Interaction of broadband quantum fields with resonant atoms in second-order algebraic perturbation theory

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Abstract. The second-order algebraic perturbation theory provides a correct and conventional description of the interaction of a single-mode quantized field and a broadband field with resonant particles, and in case of interacting two different broadband fields in the Markov approximation the generalization of the Hudson-Parthasarathy algebra is required to derive the Schrödinger equation.

1. Introduction
The development of media preparation technologies, artificial emitters and quantum particles has led to an experimental study of numerous interactions of various electromagnetic fields with quantum particles in various resonance conditions. In nonlinear optics [1], the interaction of classical fields with $N$-level (multilevel) quantum particles is effectively considered by the algebraic perturbation theory [2-4], which is an algebraic statement of the Bogolyubov-Krylov-Mitropolsky averaging method [5,6]. If one describes the effect of quantized broadband fields along with classical fields, then within the algebraic perturbation theory a broadband field is represented as a collection of independent sources [7]. Such a representation is important in view of the fact that the Markov approximation is used to solve the problems dealing with the effect of broadband fields, with the latter acting as noise sources and the targeted objects as open quantum systems. The broadband field as a set of independent noise sources was represented by Lax [8]. However, such a natural partitioning is not observed in other approaches [9, 10] to the problem of the resonant interaction of broadband fields with $N$-level quantum particles.

The description of processes of broadband field resonance interaction in the Markov approximation leads to either a restatement of the Schrödinger equation in terms of stochastic differential equations (SDEs) [11] or to a transition to master equations, with the master equations directly follow from SDE. So far the interaction of broadband fields in the Markov approximation has been considered in terms of Wiener-type SDEs. These equations were derived when using the resonance approximation (the first-order algebraic perturbation theory) and the algebra of quantum random processes introduced by Gardiner and Colet [11]. In the case of the action of two different broadband fields, according to the first-order algebraic perturbation theory, rescattering quanta from one broadband field to the other is described as a real absorption process of one quantum with the atomic quantum transition and the emission of another quantum with an inverse atomic quantum transition. However, it is possible to rescatter the quantum even without atomic quantum transitions. Such processes are described by the second-order algebraic perturbation theory.
The second order of algebraic perturbation theory occurred [7] to describe the resonant interactions of a quantum system with coherent classical and broadband quantized fields. Models of a Raman spontaneous transition between two energy levels of a quantum particle with the absorption of a quantum of a coherent field and the emission of a quantum into a broadband quantized field were constructed [7] on the basis of Wiener-type SDEs. However, in the second-order algebraic perturbation theory, in describing the effect of only one broadband resonant quantized field there are terms occurring in the effective Hamiltonian, which can not be described by means of Wiener-type quantum random processes and which, can not be neglected in spite of their smallness. In [12] these terms are represented by a quantum counting process, with SDE being derived in terms of the algebra of quantum random processes introduced by Hudson and Parthasarathy [13]. It was suggested that such SDEs should be called non-Wiener-type ones as in [12].

This paper considers the resonant interaction of two different quantized broadband fields of zero photon density with an ensemble of identical multilevel quantum particles. The effective Hamiltonian has been determined up to the second order of the algebraic perturbation theory. It has also been shown that for the correct statement of Schrödinger equations in the Markov approximation in terms of SDEs, the Hudson-Parthasarathy algebra is no longer sufficient to describe all interactions occurring in the processes. The Hudson-Parthasarathy algebra is generalized to determine a SDE describing a system of an ensemble of identical atoms and broadband fields interacting resonantly with it.

2. Second-order algebraic perturbation theory

Resonant interaction of two broadband electromagnetic fields with a localized ensemble of identical quantum particles will be considered in an approximation in which quantum transitions between energy levels of particles can be realized only between the allocated pair of energy levels, say, $|E_1>$ and $|E_2>$. This assumption would sound reasonable if the broadband fields under consideration had a zero photon density, the atoms occupied only the indicated pair of energy levels, with the lower of which being the ground level, and there being no other energy level between these levels. All the rest levels $|E_k>, k \neq 1,2$ could be regarded as being non-resonant. This fact does not mean that the nonresonance levels can be neglected - they determine the parameters of the effective Hamiltonian of the system in the second-order algebraic perturbation theory. The energy levels $|E_1>$ and $|E_2>$ are assumed to have parity, so that the transition can be considered as optically allowed.

