===================== **NANOPHOTONICS** =====================

Interaction of 3-Level Atom with Radiation[1]

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Abstract—The stochastic limit of quantum theory suggests a new, constructive, approach to nonequilibrium phenomena. We illustrate this approach when considering the interaction of 3-level system with a quantum field in a nonequilibrium state. We describe a class of states of the quantum field for which a stationary state drives the system to an inversely populated state. We find that the quotient of the population of the energy levels in the simplest case is described by the double Einstein formula which involves products of two Einstein emission/absorption factors. Emission and absorption of radiation by 3-level atom in nonequilibrium stationary state is described. © *2003 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

Recent developments of technology greatly improved our ability to control individual quantum systems. This brings quantum technology beyond academic research to the level of concretes industrial programs. For the requirements of such quantum technologies the requirement of stability is essential: it is not only required that at time T the system is in a given quantum state, but also that it remains in this state sufficiently long time to allow the manipulations required by quantum computation. One possible way to achieve this goal is to exploit a general principle of the stochastic limit [1, 2], namely: the interaction of a quantum field with a discrete system (e.g., an N-level atom) drives the system to a stationary state which is uniquely determined by the state of the field. Already now many manipulations on microscopic objects are achieved through their interaction with appropriate fields. Thus the scenario we are proposing simply integrates this approach with the additional requirement of stability. The advantage of the stochastic limit approach is that it gives a quite explicit description of the parameters which control the final state of the system. Therefore, if we are able to act on these parameters with suitably chosen initial state of the field and the interaction we could drive the system in a stable way to a large class of preassigned states.

In the present paper we consider a 3-level system (for example an atom) interacting with radiation. Several nontrivial phenomena arising in this model situation have been already described in different contexts, especially in quantum optics [3–6].

We show that for such a system application of the stochastic limit allows to obtain an interesting effect of inversion of population: for a special choice of the state of the reservoir the system relaxes to a stationary state where the population of the level of the system with higher energy will be larger than the population of the level with lower energy.

We obtain an equation for the number of photons emitted and absorbed by the system.

We investigate examples of 2- and 3-level systems. We show that for a 2-level atom emission in the stationary regime equals absorption. For a 3-level system emission and absorption of radiation are controlled by the state of the field. We find that for a 3-level system in a stationary nonequilibrium state two regimes—emission and absorption—are possible. In the emission regime the total number of quanta in the system increases, and in the absorption regime it decreases. These regimes are controlled by a function $\beta(\omega)$ (cf. (4) below). For example the 3-level system with energy levels $\varepsilon_1 < \varepsilon_2 < \varepsilon_3$ is in emission regime when

$$\beta(\varepsilon_2 - \varepsilon_1) + \beta(\varepsilon_3 - \varepsilon_2) > \beta(\varepsilon_3 - \varepsilon_1),$$

and it is in absorption regime when the opposite inequality holds.

In the emission regime the 3-level system converts radiation of frequency ω_2 into radiation of frequencies ω_1 and ω_3 and vice versa in the absorption regime. The analogies with parametric downconversion and second harmonic generation are suggestive and will be discussed elsewhere.

In particular the stationary state of the 3-level system gives an example of dissipative structure in the Prigogine sense [7].

Equilibrium states of the field are characterized by the property that the function $\beta(\omega)$ is linear ($= \beta\omega + \mu$). In these states the system is in equilibrium with radiation.

The interaction of a quantum system with a quantum field is described by a Hamiltonian

$$H = H_S + H_R + \lambda H_I. \tag{1}$$

The system degrees of freedom are described by the system Hamiltonian

$$H_S = \sum_j \varepsilon_j |\varepsilon_j\rangle\langle\varepsilon_j|. \tag{2}$$

The radiation degrees of freedom are described by the Hamiltonian

$$H_R = \int \omega(k) a^*(k) a(k) dk, \tag{3}$$

where $a(k)$ is a bosonic field with a Gaussian state characterized by

$$\langle a(k)\rangle = \langle a(k)a(k')\rangle = 0,$$

$$\langle a^*(k)a(k')\rangle = N(k)\delta(k-k') = \frac{1}{e^{\beta(\omega(k))}-1}\delta(k-k'), \tag{4}$$

and $\beta(\omega(k))$ is a (none necessarily linear) function.

