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Perfect excitation of a matter qubit by a single photon in free space

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Abstract – We propose a scheme for perfect excitation of a single two-level atom by a single photon in free space. The photon state has to match the time reversed photon state originating from spontaneous decay of a two-level system. Here, we discuss its experimental preparation. The state is characterized by a particular asymmetric exponentially shaped temporal profile. Any deviations from this ideal state limit the maximum absorption. Although perfect excitation requires an infinite amount of time, we demonstrate that there is a class of initial one-photon quantum states which can achieve almost perfect absorption even for a finite interaction time. Our results pave the way for realizing perfect coupling between flying and stationary qubits in free space thus opening a possibility for building scalable quantum networks.

Efficient coupling between light and matter at a single quantum level lies at the heart of scalable quantum information processing, computation, and communication [1–3]. Information encoded in a flying qubit used for its transfer has to be recorded by a localized stationary qubit (*e.g.,* an atom), *i.e.*, the photon has to excite the atom with unit probability. Experimental realization of these protocols remains challenging due to the weak coupling between a single photon and a single atom in free space. Recent approaches investigating this problem focus on the absorption of a single photon by an ensemble of atoms resulting in a distributed single-photon excitation entangling the atoms of the ensemble [4–9].

So far, close-to-perfect interaction has been achieved only in the context of cavity QED in the strong-coupling regime, where the atom is forced to interact with a single mode of the radiation field only [10–16]. Scaling up these schemes is difficult because of the requirement for high finesse cavities.

Currently, several groups are attempting to quantify [17,18] and improve light-matter coupling in the absence of any mode-selecting cavity [19,20]. A detailed study of single-atom–single-photon interaction in free space requires the control of all resonant degrees of freedom of the radiation field, *i.e.*, its spatio-temporal vector modes. Van Enk and Kimble showed theoretically that both strong focusing and increased overlap of a light beam with a dipole wave corresponding to the relevant atomic transition improve the coupling [21,22]. Strong focusing of a light beam was demonstrated by Quabis *et al.* by tailoring the polarization pattern of light in theory [23] and in experiment [24,25].

We aim at having full control over the field modes and at exciting and maximizing the coupling to the atom. A first step toward this goal was the demonstration of a significant attenuation of a laser beam by a single trapped ion [26]. Recently, several groups succeeded in improving on this result [17,19,27–29]. Other groups attempted to control the excitation of a single atom [30,31], the ultimate goal being perfect excitation with a single-photon wave packet [20,22,32]. This should be possible based on a time reversal argument applied to spontaneous emission of a single photon [23]. These two goals, however, namely maximum attenuation of light and prefect excitation of the atom, are distinctly different. On the one hand, attenuation of a weak laser beam with zero transmission should be reachable with the atom populating essentially the ground state and with narrow-band continuous-wave laser radiation. On the other hand, a perfectly excited atom cannot be realized in dynamical equilibrium but

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only in a transient process which involves the excitation by a π-pulse, for example. Our objective is to realize the equivalent of a π-pulse excitation process with a single-photon wave packet [33] properly shaped in space and time.

In this letter, we outline a scheme for perfect excitation of a single two-level atom by a single photon in free space. We show that for this purpose the photon state has to match the time reversed photon state originating from spontaneous decay of a two-level system and we discuss its experimental preparation. Any deviations from this ideal state limit the maximum absorption. Both the spontaneous decay process and its reverse process of perfect excitation require an infinite amount of time. However, we demonstrate that there is a class of one-photon quantum states with mode decompositions close to the ideal one which can achieve almost perfect absorption even for a finite interaction time. As the mean values of the electric and the magnetic field of the radiation field are zero, the excitation of the matter qubit is completely caused by its uncertainties. They reveal an exponentially increasing tail and a sharp-edged shape genuine to a time reversed dipole wave [22,23]. In contrast to other recent work, such as refs. [19,28], which is based on classical diffraction theory, we apply a full non-relativistic QED treatment.

