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Stimulated recombination in open systems

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In this comment we study the problem of the stimulated recombination in an open system from a stochastic point of view. We set up the bivariate master equation for the number of photons and ions inside the system. Then we perform a systematic expansion, with the system volume as an expansion parameter, and we obtain the fluctuations of the number of photons and ions around its macroscopic values in the linear noise approximation; the stationary solution is also investigated.

I. INTRODUCTION

Recently Lami and Rahman[1] studied stimulated recombination[2] from a stochastic point of view. In their interesting work they made a theoretical study of the stimulated recombination and photoionization processes in an isolated, closed macroscopic system. Their study was based entirely on the statistical mechanics of stochastic processes. This kind of research is interesting because it is now possible to envision powerful lasers with photon frequencies at which single photoionization can occur.

The present study is devoted to the atomic photoionization and stimulated recombination case, but can be easily extended to the molecular photodissociation and stimulated molecular recombination. We consider (i) stimulated atomic recombination

$$e^- + X^+ + h\nu \rightarrow X + 2h\nu$$

and (ii) photoionization

$$X + h\nu \rightarrow e^- + X^+ \; . \tag{1}$$

This paper extends the development of Ref. 1 to open systems, i.e., to systems where photons are introduced (for simplicity) at a constant rate, and which loses the photons at a rate which depends on the photon population inside the system. It is interesting to remark here that because the photon population inside the system is a stochastic variable, the loss rate is not constant and must be studied from a stochastic point of view.

This brief report is organized as follows. In Sec. II we derive the bivariate master equation[2] for the number of photons and the number of ions inside the system, and we justify the reasons which lead to this equation. Then we perform a systematic expansion[3] of the master equation; to this end it is necessary to know how the coefficients of the master equation depend on the expansion parameter which we have chosen equal to the system volume. In Sec. III we deduce from the systematic expansion the two macroscopic equations for the time evolution of the average number of photons and ions inside the system. Then we deduce the Fokker-Planck equation in the linear noise approximation.[3] This equation can be easily solved in this approximation, and we show that the fluctuation parameters obey a set of coupled integro-differential equations.

II. BIVARIATE MASTER EQUATION AND SYSTEMATIC EXPANSION

We follow here the nomenclature of Lami and Rahman.[1] In the case of an open system we have only one conservation law, instead of two as in Ref. 1. The conservation law of the number of particles is expressed as

$$a + a^+ = A \; , \tag{2}$$

where a and a^+ are the number of atoms and ions, respectively, and A is a constant. We assume also charge neutrality. This means that the total number of electrons n_e is equal to a^+. The second conservation law used by Lami and Rahman does not apply in our case because photons escape from the system at a rate which depends on the number of photons inside the system which is a stochastic variable.

Let us consider the number of photons n and the number of ions a^+ as the unknown stochastic time-dependent variables; the rest of the variables are related to these. The system that we consider is the same as Ref. 1, except that photons are pumped into the system at a constant rate δ and leave the system at a rate which depends on the number of photons inside the system. We denote by $P(n,a^+,t)$ the distribution function at time t, or probability to have n photons and a^+ ions at time t. Using the probability balance technique,[4] it is very easy to deduce the bivariate master equation, which is given by

$$\frac{dP(n,a^+,t)}{dt}=\alpha(n+1)a[a^+-1]P(n+1,a^+-1,t)+(\lambda(n-1)(a^++1)^2+\gamma(a^++1)^2)P(n-1,a^++1,t)$$
$$+\delta P(n-1,a^+,t)+\beta(n+1)P(n+1,a^+,t)-(\delta+\alpha na[a^+]+\lambda na^{+2}+\beta n+\gamma(a^+)^2)P(n,a^+,t) \quad (3)$$

where we use the square brackets to signify the functional dependence of a on the number of ions a^+. The physical meaning of the different terms and coefficients on the right-hand side of Eq. (3) can be better understood with the help of Fig. 1, where r_n is the probability per unit time that a jump occurs from (n,a^+) to $(n-1,a^+)$. In our case $r_n=\beta n$, where β is the loss coefficient for the system; g_n is the probability per unit time that a jump occurs from (n,a^+) to $(n+1,a^+)$. In our case $g_n=\delta$, where δ is the rate at which photons are pumped into the system. $gr(n,a^+)$, is the probability per unit time that a jump occurs from (n,a^+) to $(n+1,a^+-1)$. In our case $gr(n,a^+)=\lambda na^{+2}+\gamma a^{+2}$, where λ and γ are the stimulated and spontaneous recombination coefficients, respectively. $rg(n,a^+)$, is the probability per unit time that a jump occurs from (n,a^+) to $(n-1,a^++1)$. In our case $rg(n,a^+)=\alpha na[a^+]=\alpha n(A-a^+)$, where α is the photoionization coefficient.

