Nonlinear effect in processes of the interaction of atoms with a resonant laser field

R Kh Gainutdinov and A A Mutygullina
Department of Physics, Kazan Federal University, 18 Kremlevskaya St., Kazan 420008, Russia
E-mail: Renat.Gainutdinov@kpfu.ru

Abstract. We show that for some intensities of a driving laser field not only the laser modes but also the vacuum ones are involved into the strong atom-laser interaction and this has a significant effect on processes of resonance fluorescence.

1. Introduction

The problem of interaction of coherent light with matter is of great interest both from the fundamental point of view as well as for applications. One of the processes that are of fundamental importance to the understanding of this interaction is resonance fluorescence from atoms in an intense laser field. As was predicted by Mollow [1] and confirmed by a number of experiments [2], at relatively high laser intensities the fluorescence spectrum should consist of a symmetric triplet whose side components are separated from the central peak by a distance equal to the generalized Rabi frequency. The strong interaction of an atom with the laser field resonant with an atomic transition is successfully treated within the quantum optics formalism [3, 4]. In describing resonance fluorescence it is usually assumed that only the laser modes are involved into the processes of the strong interaction while other modes including the vacuum ones may be treated as small corrections [5, 6]. However, as has been shown in [7] the Lamb shift in the laser-dressed atoms may be much larger than the Lamb shifts in bare atoms. The aim of the present paper is to show that in some cases not only the atom-laser interaction but also the interaction of the bound electrons with the vacuum modes is strong, and this has a significant effect on the character of the atom-laser interaction.

Let us consider an electron interacting with an atomic nuclear and an intense laser field. It is natural to include the Coulomb field into the free Hamiltonian. This leads us to the Furry picture in which the eigenstates $|m\rangle$ of the Dirac-Coulomb Hamiltonian $H_0^F$ ($H_0^F|m\rangle = E_m|m\rangle$) are used as "free" states, and $H_0^F$ plays the role of a "free" Hamiltonian. When the laser mode frequency $\omega_L$ is close to the frequency $\omega_R = \omega_e - \omega_g$ of the transition between two bound states $|e\rangle$ and $|g\rangle$ the atom-laser interaction is strong. In quantum optics this interaction is usually described by using the two-level model and the rotating wave approximation (RWA). In this approximation the laser-dressed states are the eigenstates of the system Hamiltonian $H_{RW_A}$ [4] and are given by

$$|+, n\rangle = \cos \theta_n |e, n\rangle + \sin \theta_n |g, n+1\rangle;$$
$$|-, n\rangle = -\sin \theta_n |e, n\rangle + \cos \theta_n |g, n+1\rangle.$$
Here $|e,n\rangle$ ($|g,n\rangle$) denotes the state of the combined laser-atom system containing the atom in the bare state $|e\rangle$ ($|g\rangle$) and $n$ photons in the laser mode, and $\theta_n$ is the mixing angle defined by $\tan(2\theta_n) = -\Omega_n/\Delta$ with $\Omega_n$ and $\Delta$ being the Rabi frequency $\Omega_n = 2g_L\sqrt{n+1}$ and detuning, respectively. The energies of the dressed states are given by $E_{\pm,n} = (n+1/2)\omega_L + (\omega_e - \omega_g)/2 \pm \Omega_R^{(n)}/2$ with $\Omega_R^{(n)}$ being the generalized Rabi frequency $\Omega_n = \sqrt{\Omega_n^2 + \Delta^2}$. As the laser field is assumed to be sufficiently intense, $\Omega_n$, $\Omega_R^{(n)}$ and $\theta_n$ are replaced by their semiclassical entities $\Omega_0$, $\Omega_R$ and $\theta$ in the following discussion. Now we can generalize the Furry picture by adding to the above "free" states the laser-dressed states or, in other words, by adding the interaction that is described by $H_{RW,A}$ into the "free" Hamiltonian in the Furry picture $H_0^F \rightarrow H_0^D$. In this case the "free" Green operator $G_0^F(z) = (z-H_0^F)^{-1}$ is replaced by the operator $G_0^D(z) = (z-H_0^D)^{-1}$, and the eigenvectors of $H_0^D$ ($H_0^D|m\rangle = E_m|m\rangle$, $H_0^D|\pm,n;m\rangle = (E_{\pm,n}+E_m)|\pm,n;m\rangle$, $|m\rangle|\pm,n;m\rangle = 0$), can be used as basic vectors in the Fock space.

