Measurement models for time-resolved spectroscopy: a comment

Holger F. Hofmann and Günter Mahler
Institut für theoretische Physik und Synergetik
Pfaffenwaldring 57, 70550 Stuttgart, Germany

Abstract

We present an exactly solvable model for photon emission, which allows us to examine the evolution of the photon wave function in space and time. We apply this model to coherent phenomena in three level systems with special emphasis on the effects of the photon detection process.

I. INTRODUCTION

Describing the open system dynamics of a quantum object, the states of external fields such as the light field are usually represented by some form of statistical approximation: for instance, the master equation approach assumes an unchanging thermal state of the external fields, disregarding all effects which the system might have on them (“bath approximation”).

Approaches such as the Monte Carlo wave function method [1] or the subensemble density matrix [2] consider measurements of the photons at the instant of their emission, thereby arriving at an easy to handle description of the open system dynamics while avoiding a more detailed description of the light field itself.

However, in typical experimental situations, the light field is our only source of information, while the system dynamics must be deduced indirectly from the respective measurement protocols. Furthermore, the type of measurement we choose to perform on the photons may influence the dynamics of the system. Therefore, a closer look at the processes which allow
us to measure the emitted photons may be helpful. For this purpose we should start with the complete Schroedinger equation of the system and the field. The evolution of this state will then be analyzed with respect to a representation adopted to given measurement scenarios.

II. BASIC MODEL

In the most simple case, we have a localized two level system, consisting of the excited state $|E\rangle$ and the ground state $|G\rangle$ with a transition frequency $\omega_0$, and a one dimensional field with linear dispersion $\omega = c|k|$ coupled by a local interaction.

The field can be separated into two distinct branches, one with group velocity $c$ for $k > 0$ and one with group velocity $-c$ for $k < 0$. In the following, we will further simplify the problem by considering only the branch with positive group velocity, extending it to negative values of $k$ and $\omega$ as shown in figure (1). The introduction of negative frequencies can be justified on the ground that, since the emission of photons with zero or negative energy would violate energy conservation, such processes will only contribute to the dynamics on time scales smaller than $1/\omega_0$. Therefore, the amplitudes of those spurious field states should be small compared to those near $\omega = \omega_0$ [3].

The relevant states for the emission process are the product state of the excited system state and the field vacuum, $|E;\text{vac.}\rangle$, and the product states of the ground state and the 1 photon field states $|G;k\rangle$. Using these states as basis, the Hamiltonian of our model is

$$\hat{H} = \hbar\omega_0|E;\text{vac.}\rangle\langle E;\text{vac.}| + \int_{-\infty}^{+\infty} dk \hbar kc |G;k\rangle\langle G;k| + \int_{-\infty}^{+\infty} dk \hbar [g|G;k\rangle\langle E;\text{vac.}| + g^*|E;\text{vac.}\rangle\langle G;k|]$$

This Hamiltonian is a simple version of the Hamiltonian used in Wigner-Weisskopf theory. Therefore, we can proceed as in [4]. The time dependent Schroedinger equation is

$$\frac{d}{dt}\langle E;\text{vac.}|\psi(t)\rangle = -i\omega_0\langle E;\text{vac.}|\psi(t)\rangle$$
\[ -ig^* \int_{-\infty}^{+\infty} dk \langle G; k | \psi(t) \rangle \]  

\[
\frac{d}{dt} \langle G; k | \psi(t) \rangle = -i \langle k | c \langle G; k | \psi(t) \rangle - ig \langle E; \text{vac.} | \psi(t) \rangle \]  

If we choose the initial condition \( |\psi(t = 0)\rangle = |E; \text{vac}\rangle \), the result of the integrated Schroedinger equation for the one photon part is

\[
\langle G; k | \psi(t) \rangle = -ig \int_0^t dt' e^{-ikc(t-t')} \langle E; \text{vac.} | \psi(t') \rangle \]  

