text{coin}}^{(m)}}{G_{\text{coh}}^{(m)}} = \frac{(N_{\text{max}} - 1)!}{(N_{\text{max}} - m)! n_{\text{av}}^{m-1}} \approx \left(\frac{N_{\text{max}}}{n_{\text{av}}} \right)^{m-1}, \quad (10)$$

where we defined $G_{\text{coh}}^{(m)} = \langle \alpha | (\hat{a}^\dagger)^m \hat{a}^m | \alpha \rangle$, $G_{\text{coin}}^{(m)} = \langle \text{coin} | (\hat{a}^\dagger)^m \hat{a}^m | \text{coin} \rangle$, and assumed $N_{\text{max}} \gg m$ to obtain the approximate result.

Therefore, by combining the results of Ref. [27] on maximizing correlation functions on a constrained Hilbert space with the well-known scaling of multiphoton absorption rates in Eq. (6) [4,5,24–26], we have identified coin states as the optimal states to drive m -photon absorption processes in the single-mode weak-coupling limit considered here, with enhancement factor $(N_{\text{max}}/n_{\text{av}})^{m-1}$ of the absorption rate, in comparison to coherent states with the same average number of photons. Generating coin states requires the preparation of a Fock state with a large photon number, which is a formidable experimental challenge. As we show in the following, however, the same advantage can also be achieved by a classical mixture of coherent states, if $N_{\text{max}} \gg 1$.

First, we notice that, by its very definition (6), $G^{(m)}$ does not depend on the coherences (off-diagonal elements of the density matrix) of the state of the single-mode field in the Fock basis. Consequently, the same value of $G^{(m)}$, and thus the same efficiency in driving m -photon absorption processes, is obtained if, instead of the coherent superposition as given by the coin state (9), one uses the classical mixture

$$\hat{\rho}_{\text{coin}} = \frac{1}{N_{\text{max}}} [(N_{\text{max}} - n_{\text{av}}) |0\rangle \langle 0| + n_{\text{av}} |N_{\text{max}}\rangle \langle N_{\text{max}}|]. \quad (11)$$

Next, we further approximate $|N_{\text{max}}\rangle$ in the coin state $\hat{\rho}_{\text{coin}}$ by coherent states $|\alpha_{N_{\text{max}}}\rangle$ with $|\alpha_{N_{\text{max}}}|^2 = N_{\text{max}}$ (note that,

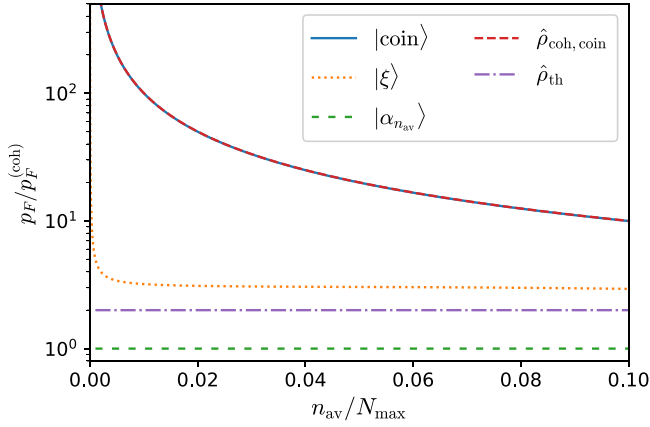


FIG. 1. Two-photon absorption rates. Probability p_F of a two-photon transition, as a function of the average number n_{av} of photons initially in the field. We consider different initial states of the light field: the coin state $|\text{coin}\rangle$, Eq. (9); the coin state approximated by a mixture of coherent states $\hat{\rho}_{\text{coin,coh}}$, Eq. (12); a squeezed vacuum state $|\xi\rangle = e^{[\xi^* \hat{a}^2 + \xi(\hat{a}^\dagger)^2]/2}|0\rangle$ determined by the squeezing parameter ξ ; a thermal state $\hat{\rho}_{\text{th}} = e^{-\hbar\omega \hat{a}^\dagger \hat{a}/k_B T} / \text{tr}\{e^{-\hbar\omega \hat{a}^\dagger \hat{a}/k_B T}\}$ with Boltzmann constant k_B and temperature T , for which $G^{(2)}/G_{\text{coh}}^{(2)} = 2$, $\forall T$; a coherent state $|\alpha_{n_{av}}\rangle$ with $|\alpha_{n_{av}}|^2 = n_{av}$. For $|\xi\rangle$, we obtained $G^{(2)}$ numerically for $\xi \in [0, 2.65]$, which leads to the values of $n_{av} = \sinh^2(|\xi|)$ shown in the Figure. If $\xi \gg 1$, we find $G^{(m)}/G_{\text{coh}}^{(m)} \approx (2m-1)!!$ [30], i.e., $G^{(2)}/G_{\text{coh}}^{(2)} \approx 3$. We restrict n_{av} to a regime in which all considered field states still lie within the restricted Hilbert space $\mathcal{H}_{N_{\text{max}}}$, with $N_{\text{max}} = 500$ (they have therefore negligible overlap with Fock states $|n\rangle$ with $n > N_{\text{max}}$).

