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Study of transitions induced by squeezed light via path integral methods

E G Thrapsaniotis

Theoretical and Physical Chemistry Institute, National Hellenic Research Foundation,
48 Vasileos Constantinou Avenue, 11635 Athens, Greece

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Abstract. In the present paper we study transitions induced by squeezed light generated by an optical device as a degenerate parametric amplifier. We adopt the dipole approximation and approach the problem by path integral methods. The light variables do not appear in the final propagator as they are integrated over. Using perturbation theory, we calculate the transition probability from the ground state $1s$ of the hydrogen atom to the state $3d$. Further we obtain the equation obeyed by a free electron in squeezed light.

1. Introduction

In recent years considerable effort has been made in applying path integral methods in quantum optics. Certain dynamical groups have been studied [1–5] and the propagators of particular forms of Hamiltonians [16–18] describing non-classical states have been obtained exactly [6–11]. Other Hamiltonians can and have been investigated only numerically via methods such as the Monte Carlo.

On the other hand, certain non-classical states have been achieved in the laboratory. For almost a decade quadrature-squeezed light has been produced experimentally. The interaction of non-classical light with matter appears as a challenging area of research.

We are going to study the influence of non-classical and particularly finite-bandwidth squeezed light generated by a degenerate parametric amplifier on an atomic or molecular bound state [12–15].

The paper proceeds in the following order. In section 2 we describe the full Hamiltonian of an electron in the non-classical field and in the presence of a potential to be specified at will, we give the full propagator and integrate over the field variables. Then we discuss the propagator obtained and obtain the equation of motion of a free electron in squeezed light. In section 3 and by application we derive the transition rate, from the state $1s$ to the state $3d$ of the hydrogen atom under the action of the squeezed light.

2. System Hamiltonian and path integration

The full Hamiltonian H can be written as the sum of three terms. The electron Hamiltonian, H_e in the potential $V(\mathbf{r})$, the squeezed field one H_f and the interaction term H_I :

$$H = H_e + H_f + H_I. \quad (1)$$

Particularly the electron Hamiltonian is given as

$$H_e = \frac{\mathbf{p}^2}{2} + V(\mathbf{r}). \quad (2)$$

The Hamiltonian of squeezed light has the form

$$H_f(t) = \omega(t)a^+a + f(t)a^2 + f^*(t)a^{+2}. \quad (3a)$$

We note that in the case of production of squeezed light by a degenerate parametric amplifier the squeezed light Hamiltonian is given as

$$H_f(t) = \omega a^+a + \kappa(e^{-2i\omega t}a^2 + e^{-2i\omega t}a^{+2}). \quad (3b)$$

Finally, the interaction Hamiltonian in the length form is given as

$$H_I = -e\mathbf{r} \cdot \mathbf{E}_f(\mathbf{r}). \quad (4)$$

The second quantized form of the field operator of squeezed light is given as

$$\mathbf{E}_f(\mathbf{r}) = \frac{1}{\sqrt{V}} i l(\omega) \hat{\varepsilon} [\hat{a} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} - \hat{a}^+ e^{-i(\mathbf{k} \cdot \mathbf{r} - \omega t)}] \quad (5)$$

where V is the quantization volume and $l(\omega)$ is a real function of frequency given as: $l(\omega) = \sqrt{\omega/2}$.

In the dipole approximation ($e^{i\mathbf{k} \cdot \mathbf{r}} \approx 1$ in (5)) which we adopt here, since the spatial dimension of the atom radiated is much less than the wavelength of the electromagnetic field inducing the transition, the field operator can be written as

$$\mathbf{E}_f = \frac{1}{\sqrt{V}} i l(\omega) \hat{\varepsilon} (\hat{a} e^{-i\omega t} - \hat{a}^+ e^{i\omega t}) \quad (6)$$

and H_I takes the form:

$$H_I = -\frac{1}{\sqrt{V}} i e l(\omega) \hat{\varepsilon} \cdot \mathbf{r} (a e^{-i\omega t} - a^+ e^{i\omega t}). \quad (7)$$