2. The Generalized Quantum Dynamics

In our investigation of the problem we do not assume a priori that the QED corrections are small. As will be shown below, a natural way to solve the problem in this case is provided by the generalized dynamical equation (GDE), which in [8] has been derived as a direct consequence of the first principles of quantum physics. Being equivalent to the Schrödinger equation in the case when the interaction in a quantum system is instantaneous, the GDE allows one to extend quantum dynamics to the case of nonlocal-in-time interactions. Generalized quantum dynamics (GQD) developed in this way provides a new insight into the many problems in quantum physics [9–13]. In the GQD a history of a physical process is represented by some version of the time evolution of the system associated with completely specified instants of the beginning and end of the interaction in the system [8]:

$$U(t, t_0) = 1 + \int_{t_0}^{t} dt_2 \int_{t_0}^{t_2} dt_1 \tilde{S}(t_2, t_1) \tag{1}$$

where $\tilde{S}(t_2, t_1)$ describes a contribution to the evolution operator $U(t, t_0)$ from the process in which the interaction begins at time $t_1$ and ends at time $t_2$. The GDE, which has the form

$$\tilde{S}(t_2, t_1) = \int_{t_1}^{t_2} dt_4 \int_{t_1}^{t_4} dt_3 \int_{t_2-t_1}^{t_4-t_3} dt_2 \tilde{S}(t_2, t_4) \tilde{S}(t_3, t_1),$$

allows one to obtain $\tilde{S}(t_2, t_1)$ for any $t_1$ and $t_2$, if the operator $\tilde{S}(t_2', t_1')$ corresponding to infinitesimal duration times $\tau = t_2' - t_1'$ of interaction are known. Most of the contribution to the evolution operator in the limit $\tau \rightarrow 0$ comes from processes associated with the fundamental interaction in the system under study, which can be parameterized as an interaction operator $H_{int}(t_2, t_1)$ [8]. This gives rise to the following boundary condition for $\tilde{S}(t_2, t_1)$:

$$\tilde{S}(t_2, t_1) \rightarrow H_{int}(t_2, t_1).$$

In the Schrödinger picture the evolution operator (1) can be represented in the form [9]

$$U_s(t, 0) = \frac{i}{2\pi} \int_{-\infty}^{\infty} dz \exp(-izt)G(z),$$

with $G(z) = G_0(z) + G_0(z)T(z)G_0$. Here $G_0(z) \equiv (z-H_0)^{-1}$ is the free Green operator and the operator $T(z)$ is defined as

$$T(z) = \int_{0}^{\infty} d\tau \exp(-iz\tau)\tilde{T}(\tau).$$
where \( \tilde{T}(t_2 - t_1) = \exp(-iH_0t_2)\tilde{S}(t_2, t_1)\exp(iH_0t_1) \). In terms of the operator \( T(z) \) the GDE and the corresponding boundary condition take the form [8]

\[
\frac{dT(z)}{dz} = -T(z)(G_0(z))^2T(z),
\]

(2)

\[
T(z) \rightarrow B(z) \equiv i \int_0^\infty d\tau \exp(iz\tau)\tilde{B}(\tau),
\]

(3)

with \( \tilde{B}(\tau) = \exp(-iH_0t_2)H_{\text{int}}(t_2, t_1)\exp(iH_0t_1) \). The contribution to the Green operator \( G(z) \), which comes from the processes associated with the self-interaction of the particles, has the same structure as the free Green operator \( G_0(z) \). For this reason it is natural to replace \( G_0(z) \) by the propagator \( \tilde{G}_0(z) \), which describes the evolution of particles interacting only with the vacuum and hence has the structure

\[
\tilde{G}_0(z) = (z - H_0 - C(z))^{-1},
\]

where the operator \( C(z) \) is determined by the equation \( C(z)|n\rangle = C_n(z)|n\rangle \) with \( |n\rangle \) being the eigenvectors of the free Hamiltonian \( H_0|n\rangle = E_n|n\rangle \) and the condition \( z - E_n - C_n(z) = 0 \) determines the physical masses of the particles. Correspondingly, the operator \( T(z) \) should be replaced by the operator \( M(z) \) which describes the evolution of particles interacting not only with the vacuum. These operators are related as follows:

\[
G_0(z) + G_0(z)T(z)G_0(z) = \tilde{G}_0(z) + \tilde{G}_0(z)M(z)\tilde{G}_0(z).
\]

By using this relation, one can rewrite (2) in terms of \( M(z) \) and \( C_n(z) \), satisfying the boundary conditions

\[
M(z) \rightarrow B_r(z), \quad C_n(z) \rightarrow \langle n|B_\delta(z)|n\rangle.
\]

Here \( B_r(z) \) is the part of the interaction operator describing the proper interaction between the particles, and the "singular" part \( B_\delta(z) \) describes their self-interaction. This trick was firstly used in [14]. The features of the above representation of the GDE is very useful for solving the bound-state problem in QED. In order to obtain the self-energy corrections to the bound states of an atom dressed by the resonant laser field one should start from the operator \( G_0^R(z) \) that in the generalized Furry picture plays the role of the "free" Green operator. In this case the equation \( z - E_m - C_m(z) = 0 \) determines the self-energy corrections to the atomic energy levels.

