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Quasi-Probability for Three-Level System and Superradiance

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A quasi-probability distribution function based on the coherent state for $U(3)$ is introduced, and its properties are then investigated. With the aid of this function it is shown that the co-operative spontaneous emission from a system of the three-level atoms can be described as a diffusion-like process on $\boldsymbol{C}^2$. In particular, the limit of validity of the semiclassical approach is examined. The case in which the semiclassical approach is not applicable can also be treated approximately by means of the Wiener-Hermite expansion method.

§ 1. Introduction

The semiclassical treatment has proved itself to be valid for an investigation of the phenomena of coherence such as photon echo[1] and self-induced transparency.[2] It has also been intended to extend the semiclassical theory for studying a spontaneous emission[3] in which quantum effect plays an essential role. In order to ascertain the extent of validity of the semiclassical theory we discuss the co-operative spontaneous emission, i.e., superradiance. Since we take this phenomenon to be originally quantal, our standpoint is quite different from the neoclassical theory developed by Jaynes et al.[4]

It is customarily believed that the system will be more classical as the photon number in a mode or the number of atoms becomes large. However, the system can never be either completely classical or semiclassical, since both of the pure classical and the semiclassical theory comprise some difficulties the quantum theory will overcome.

For such a simple system as an assembly of the two-level atoms it was shown[5),*)] that we could even interpret a co-operative spontaneous emission in terms of the classical terminologies by introducing the quasi-probability distribution function (abbreviated as QPDF) of a specific form: The effects of quantum fluctuation are completely taken into account as a spread of the QPDF at the initial stage, and each phase point corresponding to a classical dipole moves along a classical trajectory. If we interpret this spread of the QPDF as a result of the fluctuation in the ensemble of the dipole moments, we can say that the total emission from the system is given as an average of the emission from each classical dipole with respect to the ensemble.

*) Reference 5) is referred to as I hereafter.

In the present paper the problem described above is discussed for a system of the three-level atoms. In § 2 the Gilmore theory of coherent state for a multi-level system[7] is briefly reviewed. There we introduce the QPDF based on the coherent state and then derive some formulae to transform the operator master equation to the equation in a c-number phase space. Further some of the properties of the c-number equation are discussed, especially it is shown that the procedure of transformation to the c-number phase space is mathematically equivalent to taking some kind of representation of the Lie algebra $\mathfrak{u}(3)$. In § 3 an investigation of the superradiance from a system consisting of N identical atoms of three levels interacting with electromagnetic fields is made by applying the procedure developed in § 2. With the aid of the QPDF the motion of the system is described as a diffusion-like process on $\boldsymbol{C}^2$. Then we readily obtain the asymptotic behavior for a large number N of atoms by means of this equation of motion. Thus we find that the system can be well described by the classical equation of motion unless one of the three levels has extremely smaller population than the others. If this is not the case, then the quantum fluctuation exerts a good deal of influence upon the coherence properties of the system, of which we can approximately describe by using the Wiener-Hermite expansion method. In the last section the results are summarized and the unsolved problem of the limiting procedure in the theory of superradiance is discussed in connection with the rigorous result[6] obtained recently.

§ 2. Coherent state and quasi-probability

We begin with a brief review of the Gilmore theory of the coherent state for a three-level system.[7] To be definite we consider a system consisting of N identical atoms of three levels as shown in Fig. 1. Let E_{ij}^m be an operator of the mth particle represented by a 3×3 matrix whose (i,j) component is unity and all others are zero. As is easily seen the operators E_{ij}^m satisfy the commutation relation,

$$[E_{ij}^m, E_{kl}^n] = \delta_{mn}(E_{il}^m \delta_{jk} - E_{kj}^m \delta_{il}). \tag{2·1}$$

The operator E_{ij}^m $(i \neq j)$ causes a transition of the m th particle from its j th to i th energy level.

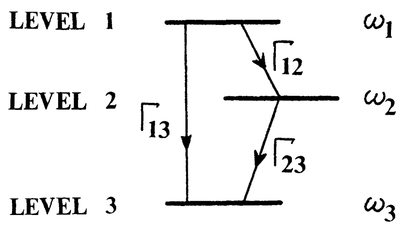

Fig. 1. Energy level diagram for the three-level atom.