Now we combine the terms (3) and (7) involving field variables in the term:

$$H_0(a^+, a; t) = H_f + H_I = \omega(t)a^+a + f(t)a^2 + f^*(t)a^{+2} + g(t)a + g^*(t)a^+ \quad (8)$$

where

$$g(t) = -\frac{1}{\sqrt{V}} i e l(\omega) \hat{\varepsilon} \cdot \mathbf{r}(t) e^{-i\omega t}. \quad (9)$$

The propagator corresponding to (8) has been derived via path integral methods by Hillery and Zubairy [6]. Here we use their result to obtain the full propagator corresponding to H , with the field variables appearing in (8) integrated, thus resulting in a path integral of only the spatial variables. It is given by the expression

$$K(\alpha_f, \mathbf{r}_f; \alpha_i, \mathbf{r}_i; t) = \int_{\mathbf{r}(0)=\mathbf{r}_i}^{\mathbf{r}(t)=\mathbf{r}_f} D\mathbf{r}(t) \times \exp \left[\begin{aligned} & -i \int_0^t d\tau \left[\frac{\dot{\mathbf{r}}^2(\tau)}{2} - V(\mathbf{r}(\tau)) \right] - i \int_0^t d\tau [2f(\tau)X(\tau) \\ & + f(\tau)Z^2(\tau) + g(\tau)Z(\tau)] - \frac{1}{2} (|\alpha_f|^2 + |\alpha_i|^2) \\ & + Y(t)\alpha_f^* \alpha_i + X(t)(\alpha_f^*)^2 - i\alpha_i^2 \int_0^t d\tau f(\tau)Y^2(\tau) + Z(t)\alpha_f^* \\ & - i\alpha_i \int_0^t d\tau [g(\tau) + 2f(\tau)Z(\tau)]Y(\tau) \end{aligned} \right] \quad (10)$$

where $X(t)$ satisfies the following Riccati differential equation:

$$\frac{dX}{dt} = -2i\omega(t)X - 4if(t)X^2 - if^*(t) \tag{11}$$

with initial condition $X(0) = 0$ which can be solved analytically if we can express $f(t)$ as

$$f(t) = h(t) \exp \left[2i \int_0^t d\tau \omega(\tau) \right]$$

where $h(t)$ is real or imaginary.

The $Y(t)$ and $Z(t)$ are given as

$$Y(t) = \exp \left[-i \int_0^t d\tau [\omega(\tau) + 4f(\tau)X(\tau)] \right] \tag{12}$$

$$Z(t) = -i \int_0^t d\tau [g^*(\tau) + 2g(\tau)X(\tau)] \exp \left[-i \int_\tau^t d\tau' [\omega(\tau') + 4f(\tau')X(\tau')] \right]. \tag{13}$$

Now we consider the special case of the Hamiltonian (3), describing squeezed light generated by a degenerate parametric amplifier, by setting

$$f(t) = \kappa e^{2i\omega t}. \tag{14}$$

Then (8) is written as

$$H_0(a^+, \alpha; t) = H_f + H_I = \omega a^+ a + \kappa e^{2i\omega t} a^2 + \kappa e^{-2i\omega t} a^{+2} + g(t)a + g^*(t)a^+ \tag{8b}$$

and by using equations (11)–(13) we obtain

$$X(t) = \frac{1}{2i} e^{-2i\omega t} \tanh(2\kappa t) \tag{15}$$

$$Y(t) = e^{-i\omega t} \operatorname{sech}(2\kappa t) \tag{16}$$

$$Z(t) = \frac{1}{\sqrt{V}} e l(\omega) \int_0^t d\tau \hat{\epsilon} \cdot \mathbf{r}(\tau) \zeta(\tau, t). \tag{17}$$