The interaction Hamiltonian H_I is of dipole type

$$H_I = \int \overline{g(k)} a(k) D^* dk + \text{h.c.}, \tag{5}$$

where D and D^* are operators on the system space.

We investigate the dynamics of this system in the stochastic limit, i.e., in the regime of weak coupling ($\lambda \longrightarrow 0$) and large times. This regime is given by time rescaling $t \mapsto t/\lambda^2$. This rescaling and the interaction (5) lead naturally to introduction the rescaled quantum fields

$$\frac{1}{\lambda} e^{-\frac{it}{\lambda^2}(\omega(k)-\omega)} a(k), \tag{6}$$

where ω are the Bohr frequencies (differences of eigenvalues of the system Hamiltonian H_S).

By the stochastic golden rule [1] the rescaled field (6) in the stochastic limit becomes a quantum white noise or master field $b_\omega(t,k)$ satisfying the commutation relations

$$[b_\omega(t,k), b_{\omega'}^*(t',k')]$$
$$= \delta_{\omega,\omega'} 2\pi\delta(t-t')\delta(\omega(k)-\omega)\delta(k-k'), \tag{7}$$

and with the mean zero and gauge invariant Gaussian state with correlations

$$\langle b_\omega^*(t,k) b_{\omega'}(t',k')\rangle$$
$$= \delta_{\omega,\omega'} 2\pi\delta(t-t')\delta(\omega(k)-\omega)\delta(k-k')N(k), \tag{8}$$

$$\langle b_\omega(t,k) b_{\omega'}^*(t',k')\rangle$$
$$= \delta_{\omega,\omega'} 2\pi\delta(t-t')\delta(\omega(k)-\omega)\delta(k-k')(N(k)+1). \tag{9}$$

In particular white noises, corresponding to different frequencies ω, are independent (stochastic resonance principle).

The Schrödinger equation becomes a white noise Hamiltonian equation, cf. [1, 2] which being put in normal order is equivalent to the stochastic Schrödinger equation

$$dU_t = (-idH(t) - Gdt)U_t, \quad t > 0 \tag{10}$$

with initial condition $U_0 = 1$, and where

(i) $dH(t)$, called the martingale term, is the stochastic differential

$$dH(t) = \int_t^{t+dt} h(s)ds$$
$$= \sum_\omega (E_\omega^*(D)dB_\omega(t) + E_\omega(D)dB_\omega^*(t)) \tag{11}$$

driven by the quantum Brownian motions

$$dB_\omega(t) = \int_t^{t+dt}\int dk \bar{g}(k) b_\omega(\tau,k)d\tau = \int_t^{t+dt} b_\omega(\tau,g)d\tau. \tag{12}$$

(ii) The operator G, called the drift, is given by

$$G = \sum_\omega ((g|g)_\omega^- E_\omega^*(D)E_\omega(D) + \overline{(g|g)}_\omega^+ E_\omega(D)E_\omega^*(D)), \tag{13}$$

where the explicit form of the constants $(g|g)_\omega^\pm$, called the generalized susceptivities, is

$$(g|g)_\omega^- = \int dk |g(k)|^2 \frac{-i(N(k)+1)}{\omega(k)-\omega-i0}$$
$$= \pi\int dk |g(k)|^2 (N(k)+1)\delta(\omega(k)-\omega) \tag{14}$$
$$- i\text{P.P.}\int dk |g(k)|^2 \frac{(N(k)+1)}{\omega(k)-\omega},$$

$$(g|g)_\omega^+ = \int dk |g(k)|^2 \frac{-iN(k)}{\omega(k)-\omega-i0}$$
$$= \pi\int dk |g(k)|^2 N(k)\delta(\omega(k)-\omega) \tag{15}$$
$$- i\text{P.P.}\int dk |g(k)|^2 \frac{N(k)}{\omega(k)-\omega},$$

where P.P. means the Gouchy principal-part integral.

