Localized Single-Photon Wave Functions in Free Space

K. W. Chan,[1] C. K. Law,[1,2] and J. H. Eberly[1]

[1]*Center for Quantum Information and Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627*

[2]*Department of Physics, The Chinese University of Hong Kong, Shatin, Hong Kong, China*

(Received 22 August 2001; published 26 February 2002)

We solve the joint open problems of photon localization and single-photon wave functions in the context of spontaneous emission from an excited atom in free space. Our wave functions are well-defined members of a discrete orthonormal function set. Both the degree and shape of the localization are controlled by entanglement mapping onto the atom wave function, even though the atom is remote from the photon.

DOI: 10.1103/PhysRevLett.88.100402 PACS numbers: 03.65.Ud, 03.70.+k, 42.50.Vk

Deterministic generation of a single photon (one and only one) is of interest both fundamentally and for applications. Single photons are of course generated individually in spontaneous emission. However, obtaining a specified photon predictably in this way is difficult because the direction of emission is typically poorly defined and the timing of emission is completely random by the nature of spontaneous quantum events. Interest arising from potential applications in quantum cryptography and computing [1] has prompted recent studies of more subtle methods of single-photon generation employing cavity QED, photonic band gap materials, nanoelectronic systems, and parametric down-conversion, in which greater control is potentially available [2–4].

Closely related issues are the wave function of a single photon and the localizability of a single photon in free space. These have been discussed since the earliest days of quantum mechanics [5], and discussions continue to the present day [6] despite the well-known existence of a proof that single photons cannot be localized. Of course, what has been proved [7] is only that there is no position operator for a photon in free space. One easily questions whether this is even relevant to photon wave functions, and we believe not. Indeed, highly localized free-space solutions of Maxwell's equations are known [8]. A complete set of such spatial modes would be more desirable but none has been presented to date.

In this Letter we present a novel analysis of the most basic example of single-photon emission, i.e., spontaneous radiative decay of an excited atom in free space. We find a unique definition for orthonormal single-photon wave functions and calculate several examples for the lowest modes. To be clear about our program and its results, we do not introduce confining structures (as might be provided by fibers, waveguides, cavities, etc.), and our derived photon wave functions are not associated with classical electromagnetic variables (vector potential, electric or magnetic fields, Hertz vector, Poynting vector, etc.) or their quantized counterparts.

To begin we remark that, in emission accompanied by recoil, conservation of momentum enforces a quantum correlation that entangles the momenta of the photon and the recoiling atom. This is important, and the first laboratory results on recoil entanglement in atomic spontaneous emission have been reported by Kurtsiefer *et al.* [9]. Further experimental work in this fundamental area can be expected because entanglement provides information that permits the photon to be localized without destruction by detection.

The spontaneous radiative decay of interest is sketched in Fig. 1(a). Here $\hbar\vec{p}$ and $\vec{r}$ are the center of mass momentum and position of a two-level atom with ground and excited states $|g\rangle$ and $|e\rangle$, mass m, transition frequency ω_0, and atomic transition operators σ_j in the usual Pauli matrix notation. Then the Hamiltonian of the system under the rotating wave approximation is given by the usual expression:

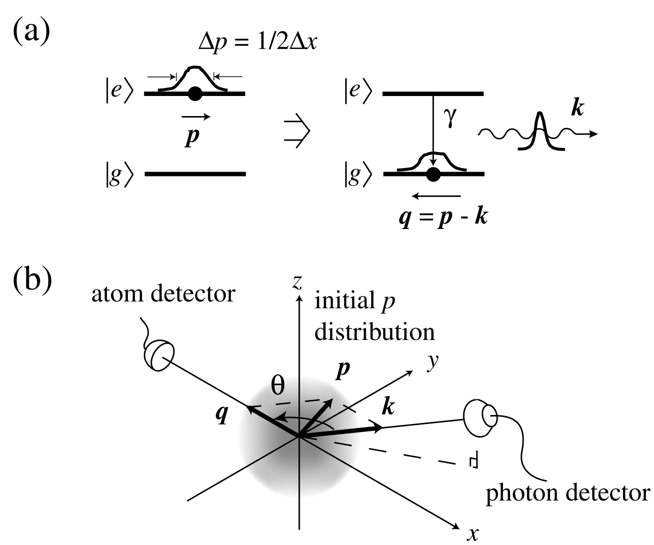

FIG. 1. (a) Spontaneous emission of a photon from a two-level atom with an initial momentum distribution $\vec{p}$. The atom gains recoil momentum $\hbar\vec{q}$ by emitting a photon of wave vector $\vec{k}$. (b) A schematic diagram of the system for a fixed observation angle.