This result can be substituted into the equation for the amplitude of the excited state. By using \( b(t) := e^{i\omega_0 t} \langle E; \text{vac.} | \psi(t) \rangle \) one obtains

\[
\frac{d}{dt} b(t) = -|g|^2 \int_0^\infty dt' b(t') \int_{-\infty}^{+\infty} dk e^{-i(kc - \omega_0)(t-t')} \]  

The integral over k, again, results in a delta function, and the evolution becomes local in time:

\[
\frac{d}{dt} b(t) = -\pi |g|^2 \int_{0}^{\infty} dt' b(t ') \int_{-\infty}^{+\infty} dk \ e^{-i(kc - \omega_0)(t-t')} \]  

\[
\langle E; \text{vac.} | \psi(t) \rangle = e^{(-i\omega_0 - \Gamma/2)t} \]  

The amplitudes calculated for the 1 photon states of our model are:

\[
\langle G; k | \psi(t) \rangle = ge^{-ikct} \frac{1 - e^{-\Gamma t/2} e^{i(kc - \omega_0)t}}{kc - \omega_0 + i\Gamma/2} \]  

We can test our assumption that the amplitudes for \( k \leq 0 \) are much smaller than those near \( k = \omega_0/c \) by calculating the k-space probability densities:

\[
|\langle G; k | \psi(t) \rangle|^2 = |g|^2 \frac{1 - 2 \cos(kct - \omega_0 t) e^{-\Gamma t/2} + e^{-\Gamma t}}{(kc - \omega_0)^2 + \Gamma^2/4} \]  

For \( t >> 1/\Gamma \), this is the familiar Lorentzian distribution. Thus, for \( \omega_0 >> \Gamma \), the contribution of field states with \( \omega < 0 \) will be negligible.
On smaller timescales, there are contributions from $k = -\infty$ to $k = +\infty$ due to the energy-time uncertainty. In a model with a more realistic dispersion relation, the $k=0$ component have a group velocity of 0. Consequently, there will be reabsorption of such components in the real space formulation, which leads to an interaction of the system with itself. The main effect of this interaction would be an energy shift in the spectrum, comparable to the Lamb shift in quantum electrodynamics. However, since our model does not include such processes, there is no such energy shift and the Lorentzian distribution of the emission centers on $\omega_0$.

It is now possible to calculate the real space amplitudes by Fourier transforming the $k$ space result:

$$\langle G; x | \psi(t) \rangle = \begin{cases} -i\sqrt{\frac{\Gamma}{2}}e^{i(\Gamma/2 + i\omega_0)(x/c - t)} & \text{for } 0 < x < ct \\ 0 & \text{otherwise} \end{cases}$$

(10)

Figure (2) shows the spatiotemporal distribution of the field state. This function can be interpreted as the real space probability density for photon detection.

III. APPLICATIONS

A. Extension to 3 Dimensions

In order to apply this result to realistic conditions in photon spectroscopy, it is necessary to give a more detailed interpretation of the $|k\rangle$ states. In typical cases, the emission will be into an unrestricted three dimensional light field. Therefore, an infinite number of degenerate modes with the same absolute value of $k$ is available. In the plane wave basis with wavevector $k = |k| \hat{k}$ and a linear polarization given by $e_p$, the interaction part of the Hamiltonian is

$$\hat{H}_{int} = \int d|k|d\hat{k} \sum_{e_p} h g(\hat{k}, e_p) |G; k, e_p\rangle \langle E; vac.| + h g^*(\hat{k}, e_p) |E; vac.\rangle \langle G; k, e_p|$$

(11)

However, instead of using this plane wave basis, we may transform to another basis in which only a single mode per energy level interacts with the system. This new mode can be
determined directly from the interaction part of the Hamiltonian:

$$\hat{H}_{\text{int}} = \int \, d|k| \hbar \bar{g} |G; k\rangle \langle E; \text{vac.}| + \hbar \bar{g} |E; \text{vac.}\rangle \langle G; k|$$  \hspace{1cm} (12)

with

$$|G; k\rangle = \int \, d\hat{k} \sum_{e_p} \frac{\bar{g}(\hat{k}, e_p)}{\bar{g}} |G; k, e_p\rangle$$  \hspace{1cm} (13)

and

$$\bar{g}^2 = \int \, d\hat{k} \sum_{e_p} |g(\hat{k}, e_p)|^2$$  \hspace{1cm} (14)