Quantum fields are considered as being narrowly directed in order to be treated as independent and to be described by two pairs of creation and annihilation operators $b_j^\dagger (\omega)$ and $b_j (\omega)$. The conditions which make possible to introduce operators $b_j^\dagger (\omega)$ and $b_j (\omega)$ satisfying the typical commutation relations $[b_j (\omega), b_j^\dagger (\omega')] = \delta_{\omega \omega'}$ are considered in [12,14]. The operators $V_j (t)$ of atomic electrodipole interaction with such fields are taken in the form (in the Dirac picture):

$$V_j (t) = -\int d\omega (b_j (\omega) e^{-i\omega t} + b_j^\dagger (\omega) e^{i\omega t}) \Gamma_j (\omega) \sum_{s,km} d_{km} e^{i\omega_n t} |E_k>^{(s)} <E_m|^n.$$  \hspace{1cm} (1)

Here, the index $s=1,..,N_a$ marks the atoms of an ensemble localized at the origin of coordinates; the sum is calculated over one-atom interaction operators with a quantized electromagnetic field. The parameters of that interaction are determined by the matrix elements $d_{km}$ of the dipole moment of the atom and the parameter $\Gamma_j (\omega)$ of the $j$-th quantized electromagnetic field. Formula (1) provides the summation over all the quantum states of an individual atom $|E_k>$, $\Omega_{km} = (E_k - E_m)/\hbar$.

One of the fundamental physical assumptions of the theory of open quantum systems is the agreement on the objects for which the Markov approximation is stated. Quantum theory is based on
the Schrodinger equation for the state vector $|\Psi^{A+F}\rangle$ of an atomic ensemble (A) and the quantized fields (F) interacting with it. In the Dirac picture we have:

$$i\hbar \frac{\partial |\Psi^{A+F}(t)\rangle}{\partial t} = (V_1(t) + V_2(t))|\Psi^{A+F}(t)\rangle.$$ (2)

However, the quantum theory has a unitary symmetry: an initial state vector $|\Psi^{hi}(t)\rangle = |\Psi^{A+F}(t)\rangle$ and the initial Hamiltonian $H^{hi}(t) = V_1(t) + V_2(t)$ can be transformed without changing the values of the observed values:

$$|\tilde{\Psi}^{A+F}(t)\rangle = U(t)|\Psi^{A+F}(t)\rangle,$$

$$\tilde{H}(t) = U(t)(V_1(t) + V_2(t))U^+(t) - i\hbar U(t)\frac{\partial}{\partial t} U^+(t).$$ (3)

One can even perform continuous unitary transformations for diagonalizing the Hamiltonian of the entire system [15-17], but to state the Markov approximation it is necessary to indicate the state vector and the effective Hamiltonian. Paper [18] provides calculation of a simple model for the interaction of a broadband field with a two-level system in the Markov approximation and shows that the result depends on whether the Markov approximation is applied to the original or transformed values.

It is reasonable to assume that the Markov approximation is applied to an effective Hamiltonian that is adequate to the situation in question. The initial Hamiltonian has all possible transitions in an atom, while the Schrödinger equation itself and the initial Hamiltonian contain rapidly time-varying terms. The rapidly varying terms are still effectively averaged over the system dynamics, so that an adequate Hamiltonian should contain only slowly varying terms (in the Dirac picture). This is the fact that distinguishes the effective Hamiltonian in the resonance approximation and can be used as the basis for the determination of the terms of the effective Hamiltonian. In the first-order algebraic perturbation theory, the Markov approximation applied in calculations gives rise to the results which are consistent with the experimental data under suitable conditions [11, 18].