The real part of the generalized susceptivities controls the line-broadening (frequency dependent refraction index, inverse life-times, rates of decoherence) and the imaginary part controls the energy or these shifts. From the stochastic Schrödinger equation, both the Langevin and the master equation are derived by means of a standard procedure.

In the present paper we consider generic quantum system, i.e., such system that for each Bohr frequency ω there exists a unique pair of eigenstates $|1_\omega\rangle$ and $|2_\omega\rangle$ corresponding to the two energy levels, ε_{1_ω}, ε_{2_ω}, so that

$$\omega = \varepsilon_{2_\omega} - \varepsilon_{1_\omega}.$$

In this case

$$E_\omega(D) = \langle 1_\omega|D|2_\omega\rangle|1_\omega\rangle\langle 2_\omega|,$$

$$E_\omega(D)E_\omega^*(D) = |\langle 1_\omega|D|2_\omega\rangle|^2|1_\omega\rangle\langle 1_\omega|, \qquad (16)$$

$$E_\omega^*(D)E_\omega(D) = |\langle 1_\omega|D|2_\omega\rangle|^2|2_\omega\rangle\langle 2_\omega|.$$

We consider a dispersion $\omega(k)$ which is ≥ 0 and, moreover, we suppose that the Lebesgue measure of the set $\{k : \omega(k) = 0\}$ equals to zero. This implies that the real part of the generalized susceptivities $\mathrm{Re}(g|g)_\omega^\pm$ is nonnegative and can be nonzero only for $\omega > 0$.

We will also use the notation $(g|g)_{ij}^\pm$ for $(g|g)_\omega^\pm$ if $\omega = \varepsilon_i - \varepsilon_j$.

In the present paper we investigate the nonequilibrium stationary states for the master equation governing evolution of the diagonal part of the density matrix of a generic quantum system. This equation was obtained in [2] as

$$\frac{d}{dt}\rho(\sigma, t) = \sum_{\sigma' : \varepsilon_{\sigma'} > \varepsilon_\sigma} (\rho(\sigma', t)2\mathrm{Re}(g|g)_{\sigma'\sigma}^-$$
$$- \rho(\sigma, t)2\mathrm{Re}(g|g)_{\sigma'\sigma}^+)|\langle \sigma, D\sigma'\rangle|^2$$
$$\qquad (17)$$
$$+ \sum_{\sigma' : \varepsilon_\sigma > \varepsilon_{\sigma'}} (\rho(\sigma', t)2\mathrm{Re}(g|g)_{\sigma\sigma'}^+$$
$$- \rho(\sigma, t)2\mathrm{Re}(g|g)_{\sigma\sigma'}^-)|\langle \sigma', D\sigma\rangle|^2,$$

where $\rho(\sigma, t) = \rho(\sigma, \sigma, t)$ and $|\sigma\rangle$ are eigenvectors of the system Hamiltonian H_S.

If the system has a finite number of energy levels, then a stationary state for the evolution, driven by the above-mentioned master equation, exists and, if the state of the reservoir is nonequilibrium, then the stationary state does not satisfy the detailed balance condition for the master equation.

The diagonal and the off-diagonal terms of the reduced density matrix evolve separately. The off-diagonal part of the density matrix evolves independently and vanishes exponentially (cf. [2]). This corresponds to the collapse of the initial quantum state to a classical mixed state, described by the diagonal part of the density matrix. The diagonal part $\rho(\sigma, \sigma, t)$ may be considered as a classical distribution function and equation (17) may be considered as a kinetic equation for it.

The structure of the present paper is as follows.

In Section 2 we describe the stationary state for a 3-level atom interacting with radiation found in [2].