$$H = \frac{(\hbar\vec{p})^2}{2m} + \int d^3k\,\hbar\omega_k a_k^\dagger a_k + \frac{1}{2}\hbar\omega_0(\sigma_z + 1) + \hbar\int d^3k\,[g(\vec{k})\sigma_- a_k^\dagger e^{-i\vec{k}\cdot\vec{r}} + \text{H.c.}], \quad (1)$$

where the photon wave vector is denoted by $\vec{k}$ and the initial center of mass wave vector of the atom is denoted by $\vec{p}$. As usual, a_k and $a_k^\dagger$ are the annihilation and creation operators of the plane-wave field mode with frequency $\omega_k = ck$. The strength of the dipole coupling with the atom and the field is given by $\hbar g(\vec{k}) = [\hbar\omega_k/2\epsilon_0(2\pi)^3]^{1/2}\vec{\epsilon}_k \cdot \vec{d}$ with $\vec{\epsilon}_k$ and $\vec{d}$ denoting the polarization vector of the field and the transition dipole moment of the atom, respectively.

At $t = 0$ the system is described by a separable product state with the field in the vacuum and the atom excited. After a sufficiently long time $t \gg \gamma^{-1}$ where γ is the atom's radiative linewidth, a photon is emitted and the atom recoils while dropping into the ground state. The asymptotic atom-photon state is not separable:

$$|\Psi(t)\rangle \rightarrow \iint d^3k\,d^3q\,C(\vec{q},\vec{k})e^{-i(E_q/\hbar+kc)t}|\vec{q};g\rangle \otimes |1_k\rangle. \quad (2)$$

Here the final wave vector of the atom is denoted by $\vec{q} \equiv \vec{p} - \vec{k}$. We have used the abbreviation $E_q \equiv (\hbar\vec{q})^2/2m$ to denote the final kinetic energy of the atom.

The absolute square of $C(\vec{q},\vec{k})$ tells us the joint probability of finding the atom and photon with the wave vectors $\vec{q}$ and $\vec{k}$. According to the solution of Rzążewski and Żakowicz [10], the amplitude $C(\vec{q},\vec{k})$ is given by

$$C(\vec{q},\vec{k}) = \frac{g(\vec{k})a_0(\vec{q} + \vec{k})}{(E_q - E_{q+k})/\hbar + kc - \omega_0 + i\gamma}. \quad (3)$$

Without loss of generality, we choose the atom to have a Gaussian distribution of motional momenta at $t = 0$, i.e., $a_0(\vec{p}) = a_0(|\vec{p}|) = \exp[-(p/\Delta p)^2]$.

As in the reported experiment [9], here we restrict our attention to the detection of the photon momentum $\vec{k}$ in a fixed direction with respect to the atom momentum $\vec{q}$, and for simplicity we choose $\theta = \pi$, as in Fig. 1(b). Further simplification can be made by noting that in realistic atomic systems, we have $\omega_0 \gg \gamma$ and $mc^2 \gg \hbar\omega_0$. Under these constraints, the photon-atom state is given by $|\Psi_\pi\rangle = \iint dq\,dk\,C_\pi(q,k)\,|q;g\rangle \otimes |1_k\rangle$, where

$$C_\pi(q,k) = \frac{N'}{\delta k + \delta q + i} \times \exp\left[-\left(\frac{mc\gamma\delta q}{\hbar\omega_0\Delta p}\right)^2\right]. \quad (4)$$

Here δk and δq are dimensionless momentum differences defined by $\delta k \equiv (ck - \omega_0)/\gamma$ and $\delta q \equiv \frac{\hbar\omega_0}{mc^2\gamma} \times (cq - \omega_0)$, and N' is a normalization factor that is nearly independent of $\vec{k}$ and includes the slowly varying $g(\vec{k})$. We point out that correlation between the photon and the atom enters through the Lorentzian term in Eq. (4). The Lorentzian enforces energy conservation, including recoil, and the Gaussian indicates the range of the amplitude $C_\pi(q,k)$ in dimensionless units appropriately defined by the emitter and the emission process.