Theory. – Let us consider a trapped atomic qubit at a fixed position $\mathbf{x}=0$ in free space. Its excited and ground state are denoted by $|e\rangle$ and $|g\rangle$, respectively. Temporally, the atom interacts almost resonantly with the quantized radiation field which contains only one photon distributed over a continuum of temporal modes centered around the optical atomic transition frequency ω_0. Its center-of-mass motion is not affected if the atom is cooled to its lowest vibrational state so that its recoil momentum is picked up by the atom-trap system as a whole, much like in the Mössbauer effect. In the interaction picture, the Hamiltonian of this matter-field system reads

$$\hat{V}(t)=\hbar\sum_{l\in I} g_l\hat{\sigma}_+\hat{a}_l e^{i\Delta_l(t-t_0)}+\text{h.c.}, \tag{1}$$

where $\Delta_l=\omega_0-\omega_l$ is the detuning between the atom and the l-th optical mode with frequency ω_l, and $g_l=-(i/\hbar)\sqrt{(\hbar\omega_l)/(2\epsilon_0)}\,\mathbf{d}\cdot\mathbf{u}_l(\mathbf{x}=\mathbf{0})$ is the l-th mode coupling constant. The orthonormal mode functions of the radiation field are denoted by $\mathbf{u}_l(\mathbf{x})$ and $\mathbf{d}$ is the atomic dipole moment. The annihilation and creation operators of the field modes are given by $\hat{a}_l$ and $\hat{a}_l^\dagger$. Similarly, $\hat{\sigma}_+=|e\rangle\langle g|$ and $\hat{\sigma}_-=|g\rangle\langle e|$ are the atomic rising and lowering operators. These latter operators are expressed in the Schrödinger picture that coincides with the interaction picture at time t_0. Only those radiation field modes $l\in I$ contribute significantly to the system dynamics which are almost resonantly coupled to the atom within a frequency interval of the order of its spontaneous decay rate $\Gamma=(4/3)\omega_0^3|\mathbf{d}|^2/(4\pi\epsilon_0\hbar c^3)\ll\omega_0$.

We aim at determining the particular one-photon state of the quantized radiation field in free space which enables perfect excitation of the two-level atom prepared in its ground state $|g\rangle$ initially. In the interaction picture, the normalized quantum state of the coupled atom-field system is of the form

$$|\psi(t)\rangle=\sum_{l\in I} f_l(t)|g\rangle\otimes\hat{a}_l^\dagger|0\rangle+f_0(t)|e\rangle\otimes|0\rangle \tag{2}$$

with $|0\rangle$ denoting the vacuum of the radiation field and with the normalization constraint $|f_0(t)|^2+\sum_{l\in I}|f_l(t)|^2=1$. The ideal one-photon wave packet is determined by a solution of the time-dependent Schrödinger equation

$$i\hbar\,\partial_t|\psi(t)\rangle=\hat{V}(t)|\psi(t)\rangle \tag{3}$$

in the time interval $(-\infty,t_0]$ with initial condition $f_0(t_0)=1,\ f_l(t_0)=0$. Within the Wigner-Weisskopf approximation [34], the unique solution of eq. (3) is given by

$$\begin{aligned} f_0(t) &= e^{-\Gamma|t-t_0|/2}, \\ f_l(t) &= \frac{g_l^*}{\Delta_l-i\frac{\Gamma}{2}\mathrm{sgn(t-t_0)}}(e^{-\Gamma|t-t_0|/2}e^{-i\Delta_l(t-t_0)}-1) \end{aligned} \tag{4}$$

with $\mathrm{sgn}(x)=x/|x|$ for $x\neq 0$. This solution is valid for time $t\in(-\infty,t_0]$ or $t\in[t_0,\infty)$ and it is continuous but not smooth at time t_0 where the specified conditions are met. According to eqs. (2) and (4), the atom-field state is separable only at times $t\to\pm\infty$ with the atom being in its ground state. Thus, in the time interval $(-\infty,\,t_0]$, eq. (4) describes perfect absorption of the photon which is completed at time t_0. Similarly, in the interval $[t_0,\,\infty)$, it describes spontaneous decay of the two-level system initially prepared in its excited state at t_0.

From the uniqueness of the solution (4) it follows that perfect excitation of the two-level atom starting from its ground state is possible only if the light is prepared in the one-photon state (expressed in the Schrödinger picture)

$$|\chi\rangle_-=-\lim_{t_{\mathrm{in}}\to-\infty}\sum_{l\in I}\frac{g_l^*}{\Delta_l+i\frac{\Gamma}{2}}\,e^{-i\omega_l(t_{\mathrm{in}}-t_0)}a_l^\dagger|0\rangle. \tag{5}$$