formally, $|\alpha_{N_{\text{max}}}\rangle \notin \mathcal{H}_{N_{\text{max}}}$ [29]):

$$\hat{\rho}_{\text{coin,coh}} = \frac{1}{N_{\text{max}}}[(N_{\text{max}} - n_{av})|0\rangle\langle 0| + n_{av}|\alpha_{N_{\text{max}}}\rangle\langle \alpha_{N_{\text{max}}}||]. \quad (12)$$

Here, we have assumed $\langle \alpha_{N_{\text{max}}} | 0 \rangle \approx 0$, which is valid for $N_{\text{max}} \gg 1$. For the state $\hat{\rho}_{\text{coin,coh}}$, we find that the m -point coherence function (normalized by $G_{\text{coh}}^{(m)}$) reads

$$\frac{G_{\text{coin,coh}}^{(m)}}{G_{\text{coh}}^{(m)}} = \left(\frac{N_{\text{max}}}{n_{av}}\right)^{m-1}, \quad (13)$$

in agreement with Eq. (10), in the limit $N_{\text{max}} \gg m$.

We have thus shown that a classical mixture of the vacuum with a coherent state optimally drives m -photon absorption processes in the perturbative regime. As illustrated in Fig. 1, this state is better suited than coherent, squeezed or thermal states with the same average number of photons (i.e., with the same intensity).

IV. DISCUSSION AND CONCLUSION

What is the origin of the advantage of the state $\hat{\rho}_{\text{coin,coh}}$ (compared to just a coherent state) for driving m -photon transitions? As a statistical mixture, the state $\hat{\rho}_{\text{coin,coh}}$ in Eq. (12)

formally describes the situation in which one averages over many experiments. In each experiment, there is a probability n_{av}/N_{max} of employing a coherent laser driving with an average photon count of $N_{\text{max}} > n_{av}$, while in the remaining cases, the driving field is blocked, exposing the sample solely to the field's vacuum state. The m -photon excitation probability p_m is thus proportional to $(n_{av}/N_{\text{max}})G^{(m)}$, where $G^{(m)}$ is the coherence function of the driving field with on average N_{max} photons, given by $G^{(m)} = N_{\text{max}}^m$. Consequently, $p_m \propto (n_{av}/N_{\text{max}})G^{(m)} = n_{av}N_{\text{max}}^{m-1}$, whereas for a coherent state with n_{av} photons, $p_m \propto n_{av}^m$.

This demonstrates that the enhancement of m -photon absorption processes via enhanced number fluctuations arises from the fact that the probability of an m -photon absorption process, with $m \geq 2$, scales faster than linearly with the number of photons in the state. For a fixed average photon count n_{av} and a restricted maximal photon number, it is thus advantageous to maintain a low, yet nonzero, probability of encountering a state with a significant number of photons $n \gg n_{av}$, compared to having a steady field state with n_{av} photons. This phenomenon is purely classical. We emphasize that our results do not apply to multimode driving of m -photon transitions, e.g., via two-photon states entangled in the frequency domain [10–13]. However, optimal control theory, as employed here, can identify whether a genuine quantum advantage is present or not also in other control schemes involving quantum states of light.

In conclusion, we have shown that quantum features provide no advantage for optimally driving m -photon transitions between two discrete electronic states with fixed-intensity, narrow-band incident light fields, with a restricted maximal photon number. Instead, a classical mixture of the vacuum with a coherent state, which approximates the optimal multiphoton absorption rate of coin states (9) very well, is found to be the optimal field state.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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