Now we define the collective operators H_i and $R_{ij}^{\pm}$ for the N identical atoms of three levels,

$$H_i=\sum_{m=1}^{N} E_{ii}^m, \quad (i=1,2,3)$$

$$R_{ij}^{+}=\sum_{m=1}^{N} E_{ij}^m, \qquad R_{ij}^{-}=\sum_{m=1}^{N} E_{ji}^m. \quad (i<j) \tag{2·2}$$

Here H_i describes the population of the i th level. The raising and lowering operator between i th and j th level are respectively denoted by R_{ij}^{+} and R_{ij}^{-}.

The Dicke state for the three-level atoms is labeled by the pattern,[7)~11),*)]

$$\left|\begin{pmatrix} m_1 & & m_2 & & m_3 \\ & p_1 & & p_2 & \\ & & q & & \end{pmatrix}\right\rangle, \tag{2·3}$$

where m_1, m_2 and m_3 are positive integers or zero which prescribe the Young frame satisfying

$$m_1+m_2+m_3=N, \qquad m_1\geq m_2\geq m_3\geq 0. \tag{2·4}$$

The parameters p_1, p_2 and q are also integers varying in the ranges,

$$m_1\geq p_1\geq m_2\geq p_2\geq m_3,$$

$$p_1\geq q\geq p_2. \tag{2·5}$$

Hereafter we confine ourselves to the totally symmetric case, i.e., $[m_1, m_2, m_3]=[N, 0, 0]$. We then use the abbreviation,

$$|p, q)\equiv\left|\begin{pmatrix} N & & 0 & & 0 \\ & p & & 0 & \\ & & q & & \end{pmatrix}\right\rangle. \tag{2·6}$$

Some of the matrix elements of the operators (2·2) with respect to state $|p, q)$ are summarized in the Appendix[8)~11)] (see (A·1)), from which we can find the physical meanings of the quantum numbers p and q, i.e., q, $p-q$ and $N-p$ are respectively the population of the 1 st, 2 nd and 3 rd levels.

Since we think of an application to superradiance, our definition of the coherent states is slightly different from Gilmore's: Coherent states are such states into which the fully excited state $|N, N)$ can evolve by a classical driving field. Let $|\mu, \nu\rangle$ be a coherent state which is defined by the following equation:

$$|\mu, \nu\rangle\equiv U|N, N)\equiv\exp(\alpha R_{12}^{-}+\beta R_{13}^{-}-\text{h.c.})|N, N) \tag{2·7a}$$

$$=(1+r^2)^{-N/2}\exp(\mu R_{12}^{-}+\nu R_{13}^{-})|N, N) \tag{2·7b}$$

$$=(1+r)^{-N/2}\sum_{\substack{0\leq m+n\leq N\\ 0\leq n\leq N}}\mu^m\nu^n\left(\frac{N!}{m!n!(N-m-n)!}\right)^{1/2}|N-n, N-m-n), \tag{2·7c}$$

*) We need Yamanouchi symbols in addition to the quantum numbers in (2·3) to specify the state uniquely. See Ref. 7).

where α and β are the complex amplitudes of the classical driving field. The parameters μ, ν and r are related to α and β through the equations

$$\mu=\alpha a^{-1}\tan a\,,\qquad \nu=\beta a^{-1}\tan a\,,$$

$$r=(|\mu|^2+|\nu|^2)^{1/2}\,,\qquad a=(|\alpha|^2+|\beta|^2)^{1/2}\,. \tag{2·8}$$

Operating the unitary operator U defined in Eq. (2·7a) on the one-particle state $(N=1)$,

$$|1,1)={}^t(1,0,0), \tag{2·9}$$

we can find the meanings of μ and ν. In (2·9) t indicates a transpose, i.e., $|1,1)$ is a column vector. The geometrical interpretation is as follows: a stereographic projection from south pole maps the hypersphere S^4 on $\boldsymbol{C}^2$ ($=\boldsymbol{C}\times\boldsymbol{C}$: $\boldsymbol{C}$ indicates a complex plane). The hypersphere S^4 is in one to one correspondence with the set of coherent states,[7] and $\boldsymbol{C}^2$ is, in this case, just (μ,ν) plane. On the other hand the physical interpretation is that a point (μ,ν) corresponds to a state whose complex amplitudes of the 1 st, 2 nd and 3 rd energy levels are in the ratios $1:\mu:\nu$.