In Section 3 we investigate the properties of this stationary state and find that the quotient of the populations of energy levels in the stationary state does not obey the Einstein emission/absorption relations. If the atom is in Λ-configuration the quotient of the populations will obey a new relation that we call the Double Einstein relation.

In Section 4 we use the form of this state to describe the inversion of population in our 3-level system.

In Section 5 we derive a master equation for the density of photons.

In Section 6 we use this equation to investigate emission and absorption of radiation by the system in the obtained nonequilibrium stationary state.

2. STATIONARY LEVEL FOR 3-LEVEL SYSTEM

For a 3-level system with energy states $|1\rangle$, $|2\rangle$, $|3\rangle$, energies $\varepsilon_1 < \varepsilon_2 < \varepsilon_3$, and Bohr frequencies

$$\omega_1 = \varepsilon_2 - \varepsilon_1, \quad \omega_2 = \varepsilon_3 - \varepsilon_1, \quad \omega_3 = \varepsilon_3 - \varepsilon_2,$$

the master equation describes relaxation of the system to a diagonal stationary state whose diagonal elements have the form

$$\rho_1 = |\langle 1|D|2\rangle|^2|\langle 1|D|3\rangle|^2 \frac{I(\omega_1)}{1 - e^{-\beta(\omega_1)}}\frac{I(\omega_2)}{1 - e^{-\beta(\omega_2)}}$$
$$+ |\langle 1|D|2\rangle|^2|\langle 2|D|3\rangle|^2 \frac{I(\omega_1)}{1 - e^{-\beta(\omega_1)}}\frac{I(\omega_3)}{1 - e^{-\beta(\omega_3)}} \qquad (18)$$
$$+ |\langle 1|D|3\rangle|^2|\langle 2|D|3\rangle|^2 \frac{I(\omega_2)}{1 - e^{-\beta(\omega_2)}}\frac{I(\omega_3)}{e^{\beta(\omega_3)} - 1},$$

$$\rho_2 = |\langle 1|D|2\rangle|^2|\langle 1|D|3\rangle|^2 \frac{I(\omega_1)}{e^{\beta(\omega_1)} - 1}\frac{I(\omega_2)}{1 - e^{-\beta(\omega_2)}}$$
$$+ |\langle 1|D|2\rangle|^2|\langle 2|D|3\rangle|^2 \frac{I(\omega_1)}{e^{\beta(\omega_1)} - 1}\frac{I(\omega_3)}{1 - e^{-\beta(\omega_3)}} \qquad (19)$$
$$+ |\langle 1|D|3\rangle|^2|\langle 2|D|3\rangle|^2 \frac{I(\omega_2)}{e^{\beta(\omega_2)} - 1}\frac{I(\omega_3)}{1 - e^{-\beta(\omega_3)}},$$

$$\rho_3 = |\langle 1|D|2\rangle|^2|\langle 1|D|3\rangle|^2 \frac{I(\omega_1)}{1 - e^{-\beta(\omega_1)}}\frac{I(\omega_2)}{e^{\beta(\omega_2)} - 1}$$
$$+ |\langle 1|D|2\rangle|^2|\langle 2|D|3\rangle|^2 \frac{I(\omega_1)}{e^{\beta(\omega_1)} - 1}\frac{I(\omega_3)}{e^{\beta(\omega_3)} - 1} \qquad (20)$$
$$+ |\langle 1|D|3\rangle|^2|\langle 2|D|3\rangle|^2 \frac{I(\omega_2)}{e^{\beta(\omega_2)} - 1}\frac{I(\omega_3)}{e^{\beta(\omega_3)} - 1},$$

where $\rho_i = \langle i|\rho|i\rangle$, and

$$I(\omega) = \int|g(k)|^2\delta(\omega(k) - \omega)dk. \qquad (21)$$

3. THE DOUBLE EINSTEIN FORMULA

We consider the quotient

$$\frac{\mathrm{Re}(g|g)_\omega^-}{\mathrm{Re}(g|g)_\omega^+} = \frac{N(\omega) + 1}{N(\omega)}. \qquad (22)$$