In order to derive the effective Hamiltonian of the problem, we expand the transformed Hamiltonian $\tilde{H}(t)$ and the generator $S(t)$ of transformation $U(t) = \exp(-iS(t))$ in a series in the interaction parameters with broadband fields

$$S(t) = S^{(1,0)}(t) + S^{(0,1)}(t) + \ldots,$$

then we obtain (taking into account the Baker-Hausdorff formula [3])

$$\tilde{H}^{(1,0)}(t) = V_1(t) + \hbar \frac{\partial S^{(1,0)}(t)}{\partial t}, \quad \tilde{H}^{(0,1)}(t) = V_2(t) + \hbar \frac{\partial S^{(0,1)}(t)}{\partial t},$$

$$\tilde{H}^{(2,0)}(t) = -\frac{1}{2}[S^{(1,0)}(t), V_1(t)] - \frac{1}{2}[S^{(1,0)}(t), \tilde{H}^{(0,1)}(t)] + \hbar \frac{\partial S^{(2,0)}(t)}{\partial t},$$

$$\tilde{H}^{(0,2)}(t) = -\frac{1}{2}[Q^{(0,1)}(t), V_2(t)] - \frac{1}{2}[Q^{(0,1)}(t), \tilde{H}^{(0,1)}(t)] + \hbar \frac{\partial S^{(0,2)}(t)}{\partial t},$$

$$\tilde{H}^{(1,1)}(t) = -\frac{1}{2}([S^{(1,0)}(t), V_1(t)] + [S^{(1,0)}(t), \tilde{H}^{(0,1)}(t)] + [Q^{(0,1)}(t), V_2(t)] + [Q^{(0,1)}(t), \tilde{H}^{(0,1)}(t)]) + \hbar \frac{\partial S^{(1,1)}(t)}{\partial t}.$$ (4)

The major idea for finding the terms $S^{(k,k)}$ and the first terms of the transformed Hamiltonian $\tilde{H}^{(1,0)} + \tilde{H}^{(1,0)} + \tilde{H}^{(2,0)} + \tilde{H}^{(0,2)} + \tilde{H}^{(1,1)}$ is the absence of rapidly time-varying terms in $\tilde{H}^{(1,1)}(t)$ in the interaction representation. This requirement of algebraic perturbation theory distinguishes the approach [2-4] from other approaches to constructing the effective Hamiltonian of the theory of open quantum systems [9, 10].

As a result of conventional [2-4, 7, 12, 14] calculations, there emerge terms in the second order of the effective Hamiltonian that are diagonal in atomic variables and do not depend on photon variables.
(Lamb shifts). They can be conveniently included into the energies of atomic quantum states. Besides, there occurs an operator of the dipole-dipole interaction of atoms \( V^{D-D}(t) \):

\[
V^{D-D}(t) = -\int \frac{(\Gamma_1^2(\omega) + \Gamma_2^2(\omega)) d\omega}{h(\omega + \Omega_{21})} |d_{21}|^2 (R_R + R_i R_e - N_u),
\]

The operators \( R_\perp \) and \( R_\parallel \) take the form:

\[
R_3 = \frac{1}{2} \sum_s (|E_2 >^{(s)} < E_1|^{(s)} - |E_1 >^{(s)} < E_2|^{(s)}), \quad R_2 = \sum_s |E_1 >^{(s)} < E_2|^{(s)}, \quad R_1 = \sum_s |E_2 >^{(s)} < E_1|^{(s)}
\]

and obey the commutation relation of the \( su(2) \) algebra

\[
[R_1, R_2] = \pm R_3, \quad [R_2, R_1] = 2R_3.
\]

Projection operators associated with the non-resonant energy levels can be omitted in the rest second-order terms \( \tilde{H}^{(0,1)} + \tilde{H}^{(1,0)} + \tilde{H}^{(2,0)} + \tilde{H}^{(0,2)} + \tilde{H}^{(1,1)} \). Therefore, on the basis of the terms \( \tilde{H}^{(0,1)} + \tilde{H}^{(1,0)} + \tilde{H}^{(2,0)} + \tilde{H}^{(0,2)} + \tilde{H}^{(1,1)} \) it is not difficult to derive the effective Hamiltonian in the Dirac picture

\[
H_{eff}^{T}(t) = \sum_{j=1,2} (V_j^R(t) + H_j^{St}(t)) + V^{D-D}(t) + V_{12}^{int}(t),
\]

where \( V_j^R(t) \) is the resonant interaction operator of electromagnetic fields with an atomic ensemble

\[
V_j^R(t) = -\int \Gamma_j(\omega) d\omega \int d\omega' \int d\omega'' \int d\omega''' \sum_{i,k} \frac{1}{2} (\Pi_k(\omega) + \Pi_k(\omega')) |E_k >^{(i)} < E_k|^{(i)},
\]