The coherent state $|\mu,\nu\rangle$ bears the two significant properties such as (1) overcompleteness,

$$\frac{(N+1)(N+2)}{\pi^2}\int_{\boldsymbol{C}^2}|\mu,\nu\rangle\langle\mu,\nu|\frac{d^2\mu d^2\nu}{(1+|\mu|^2+|\nu|^2)^3}=\mathbf{1}\,. \tag{2·10}$$

which is readily verified by means of Eq. (2·7c), and (2) the fulfilment of the seven eigenvalue equations. These eigenvalue equations are summarized in the Appendix (see Eqs. (A·3)).

We are now in a position to introduce the QPDF. Let σ be the density operator. The c-number function, the so-called Q function,[12] is then defined as

$$Q(\mu,\nu)=\langle\mu,\nu|\sigma|\mu,\nu\rangle. \tag{2·11}$$

With the use of Eq. (2·7b) and of the eigenvalue equations (A·3) we obtain the realization of operators R^{+}_{ij} etc., e.g., the differential operator, $\partial/\partial\mu^*+N\mu/(1+r^2)$, can be regarded as a realization of R^{+}_{12} in (A·4b). Since this procedure is quite analogous to that developed in I, we just summarize the results in (A·4) instead of repeating it here.

We shall briefly elucidate the mathematical background of this realization. Let V be a $(N+1)(N+2)/2$-dimensional vector space whose bases are given by

$$v_{ij}=(\mu^*)^i(\nu^*)^j/(1+r^2)^N\,.$$

$$(\mu,\nu \text{ are fixed, } r^2=|\mu|^2+|\nu|^2,\quad 0\leq i+j\leq N) \tag{2·12}$$

We take $g=(\alpha_{ij})\in U(3)$, $f\in V$ and consider the following irreducible representation (ρ, V) of $U(3)$:

$$\rho(g)f(\mu^*,\nu^*)=x^N h(y/x,z/x)/(1+r^2)^N\,, \tag{2·13}$$

where

$$(x, y, z) = (1, \mu^*, \nu^*) \cdot (\alpha_{ij}),\ h = (1+r^2)^N f. \qquad (2\cdot 14)$$

Differentiating the representation (2·13), we obtain the differential representation[13),*)] (ρ_*, V) for the Lie algebra $\mathfrak{u}(3)$ as

$$\rho_*(X)f = \lim_{t\to 0} t^{-1}(\rho(e^{tX}) - 1)f, \qquad X \in \mathfrak{u}(3), \qquad (2\cdot 15)$$

which essentially coincides with (A·4). Note that R_{ij}^+ etc. do not belong to $\mathfrak{u}(3)$ but to its complex Lie algebra. Further, it should be noted that the factor $1/(1+r^2)^N$ is of importance, since this factor enables us to extract the classical parts out of the generators of $U(3)$, e.g., the second term in the operator $\partial/\partial\mu^* + N\mu/(1+r^2)$ can be regarded as its classical part. This significant property is fully utilized in the next section.

§ 3. Superradiance from the three-level atoms

Now we study the co-operative spontaneous emission from N identical atoms of three levels confined in a small region following the procedure developed in the preceding section. The basic Hamiltonian of this system is written as[15),16)]

$$H = \sum_{i=1}^{3} \omega_i H_i + \sum_k \omega_k a_k{}^\dagger a_k + \sum_{k,\alpha} (g_{k\alpha} R_\alpha{}^+ + g_{k\alpha}^* R_\alpha{}^-)(a_k + a_k{}^\dagger). \qquad (3\cdot 1)$$

Here $a_k{}^\dagger$ and a_k are creation and annihilation operators of photon with momentum k. H_i denotes the population of the i th level, and $R_\alpha{}^+$ and $R_\alpha{}^-$ ($\alpha = (i, j)$) are the operators causing the transition between i th and j th level. Summation over α stands for all the pairs $(i, j); 1 \leq i < j \leq 3$. We have neglected in Eq. (3·1) the position dependence of the coupling constant $g_{k\alpha}$.