Recalling that $N(\omega)$ is the density of field quanta (photon, phonons, …) at frequency ω, and comparing formula (22) with the well known Einstein formula of radiation theory

$$\frac{W_{\text{emission}}}{W_{\text{absorption}}} = \frac{\bar{n}_\omega + 1}{\bar{n}_\omega} \qquad (23)$$

giving the quotient of the probability of emission and absorption of a light quantum by an atom (cf. [8]), we gain some physical intuition of the meaning of the generalized susceptivities. In fact the quotient (23) *is just that which is necessary to preserve the correct thermal equilibrium of the radiation with the gas* ([8], p. 180).

In the stochastic limit approach this statement can be proved using the master equation (17): if the state of the reservoir is equilibrium, then the dynamics generated by the master equation describes the relaxation to equilibrium state of the system obeying the detailed balance condition for (17), i.e.,

$$\frac{\rho_{\sigma'}}{\rho_\sigma} = \frac{\text{Re}(g|g)_\omega^-}{\text{Re}(g|g)_\omega^+} = \frac{N(\omega) + 1}{N(\omega)}, \qquad (24)$$

$$\omega = \varepsilon_\sigma - \varepsilon_{\sigma'} > 0.$$

For equilibrium state the quotient of populations of the two levels with energy difference ω is equal to the Einstein emission–absorption quotient for quanta with energy ω. This suggests that the quotients (22) may play a similar role for some stationary nonequilibrium states.

Let us give an example of such a state for which we shall get a generalization of condition (24). Consider the state (18)–(20). For simplicity we consider the case when the matrix element $\langle 1|D|2\rangle$ is negligible (direct transitions between levels 1 and 2 are prohibited). In this case

$$\frac{\rho_1}{\rho_2} = \frac{\text{Re}(g|g)_{\omega_2}^+ \text{Re}(g|g)_{\omega_3}^-}{\text{Re}(g|g)_{\omega_2}^- \text{Re}(g|g)_{\omega_3}^+}$$

$$= \frac{N(\omega_2)}{N(\omega_2) + 1} \frac{N(\omega_3) + 1}{N(\omega_3)} = e^{-\beta(\omega_2)} e^{\beta(\omega_3)}. \qquad (25)$$

Comparing with (24) and the Einstein emission–absorption relation we call this formula the Double Einstein formula.

The relation (25) for the system under study is reasonable, since direct transitions from level 2 to level 1 are prohibited ($\langle 1|D|2\rangle$ is negligible). In this case to jump from level 2 to level 1 the system have to make two sequential jumps: from level 2 to level 3 and then

from level 3 to level 1. Therefore it is reasonable to represent (25) in the following form:

$$\frac{\rho_1}{\rho_2} = \left.\frac{W_{\text{absorption}}}{W_{\text{emission}}}\right|_{2-3} \left.\frac{W_{\text{emission}}}{W_{\text{absorption}}}\right|_{1-3}$$

$$= \frac{N(\omega_2) + 1}{N(\omega_2)} \frac{N(\omega_3)}{N(\omega_3) + 1}.$$

Note that this formula is valid for a special choice of the system ($\langle 1|D|2\rangle = 0$). Moreover, for a Gibbs state, β is linear and (25) coincides with the Einstein equation (23) with $\bar{n}_\omega = N(\omega_1)$ at the right-hand side.

4. INVERSE POPULATION STATE

Let us consider the following question: for a Gibbs distribution we have $\rho_1 > \rho_2 > \rho_3$. That is the number of particles at the level decreases with increasing of the level energy. Can we find such a stationary state where at least one pair of the levels has inversed order: the number of particles increases when energy increases? Such kind of states are important in quantum optics (laser theory).

Let us apply the method of the previous section to construct a stationary state where $\rho_2 > \rho_1$ (population of level 2 is larger than population of level 1).