The solution also shows that the completion of both perfect absorption and spontaneous emission of a photon requires an infinite amount of time. This fact is not surprising in view of the well-known exponential decay law since perfect excitation might be considered as the reverse process of spontaneous emission. The final state of the radiation field resulting from spontaneous decay of the atomic qubit starting at time t_0 reads

$$|\chi\rangle_+=-\lim_{t_{\mathrm{in}}\to\infty}\sum_{l\in I}\frac{g_l^*}{\Delta_l-i\frac{\Gamma}{2}}\,e^{-i\omega_l(t_{\mathrm{in}}-t_0)}a_l^\dagger|0\rangle. \tag{6}$$

In view of eq. (5), the natural question arises what excitation probabilities can be obtained during a finite

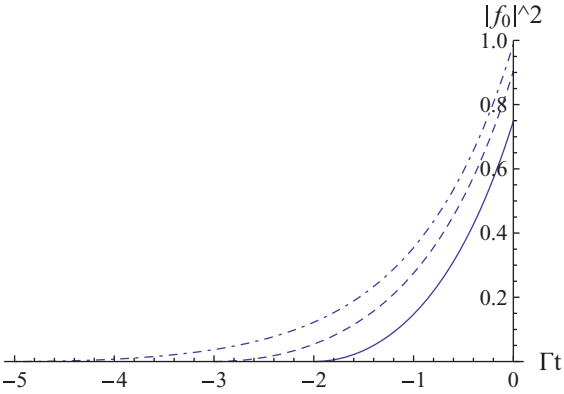

Fig. 1: (Colour on-line) Excitation probability $|f_0(t)|^2$ of eq. (8) evaluated for $\Gamma T = 2$ (full), $\Gamma T = 3$ (dashed) and $\Gamma T = 5$ (dash-dotted). The corresponding maximum excitation probabilities are 0.75, 0.90, and 0.99.

interaction time $t \in [t_{\rm in},\, t_0]$ and how do they depend on the spatio-temporal properties of exciting wave packet? For this purpose, let us assume that initially at time $t_{\rm in}$ we prepare the atom in its ground state and the radiation field in the state

$$|\chi(t_{\rm in})\rangle = -\sum_{l\in I} \frac{g_l^*}{\Delta_l + i\frac{\Gamma}{2}}\, e^{-i\omega_l(t_{\rm in}-t_0)} a_l^\dagger |0\rangle. \qquad (7)$$

This state has the same probability distribution for the occupied modes of the radiation field as the ideal state (5) but the phases of the corresponding probability amplitudes are different. For $t \geqslant t_{\rm in}$, the solution of the Schrödinger equation yields the excited-state probability amplitude

$$\begin{aligned} f_0(t) \;&=\; \int_{t_{\rm in}}^{t} {\rm d}t' e^{-\Gamma(t-t')/2}\Gamma e^{-\Gamma(t_0-t')/2}\Theta(t_0-t') \\ &= \Theta(t_0-t_{\rm in})\Big(\Theta(t_0-t)e^{-\Gamma(t_0-t)/2} \\ &\quad \times(1-e^{-\Gamma(t-t_{\rm in})}) + \Theta(t-t_0)e^{-\Gamma(t-t_0)/2} \\ &\quad \times(1-e^{-\Gamma(t_0-t_{\rm in})})\Big). \end{aligned} \qquad (8)$$

The time evolution of the excitation probability $|f_0(t)|^2$ is depicted in fig. 1. It approaches unity only in the limit of $t_{\rm in} \to -\infty$. However, for a finite interaction time $T = t_0 - t_{\rm in}$, with $T \gg 1/\Gamma$, the excitation probability approaches unity exponentially fast, *i.e.*, $|f_0(t_0)|^2 = (1-\exp(-\Gamma T))^2$. According to eq. (8), the probability amplitude $f_0(t)$ results from a constructive interference of all probability amplitudes associated with the excitation of the atom by the uncertainties of the radiation field at times $t', \in [t_{\rm in}, t]$. The probability amplitude of the field-induced excitation during a time interval $[t',\, t' + {\rm d}t']$ is given by ${\rm d}t'\,\Gamma \exp\{-\Gamma(t_0-t')/2\}\Theta(t_0-t')$ and the atomic probability amplitude to remain in the excited state $|e\rangle$ during a time interval $[t',\, t]$ by $\exp\{-\Gamma(t-t')/2\}$. This equation also shows explicitly that the field uncertainties can excite the atom only for $t' \leqslant t_0$.