Now with the help of the step operators E_p and E_i for photons and ions, respectively, we can rewrite Eq. (3) as follows:

$$\frac{dP(n,a^+,t)}{dt}=\alpha(E_pE_i^{-1}-1)n(A-a^+)P(n,a^+,t)+(E_p^{-1}E_i-1)(\lambda n+\gamma)a^{+2}P(n,a^+,t)$$
$$+\delta(E_p^{-1}-1)P(n,a^+,t)+\beta(E_p-1)nP(n,a^+,t)\ . \quad (4)$$

To perform a systematic expansion of the master equation (4), we need to choose some expansion parameter. By physical considerations,[5] it is easily deduced that the appropiate expansion parameter is the system volume Ω. Now following Van Kampen[3] and with the help of the considerations of Lami *et al.*,[2,5] we rewrite Eq. (4) as follows:

$$\frac{dP(n,a^+,t)}{dt}=\frac{\alpha'}{\Omega}(E_pE_i^{-1}-1)n(\Omega\rho_i-a^+)P(n,a^+,t)+(E_p^{-1}E_i-1)\left[\frac{\lambda'}{\Omega^2}n+\frac{\gamma'}{\Omega}\right]a^{+2}P(n,a^+,t)$$
$$+\Omega\rho_p(E_p{}^{-1}-1)P(n,a^+,t)+\beta(E_p-1)nP(n,a^+,t)\ , \quad (5)$$

where

$$\alpha'=\alpha\Omega\ , \quad (6a)$$

$$\lambda'=\lambda\Omega^2\ , \quad (6b)$$

$$\gamma'=\gamma\Omega\ , \quad (6c)$$

and $\rho_i=A/\Omega$ is the initial atomic density per unit volume, and $\rho_p=\delta/\Omega$ is the rate at which photons are injected into the system per unit volume.

The next step in the expansion is to set

$$n=\Omega\phi(t)+\Omega^{1/2}\xi\ , \quad (7a)$$

$$a^+=\Omega\psi(t)+\Omega^{1/2}\eta\ , \quad (7b)$$

where $n^*(t)=\Omega\phi$ and $I^*(t)=\Omega\psi$, are the macroscopic values of the number of photons and ions, respectively, at time t, and ξ and η are the new stochastic variables. Now if (7a) and (7b) are substituted into $P(n,a^+,t)$, we obtain the distribution function of ξ and η denoted by $\Pi(\xi,\eta,t)$. Now we consider the following expansions of the step operators:

$$E_p=1+\Omega^{-1/2}\frac{\partial}{\partial\xi}+\tfrac{1}{2}\Omega^{-1}\frac{\partial^2}{\partial\xi^2}+\cdots\ , \quad (8a)$$

$$E_p^{-1}=1-\Omega^{-1/2}\frac{\partial}{\partial\xi}+\tfrac{1}{2}\Omega^{-1}\frac{\partial^2}{\partial\xi^2}-\cdots\ , \quad (8b)$$

$$E_i=1+\Omega^{-1/2}\frac{\partial}{\partial\eta}+\tfrac{1}{2}\Omega^{-1}\frac{\partial^2}{\partial\eta^2}+\cdots\ , \quad (8c)$$

$$E_i^{-1}=1-\Omega^{-1/2}\frac{\partial}{\partial\eta}+\tfrac{1}{2}\Omega^{-1}\frac{\partial^2}{\partial\eta^2}-\cdots\ . \quad (8d)$$