\( H_j^{St}(t) \) is the operator of the Stark interaction of an atomic ensemble with the \( j \)-th vacuum field where the approximation of a localized atomic ensemble is essential:

\[
H_j^{St}(t) = \int d\omega d\omega' \Gamma_j(\omega) \Gamma_j(\omega') \int d\omega'' \int d\omega''' \sum_{i,k} \frac{1}{2} (\Pi_k(\omega) + \Pi_k(\omega')) |E_k >^{(i)} < E_k|^{(i)},
\]

at last, operator \( V_{12}^{int}(t) \) describes the mutual interaction of broadband fields on the atoms of the ensemble:

\[
V_{12}^{int}(t) = \int d\omega d\omega' \Gamma_1(\omega) \Gamma_2(\omega') \int d\omega'' \int d\omega''' \sum_{i,k=1,2} \frac{1}{2} (\Pi_k(\omega) + \Pi_k(\omega')) |E_k >^{(i)} < E_k|^{(i)} + H.c.
\]

The letters \( H.c. \) denote the term that is Hermitian conjugate to the previous one. Atomic operator \( \sum_i |E_k >^{(i)} < E_k|^{(i)} \) for resonance levels is expressed in terms of generators \( su(2) \) of algebra as in

\[
\sum_i |E_k >^{(i)} < E_k|^{(i)} = \frac{1}{2} \sum_i (|E_1 >^{(i)} < E_1|^{(i)} + |E_2 >^{(i)} < E_2|^{(i)}) - \frac{1}{2} \sum_i (|E_2 >^{(i)} < E_2|^{(i)} - |E_2 >^{(i)} < E_1|^{(i)}),
\]

\[
\sum_i |E_2 >^{(i)} < E_1|^{(i)} = \frac{1}{2} \sum_i (|E_1 >^{(i)} < E_1|^{(i)} + |E_2 >^{(i)} < E_2|^{(i)}) + \frac{1}{2} \sum_i (|E_2 >^{(i)} < E_2|^{(i)} - |E_2 >^{(i)} < E_2|^{(i)}).
\]

According to the accepted assumption, atoms populate only resonance levels, hence

\[
\sum_i |E_1 >^{(i)} < E_1|^{(i)} = \frac{N_u}{2} - R_3, \quad \sum_i |E_2 >^{(i)} < E_2|^{(i)} = \frac{N_u}{2} + R_3.
\]

In the second-order algebraic perturbation theory, there occur typical parameters

\[
\Pi_k(\omega) = \sum_j \frac{|d_{kj}|^2}{\hbar} \left( \frac{1}{\Omega_{kj} + \omega} + \frac{1}{\Omega_{kj} - \omega} \right), \quad \Omega_{km} = |E_k - E_m| / \hbar.
\]

The prime at the summation sign means that the summation eliminates the resonance terms.
If we additionally leave only the diagonal terms in terms $H_j^{Si}(t)$ with respect to the field operators, i.e. instead of $H_j^{Si}(t)$ we consider the following values

$$H_j^{(2)Diag}(t) = \int d\omega \left| \sum_{i,k=1,2} \left\{ b_j^\dagger(\omega) b_j(\omega) \Pi_k(\omega) \right\} |E_k >^{(i)} < E_j|^{(i)} \right.,$$

then these values comply with the results of [9, 10]. However, from the point of view of algebraic perturbation theory, there are no grounds to introduce additional requirements of this kind for values of the second order of smallness in interaction constants. Moreover, if one of the fields, say, $j = 2$ is replaced by a single-mode field in the cavity, the pump/relaxation operator for the photon field $\Gamma^h(t)$, i.e. instead of $V_2(t)$ the following value is considered

$$V_2(t) \rightarrow V_2(t) = -g_c(c_c^* e^{i\Omega t} + c_c e^{-i\Omega t}) \sum_{i,j} d_{i} e^{it\Omega_{i,j}} |E_k)^{>^{(i)}} < E_j|^<^{(i)},$$

then, as a result of the algebraic perturbation theory, the pumping/relaxation operator for the photon mode of a single-mode cavity will follow from $V_{12}^{hi}(t)$:

$$V_{12}^{hi}(t) \rightarrow g_c \int d\omega \Pi_i(\omega) \Pi_j(\omega) e^{-i\Omega_{i,j} t} \sum_{i,k=1,2} \left\{ \Pi_i(\omega) + \Pi_k(\omega) \right\} |E_k >^{(i)} < E_j|^<^{(i)} + H.c.$$

The operator $c_c$ mentioned above was the annihilation operator for the photon of the frequency cavity mode $\omega_c$, $g_c$ - the parameter of the quantized field of a single-mode cavity.