Before discussing the theory further we want to make at least nominal connection with experimental considerations. We suppose there should be a fairly wide region where C is not too small. This means that if we write the exponential in (4) as $\exp[-(\delta q/\eta)^2]$, the parameter η should roughly fall in the range of 1 and 10. Thus we are regarding η as a kind of "observability index." This significance for η makes an intuitive interpretation of it desirable, and this can be obtained easily by expressing $\hbar\Delta p$ in terms of the velocity dispersion $m\Delta v$, and then replacing $\Delta v/c$ by $\Delta\omega/\omega_0$, the corresponding relative Doppler width of the atomic transition that would be associated with the velocity spread. Then the "large" parameter in the exponent of (4) can be rewritten as:

$$\eta \equiv \frac{\hbar\omega_0\Delta p}{mc\gamma} = \frac{\omega_0 m\Delta v}{mc\gamma} = \frac{\Delta\omega}{\gamma}, \quad (5)$$

i.e., η is simply the ratio of motional to radiative linewidths. The nature of $C_\pi(q,k)$ is shown in the inset of Fig. 2 for $\eta = 10$.

We now show that both photon localization and a set of well-defined photon wave functions are determined quantitatively by $C_\pi(q,k)$ without further approximation. Remarkably, in contrast to the conclusions reached in previous studies, *localization need not be related to photon wavelength nor necessarily associated with a wavelength-scale spatial range.* The key to our new conclusion is a Schmidt-mode analysis [11] of the quantum nonseparability between the emitting atom and emitted photon.

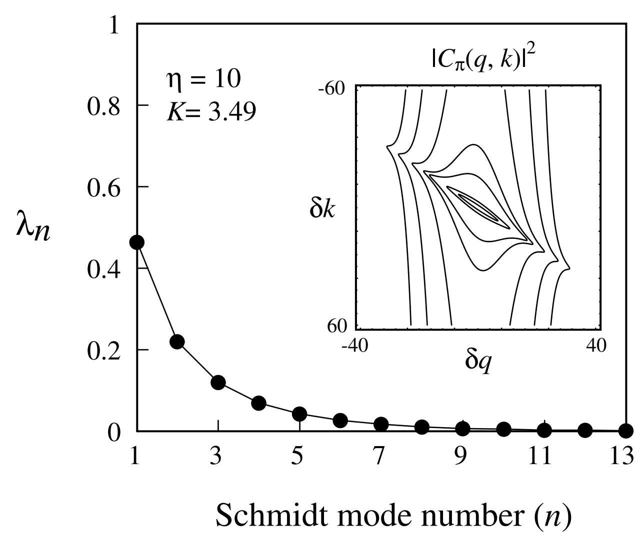

FIG. 2. Distribution of the eigenvalues λ_n for $\eta = 10$. The inset shows a contour plot of the absolute square of amplitude $C_\pi(q,k)$, where the dimensionless axis labels δq and δk are defined in the text.