Applying the Born-Markov and the rotating wave approximations and ignoring the effects of the frequency shift as well as the thermal field, we obtain the master equation in the interaction picture for the reduced density operator σ describing the states of the atomic system.[16)] Let us investigate only the case in which energy spectrum is not degenerate $(\mathrm{Min}_{i\neq j}|\omega_i - \omega_j| \gg \mathrm{Max}_\alpha \Gamma_\alpha{}^{-1})$.

The reduced density operator σ satisfies the equation

$$\dot{\sigma} = \sum_\alpha \frac{\Gamma_\alpha}{2} \{[R_\alpha{}^-, \sigma R_\alpha{}^+] + [R_\alpha{}^- \sigma, R_\alpha{}^+]\} \equiv \sum_\alpha \mathcal{L}_\alpha \sigma, \qquad (3\cdot 2)$$

where

$$\Gamma_\alpha = 2\pi \sum_k |g_{k\alpha}|^2 \delta(\omega_k - \omega_\alpha), \qquad \omega_\alpha \equiv \omega_{(i,j)} \equiv |\omega_i - \omega_j|. \qquad (3\cdot 3)$$

Using (A·4) we transform the operator master equation for σ into the associated c-number Fokker-Planck equation for R defined by

*) The fact that the coherent states are closely related to some kind of differential representation is first pointed out Hioe[14)] concerning $SU(2)$.

$$R=\langle\mu,\nu|\sigma|\mu,\nu\rangle/(1+r^2)^3,$$
$$r^2=|\mu|^2+|\nu|^2. \tag{3·4}$$

The results are shown in the Appendix (A·7)~(A·9).

Let us first investigate the relation between our approach and the semiclassical theory developed by Chrisp and Jaynes.[3] Estimating an order of magnitude of the moments, we can easily understand that the diffusion terms in the Fokker-Planck operators (A·7)~(A·9) can be neglected, so long as the conditions

$$\langle|\mu|^2\rangle\ll N, \quad \langle|\nu|^2\rangle\ll N \tag{3·5}$$

and

$$\langle|\nu/\mu|^2\rangle\ll N \tag{3·6}$$

are all satisfied.*) We set $X=N/(1+r^2)$, $Y=N|\mu|^2/(1+r^2)$ and $Z=N|\nu|^2/(1+r^2)$. These are respectively the classical parts of the population operators, H_1, H_2 and H_3. Thus we arrive at the set of equations of motion:

$$\dot{X}=-\Gamma_{12}XY-\Gamma_{13}ZX,$$
$$\dot{Y}=\Gamma_{12}XY-\Gamma_{23}YZ,$$
$$\dot{Z}=\Gamma_{23}YZ+\Gamma_{13}ZX. \tag{3·7}$$

These are in agreement with the semiclassical equations of motion.[3] This semi-classical equation completely lacks the effect of quantum fluctuation, as we see from the following discussion of the two typical cases:

Case a) $X(0)=N, \quad Y(0)=Z(0)=0$

No emission occurs because there exists no atomic dipole initially. On the other hand the quantum theory involves no such difficulty, since the initial value is always accompanied with the spread of the QPDF due to the quantum fluctuation of the atomic dipole given by

$$\langle\mu,\nu|N,N)(N,N|\mu,\nu\rangle=1/(1+r^2)^N. \tag{3·8}$$

Since we have already met such a problem in the two-level system of atoms[5] and the argument for the three-level system of atoms is quite analogous, we shall not discuss it any longer.

Case b) $X(0)/Y(0), \quad Z(0)/Y(0)\sim O(N)$

This situation is peculiar to the multi-level system of atoms. Let us look at what the semiclassical theory predicts. For simplicity let $\Gamma_{13}=0$, $\Gamma_{12}=\Gamma_{23}=\Gamma$. This Eqs. (3·7) have another conserved quantity $XZ=$constant in addition to $X+Y$

*) In case Γ_{12}/Γ_{23}, $\Gamma_{13}/\Gamma_{23}\lesssim 0(1/N)$, we should take account of the diffusion terms in $\mathcal{L}_{23}$ when $\langle|\mu|^2\rangle$, $\langle|\nu|^2\rangle\sim O(1/N)$. However such a case will not actually occur, so that we shall not discuss it.