Thus, the operators of the effective second-order Hamiltonian (4) agree with the known effective Hamiltonians of the problems of pumping by a broadband quantized field of a single-mode cavity [11,19].

3. Markov approximation and stochastic differential equation

The Markov conditions are formulated for the transformed states as the following:

1. At the initial instant of time, the states of the atomic ensemble $|\bar{\Psi}^A(0)\rangle$ and vacuum electromagnetic fields $|\bar{\Omega}^F(0)\rangle$ and $|\bar{\Omega}^F(0)\rangle$ are not correlated, i.e. the state function of the open system and the environment is factorized

$$|\bar{\Psi}^{A+F}(0)\rangle = |\bar{\Psi}^A(0)\rangle \otimes |\bar{\Psi}^F(0)\rangle \otimes |\bar{\Omega}^F(0)\rangle >,$$

and the electromagnetic field modes are statistically independent of each other (the delta-correlation property of the modes). Only the following mean value is other than zero

$$<\bar{\Omega}^F(0)|b_j(\omega) b_j^\dagger(\omega')|\bar{\Omega}^F(0)>> \delta(\omega - \omega').$$

2. The parameters of interaction between the atomic ensemble and the medium, determined by the values $\Gamma_j(\omega)$ and $\Pi_k(\omega)$, are independent of frequencies and are permanent values

$$\Gamma_j(\omega) = const = \Gamma_j,~~\Pi_k(\omega) = const = \Pi_k.$$

3. The limits of integration over frequencies range from $-\infty$ to $+\infty$.

Under these assumptions, the effective Hamiltonian is expressed in terms of the operator values

$$b_j(t) = \int_{-\infty}^{\infty} \frac{d\omega}{\sqrt{2\pi}} e^{-i(A - 2\omega_0) t} b_j(\omega), B_j(t) = \int_{0}^{t} dt' b_j(t'), dB_j(t) = B_j(t + dt) - B_j(t),$$

$$\Lambda_{b_j}(t) = \int_{0}^{t} dt' b_j^\dagger(t') b_j(t'), d\Lambda_{b_j}(t) = \Lambda_{b_j}(t + dt) - \Lambda_{b_j}(t).$$

Henceforward all values are assumed to be dimensionless; a non-dimensionalized procedure is performed according to work [12].
An important circumstance is the need to define integrals with respect to differentials $dB_j(t)$ and $d\Lambda_{j\ell}(t)$ in terms of Ito integrals [11-13]. Then the Ito differentials $dB_j(t)$ and $d\Lambda_{j\ell}(t)$ that describe only one $j$-th broadband field satisfy the Hudson-Parthasarathy algebra [13]:

\[
d\Lambda_{j\ell}(t)d\Lambda_{j\ell}(t) = d\Lambda_{j\ell}(t), \quad d\Lambda_{j\ell}(t)dB_j^+(t) = dB_j^+(t), \quad dB_j(t)d\Lambda_{j\ell}(t) = dB_j(t), \quad dB(t)dB^+(t) = dt, \quad (11)
\]

Then the Schrödinger equation for the evolution operator with respect to $\Lambda_{j\ell}(t)$ and $\Lambda_{j\ell}(t)$, apart from the counting process $\Lambda_{j\ell}(t)$ for the $j$-th field. To obtain the relations involving these processes, we need a generalization of the Hudson-Parthasarathy algebra which is taken in the following form:

\[
dB_j(t)dB_j^+(t) = d\delta_{j\ell}, \quad d\Lambda_{j\ell}(t)d\Lambda_{j\ell}(t) = d\Lambda_{j\ell}(t)d\delta_{j\ell}, \quad d\Lambda_{j\ell}(t)d\Lambda_{j\ell}(t) = d\Lambda_{j\ell}(t), \quad d\Lambda_{j\ell}(t)dB_j^+(t) = d\Lambda_{j\ell}(t), \quad dB_j(t)d\Lambda_{j\ell}(t) = dB_j(t).
\]