We will consider the same system considered in the previous section. In particular, we take

$$\langle 1|D|2\rangle = 0.$$

Then from (25) we have $\rho_2 > \rho_1$ if and only if

$$e^{-\beta(\omega_2)} e^{\beta(\omega_3)} > 1.$$

This inequality is equivalent to

$$\beta(\omega_3) > \beta(\omega_1 + \omega_3). \qquad (26)$$

This means that the local temperature function is non-monotonic and can decrease when energy increases. Let us note that the quotient ρ_2/ρ_1 does not depend on $\langle 1|D|3\rangle$, $\langle 2|D|3\rangle$ when equation $\langle 1|D|2\rangle = 0$ is valid (metastable level 2). We found that for nonmonotonic temperature functions we can have the inverse population effect: population of level with higher energy is larger than population of level with lower energy. In the theory of lasers the inverse population effect is discussed sometimes as an effect of negative temperature [9]. Indeed if we suppose that the state of the field is equilibrium and therefore the local temperature function is linear $\beta(\omega) = \beta\omega$ then (26) takes the form

$$\beta\omega_1 < 0$$

for $\omega_1 > 0$.

In present approach we found the inverse population effect without introduction of negative temperature. This effect follows from the fact that the reservoir is

highly nonequilibrium and the temperature function can decrease with energy.

5. MASTER EQUATION FOR THE NUMBER OPERATOR

We consider the number operator $n(k) = a^*(k)a(k)$. This operator has constant free evolution

$$e^{itH_0}n(k)e^{-itH_0} = n(k),$$

and therefore in the stochastic limit it is not changed. The relation with the master field is as following

$$[b_\omega(t, k), n(k')] = \lim_{\lambda \to 0}\frac{1}{\lambda}e^{-\frac{it}{\lambda^2}(\omega(k) - \omega)}[a(k), n(k')]$$

$$= \lim_{\lambda \to 0}\frac{1}{\lambda}e^{-\frac{it}{\lambda^2}(\omega(k) - \omega)}a(k)\delta(k - k') = b_\omega(t, k)\delta(k - k').$$

This means that the number operator extends the quantum noise algebra.

Let us find the master equation for the number operator. Since the number operator does not commute with the noises we cannot apply the master equation [2] directly. Applying the quantum stochastic differential equation for the evolution operator we get

$$d\langle U_t^* n(k) U_t\rangle$$

$$= \sum_\omega \langle U_t^*(E_\omega^*(D)dB_\omega(t)n(k)E_\omega(D)dB_\omega^*(t)$$

$$+ E_\omega(D)dB_\omega^*(t)n(k)E_\omega^*(D)dB_\omega(t)$$

$$- dt n(k)2\mathrm{Re}\,G)U_t\rangle$$

$$= \sum_\omega \langle U_t^*(E_\omega^*(D)E_\omega(D)[dB_\omega(t), n(k)]dB_\omega^*(t)$$

$$+ E_\omega(D)E_\omega^*(D)[dB_\omega^*(t), n(k)]dB_\omega(t))U_t\rangle$$

$$= dt\sum_\omega \langle U_t^*(E_\omega^*(D)E_\omega(D)|g(k)|^2$$

$$\times 2\pi\delta(\omega(k) - \omega)(N(k) + 1)$$

$$- E_\omega(D)E_\omega^*(D)|g(k)|^2 2\pi\delta(\omega(k) - \omega)N(k))U_t\rangle.$$

Denoting

$$\langle X\rangle_t = \langle U_t^* X U_t\rangle,$$

we arrive to master equation

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi\sum_\omega\delta(\omega(k) - \omega)$$

$$\times (|g(k)|^2(N(k) + 1)\langle E_\omega^*(D)E_\omega(D)\rangle_t$$

$$- |g(k)|^2 N(k)\langle E_\omega(D)E_\omega^*(D)\rangle_t). \tag{27}$$

This is a completely general master equation for the number operator $n(k)$. Therefore to find $\langle n(k)\rangle_t$ it is sufficient to determine $\langle E_\omega^*(D)E_\omega(D)\rangle_t$ and $\langle E_\omega(D)E_\omega^*(D)\rangle_t$.