$+Z=N$. From this we see that the atomic energy is only partially released. This false prediction can be revised in the quantum theory. Note that the second condition (3·6) is no longer satisfied in this case. Hence in the Fokker-Planck operator $\mathcal{L}_{23}$ the diffusion term $|\nu|^2\partial^2/\partial\mu\partial\mu^*$, which is of the order of magnitude of $\langle|\nu/\mu|^2\rangle\sim O(N)$, should be included to describe the system correctly.

The associated Langevin equation is given by

$$\dot{\mu}=\tfrac{1}{2}N\Gamma\mu(1+|\mu|^2-|\nu|^2)/(1+r^2)+\tfrac{1}{2}\Gamma\nu F,$$
$$\dot{\nu}=N\Gamma|\mu|^2\nu/(1+r^2),$$
$$F(t)=f(t)+ig(t),\quad r^2=|\mu|^2+|\nu|^2,$$
$$\langle f(t)g(t')\rangle=0,\ \langle f(t)f(t')\rangle=\langle g(t)g(t')\rangle=\delta(t-t'). \tag{3·9}$$

Using the classical variables X, Y, Z and $W\equiv N\dot{\mu}\nu^*/(1+r^2)\equiv U+iV$ relating to the dipole operator R_{23}^+, we rewrite Eqs. (3·9) in the following form:*)

$$\dot{X}=-\Gamma XY-N^{-1}\Gamma X\,\mathrm{Re}(WF), \tag{3·10a}$$
$$\dot{Z}=\Gamma YZ-N^{-1}\Gamma Z\,\mathrm{Re}(WF), \tag{3·10b}$$
$$\dot{W}=\Gamma W(N/2-Z)+\Gamma ZF^*/2, \tag{3·10c}$$

where we have omitted the small terms in comparison with $ZF^*/2$ on the right-hand side of Eq. (3·10c). The expression for Y need not be considered explicitly because the relation $Y=N-X-Z$ always holds. From Eqs. (3·10) we obtain the equation

$$\frac{d}{dt}(XZ)=-\frac{2\Gamma}{N}XZ\,\mathrm{Re}(WF). \tag{3·11}$$

Equation (3·11) shows that XZ is no longer conserved but randomly modulated. In general it is hard to solve such nonlinear Langevin equations as Eqs. (3·10). Here we treat it using Wiener-Hermite expansion method.[19),20)] We expand X, Y, Z, U and V as

$$X(t)=\sum_{\substack{i=0\\j'=0}}^{\infty}\underbrace{\int_0^t\cdots\int_0^t}_{i+j'}d1\,d2\cdots di\,d1'\,d2',\cdots dj'\,K_{ij'}^X(t;1,2,\cdots i;1',2',\cdots j')$$
$$\times F_i(1,2,\cdots i)G_{j'}(1',2',\cdots j'). \tag{2·12}$$

We obtain the similar expansions for Y, Z, U and V. The functions $F_1, F_2, \cdots$ and $G_1, G_2, \cdots$ are defined by

$$F_1(t)=f(t),\quad F_2(t,t')=f(t)f(t')-\delta(t-t'),\ \cdots,$$
$$G_1(t)=g(t),\quad G_2(t,t')=g(t)g(t')-\delta(t-t'),\ \cdots. \tag{3·13}$$

Keeping the terms up to zeroth order of X, Y and Z, and up to first order of

*) The Langevin equations (3·9) and (3·10) are assumed not to be the Ito-Doob type[17)] but to be of the Stratonovich type.[18)]