Now direct substitution of (7a) into the bivariate master equation (5) yields, after some algebra,

$$\frac{\partial\Pi}{\partial t}-\Omega^{1/2}\frac{\partial\Pi}{\partial\xi}\frac{d\phi}{dt}-\Omega^{1/2}\frac{\partial\Pi}{\partial\eta}\frac{d\psi}{dt}=\frac{\alpha'}{\Omega}\left[\Omega^{-1/2}\left[\frac{\partial}{\partial\xi}-\frac{\partial}{\partial\eta}\right]+\frac{\Omega^{-1}}{2}\left[\frac{\partial}{\partial\xi}-\frac{\partial}{\partial\eta}\right]^2\right](\Omega\phi+\Omega^{1/2}\xi)$$
$$\times[\Omega\rho_i-(\Omega\psi+\Omega^{1/2}\eta)]\Pi$$
$$+\left[\Omega^{-1/2}\left[\frac{\partial}{\partial\eta}-\frac{\partial}{\partial\xi}\right]+\frac{\Omega^{-1}}{2}\left[\frac{\partial}{\partial\eta}-\frac{\partial}{\partial\xi}\right]^2\right]\left[\frac{\lambda'}{\Omega^2}(\Omega\phi+\Omega^{1/2}\xi)+\frac{\gamma'}{\Omega}\right]$$
$$\times(\Omega^2\psi^2+2\Omega^{3/2}\psi\eta+\Omega\eta^2)\Pi+\Omega\rho_p\left[-\Omega^{-1/2}\frac{\partial}{\partial\xi}+\frac{\Omega^{-1}}{2}\frac{\partial^2}{\partial\xi^2}\right]\Pi$$
$$+\beta\left[\Omega^{-1/2}\frac{\partial}{\partial\xi}+\frac{\Omega^{-1}}{2}\frac{\partial^2}{\partial\xi^2}\right](\Omega\phi+\Omega^{1/2}\xi)\Pi+\cdots\ . \quad (9)$$

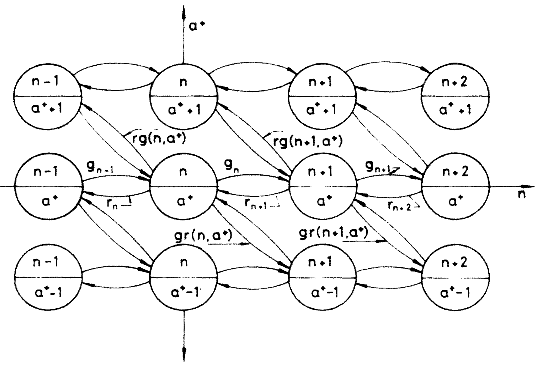

FIG. 1. Transition probabilities in the state space (n,a^+).

Equation (9) is the systematic expansion of the master equation which will serve as the starting point for the next section.

III. THE LINEAR-NOISE APPROXIMATION AND THE EMERGENCE OF THE MACROSCOPIC LAW

At first sight, Eq. (9) is not a proper expansion for large Ω, because several terms of order $\Omega^{1/2}$ appear. However, one notices immediately that the terms involving $\Omega^{1/2}$ are of two classes: the first one involves Π only through $\partial\Pi/\partial\xi$ and the second one only through $\partial\Pi/\partial\eta$; it is therefore possible to let them cancel by choosing ψ and ϕ such that the coefficients of $\Omega^{1/2}\partial\Pi/\partial\xi$ and $\Omega^{1/2}\partial\Pi/\partial\eta$ vanish. After some algebra one finds that the cancellation of the coefficients of $\Omega^{1/2}\partial\Pi/\partial\xi$ gives

$$\frac{d\phi}{dt}=\lambda'\phi\psi^2+\gamma'\psi^2+\rho_p-\alpha'\phi(\rho_i-\psi)-\beta\phi \ . \tag{10a}$$

In the same way the cancellation of the coefficients of $\Omega^{1/2}\partial\Pi/\partial\eta$ yields

$$\frac{d\psi}{dt}=\alpha'\phi(\rho_i-\psi)-\lambda'\phi\psi^2-\gamma'\psi^2 \ . \tag{10b}$$

Equations (10a) and (10b) can be written using (6a)–(6c) in the form

$$\frac{dn^*}{dt}=\lambda n^*I^{*2}+\gamma I^{*2}+\delta-\alpha n^*(A-I^*)-\beta n^* \ , \tag{11a}$$

$$\frac{dI^*}{dt}=\alpha n^*(A-I^*)-\lambda n^*I^{*2}-\gamma I^{*2} \ . \tag{11b}$$

Equations (11a) and (11b) form a system of coupled nonlinear differential equations, the so-called macroscopic equations, but we are really interested in the fluctuations around the macroscopic values n^* and I^* for the number of photons and ions, respectively. It is interesting to point out that the macroscopic equations can be deduced directly from physical considerations without any recourse to systematic expansion. However, the fact that Eqs. (11a) and (11b) are the same as are obtained from physical considerations means that we are on the right path.