Where the function $\zeta(\tau, t)$ in (17) is given as

$$\zeta(\tau, t) = [e^{2i\omega\tau} + ie^{-2i\omega\tau} \tanh(2\kappa t)] e^{-i\omega t} \frac{\cosh(2\kappa\tau)}{\cosh(2\kappa t)}. \tag{18}$$

At this point we perform the integration over the field variable α to obtain the reduced propagator which describes the electron for a field transition from vacuum to vacuum. It is given as

$$\tilde{K}(\mathbf{r}_f, \mathbf{r}_i, t) = \int d^2\alpha K(\alpha_f, \mathbf{r}_f; \alpha_i, \mathbf{r}_i; t)_{\alpha_f=\alpha_i=\alpha}. \tag{19}$$

The propagator of (10) with diagonal field variables can be written as

$$K(\alpha_f, \mathbf{r}_f; \alpha_i, \mathbf{r}_i; t)_{\alpha_f=\alpha_i=\alpha} = \int_{\mathbf{r}(0)=\mathbf{r}_i}^{\mathbf{r}(t)=\mathbf{r}_f} D\mathbf{r}(t) \exp \left[-i \int_0^t d\tau \left[\frac{\mathbf{v}^2}{\tau} (u - V(\mathbf{v}(\tau))) \right. \right. \\ \left. \left. - \frac{1}{2} \ln \cosh(2\kappa t) + A - B|\alpha|^2 + \frac{C}{2}\alpha^{*2} + \frac{C_1}{2}\alpha^2 + D_1\alpha + D\alpha^* \right] \right] \tag{20}$$

where

$$D(t) = \frac{1}{\sqrt{V}} e l(\omega) \int_0^t d\tau \hat{\epsilon} \cdot \mathbf{r}(\tau) \zeta(\tau, t) \tag{21a}$$

$$D_1(t) = -\frac{1}{\sqrt{V}} e l(\omega) \int_0^t d\tau \hat{\epsilon} \cdot \mathbf{r}(\tau) [Y(\tau) e^{-i\omega t} + i\theta(\tau, t)] \tag{21b}$$

$$C_1(t) = -i \tanh(2\kappa t) \tag{21c}$$

$$C(t) = -ie^{-2i\omega t} \tanh(2\kappa t) \tag{21d}$$

$$B(t) = 1 - Y(t) \tag{21e}$$

$$A(t) = -\frac{1}{V} e^2 l^2(\omega) \int_0^t d\tau \int_0^\tau d\rho \hat{e} \cdot \mathbf{r}(\tau) \hat{e} \cdot \mathbf{r}(\rho) [\zeta(\rho, \tau) e^{-i\omega\tau} + i\lambda(t, \tau, \rho)] \tag{21f}$$

$\theta(\tau, t)$ and $\lambda(t, \tau, \rho)$ are given as

$$\theta(\tau, t) = [e^{2i\omega\tau} + ie^{-2i\omega\tau} \tanh(2\kappa\tau)] \cosh(2\kappa\tau) [\tanh(2\kappa t) - \tanh(2\kappa\tau)] \tag{21g}$$

$$\lambda(t, \tau, \rho) = [e^{2i\omega\tau} + ie^{-2i\omega\tau} \tanh(2\kappa\tau)] [e^{2i\omega\rho} + ie^{-2i\omega\rho} \tanh(2\kappa\rho)] \times \cosh(2\kappa\tau) \cosh(2\kappa\rho) [\tanh(2\kappa t) - \tanh(2\kappa\rho)]. \tag{21h}$$