Since we consider a generic system according (16) equation (27) takes the form

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi|g(k)|^2\sum_\omega\delta(\omega(k) - \omega)$$

$$\times |\langle 1_\omega|D|2_\omega\rangle|^2((N(k) + 1)\rho_{2_\omega} - N(k)\rho_{1_\omega}). \tag{28}$$

6. INTERACTION OF ATOM WITH RADIATION IN THE STATIONARY STATE

For a 2-level atom there is only one term in the ω-summation in (28) and the stationary state is given by (up to normalization)

$$\rho_1 = \mathrm{Re}(g|g)_\omega^-, \quad \rho_2 = \mathrm{Re}(g|g)_\omega^+.$$

Therefore if $N(k) = N(\omega(k))$ the quotient obeys the Einstein relation

$$\frac{\rho_1}{\rho_2} = \frac{N(\omega) + 1}{N(\omega)} = e^{\beta(\omega)}. \tag{29}$$

Equation (28) takes the form

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi\delta(\omega(k) - \omega)|g(k)|^2|\langle 1_\omega|D|2_\omega\rangle|^2$$

$$\times (N(\omega)(N(\omega) + 1) - (N(\omega) + 1)N(\omega)) = 0. \tag{30}$$

It means that when the atom is in its stationary state the mean number of photons is constant in each mode. Vanishing of (30) is equivalent to the Einstein relation. Let us emphasize that this conclusion does not require the state of the field to be equilibrium.

For a 3-level system the master equation for the number of photons (27) takes the form

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi|g(k)|^2(\delta(\omega(k) - \omega_1)|\langle 1|D|2\rangle|^2$$

$$\times ((N(\omega_1) + 1)\rho_2 - N(\omega_1)\rho_1)$$

$$+ \delta(\omega(k) - \omega_2)|\langle 1|D|3\rangle|^2((N(\omega_2) + 1)\rho_3 - N(\omega_2)\rho_1)$$

$$+ \delta(\omega(k) - \omega_3)|\langle 2|D|3\rangle|^2((N(\omega_3) + 1)\rho_3 - N(\omega_3)\rho_2)). \tag{31}$$

This equation describes the balance of radiation with the 3-level atom. We consider now the stationary state of the atom. When the stationary state is equilibrium then in the right-hand side of (31) each term vanishes separately. This implies that in an equilibrium state the system is in detailed equilibrium with the radiation: emission equals absorption for each frequency. This is not true when the stationary state is nonequilibrium. This implies that the atom can absorb radiation at certain frequency and emit it at another ones (parametric downconversion) and can perform simultaneous absorptions at different frequencies into a single jump. For example (18)–(21) imply

$$|\langle 1|D|2\rangle|^2 ((N(\omega_1)+1)\rho_2 - N(\omega_1)\rho_1)$$

$$= |\langle 1|D|2\rangle|^2 |\langle 1|D|3\rangle|^2 |\langle 2|D|3\rangle|^2 I(\omega_2) I(\omega_3) \tag{32}$$

$$\times \frac{e^{\beta(\omega_1)-\beta(\omega_2)+\beta(\omega_3)}-1}{(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_3)}-1)},$$

$$|\langle 1|D|3\rangle|^2 ((N(\omega_2)+1)\rho_3 - N(\omega_2)\rho_1)$$

$$= |\langle 1|D|3\rangle|^2 |\langle 1|D|2\rangle|^2 |\langle 2|D|3\rangle|^2 I(\omega_1) I(\omega_3) \tag{33}$$

$$\times \frac{1-e^{-\beta(\omega_2)+\beta(\omega_1)+\beta(\omega_3)}}{(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_1)}-1)(e^{\beta(\omega_3)}-1)},$$

$$|\langle 2|D|3\rangle|^2 ((N(\omega_3)+1)\rho_3 - N(\omega_3)\rho_2)$$

$$= |\langle 2|D|3\rangle|^2 |\langle 1|D|2\rangle|^2 |\langle 1|D|3\rangle|^2 I(\omega_1) I(\omega_2) \tag{34}$$

$$\times \frac{e^{\beta(\omega_3)+\beta(\omega_1)-\beta(\omega_2)}-1}{(e^{\beta(\omega_3)}-1)(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})},$$