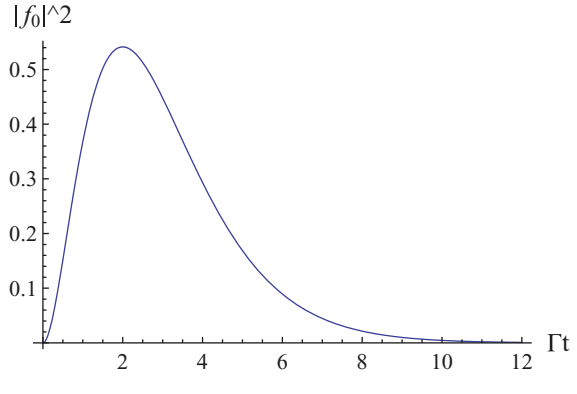

Fig. 2: (Colour on-line) Excitation probability $|f_0(t)|^2$ of eq. (10). The maximum amounts to $|f_0(t)|^2 = 0.54$.

The form of the quantum state of eq. (7) is crucial for achieving almost perfect excitation during a finite interaction time. This state corresponds to a time-reversed dipole wave [20,22]. If the initial state of the light was replaced by a reflected but not time-reversed dipole wave, such as

$$|\sigma(t_{\rm in})\rangle = -\sum_{l\in I} \frac{g_l^*}{\Delta_l - i\frac{\Gamma}{2}}\, e^{-i\omega_l(t_{\rm in}-t_0)} a_l^\dagger |0\rangle, \qquad (9)$$

the resulting excitation of the atomic qubit would change drastically. This initial condition yields the excitation amplitude

$$\begin{aligned} f_0(t) \;&=\; \int_{t_{\rm in}}^{t} {\rm d}t' e^{-\Gamma(t-t')/2}\Gamma e^{-\Gamma(t'-t_0)/2}\Theta(t'-t_0) \\ &= -\Theta(t-t_{\rm in})\Big(\Theta(t_{\rm in}-t_0)\Gamma(t-t_{\rm in})\, e^{-\Gamma(t-t_0)/2} \\ &\quad + \Theta(t_0-t_{\rm in})\Gamma(t-t_0)e^{-\Gamma(t-t_0)/2}\Big). \end{aligned} \qquad (10)$$

This probability amplitude $f_0(t)$ also results from a constructive interference of all probability amplitudes associated with the excitation of the atom at times $t' \in [t_{\rm in}, t]$. The probability amplitude of the atomic excitation in a time interval $[t',\, t' + {\rm d}t']$ is given by ${\rm d}t'\,\Gamma \exp\{-\Gamma(t'-t_0)/2\}\Theta(t'-t_0)$ and the probability amplitude of the atom to remain in the excited state during a time interval $[t',\, t]$ by $\exp\{-\Gamma(t-t')/2\}$. The field uncertainties responsible for the excitation are non-zero only for $t' \geqslant t_0$. This implies $f_0(t) = 0$ for $t_{\rm in} \leqslant t \leqslant t_0$.

A typical time evolution of eq. (10) is depicted in fig. 2. The atomic excitation reaches its maximum at the interaction time $t_{\rm max} = t_0 - t_{\rm in} + 2/\Gamma$ with $t_0 \geqslant t_{\rm in}$, which equals $|f_0(t = t_0 + 2/\Gamma)|^2 = 0.54$. This maximum value cannot be increased further by any choice of the interaction time. In other words, perfect excitation cannot be achieved with the single-photon state of eq. (9). It is interesting to note that the time dependence described by eq. (10) is also obtained in the case of spontaneous decay of an initially excited two-level atom positioned in the center of a very large spherically symmetric metallic

cavity of radius R for times $1/\Gamma \ll t \ll 2t_0$ [35]. Here, $t_0 = 2R/c$ denotes the time which the spontaneously emitted photon takes to return again to the atom after reflection at the metallic boundary of the cavity. The subsequent reabsorption of the photon is described by the probability amplitude given by eq. (10). Recently related experimental results were reported [36].

Similarly, a single-photon quantum state with a Gaussian mode distribution

$$|\Phi(t_{\rm in})\rangle = \mathcal{N} \sum_{l\in I} g_l^* e^{-\Delta_l^2/\sigma^2} e^{-i\omega_l(t_{\rm in}-t_0)}\, a_l^\dagger |0\rangle \qquad (11)$$

will not achieve perfect excitation. The normalization constant is given by $\mathcal{N} = (8\pi)^{1/4}/\sqrt{\Gamma\sigma}$ and σ measures the width of the mode distribution. For this state, the maximum value of the excitation probability is equal to $|f_0(t_{\rm max})|^2 = 0.80$ and is obtained for a width $\sigma = 1.46\Gamma$ at time $t_{\rm max} = t_0 + 1/\Gamma$.