In many situations, it may suffice to inspect the symmetry of the system and of the field. For example, the spherical symmetry of an atomic system allows us to classify both the system and the field states according to the quantum numbers of the angular momentum, \(l\) and \(m\) (for details, see [7], [8]). In the following, we restrict the atomic states to \(|S\rangle\) (\(l=0\)) and \(|P\rangle\) (\(l=1\)). A dipole transition emits only photons of \(l = 1\) with \(m = 0, \pm 1\). Since the total angular momentum is conserved, the system must change its quantum numbers \(l\) and \(m\) accordingly.

Since for radii much larger than the wavelength spherical waves approach plane waves, the previous interpretation of the wave function in real space still applies, except in the immediate vicinity of the object. Replacing \(x\) by \(r\), the \(|r\rangle\) states now describe a photon at a distance \(r\) from the system, while the angular dependence and the polarization of the photon is given by the photon state quantum number \(m\). With the \(z\)-axis as quantization axis, the amplitudes are

$$\langle k, e_p|k'; m = 0 \rangle = \frac{1}{4\pi} \delta(|k| - k') e_p e_z$$  \hspace{1cm} (15)

$$\langle k, e_p|k'; m = \pm 1 \rangle = \frac{1}{4\pi} \delta(|k| - k') \frac{1}{\sqrt{2}} (e_p e_x \pm ie_p e_y)$$  \hspace{1cm} (16)

The condition for transversality of the light field, \(e_p k = 0\), yields the angular dependence of the emitted intensity characteristic for dipole radiation.

**B. Three Level System**

A simple application of this model is the standard quantum beat scenario (three level system) in which an atom in a magnetic field is excited from the groundstate \(|G\rangle = |S\rangle\)
with a short laser pulse polarized in a direction orthogonal to the magnetic field. The atom is left in a superposition of the $m = +1$ and the $m = -1$ sublevels of the excited P state $|E_\pm\rangle = |P, m = \pm 1\rangle$. Since we can apply our theory to both the $\Delta m = +1$ and the $\Delta m = -1$ transitions separately and since we are dealing with the linear dynamics of the Schroedinger equation, we can immediately write down the complete evolution of both the field and the system in local representation:

$$|\psi(t = 0)\rangle = \frac{1}{\sqrt{2}}(|P, m = +1; vac.\rangle + |P, m = -1; vac.\rangle) \quad (17)$$

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}}e^{-\Gamma_+ t/2 - i\omega_+ t}|P, m = +1; vac.\rangle$$

$$+ \frac{1}{\sqrt{2}}e^{-\Gamma_- t/2 - i\omega_- t}|P, m = -1; vac.\rangle$$

$$- \frac{i}{\sqrt{2}} \int_0^{ct} \left( \sqrt{\frac{\Gamma_+}{c}} e^{(\Gamma_+/2 + i\omega_+)(r/c - t)} |S; r, m = +1\rangle \right) dr$$

$$+ \sqrt{\frac{\Gamma_-}{c}} e^{(\Gamma_-/2 + i\omega_-)(r/c - t)} |S; r, m = -1\rangle) \quad (18)$$

Written in the $m$ basis of the light field and the $m$ basis of the system, quantum beats do not appear in the evolution of either subsystem. A simple transformation to another basis in atom-,

$$|P_x\rangle = \frac{1}{\sqrt{2}}(|P, m = +1\rangle + |P, m = -1\rangle) \quad (19)$$

$$|P_y\rangle = \frac{i}{\sqrt{2}}(|P, m = +1\rangle - |P, m = -1\rangle) \quad (20)$$

and field-states

$$|r, d_x\rangle = \frac{1}{\sqrt{2}}(|r, m = +1\rangle + |r, m = -1\rangle) \quad (21)$$