U and V, we obtain a closed set of the ordinary differential equations,

$$\begin{aligned}
&\langle\dot{X}\rangle=-\langle X\rangle(N-\langle X\rangle-\langle Z\rangle)-\langle X\rangle\langle Z\rangle/2N,\\
&\langle\dot{Z}\rangle=\langle Z\rangle(N-\langle X\rangle-\langle Z\rangle)-\langle Z\rangle^2/2N,\\
&\langle\dot{W}\rangle=\langle W\rangle(N/2-\langle Z\rangle),\\
&\Delta\dot{W}_1(t;1)=(N-2\langle Z\rangle)\Delta W_1(t;1),\\
&\Delta W_1(t;t)=\langle Z(t)\rangle(1-i)/2,\\
&K^U_{01}(t;1)=K^V_{10}(t;1)\equiv 0,
\end{aligned}\tag{3·14}$$

where t is scaled by $1/\Gamma$, i.e., $\Gamma t\to$t. The expectation value $\langle X\rangle$ is defined by the equation

$$\langle X(t)\rangle=K^X_{00}(t).\tag{3·15}$$

The other expectation values, $\langle Z\rangle$, $\langle U\rangle$ and $\langle V\rangle$, are similarly defined. Moreover $\Delta W_1(t,1)$ is defined as

$$\Delta W_1(t;1)\equiv K^U_{10}(t;1)+iK^V_{01}(t;1).\tag{3·16}$$

If the atomic system is initially excited to a coherent state, for which the initial spread of QPDF is negligible, then we can calculate the fluctuation of the dipole moment between 2nd and 3rd level as follows:

$$\begin{aligned}
\Delta(U^2+V^2)&\equiv\langle U^2+V^2\rangle-(\langle U\rangle^2+\langle V\rangle^2)\\
&=\int_0^t|K^U_{10}(t;1)|^2d1+\int_0^t|K^V_{01}(t;1)|^2d1.
\end{aligned}\tag{3·17}$$

A typical solution of Eqs. (3·14) is illustrated in Fig. 2. The figure shows that the atomic system loses its coherence rapidly before the expectation value of the population deviates remarkably from the classical prediction. It is worth while noting that such an emission is not a co-operative emission because it is induced

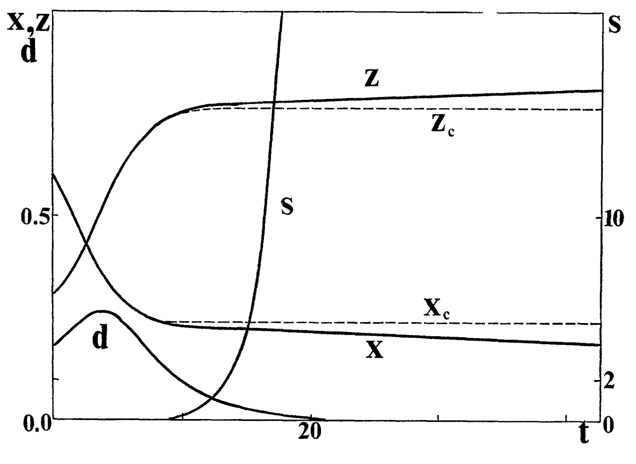

Fig. 2. Time development of the population, the dipole moment and its fluctuation (per atom). X and Z are the population of the 1st and 3rd energy level. Their semiclassical counterparts are denoted by X_c and Z_c. $d=\langle|\mu_{23}|^2\rangle^{1/2}$, $S=(d^2-|\langle\mu_{23}\rangle|^2)/|\langle\mu_{23}\rangle|^2$. Here μ_{23} is a dipole moment between 2nd and 3rd level. S denotes the square of the ratio of the variance to the mean of the dipole moment. N=200.

rather by quantum fluctuation. It will be further possible to evaluate the atomic dipole R_{12}^{+} or the higher order corrections.

If we are concerned only with the motion of the mean value of X, Y and Z, then we may study a rate equation including the quantum effects as an additional decay rate whose order of magnitude is $1/N$. This approach was carried out by Cho et al.[15] but their result is slightly different from ours (Eqs. (3·14)).

§ 4. Conclusion

We have studied superradiance from many three-level atoms by introducing a QPDF based on the coherent state for $U(3)$. We have shown that for large N the system can be described well by the classical equation of motion unless one of the three levels has extremely smaller population than the others (see Eqs. (3·5)～(3·7)). When this condition is not satisfied, the feature of the emission is far from the co-operative spontaneous emission because the emission is induced by quantum fluctuation. We have also discussed such an aspect (see Fig. 2).