It is worth noting that the interaction of a two-level atom with a photon is governed completely by its electric and magnetic field uncertainties since the mean values of the fields vanish for the atom-field state (2). The time evolution of the excitation probability $|f_0(t)|^2$ probes the dynamics of the uncertainties of the quantized radiation field in the immediate vicinity of the atom. For the case of perfect absorption these uncertainties are given explicitly by eq. (4). In order to obtain also insight into the uncertainties of the radiation field at a space-time point $(\mathbf{x}, t)$ far away from the atom $|\mathbf{x}|\omega_0/c \gg 1$, one has to investigate the normally ordered variances of the electric $\hat{\mathbf{E}}$ and the magnetic $\hat{\mathbf{B}}$ fields. Before the completion of the ideal excitation process $t \leqslant t_0$ eqs. (2) and (4) yield the result ($\hat{\mathbf{F}} = \hat{\mathbf{E}}$ or $(c\hat{\mathbf{B}})$)

$$\langle :(\mathbf{e}\cdot\hat{\mathbf{F}}(\mathbf{x}))^2: \rangle = \hbar\omega_0 \frac{6\Gamma \sin^2\theta}{16\pi\epsilon_0 c|\mathbf{x}|^2} (\mathbf{e}\cdot\mathbf{e}_\theta)^2 \times e^{-\Gamma(t_0-t-|\mathbf{x}|/c)} \Theta\left(t_0 - t - |\mathbf{x}|/c\right) \qquad (12)$$

in the limit of interest $\omega_0 \ll \Gamma$. Here, θ denotes the angle between the atomic dipole operator $\mathbf{d}$ and $\mathbf{x}$, $\mathbf{e}_\theta$ and $\mathbf{e}_\varphi$ are unit vectors along the corresponding coordinate lines of spherical coordinates and $\mathbf{e}$ is an arbitrary unit vector. These variances reveal the characteristic shape of the time-reversed dipole wave which leads to asymptotically perfect excitation under the model assumptions made above.

Planned experiment. – In our planned experiment [20], we will use a $^{174}Yb^{2+}$ ion as a two-level system with $^1S_0$ and $^3P_1^0$ electronic levels as the ground and the excited state, respectively, and no hyperfine structure. The atomic transition frequency $\omega_0 = 251.8\,$nm is in the ultraviolet regime. The ion will be trapped at the focus $f = 2.1\,$mm of a metallic parabolic mirror, being one electrode of a Paul trap. The rf needle-shaped electrode will come from the back of the mirror through a small hole. This trap design will ensure almost full $4\,\pi$ angle of ion-light interaction in the strong focusing regime. The aberration corrections will be done using a diffractive element located in front of the mirror. Since the ion has only one decay channel and its dipole moment will be parallel to the mirror axis we obtain free space geometry. There are several methods which allow for single-photon pulse generation with the desired spatio-temporal shape and spectral distribution. The first relies on electro-optic modulation of a single-photon wave packet [37]. Another experimentally more accessible method applies a strongly attenuated laser pulse containing $\overline{n} \ll 1$ photons on average. This technique is widely used in quantum key distribution [38]. We can shape a pulsed temporal mode electronically with modulators starting from a continuous-wave laser. Next, we will turn it into a radially polarized spatial doughnut mode. After reflection from the mirror surface, its polarization, at the focal point, will only contribute to polarization parallel to the axis of the mirror and, therefore, will excite a linear dipole oscillating parallel to this axis. Using this simpler method, perfect coupling is achieved if the probability of excitation matches the probability of finding a single photon in the pulse. In addition, as a third option one can generate the properly shaped single-photon Fock state wave function conditionally using photon pairs from parametric down conversion. This method is similar to ghost imaging in the time domain.

Of course, none of these methods will produce an infinitely long pulse. This is not an obstacle for our experiment, however, because one can truncate somewhat the exponential tail of the one-photon wave packet of eq. (7). For example, truncating the pulse to a duration of five lifetimes the excitation probability can be as high as 0.99. For quantum-storage applications it is straightforward to expand this scheme to a lambda transition between two long-lived states [32]. Furthermore, efficient coupling in free space opens the possibility for non-linear optics at the single-photon level.

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