At this point we give the identity

$$\int \exp \left[-B|\beta|^2 + \frac{C}{2} \beta^{*2} + \frac{C_1}{2} \beta^2 + D_1\beta + D\beta^* \right] d^2\beta = \frac{\pi}{\sqrt{K}} \exp \left\{ \frac{1}{K} \left[DD_1B + D^2 \frac{C_1}{2} + D_1^2 \frac{C}{2} \right] \right\} \tag{22a}$$

where

$$K = B^2 - CC_1 = 1 - 2e^{-i\omega t} \operatorname{sech}(2\kappa t) + e^{-2i\omega t} \tag{22b}$$

and which is valid under the restrictions

$$\operatorname{Re} K > 0 \quad \text{and} \quad \operatorname{Re} \left[B + \frac{C_1 + C}{2} \right] > 0. \tag{22c}$$

Then the produced propagator can be written as

$$\tilde{K}(\mathbf{r}_f, \mathbf{r}_i, t) = \frac{\pi}{\sqrt{N(t)}} \int_{\mathbf{r}(0)=\mathbf{r}_i}^{\mathbf{r}(t)=\mathbf{r}_f} D\mathbf{r}(t) \exp\{iS_{\text{tot}}[\mathbf{r}(t)]\} \tag{23a}$$

where

$$N(t) = \cosh(2\kappa t) - 2e^{-i\omega t} + \cosh(2\kappa t)e^{-2i\omega t} \tag{23b}$$

and

$$S_{\text{tot}}[\mathbf{r}(t)] = \int_0^t \left[\frac{\dot{\mathbf{r}}^2(\rho)}{2} - V(\mathbf{r}(\rho)) \right] d\rho + \frac{1}{V} e^2 l^2(\omega) \int_0^t d\rho \int_0^\rho d\sigma \hat{e} \cdot \mathbf{r}(\rho) \hat{e} \cdot \mathbf{r}(\sigma) \phi(t, \rho, \sigma) \tag{23c}$$

$\phi(t, \rho, \sigma)$ is given as

$$\phi(t, \rho, \sigma) = -\zeta(\sigma, \rho) e^{-i\omega\rho} - i\lambda(t, \rho, \sigma) - \frac{1}{K} \begin{bmatrix} B(t)\zeta(\rho, t)[Y(\sigma) e^{-i\omega\sigma} + i\theta(\sigma, t)] \\ + B(t)\zeta(\sigma, t)[Y(\rho) e^{-i\omega\rho} + i\theta(\rho, t)] \\ + C_1(t)\zeta(\rho, t)\zeta(\sigma, t) \\ + C(t)[Y(\rho) e^{-i\omega\rho} + i\theta(\rho, t)][Y(\sigma) e^{-i\omega\sigma} + i\theta(\sigma, t)] \end{bmatrix}. \tag{23d}$$

The above path integral can be treated perturbatively to obtain any transition probability we require. Additionally as a by-product we obtain the equation of motion of a free electron in squeezed light from the first variation of the action (23c) without a potential term. It is given as

$$\ddot{\mathbf{r}}(\rho) = \frac{1}{V} 2e^2 l^2(\omega) \hat{e} \int_0^\rho d\sigma \hat{e} \cdot \mathbf{r}(\sigma) \phi(t, \rho, \sigma). \tag{24}$$

3. Application to the hydrogen atom

Now as an application we calculate the transition probability from the ground state 1s of the hydrogen atom to the state 3d. This corresponds to the calculation of a transition amplitude, i.e. the calculation of a certain matrix element between the initial and the final state. Subsequently the limit $t \rightarrow \infty$ is taken so that the transition has been achieved. In the calculations we use for κ appearing in (3b) the value of 0.9 MHz.