3. Vacuum modes and the strong atom-laser interaction

The resonant laser field has an effect on the self-interaction of the atom only in the states \(|e\rangle \) and \(|g\rangle \). For this reason in solving the problem we can include the self-energy corrections to the energies of other states from the very beginning. In this case only the self-energy functions \( C_{\pm,n}(z) \) do not equal to zero, and the above reduction of Eq. (2) gives rise to the equations

\[
\frac{dC_{\pm,n}(z)}{dz} = -(\pm, n|M(z)(\tilde{G}_0(z))^2M(z)|\pm, n),
\]

(4)

\[
\frac{dM(z)}{dz} = -(1 - P(z)\tilde{G}_0(z))M(z)(\tilde{G}_0(z))^2M(z)(1 - \tilde{G}_0(z)P(z)) + P(z)\tilde{G}_0(z)M(z)(\tilde{G}_0(z))^2M(z)\tilde{G}_0(z)P(z),
\]

(5)

where

\[
P(z) = \sum_{n,m}(P^+_{m}M(z)P^-_{m} + P^-_{m}M(z)P^+_{m}),
\]

\[
P^\pm_{m} = |\pm, n;m\rangle\langle \pm, n;m|,
\]
\[ \tilde{G}_0(z) = (z - H_0^D - C(z))^{-1}, \]
and \( C(z) |\pm, n, m \rangle = C_{\pm,n}(z - E_m)|\pm, n, m \rangle \). In this case the interaction operator in the boundary condition (3) for these equations is of the form \( B(z) = H_I + B_D^F(z) \), where \( H_I = e \int d^3x : A_\mu(0, x)\psi(0, x)\gamma^\mu \psi(0, x) : \); \( \psi(0, x) \) is the Dirac field in the Furry picture, \( A^\mu(x) \) is the electromagnetic field, the symbol : : denotes the normal ordering, and \( B_D^F(z) \) is a nonlocal operator describing the self-interaction of electrons in the generalized Furry picture. In some cases Eqs. (4) and (5) can be solved iteratively. Putting \( M(z) = H_I \) on the right-hand part of Eqs. (4) and (5), for \( (\pm, n|M(z)|\mp, n \rangle \equiv (\pm, n|\Sigma(z)|\mp, n \rangle \) and \( C_{\pm,n}(z) \equiv (\pm, n|\Sigma(z)|\pm, n \rangle \), we get the equation
\[
\frac{d(\psi_2|\Sigma^{(0)}(z)|\psi_1)}{dz} = -\langle \psi_2|H_I(G_0^D(z))^2H_I|\psi_1 \rangle. \tag{6}
\]
In calculating with the accuracy up to order \( o\{\Lambda^2\} \), where \( \Lambda = \omega_R/\alpha^2Z^2m_e \) (in general this parameter is extremely small), \( G_0^F \) in Eq. (6) can be replaced by \( G_0^D(z) \) and the operator \( B_D^F \) is a simple redistribution of \( B_e(z) \) and \( B_g(z) \) being the eigenvalues of the operator \( B_D^F(z) \) describing the self-interaction of the bare atoms \( (B_D^F(z)|\epsilon) = B_{\delta,e}(z)|\epsilon \rangle, B_D^F(z)|g) = B_{\delta,g}(z)|g) \). In this approximation the solution of Eq.(6) is
\[
\begin{align*}
\langle +, n|\Sigma^{(0)}(z)|+, n \rangle &= \cos^2\theta C_c(z') + \sin^2\theta C_g(z''), \\
\langle -, n|\Sigma^{(0)}(z)|-, n \rangle &= \sin^2\theta C_c(z') + \cos^2\theta C_g(z''), \\
\langle \pm, n|M^{(0)}(z)|\mp, n \rangle &= \frac{1}{2}\sin 2\theta (C_c(z') - C_g(z'')) \tag{7},
\end{align*}
\]
where \( z' = z - n\omega_L, z'' = z - (n+1)\omega_L \), and
\[
C_i(z) = \lim_{\mu \to \infty} \langle i|H_I(G_0^F(z) - G_0^D(\mu))H_I|i \rangle + B_{\delta,i}(\mu), \quad i = e, g \tag{9}
\]
Here \( \mu \) plays the role of the regularization parameter, and \( \langle i|B_D^F(\mu)|i \rangle \) manifests itself as a sum of \( \mu \)-dependent counterterms. Thus, the leading order solution of Eqs. (4) and (5) is
\[
\begin{align*}
C^{(0)}_{\pm,n}(z) &\equiv \langle \pm, n|\Sigma^{(0)}(z)|\pm, n \rangle, \\
M^{(0)}(z) &= H_I + L(z), \\
L(z) &= \sum_{n,m} P^+ m(\Sigma^{(0)}(z)P^- m + P^- m\Sigma^{(0)}(z)P^+ m) \tag{10},
\end{align*}
\]
with \( \langle \pm, n|\Sigma^{(0)}(z)|\pm, n \rangle \) being defined by Eqs. (7). The next iteration yields
\[
\begin{align*}
C^{(1)}_{\pm,n}(z) &= C^{(0)}_{\pm,n}(z) + \langle \pm, n|M^{(1)}(z)\tilde{G}_0(z)M^{(1)}(z) - H_I\tilde{G}_0(z)H_I|\pm, n \rangle, \\
M^{(1)}(z) &= M^{(0)}(z) + H_I\tilde{G}_0(z)L(z) + L(z)\tilde{G}_0(z)H_I, \tag{11}
\end{align*}
\]
where \( \tilde{G}_0(z) = z - H_0^D - C^{(0)} \). As it follows from these equations, the correction to the leading order solution is of the relative order \( O(\Lambda_0) \) where \( \Lambda_0 \equiv -\sin(2\theta)(\delta E_g - \delta E_e)/(2\Omega_R) \), and the iterative scheme yields the expansion of the solution of Eqs. (4) and (5) in powers of this parameter. In the case \( \Omega \gg \Gamma_e \) the decay law may be close to an exponential one and for describing the fluorescence spectrum one can use the standard master equations even if the interaction of an atom with the vacuum modes is effectively strong. In this way one can use the simple rules derived in Ref. [15] for evaluating the positions, the heights and the widths of the
various components of the fluorescence spectrum. In particular, these rules relate the widths $\Gamma_{ij}$ of the transitions from $|i, n\rangle$ to $|j, n-1\rangle$ (here and below we use the notation $|1, n\rangle \equiv |-, n\rangle$ and $|2, n\rangle \equiv |+, n\rangle$) with the widths of the central and the sideband components of the fluorescence spectrum respectively $L_{ii} = \Gamma_{ii} + \Gamma_{jj}$, $L_{ij} = L_{ji} = \frac{1}{2} (\Gamma_{i} + \Gamma_{j}) - d_{ij}^{+} \cdot d_{jj}^{+}$, where