Then the Schrödinger equation for the evolution operator $U(t)$ of the transformed vector $[\hat{\Phi}^{A+\ell}(t) = U(t)\hat{\Phi}^{A+\ell}(0)]$ can be restated in terms of the introduced quantum stochastic processes and expressed as ($\chi_{j\ell}, \kappa_j, \kappa_0$ are dimensionless interaction parameters):

\[
dU(t) = [\exp[-i(V^{D-D}dt + \sum_j(Y_jdB_j(t) + Y_jdB_j^+(t)) + \frac{N_0}{2} + \eta_0R_j)(\sum_j \kappa_jd\Lambda_{j\ell}(t) + \sqrt{\kappa_1\kappa_2}(d\Lambda_{12}(t) + d\Lambda_{21}(t))) - 1])U(t), \quad Y_j = \chi_jR_j.
\]

Providing that

\[
\chi_1 = \chi_2 = \chi, \quad \kappa_1 = \kappa_2 = \kappa,
\]

One can introduce effective stochastic processes instead of the processes $B_j(t)$ and $\Lambda_{j\ell}(t)$:

\[
B(t) = (B_1(t) + B_2(t))/\sqrt{2}, \quad \Lambda(t) = [\Lambda_{11}(t) + \Lambda_{22}(t) + \Lambda_{12}(t) + \Lambda_{21}(t)]/2.
\]

For the introduced processes we have the relations of the conventional Hudson-Parthasarathy algebra (11):

\[
dB(t)dB^+(t) = dt, \quad d\Lambda(t)d\Lambda(t) = d\Lambda(t), \quad d\Lambda(t)dB^+(t) = dB^+(t), \quad dB(t)d\Lambda(t) = dB(t).
\]

All the terms in the expansion of the exponential can be fully summed up and the SDE can be written in a form that coincides with the SDE of [12] for some particular case (14) in expression (13):

\[
dU(t) = -iH^{D-D}(t)dtU(t) + (Y^+ \frac{Y^+ + iY^\Lambda}{(Y^\Lambda)^2} Y^\Lambda dB(t) + \frac{Y^\Lambda}{Y^\Lambda} dB(t) + \frac{Y^\Lambda}{Y^\Lambda} Y^\Lambda dB^+(t) + Y^\Lambda d\Lambda(t))U(t),
\]

\[
Y^\Lambda = \sqrt{2}Y\Lambda, \quad Y\Lambda = 2\kappa(\frac{N_0}{2} + \eta_0R_j), \quad Y^\Lambda = e^{-i\Lambda} - 1.
\]

Similar to works [4,12,14,19], equation (16) describes a non-Wiener decay of an excited atomic ensemble of identical atoms with a typical stabilization effect of excited states with respect to collective decay.

4. Conclusion

The described application of algebraic perturbation theory [2-4] in relation to the interaction of broadband fields with a quantum multilevel system has revealed the consistency of various quantum optical models obtained using the methods of algebraic perturbation theory. There are quite a number of examples of consistency, e.g. a particular case of interaction between single-mode and broadband
quantized fields, which also describes the interaction of fields on the mirror of a single-mode cavity [19], can be added by models of quasi-resonance interaction in the dispersion limit [20,21] and others [7,12,18-23]. They have proved the effectiveness of the problem statement and solution in the theory of open quantum systems in the effective Hamiltonian representation obtained by methods of algebraic perturbation theory, followed by the statement of the Markov conditions and the appropriate SDEs. In the second-order algebraic perturbation theory, quantum SDEs (referred to as non-Wiener SDEs) are those of the types of (13), or (16), which are both controlled by the quantum counting process and the Wiener stochastic processes. The increased role of the quantum reading process under second-order smallness of the terms of perturbation theory could be due to the counting property \( d\lambda(t) d\lambda(t) = d\lambda(t) \) and was proved in [12] for a localized ensemble of identical atoms. Generalization of the theory to extended medium is important. An example of such generalization is presented in [23].

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