We finally remark that there remains an unsolved problem in the limiting procedure with which we get a classical equation of motion. We make first Markoffian and 1st Born approximations (the lowest order approximation of the perturbation series), then let $N\to\infty$. As pointed out by Wills and Picard,[21] this procedure gives rise to the difficulty of divergence which the superradiance master equation is suffering from. The perturbation series for the density operator is, at the same time, an increasing power series of N. Although the recent development of statistical mechanics[6] clarifies the basis of Born and Markoffian approximation, such a divergence problem is not taken into consideration as yet. This is a problem to be resolved in the future.

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Appendix

1) *Matrix elements*[8)~11)]

$$H_1|p,q\rangle = q|p,q\rangle, \qquad R_{12}^{-}|p,q\rangle = \sqrt{q(p-q+1)}\,|p,q-1\rangle,$$
$$H_2|p,q\rangle = (p-q)|p,q\rangle, \qquad R_{23}^{-}|p,q\rangle = \sqrt{(N-p+1)(p-q)}\,|p-1,q\rangle,$$
$$H_3|p,q\rangle = (N-p)|p,q\rangle, \qquad R_{13}^{-}|p,q\rangle = \sqrt{q(N-p+1)}\,|p-1,q-1\rangle. \qquad (\mathrm{A}\cdot 1)$$

Matrix elements of R_{12}^{+}, R_{23}^{+} and R_{13}^{+} are easily obtained from R_{12}^{-}, R_{13}^{-} and R_{23}^{-} by using $(R_{12}^-)^+ = R_{12}^+$ etc.

2) *Eigenvalue equations*

Let U be a unitary operator defined by $|\mu, \nu\rangle = U|N, N\rangle$. Then the following seven identities hold:

$$UR_{ij}^+U^{-1}|\mu, \nu\rangle = 0\,, \quad UR_{23}^-U^{-1}|\mu, \nu\rangle = 0\,,$$
$$UH_kU^{-1}|\mu, \nu\rangle = 0\,, \quad UH_1U^{-1}|\mu, \nu\rangle = N|\mu, \nu\rangle, \tag{A·2}$$

where $(i,j) = (1,2)$, $(2,3)$, $(1,3)$ and $k=2,3$. The explicit forms are as follows:

$$(H_1 + \mu R_{12}^- + \nu R_{13}^-)\,|\mu, \nu\rangle = N|\mu, \nu\rangle,$$
$$(R_{12}^+ + \mu^2 R_{12}^- + \mu\nu R_{13}^-)\,|\mu, \nu\rangle = N\mu|\mu, \nu\rangle,$$
$$(R_{13}^+ + \mu\nu R_{12}^- + \nu^2 R_{13}^-)\,|\mu, \nu\rangle = N\nu|\mu, \nu\rangle,$$
$$(H_2 - \mu R_{12}^-)\,|\mu, \nu\rangle = 0\,, \quad (R_{23}^+ - \nu R_{12}^-)\,|\mu, \nu\rangle = 0\,,$$
$$(R_{23}^- - \mu R_{13}^-)\,|\mu, \nu\rangle = 0\,, \quad (H_3 - \nu R_{13}^-)\,|\mu, \nu\rangle = 0\,. \tag{A·3}$$

3) *Realization of operators R_{ij}^+ etc. (see Eqs. (2·2).)*

$$\langle \mu, \nu|H_1\sigma|\mu, \nu\rangle = \left(-\mu^*\frac{\partial}{\partial\mu^*} - \nu^*\frac{\partial}{\partial\nu^*} + \frac{N}{1+r^2}\right)Q\,, \tag{A·4a}$$

$$\langle \mu, \nu|R_{12}^+\sigma|\mu, \nu\rangle = \left(\frac{\partial}{\partial\mu^*} + \frac{N\mu}{1+r^2}\right)Q\,, \tag{A·4b}$$

$$\langle \mu, \nu|R_{12}^-\sigma|\mu, \nu\rangle = \left(-\mu^{*2}\frac{\partial}{\partial\mu^*} - \mu^*\nu^*\frac{\partial}{\partial\nu^*} + \frac{N\mu^*}{1+r^2}\right)Q\,, \tag{A·4c}$$

$$\langle \mu, \nu|R_{23}^+\sigma|\mu, \nu\rangle = \left(\mu^*\frac{\partial}{\partial\nu^*} + \frac{N\mu^*\nu}{1+r^2}\right)Q\,, \tag{A·4d}$$

$$\langle \mu, \nu|H_2\sigma|\mu, \nu\rangle = \left(\mu^*\frac{\partial}{\partial\mu^*} + \frac{N|\mu|^2}{1+r^2}\right)Q\,. \tag{A·4e}$$