$$|r, d_y\rangle = \frac{i}{\sqrt{2}}(|r, m = +1\rangle - |r, m = -1\rangle) \quad (22)$$

shows, however, that beats do occur then, contrary to the statement made in [6], that orthogonal dipoles should definitely exclude beats. The wave function written in this basis for the case of $\Gamma_+ = \Gamma_- := \Gamma$ now reads:
\[ |\psi(t)\rangle = e^{-\Gamma t/2-i\tilde{\omega}t}(\cos(\delta\omega t)|P_x;\text{vac.}\rangle + \sin(\delta\omega t)|P_y;\text{vac.}\rangle) \\
- i\sqrt{\Gamma/c} \int_0^t e^{(r/2+i\tilde{\omega})(r/c-t)}(\cos(\delta\omega (r/c - t))|S;r,d_x) \\
+ \sin(\delta\omega (r/c - t))|S;r,d_y) \rangle \, dr \]  

(23)

where \( \tilde{\omega} = \frac{\omega_+ + \omega_-}{2} \) and \( \delta\omega = \frac{\omega_+ - \omega_-}{2} \). This representation of the evolution clearly shows beats in both the system and the field. Figure (3) shows the real space probability distribution of this linearly polarized photon. Of course, whether beats are observed or not depends on the type of measurement performed.

C. System-Field Correlations

Another application of this model is the description of entangled states between field and system if the decay is from a single excited level \(|E\rangle = |S\rangle\) into two (or possibly more) alternative groundstates \(|G_{\pm}\rangle = |P,m = \pm 1\rangle\). Although the situation may at first appear very similar to the quantum beat scenario, we cannot single out two paths, since the initial state cannot be separated into two components, each belonging to only one path. A possibility is to view the decay as a single path, leading from the excited state to a linear combination of the two groundstates, \( \frac{\Gamma_+}{\sqrt{\Gamma_+^2 + \Gamma_-^2}} |P,m = +1\rangle + \frac{\Gamma_-}{\sqrt{\Gamma_+^2 + \Gamma_-^2}} |P,m = -1\rangle \). However, the two groundstates will generally not be degenerate. Therefore, the state of the system will start to evolve in time at the instant of the emission, \( t-r/c \), causing a dependence of the system state on the field state \(|r\rangle\). The measurement of a photon at \( r \) implies that a time of \( r/c \) has elapsed since emission, so that the state of the total system would be

\[ |G;r\rangle := \frac{1}{\sqrt{\Gamma_+^2 + \Gamma_-^2}}(\Gamma_+ e^{i\omega_+ r/c}|P,m = +1; r, m = -1\rangle \\
+ \Gamma_- e^{i\omega_- r/c}|P,m = -1; r, m = +1\rangle) \]  

(24)

Thus we can use the fact that our model treats emissions as instantaneous to replace the ground state with the temporal evolution of this two level subsystem.

With \( \Gamma = \Gamma_+ + \Gamma_- \) we obtain the result of the complete evolution as
The one photon part of this wave function carries a strong system-field correlation. In the case $\Gamma_+ = \Gamma_- = \Gamma/2$, any linear combination of $|P, m = +1\rangle$ and $|P, m = -1\rangle$ states may be observed, depending on the measurement results in the field. For example, we may transform to the field basis $|r, dx\rangle$, $|r, dy\rangle$ and the atomic basis $|Px\rangle$, $|Py\rangle$ as in section 3.2:

\[
|\psi(t)\rangle = e^{-\Gamma t/2+i\omega t}|S; vac.\rangle
- i\sqrt{\Gamma/2} \int_0^t e^{(\Gamma/2+i\omega)(r/c-t)}|G; r\rangle dr
\]

\[
= e^{-(\Gamma_+ t/2+\Gamma_- t/2+i\omega t)}|S; vac.\rangle
- i\sqrt{\Gamma_+ + \Gamma_-} \int_0^t e^{(\Gamma_+ t/2+\Gamma_- t/2+i\omega t)}(r/c-t) \frac{1}{\sqrt{\Gamma_+^2 + \Gamma_-^2}}
\]

\[
(\Gamma_+ e^{i\omega_+(r/c-t)}|P, m = +1; r, m = -1\rangle + \Gamma_- e^{i\omega_-(r/c-t)}|P, m = -1; r, m = +1\rangle) dr
\]