In that case the potential in (23c) is given as

$$V(\mathbf{r}) = -\frac{1}{r}. \quad (25)$$

We proceed by first giving the fixed-energy amplitude corresponding to the first two terms in (23c) as [19]

$$G'_0(\mathbf{r}, \mathbf{r}'; E) = \sum_{l=0}^{\infty} g'_l(r, r'; E) \sum_{m=-l}^l Y_{lm}(\hat{r}) Y_{lm}^*(\hat{r}') \quad (26a)$$

where

$$g'_l(r, r'; E) = -\frac{u}{rr'} \frac{\Gamma(1+l-u)}{\Gamma(2l+2)} M_{u, l+\frac{1}{2}}\left(\frac{2r_{>}}{u}\right) W_{u, l+\frac{1}{2}}\left(\frac{2r_{<}}{u}\right) \quad (26b)$$

$$r_{>} = \max(r, r') \quad \text{and} \quad r_{<} = \min(r, r') \quad (26c)$$

$$u = \frac{1}{(-2E)^{1/2}} \quad (26d)$$

in atomic units.

$M_{u, l+\frac{1}{2}}(r)$ is the regular and $W_{u, l+\frac{1}{2}}(r)$ the irregular Whittaker function.

The bound-state part of the radial fixed energy amplitude can be written as

$$g'_{l, \text{bound}}(r, r'; E) = \sum_{n=l+1}^{\infty} \frac{i}{E - E_n} R_{nl}(r) R_{nl}(r'). \quad (27)$$

To a first approximation we use the fixed energy amplitude in the form (27).

The time-dependent function $N(t)$ appearing in (23a) can be incorporated in the fixed energy amplitude by using equations (26a) and (27) in the transform (28a) and the numerical fast Fourier transform in (28b) given below

$$K'_0(\mathbf{r}_f, \mathbf{r}_i, t) = \int_{-\infty}^{+\infty} \frac{dE}{2\pi} e^{-iEt} G'_0(\mathbf{r}_f, \mathbf{r}_i, E) \quad (28a)$$

$$G_0(\mathbf{r}_f, \mathbf{r}_i, E) = \int_0^{+\infty} dt e^{-iEt} \frac{\pi}{\sqrt{N(t)}} K'_0(\mathbf{r}_f, \mathbf{r}_i, t). \quad (28b)$$

The total fixed energy amplitude is given as

$$G(\mathbf{r}, \mathbf{r}'; E) = G_0(\mathbf{r}, \mathbf{r}'; E) - i \int G_0(\mathbf{r}, \mathbf{r}''; E) W(\mathbf{r}''; E - E') G(\mathbf{r}'', \mathbf{r}'; E') d^3r'' dE' \quad (29)$$

where

$$W(\mathbf{r}, E) = \int d\rho e^{iE\rho} W(\mathbf{r}, \rho) \quad (30a)$$

and

$$W(\mathbf{r}, \rho) = \frac{1}{V} \alpha l^2(\omega) \hat{\mathbf{e}} \cdot \mathbf{r}(\rho) \int_0^\rho d\sigma \hat{\mathbf{e}} \cdot \mathbf{r}(\sigma) \phi(t, \rho, \sigma) \quad (30b)$$

where α is the fine structure constant.

Now we perform the Markov approximation on (30b) and obtain the expression

$$W(\mathbf{r}, \rho) = \frac{1}{V} \alpha l^2 (\omega) (\hat{\mathbf{e}} \cdot \mathbf{r}(\rho))^2 v(t, \rho) \quad (31a)$$

where

$$v(t, \rho) = \int_0^\rho d\sigma \phi(t, \rho, \sigma). \quad (31b)$$

Up to first order in W the fixed energy propagator is given as

$$G(\mathbf{r}, \mathbf{r}'; E) = G_0(\mathbf{r}, \mathbf{r}'; E) - i \int G_0(\mathbf{r}, \mathbf{r}''; E) W(\mathbf{r}'', E - E') G_0(\mathbf{r}'', \mathbf{r}'; E') d^3 r'' dE' \quad (32)$$

and the transition amplitude $A_{fi}(t)$ from the state $\psi_i(\mathbf{r})$ to $\psi_f(\mathbf{r})$ is

$$A_{fi}(t) = -\frac{1}{2\pi i} \int_{-\infty}^{+\infty} dE \langle \psi_f(\mathbf{r}) | G(\mathbf{r}, \mathbf{r}'; E) | \psi_i(\mathbf{r}') \rangle e^{-i(E-E_f)t} \quad (33)$$

where E_f is the unperturbed energy of the state $|\psi_f\rangle$.