(note that (32)–(34) contain the combination $\beta(\omega_3)+\beta(\omega_1)-\beta(\omega_2)$ that vanishes for an equilibrium state of the reservoir) and equation (31) takes the form

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi |g(k)|^2 |\langle 1|D|2\rangle|^2 |\langle 1|D|3\rangle|^2 |\langle 2|D|3\rangle|^2$$

$$\times \frac{e^{\beta(\omega_1)-\beta(\omega_2)+\beta(\omega_3)}-1}{(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_3)}-1)}$$

$$\times \Big((I(\omega_2)I(\omega_3)\delta(\omega(k)-\omega_1) - \tag{35}$$

$$- I(\omega_1)I(\omega_3)\delta(\omega(k)-\omega_2)$$

$$+ I(\omega_1)I(\omega_2)\,\delta\,(\omega(k)-\omega_3)\Big).$$

Equation (35) shows that being in nonequilibrium stationary state the 3-level system converts radiation with energy $\omega_2 = \omega_1 + \omega_3$ into radiation with energies ω_1 and ω_3 if

$$\beta(\omega_1)+\beta(\omega_3) > \beta(\omega_1+\omega_3)$$

and vice versa in the case of the opposite inequality.

Integrating (35) over k we get for arbitrary function $E(\omega(k))$

$$\frac{d}{dt}\int E(\omega(k))\langle n(k)\rangle_t dk$$

$$= 2\pi |\langle 1|D|2\rangle|^2 |\langle 1|D|3\rangle|^2 |\langle 2|D|3\rangle|^2$$

$$\times I(\omega_1)I(\omega_2)I(\omega_3)(E(\omega_1)-E(\omega_2)+E(\omega_3)) \tag{36}$$

$$\times \frac{e^{\beta(\omega_1)-\beta(\omega_2)+\beta(\omega_3)}-1}{(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_3)}-1)}.$$

When $E(\omega)$ is the dispersion of the field, i.e.,

$$E(\omega) = \omega,$$

then

$$E(\omega_1)-E(\omega_2)+E(\omega_3) = 0,$$

and (36) implies the conservation of energy

$$\frac{d}{dt}\int \omega(k)\langle n(k)\rangle_t dk = 0.$$

For the time derivative of the number operator we get

$$\int \frac{d}{dt}\langle n(k)\rangle_t dk = 2\pi |\langle 1|D|2\rangle|^2 |\langle 1|D|3\rangle|^2 |\langle 2|D|3\rangle|^2$$

$$\times I(\omega_1)I(\omega_2)I(\omega_3)\frac{e^{\beta(\omega_1)-\beta(\omega_2)+\beta(\omega_3)}-1}{(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_3)}-1)}.$$

Using that $\omega_2 = \omega_1 + \omega_3$ we obtain that if

$$\beta(\omega_1)+\beta(\omega_3) < \beta(\omega_1+\omega_3), \tag{37}$$

the derivative is negative and the system absorbs the radiation (total number of absorbed photons is larger than the total number of emitted photons). In this case (35) implies that the system absorbs photons with frequencies ω_1 and ω_3 and emits photons with frequency ω_2.

On the other hand, if

$$\beta(\omega_1)+\beta(\omega_3) > \beta(\omega_1+\omega_3), \tag{38}$$

then the derivative is positive and the system emits the radiation (the total number of absorbed photons is smaller than the total number of emitted photons). In this case (35) implies that the system emits photons with frequencies ω_1 and ω_3 and absorbs photons with frequency ω_2. For instance in the case of inverse population (26) the condition of emission regime (38) is satisfied.

These regimes of emission and absorption are controlled only by the difference $\beta(\omega_1)+\beta(\omega_3) - \beta(\omega_1+\omega_3)$.

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