(25)

Again, we use the notation $\bar{\omega} = \frac{\omega_+ + \omega_-}{2}$ and $\delta\omega = \frac{\omega_+ - \omega_-}{2}$. While the total probability of measuring a photon with $d_x$ polarization does not oscillate, those photon detections correlated with an atomic state of $|Px\rangle$ and $|Py\rangle$ both display beats as shown in Figure (4).

The beats could therefore be seen by measuring the atomic state and separating out the photon detections accordingly. This seems to be at variance with the expectation that in this type of three level system, beats should be absent because a measurement of the atomic state would reveal the decay channel, $\omega_+$ or $\omega_-$. However, measuring the $|Px\rangle$ and $|Py\rangle$ states of the atomic system reveals only the phase of the beats, preventing the determination of the decay channel. Coincidence measurements of this type can therefore be considered as quantum eraser measurements as described in [10] and [11] for the case of 2 spatially

8
separated sources of photonscattering, since the measurement of $|P_x\rangle$ and $|P_y\rangle$ effectively erases the information to be gained by measuring $|P, m = +1\rangle$ and $|P, m = -1\rangle$.

The two cases described above and in section 3.2, respectively, may be combined into a single four level system with a cascade decay leading from an excited level through two (or more) intermediate levels to the ground state. This model could thus be used to describe the quantum beats observed in cascade decays [12]. The resulting wave function will include a two photon part, so it may be necessary to consider the boson nature of the photons when transforming to another measurement base.

Since the emission process is local in time and there is no interaction between the photons, our present model can be adapted to any other scenario by simply adding the effects of the possible channels of decay. In this way, it should be possible to develop a better understanding of the role which the optical measurement apparatus plays in quantum measurements.

IV. MEASUREMENT PROCESSES

For actual measurements the atomic object and the field is embedded into a dissipative environment, usually a filter and a detector, which, again, may be specified by its interaction with the light field. The type of measurement process can be included in the form of a projection acting on the field part of the wavefunction. The choice of base states used should thus be made with a special experimental setup in mind. For example, a polarizer-detector setup can be considered as a projective measurement on $|r; d_x\rangle$ states, where $r$ is the distance between the system and the detector. With the 3-level system described by equation (23), this would allow us to observe quantum beats. A more realistic representation would consider the limited area covered by the detector, meaning that actually a state of the form $(\sqrt{\sigma}|r, d_x\rangle + \sqrt{1-\sigma}|r, l > 1\rangle)$ is measured, where $\sigma$ is the proportion of photons emitted into the angle covered by the detector. $|r, l > 1\rangle$ represents a superposition of multipole states with $l > 1$. It ensures, that the measured state exists only in a limited area
by interfering with $|r, d_x\rangle$ in such a way, that real space components outside the emission angles covered by the detector vanish.

In our model, the field dynamics in real space rigidly move the field states away from the system at the speed of light (see Fig.1). Therefore, the state of the field represents a temporal record of the field dynamics at the system. It does not matter at what distance $r$ we choose to measure, since a shift in $r$ is equivalent to a delay time $\Delta t = r/c$. If we are only interested in time resolved spectroscopy, we may simulate the photon measurement directly at the system. This corresponds to the measurement approaches of [1] and [4].

However, as we have calculated the whole wavefunction of the field, we can also consider frequency resolved measurements. Most frequency selective filters are based on an interference between optical paths of different length. This corresponds to a linear combination of different $|r\rangle$ states of the field. A Michelson interferometer would produce a measurement of $\frac{1}{\sqrt{2}}(|r_1\rangle + |r_2\rangle)$ in the detector, while a Fabry-Perot interferometer measures $\frac{1}{\sqrt{1-R^2}} \sum_{n=1}^{\infty} R^{2n} |r = nd\rangle$, $R$ being the reflectivity and $d$ the distance between the panels.