Schmidt decomposition of the nonseparable $C_\pi(q,k)$ function converts it uniquely to a sum of product functions:

$$C_\pi(q,k) = \sum_n \sqrt{\lambda_n}\, \psi_n(q)\phi_n(k), \tag{6}$$

where the λ's are the joint eigenvalues of the atomic and photonic density matrices (thus $\sum \lambda_n = 1$) and $\psi_n(q)$ and $\phi_n(k)$ are the corresponding atom and photon eigenfunctions. In Fig. 2 a plot of λ's is shown for $\eta = 10$. For our purposes, the Schmidt decomposition has three important properties. The first is obvious—it provides a complete set of orthonormal functions $\phi_n(k)$ for the photon that is specific to the generation process, just what one would want for photon wave functions. Second, it provides an exact and unique association of the ψ's with the ϕ's in pairs. Each λ_n determines the occupation probability of its mode pair. Third, one sees that the atom-photon amplitude C is expressed in a discrete basis. It is perhaps surprising, even startling, that spontaneous emission involves only a countable number of modes, despite its conventional formulation in a continuous Hilbert space (recall the Hamiltonian). However, it may be even more startling that the total number of modes that can play an important role can be very small (in Fig. 2 it is only four or five).

In principle, the second Schmidt property mentioned above is the most important. It ensures that observing the atom in mode $\psi_n(q)$ by a standard (projective) measurement one guarantees that the photon wave function is the unique Schmidt partner $\phi_n(k)$. In this way one identifies and at the same time classifies [12] a single photon by an observation on the atom, and does not destroy the photon. The role of the third Schmidt property will be seen after we count the number of active modes. Note that the number of active modes is a measure of the degree of entanglement. The participation ratio K provides this number [13]:

$$K \equiv 1 \Big/ \sum_n \lambda_n^2 . \tag{7}$$

It is important to link the degree of entanglement (number of active modes) with the observability index. We have found numerically a simple empirical relation between K and η appropriate to spontaneous emission with recoil. For $\eta \lesssim 1$ we find $K \approx 1$, and for $\eta > 1$ we find $K \approx 1 + 0.28(\eta - 1)$. Taken along with the linewidth interpretation of η, this tells us two things—that we require a substantial degree of entanglement (the participation of at least several Schmidt modes) to meet the observability criterion $\eta = 1$ to 10 mentioned above, and that the effective number of modes is roughly 1/4 to 1/3 the number of natural linewidths fitting under the motional linewidth [the fractional value is related to the choice of the initial momentum distribution $a_0(\vec{p})$].

Our procedure provides the explicit form of the Schmidt mode functions associated with the photon and the atom. In Fig. 3, we show both atom and photon Schmidt functions in position space at $t = 5/\gamma$. These spatial functions are essentially the Fourier transforms of $\phi_n(k)$ and $\psi_n(q)$:

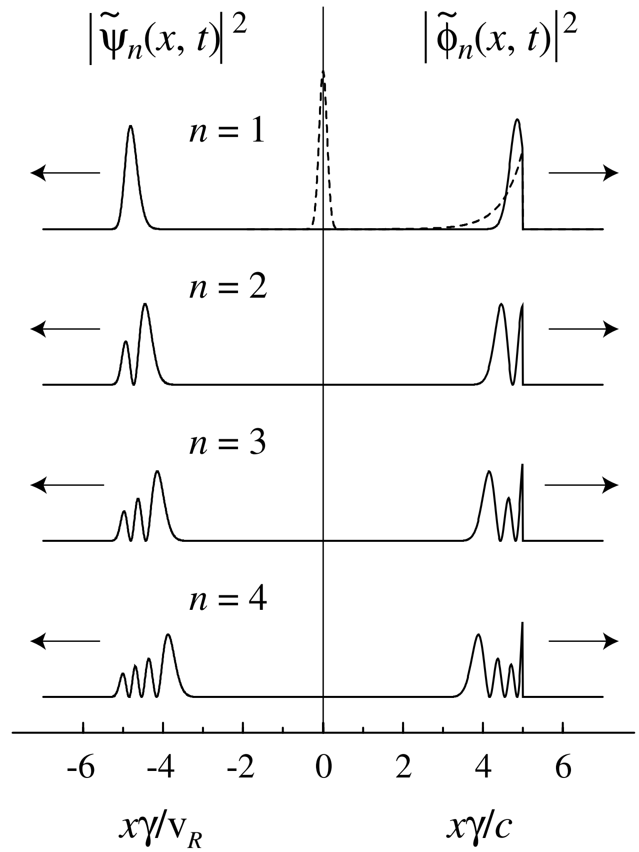

FIG. 3. The first four Schmidt pairs in position space at the time $t = 5\gamma^{-1}$. The left column gives the atom modes and the right column gives the photon modes, with arrows indicating the propagation directions. Here $\eta = 10$ and $\omega_0/\Delta p = 20$ are used. The spatial axes of the atom and photon modes are in different dimensionless units. The mean recoil velocity of the atom $\hbar\omega_0/mc = \eta\gamma/\Delta p$ is denoted by v_R. The dotted lines in the first row show the atom and photon states in the zero recoil limit. They are plotted using the same scale as the entangled case.

$$\tilde{\phi}_n(x,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dk\, \phi_n(k) e^{ik(x-ct)}, \tag{8}$$

$$\tilde{\psi}_n(x,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dq\, \psi_n(q) e^{iq(x-\hbar qt/2m)}. \tag{9}$$

Note that the photon wave functions all preserve a sharp edge at exactly $x\gamma/c = 5$, corresponding to the initiation of decay at $t = 0$. We see that both photon and atom wave functions have nodes that divide them into pulses. A distinct feature is that the first mode is a one-pulse form, the second mode is a two-pulse form, and so on. This is observed over a wide range of η in our numerical calculations, and a similar finding was noted in Schmidt analysis of down-conversion (see Huang and Eberly [4]).

The connections between localization and entanglement and the emission process are also clear as illustrated in Fig. 3. The photon functions are entangled uniquely pairwise with their atomic counterparts according to the Schmidt prescription. It is remarkable that the shapes of the atom modes and the photon modes are nearly exactly the same, except for the regions near their respective wave fronts. Therefore the "shape" of the photon is essentially controlled by a mapping onto the atom shape, even though the atom and photon are remote from, and exert no force on, each other. The ratio of widths of the atom and photon modes is approximately $\hbar\omega_0/mc^2 \ll 1$. The degree of localization is characterized by how rapidly the tail of the photon wave function drops to zero. We find that the tail of each photon wave function behaves as a Gaussian, and this corresponds to a stronger form of localization than one expects from exponential decay. It is a consequence of the correlation mixing of the Gaussian and Lorentzian signatures of the emission process [see Eq. (4)]. In fact, we find that the width of the Gaussian tail is inversely proportional to $\Delta\omega/c$ when η is large. Therefore the photon localization is governed by the motional linewidth of the atom.

In summary, we have described a new procedure to investigate atomic spontaneous emission, taking account of the photon-atom entanglement generated. The heart of this procedure is the systematic exploitation of the properties of the Schmidt method for expressing joint two-particle states. One result is what we believe to be the first workable prescription for a discrete orthonormal set of mode functions for a single photon in free space, i.e., a complete and countable set of photon wave functions despite the intuitively continuous character of spontaneous emission and recoil. Note that the nature of spontaneous emission itself ensures that the mode functions are associated with a single photon. They have finite extent that is determined by the physical details of the emission process and are clearly predictably localized (both spatially and temporally) in principle in a natural way, by projective measurement of the atom. Thus we suggest that the Schmidt analysis provides the most natural understanding of photon localization as well as providing the photon wave functions that quantify the extent of localization.

To conclude, we remark that we have also isolated η, a new control parameter for spontaneous emission entanglement. It can be interpreted as the ratio of motional linewidth to natural linewidth. The strength of entanglement is quantified by the participation number K, and we found empirically that K is approximately a linear function of η when $\eta > 1$. In typical two-level atomic systems, the value of η is of order 0.1 or smaller [14], but we note that our result can be generalized to Raman scattering situations where the value of η can be substantially increased because of the much narrower spontaneous Raman linewidths. Finally, we note that our analysis was entirely focused on the final state of the system. It is interesting to ask about the time development of entanglement as the system evolves [13]. Our numerical results indicate that for the open system of spontaneous emission K is always an increasing function of time. This raises interesting questions about the existence of quantum rules for irreversibility of entanglement analogous to entropy increase in the second law of thermodynamics. We hope to address this issue in the future.