In the present applied case of the hydrogen atom and in atomic units the initial 1s state is given as

$$|\psi_i(\mathbf{r})\rangle = R_{10}(r) Y_{00}(\hat{r}) = 2e^{-r} \frac{1}{\sqrt{4\pi}} \quad (34a)$$

while the final 3d state is given as

$$|\psi_f(\mathbf{r})\rangle = R_{32}(r) Y_{20}(\hat{r}) = \frac{4}{81\sqrt{30}} e^{-r/3} r^2 \sqrt{\frac{5}{4\pi}} \left(\frac{3}{2} \cos^2 \theta - \frac{1}{2} \right). \quad (34b)$$

The presence of only the $m = 0$ quantum number in the final state is explained via the application of the Wigner–Eckart theorem (see later).

Now we study the matrix element appearing in (33). On substituting the expression (32) in (33) we observe that the first term on the right-hand side of (32) is reduced to zero as it contains only diagonal matrix elements. As far as the second term is concerned, on taking the matrix elements in (33) only one pole survives from each Green function in (26a–d) and (27) and the calculations in (28a, b) are straightforward. Taking into account that the non-diagonal matrix elements of the Green function in (26a–d) and (27) are zero we have simply to evaluate the matrix element

$$\langle 320 | W(\mathbf{r}, E) | 100 \rangle = \int d\rho e^{iE\rho} \langle 320 | W(\mathbf{r}, \rho) | 100 \rangle \quad (35)$$

where

$$|nlm\rangle = R_{nl}(r) Y_{lm}(\hat{r}). \quad (36)$$

As the time dependence of the radial function is concerned, it can be transferred to the bra and ket states with which it is bracketed via the relation

$$\mathbf{r}(t) = \exp(iH_0 t) \mathbf{r}(0) \exp(-iH_0 t) \quad (37)$$

where H_0 is the Hamiltonian related with the action

$$S[\mathbf{r}(t)] = \int_0^t \left[\frac{\dot{\mathbf{r}}^2(\rho)}{2} - V(\mathbf{r}(\rho)) \right] d\rho. \quad (38)$$

On choosing the z -axis in the direction of the polarization vector, and inserting a complete set of states between the two radial vectors in (31a) we obtain the usual selection rules $l' = l \pm 1$, $m' = m$ for each matrix element after performing the proper integrations.

We see posteriorly that the final state has correctly been chosen. Use has been made of the linear polarization case in the Wigner–Eckart theorem, i.e.

$$\langle nlm|\hat{\epsilon} \cdot \mathbf{r}(0)|n'l'm'\rangle = \frac{1}{\sqrt{2l'+1}} \langle lm; 10|l'm'\rangle \langle n|r(0)|n'\rangle. \quad (39)$$

Now after performing the remaining integrations and using the fact that the transition probability per second is given by $d|A_{fi}(t)|^2/dt$, we obtain a value of this equal to $1.49 \times 10^{16} \text{ s}^{-1}$ or equal to $3.61 \times 10^{-1} \text{ a.u.}$ for the transition $1s \rightarrow 3d$ mentioned above.

4. Conclusions

In the present paper we investigate transitions induced by squeezed light, generated, for example, by a degenerate parametric amplifier. In such a case two photons appear almost ‘simultaneously’ called the signal and idler photons respectively. We use path integral methods and treat the component of the action involving the squeezed electromagnetic field terms using perturbation theory. In fact in the calculations we have made the Born–Markov approximation. We apply the author’s methods to the case of a transition in the hydrogen atom. These methods are tractable and we believe that they give a new aspect on the interaction of radiation with matter. In the future, we plan to extend the present theory to the case of many electron atoms as well as many squeezed modes.

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