The stationary point in the phase plane (n^*,I^*) is obtained by setting the time derivative of the macroscopic equations equal to zero. In this way we obtain $n^{*s}=\delta/\beta$ and

$$I^{*s}=\frac{[\alpha^2\delta^2+4(\beta\gamma+\lambda\delta)\alpha\delta A]^{1/2}-\alpha\delta}{2(\beta\gamma+\lambda\delta)} \ . \tag{12}$$

We observe that the macroscopic equations (11a) and (11b) do not give rise to multiple stationary solutions which one gets in considering closed systems.

The terms of order Ω^0 in Eq. (9) yield after some algebra the following bivariate linear Fokker-Planck equation:

$$\frac{\partial\Pi(\xi,\eta,t)}{\partial t}=[-\alpha'(\rho_i-\psi)+\lambda'\psi^2]\left[\frac{\partial}{\partial\eta}-\frac{\partial}{\partial\xi}\right]\xi\Pi+(\alpha'\phi+2\lambda'\phi\psi+2\gamma'\psi)\left[\frac{\partial}{\partial\eta}-\frac{\partial}{\partial\xi}\right]\eta\Pi$$
$$+\tfrac{1}{2}[\alpha'\phi(\rho_i-\psi)+(\lambda'\phi+\gamma')\psi^2]\left[\frac{\partial}{\partial\eta}-\frac{\partial}{\partial\xi}\right]^2\Pi+\tfrac{1}{2}(\rho_p+\beta\phi)\frac{\partial^2\Pi}{\partial\xi^2}+\beta\frac{\partial}{\partial\xi}\xi\Pi \ . \tag{13}$$

The solution of Eq. (13) is well known, and is given by

$$\Pi(\xi,\eta,t)=(2\pi)^{-1}(\det\Xi)^{-1/2}\times\exp[-\tfrac{1}{2}(y^t-\langle y\rangle^t)\Xi^{-1}(y-\langle y\rangle)] \ , \tag{14}$$

where $y_1=\xi$ and $y_2=\eta;y\equiv[y_i]$, t means transpose, and Ξ is the variance-covariance matrix given by $\Xi_{ij}=(\langle y_iy_j\rangle-\langle y_i\rangle\langle y_j\rangle)$.

In order to know the distribution function, we need to calculate the averages $\langle\xi\rangle,\langle\eta\rangle$ and the moments, $\langle\xi^2\rangle,\langle\eta^2\rangle,\langle\xi\eta\rangle$ versus time. $\langle\xi\rangle$ and $\langle\eta\rangle$ satisfy a pair of coupled differential equations. The first one is obtained by multiplying Eq. (13) by ξ and then integrating over ξ and η. The second one is obtained by the same method but multiplying by η. In this way after integrations by parts we obtain

$$\begin{aligned}\partial_t\langle\xi\rangle&=[-\alpha'(\rho_i-\psi)+\lambda'\psi^2-\beta]\langle\xi\rangle\\&\quad+(\alpha'\phi+2\lambda'\phi\psi+2\gamma'\psi)\langle\eta\rangle \ ,\\ \partial_t\langle\eta\rangle&=[\alpha'(\rho_i-\psi)-\lambda'\psi^2]\langle\xi\rangle\\&\quad-(\alpha'\phi+2\lambda'\phi\psi+2\gamma'\psi)\langle\eta\rangle \ .\end{aligned} \tag{15}$$