This research was partially supported by ARO-D Grant No. DAAD19-99-1-0215 and by NSF Grant No. PHY-0072359.

[1] For a review of quantum information and early proposals of implementations, see H. E. Brandt, Prog. Quantum Electron. **22**, 257 (1998), and references therein.

[2] C. K. Law and J. H. Eberly, Phys. Rev. Lett. **76**, 1055 (1996); C. K. Law and H. J. Kimble, J. Mod. Opt. **44**, 2067 (1997); A. Imamoglu *et al.,* Phys. Rev. Lett. **79**, 1467 (1997); A. Kuhn *et al.,* Appl. Phys. B, **69**, 373 (1999).

[3] J. Kim *et al.,* Nature (London) **397**, 500 (1999); P. Michler *et al.,* Science **290**, 2282 (2000); B. T. H. Varcoe *et al.,* Nature (London) **403**, 743 (2000); C. Santori *et al.,* Phys. Rev. Lett. **86**, 1502 (2001).

[4] C. K. Hong and L. Mandel, Phys. Rev. Lett. **56**, 58 (1986); H. Huang and J. H. Eberly, J. Mod. Opt. **40**, 915 (1993); C. K. Law, I. A. Walmsley, and J. H. Eberly, Phys. Rev. Lett. **84**, 5304 (2000); H. H. Arnaut and G. Barbosa *ibid.* **85**, 286 (2000).

[5] P. A. M. Dirac, Proc. R. Soc. London A **114**, 243 (1928); J. R. Oppenheimer, Phys. Rev. **38**, 725 (1931).

[6] J. Sipe, Phys. Rev. A **52**, 1875 (1995); I. Bialynicki-Birula, in *Progress in Optics,* edited by E. Wolf (Elsevier, Amsterdam, 1998), Vol. XXXVIII, p. 245; M. O. Scully and M. H. S. Zubairy, *Quantum Optics* (Academic, New York, 1997).

[7] See M. H. L. Pryce, Proc. R. Soc. London A **195**, 62 (1948); T. D. Newton and E. P. Wigner, Rev. Mod. Phys. **21**, 400 (1949); A. S. Wightman, Rev. Mod. Phys. **34**, 845 (1962); L. Mandel, Phys. Rev. **144**, 1071 (1966).

[8] W. O. Amrein, Helv. Phys. Acta **42**, 149 (1969); R. W. Hellwarth and P. Nouchi, Phys. Rev. E **54**, 889 (1996); C. Adlard, E. R. Pike, and S. Sarkar, Phys. Rev. Lett. **79**, 1585 (1997); I. Bialynicki-Birula, Phys. Rev. Lett. **80**, 5247 (1998).

[9] C. Kurtsiefer *et al.,* Phys. Rev. A **55**, R2539 (1997); for earlier experiments addressing the effects of recoil entanglement, see T. Pfau *et al.,* Phys. Rev. Lett. **73**, 1223 (1994); M. S. Chapman *et al.,* Phys. Rev. Lett. **75**, 3783 (1995).

[10] K. Rzążewski and W. Żakowicz, J. Phys. B **25**, L319 (1992).

[11] Schmidt modes are explained in A. Ekert and P. L. Knight, Am. J. Phys. **63**, 415 (1995), and references therein.

[12] In parametric down-conversion, observation of the signal photon can approximately localize the idler photon without destroying it but, under conditions examined previously, cannot classify it. This weak type of localization motivated the Hong-Mandel experiment [4]. Down-conversion has recently been analyzed from the Schmidt viewpoint: see Huang and Eberly, and Law *et al.,* in [4].

[13] R. Grobe, K. Rzążewski, and J. H. Eberly, J. Phys. B **27**, L503 (1994).

[14] For sodium, $\hbar\omega_0 \approx 2.14$ eV, $mc^2 \approx 10^{10}\hbar\omega_0$, $\gamma \approx 10^7\ \mathrm{s}^{-1} \approx 10^{-9}\omega_0$, and $\Delta pc \approx 0.46\omega_0$, we find $\eta \approx 0.05$.