In the case of cascades, the consecutive decays will produce a many photon wavefunction. Nevertheless, we can still apply our theory by using the many photon real space base $|r_1, r_2, ...\rangle$. Coincidence measurements such as applied in [12] can now be represented by a measurement base of $|r_0, r_0 + c\tau\rangle$, with $\tau$ as the delay time.

V. CONCLUSIONS

We have presented an exactly solvable model for the unitary temporal evolution of the total system-field state. The assumptions used in the model are equivalent to the approximations of Wigner-Weisskopf theory. However, our approach enables us to provide a physical interpretation of these approximations in terms of locality and constant group velocity.

Also, it is possible to visualize the evolution of the wave function in space and time, allowing us to take a closer look both at time resolved spectroscopy and at the effect of interferometric filters. This clearly reveals the special role of the measurement process in
quantum mechanics, which is often concealed by the application of approximations such as the bath approximation or the omission of correlations.

The possibility of adapting the model to different situations may provide an opportunity to study the emission process in complex quantum systems such as molecules and nanostuctures.
REFERENCES

* Present adress: Institute of Technical Physics, Pfaffenwaldring 38-40, 70569 Stuttgart, Germany

[1] J.Dalibard, Y.Castin and K.Molmer, Phys. Rev. Lett. 68, 580 (1992)

[2] M.Keller, G.Mahler, J. mod. Optics 1994 (in press); G.C. Hegerfeldt, Phys. Rev. A 47, 449 (1993)

[3] It should be noted, that a limitation to positive energies may also cause nonlocal effects as discussed in G.C.Hegerfeldt, Phys. Rev. Lett. 72, 596 (1994). However, we will neglect such effects, since they will be quite small for the reasons already mentioned.

[4] C.Cohen-Tanoudji, B.Diu, F.Laloé, Quantum Mechanics 2 (John Wiley & Sons, 1977)

[5] The local interpretation of the photon wavefunction is not uncrirical, since there is no transversal delta function. However, the one dimensional model should represent the propagation of a photon in a light wave guide quite well. In three dimensions, we transform only the radial component to real space, while the angular dependence, which ensures transversality, remains unchanged.

The local interpretation can further be justified on the ground that the coupling between system and field is local. Since the measurement apparatus will interact with the field in a similar fashion, it is natural to assume real space locality also for this process.

[6] G.C.Hegerfeldt, M.B.Plenio, Quantum Optics 6, 15 (1994)

[7] Blum, Density Matrix Theory and Applications (Plenum Press, New York and London, 1981)

[8] L.D.Landau, E.M.Lifschitz, Lehrbuch der Theoretischen Physik (Akademie Verlag, 1991)

[9] J.Gea-Banacloche, M.O.Scully and M.S.Zubairy, Physica Scripta T 21, 81 (1988)

[10] M.O.Scully and K.Druelle, Phys. Rev. A 25(4), 2208 (1982)
[11] M. Hillery and M. O. Scully in P. Meystre, M. O. Scully (ed.) *Quantum Optics, Experimental Gravitation and Measurement Theory*, (Plenum Press, 1983)

[12] A. Aspect, J. Dalibard, P. Grangier and G. Roger, Opt. Com. 49 (6), 429 (1984)
FIGURES

Fig. 1. Extension of the linear dispersion $\omega = |k|c$ (solid line) to negative Frequencies (dashed line). The dotted line marks the resonant frequency of the system.

Fig. 2. Spatial distribution of the probability of photon detection for $t = ln2/\Gamma$ (solid line), $t = 2ln2/\Gamma$ (dashed line) and $t = 3ln2/\Gamma$ (dotted line).

Fig. 3. Spatial distribution of the probability of photon detection for linearly polarized photons in a quantum beat experiment.

Fig. 4. Beats correlated with the $|P_x\rangle$ state (solid line) and the $|P_y\rangle$ state (dashed line) of the atomic system. The dotted line shows the sum of both probability densities, which displays no beats.