These equations are the variational equations associated with the macroscopic equations which again confirm the correctness of the master-equation expansion. In order to find the set of differential equations satisfied by $\langle\xi^2\rangle,\langle\xi\eta\rangle,\langle\eta^2\rangle$, Eq. (13) is first multiplied by ξ^2 and then integrated over ξ and η, and the same procedure is applied with $\xi\eta$ and η^2. Finally after some calculations one arrives at the following set of coupled differential equations:

$$\partial_t\langle \xi^2\rangle = 2[-\alpha'(\rho_i-\psi)+\lambda'\psi^2-\beta]\langle \xi^2\rangle + 2(\alpha'\phi+2\lambda'\phi\psi+2\gamma'\psi)\langle \xi\eta\rangle + [\alpha'\phi(\rho_i-\psi)+(\lambda'\phi+\gamma')\psi^2+\rho_p+\beta\phi] , \tag{16a}$$

$$\partial_t\langle \eta^2\rangle = -2(\alpha'\phi+2\lambda'\phi\psi+2\gamma'\psi)\langle \eta^2\rangle - 2[-\alpha'(\rho_i-\psi)+\lambda'\psi^2]\langle \xi\eta\rangle + [\alpha'\phi(\rho_i-\psi)+(\lambda'\phi+\gamma')\psi^2] , \tag{16b}$$

$$\partial_t\langle \xi\eta\rangle = [\alpha'(\rho_i-\psi)-\lambda'\psi^2]\langle \xi^2\rangle + (\alpha'\phi+2\lambda'\phi\psi+2\gamma'\psi)\langle \eta^2\rangle + [-\alpha'(\rho_i-\psi) + \lambda'\psi^2-(\alpha'\phi+2\lambda'\phi\psi+2\gamma'\psi)-\beta]\langle \xi\eta\rangle - [\alpha'\phi(\rho_i-\psi)+(\lambda'\phi+\gamma')\psi^2] . \tag{16c}$$

Equations (16a)—(16c) determine the variances and the covariances of the fluctuations of n and a^+ around the macroscopic values n^* and I^*. To solve this set of equations it is necessary to know the initial distribution at some initial time. For instance, if at time $t=0$, we start to pump photons into the system, obviously at time 0, $\langle \xi^2\rangle = \langle \xi\eta\rangle = \langle \eta^2\rangle = 0$.

We have solved the set of Eqs. (16a)—(16c) in the stationary state, and we have found that the correlation between ξ and η is negative. This is a direct consequence of the physical fact that each time we have one stimulated atomic recombination, n increases and a^+ decreases, and if we have one photoionization process n decreases and a^+ increases; so that in both cases we have a negative correlation.

We have also found that the variances, $\langle\langle n^2\rangle\rangle^S$ and $\langle\langle a^{+2}\rangle\rangle^S$, for the number of photons and ions inside the system at the stationary state are not Poisson variances. Finally, from the stationary solutions it is easily deduced that the covariance $\langle\langle na^+\rangle\rangle^S$ should be bigger in absolute value than n^{*S}, i.e., the macroscopic number of photons at the stationary state.

IV. CONCLUSIONS

In this paper we have studied the problem of the atomic stimulated recombination and photoionization in an open system in which photons are pumped into the system at a constant rate, but which leave the system at a rate which depends on the number of photons inside the system. We have seen that in this case we have only one conservation law instead of two like in Ref. 1. This fact has forced us to set up a bivariate master equation for the number of photons and ions, respectively.

Due to the nonlinear character of this master equation, it is not possible to apply the Kolmogorov[3,4] methodology, and we have used a systematic expansion of the master equation in terms of the system volume Ω. First we have studied, with the help of the works of Van Kampen and Lami and co-workers[2,5] how the various coefficients depend on the volume Ω. Then we have set up the master equation in a suitable form to perform the systematic expansion. From this expansion we have obtained the macroscopic equations, which are the same ones that are obtained directly from physical considerations. Then we have obtained in the linear-noise approximation the bivariate linear Fokker-Planck equation satisfied by the fluctuation of the number of photons and ions around its macroscopic values. We have shown that the average of these fluctuations satisfy the variational equations associated with the macroscopic equations (10a) and (10b), which again confirms the validity of the systematic expansion.

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[5]See N. G. Van Kampen in Ref. 3, Chap. IX. See also A. Lami, N. K. Rahman, and F. H. M. Faisal in Ref. 2. Of special interest are Sec. II and Appendix A.