Expressions for $\langle \mu, \nu|R_{13}^+\sigma|\mu, \nu\rangle$, $\langle \mu, \nu|R_{13}^-\sigma|\mu, \nu\rangle$, $\langle \mu, \nu|R_{23}^-\sigma|\mu, \nu\rangle$ and $\langle \mu, \nu|H_3\sigma|\mu, \nu\rangle$ are given by exchanging μ and ν in the expressions (A·4b), (A·4c) (A·4d) and (A·4e), respectively.

4) *Formula for evaluating the expectation value*

Let $\hat{A}$ be an operator for the atomic system only. The expectation value of $\hat{A}$ is given by

$$\langle \hat{A}\rangle \equiv \mathrm{tr}\,(\sigma\hat{A}) = \frac{(N+1)(N+2)}{\pi^2}\int_{c^2}\langle \mu, \nu|\hat{A}\sigma|\mu, \nu\rangle\frac{d^2\mu\, d^2\nu}{(1+r^2)^3}$$
$$= \frac{(N+1)(N+2)}{\pi^2}\int_{c^2}(\underset{\sim}{A}Q)\,\frac{d^2\mu\, d^2\nu}{(1+r^2)^3}\,, \tag{A·5}$$

where we have made use of the overcompleteness relation (2·9), and $\underset{\sim}{A}$ stands for the realization of $\hat{A}$ in the sense of (A·4). When the classical part A of $\underset{\sim}{A}$ is dominant, $\langle\hat{A}\rangle$ can be approximately written as

$$\langle\hat{A}\rangle \simeq \frac{(N+1)(N+2)}{\pi^2}\int_{c^2} AR\, d^2\mu\, d^2\nu \simeq \frac{N^2}{\pi^2}\int_{c^2} AR\, d^2\mu\, d^2\nu\,, \qquad (\text{A}\cdot 6)$$

where $R=Q/(1+r^2)^3$.

5) *Fokker-Planck operator associated with the damping Liouvillians* (3·3)

(i) $$\mathcal{L}_{12}\sigma:\Big\{\frac{\partial^2}{\partial\mu\partial\mu^*}|\mu|^4+2\frac{\partial^2}{\partial\mu\partial\nu^*}|\mu|^2\mu\nu^*+\frac{\partial^2}{\partial\nu\partial\nu^*}|\mu|^2|\nu|^2+\frac{\partial^2}{\partial\mu\partial\nu}\mu\nu$$

$$+\frac{\partial^2}{\partial\mu^2}\mu^2-(N+3)\frac{\partial}{\partial\mu}\frac{1+|\mu|^2}{1+r^2}\mu-(N+3)\frac{\partial}{\partial\nu}\frac{|\mu|^2\nu}{1+r^2}\Big\}R+\text{c.c.} \qquad (\text{A}\cdot 7)$$

(ii) $\mathcal{L}_{13}\sigma$: μ and ν are to be exchanged in (A·7). (A·8)

(iii) $$\mathcal{L}_{23}\sigma:\Big\{\frac{\partial^2}{\partial\mu\partial\mu^*}|\nu|^2-\frac{\partial^2}{\partial\mu\partial\nu}\mu\nu+\frac{\partial}{\partial\nu}\nu+(N+3)\frac{\partial}{\partial\mu}\frac{\mu|\nu|^2}{1+r^2}$$

$$-(N+3)\frac{\partial}{\partial\nu}\frac{|\mu|^2\nu}{1+r^2}\Big\}R+\text{c.c.} \qquad (\text{A}\cdot 9)$$

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