$$
\Gamma_{ij} = |d_{ij}^{+}|^{2}, \quad d_{ij}^{\pm} = \langle i, n | D | j, n \mp 1 \rangle,
$$

and $D$ is the dipole momentum operator. Here the factor $\alpha = \sqrt{\frac{2 \hbar}{\pi m}}$ is reincluded in the definition of $D$ in order to get a simple expression for $\Gamma$. In deriving these formulas it is assumed that with the accuracy of order $O(\alpha)$ the transitions between the states $|i, n\rangle$ and $|j, n-1; k, \varepsilon_{\lambda}\rangle$ are described by the corresponding matrix elements of the interaction Hamiltonian $H_I$ in the dipole approximation $H_I = -a^{-1} D \cdot \varepsilon_{\lambda}$ where $k$ and $\varepsilon_{\lambda}$ are the photon momentum and polarization respectively. However, taking into account vacuum modes implies that the coupling of the laser-dressed atom to the radiation field is described by the corresponding matrix elements of the operator $M(z)$, and hence the dipole momentum in Eq. (12) is defined as $\langle j, n-1; k, \varepsilon_{\lambda}| M^{(1)} | i, n\rangle = -a^{-1} \langle j, n-1 | D_{\mu} \cdot \varepsilon_{\lambda}| i, n\rangle$. From Eq. (10) it follows that $M(z)$ is reduced to $H_I$ only at leading order and the higher order corrections can be significant.

4. Outlook

In summary, we have shown that the vacuum modes may be involved into the strong interaction of an atom with a resonant laser field. As it follows from Eq. (8), the influence of this nonlinear effect on resonance fluorescence depends on the self-energy functions $C_{\epsilon}(z)$ and $C_{\eta}(z)$ of the relevant states of the bare atom. In free space the dependence of $C_{\epsilon}(z)$ on $\varepsilon$ in the energy interval between $E_{-, n}$ and $E_{+, n}$ is negligible. This allows one to diagonalize $\Sigma^{(0)}(z)$ in Eq. (10) and to get the solution without further iterations. The solution obtained in this way is reduced to a change of the mixing angle in the definition of the dressed states $\theta_n$. The situation is cardinally changed in the case when the atom is placed in a photonic crystal (PC) being periodic nanostructures. Because of the strong modification of the interaction of an electron in a PC with the radiation field which can even result in the electron mass changes its value [16], the corresponding self-energy functions are significantly changed in the above energy interval and hence in this case the effect can be significant.

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