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# Stochastic Processes in Raman Scattering

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## Abstract

The fluctuations in Raman scattering are considered using an adiabatic elimination from the Fokker-Planck equation corresponding to the Langevin equations contained the non-linear coupling in irrelevant macrovariables.

## § 1. Introduction

In recent years there has been great interest shown in non-linear systems which are maintained in non-equilibrium steady states. The study of non-linear systems has shown the existence of a variety of instability.<sup>1-2)</sup> We have studied the ordered structures in non-linear chemical reactions and in non-linear optics.<sup>3-5)</sup> In this paper we consider the stochastic processes of Raman scattering, which involved non-linear coupling in an irrelevant set.

Macroscopic properties are described by a relevant subset of macrovariables of the system. It is necessary to obtain closed equations of motion for the subset by eliminating the irrelevant set. Multiplicative stochastic forces appear when one replaces external parameters in equations of motion by fluctuating ones in order to take into account fluctuations of the surrounding, and eliminates irrelevant variables in coupled Langevin equations by an adiabatic procedure.

In § 2, we develop an adiabatic elimination from the Fokker-Planck equation corresponding to the Langevin equations contained the non-linear coupling in the irrelevant set. In § 3 we obtain the Fokker-Planck equation and the Langevin equation for the photon number in Raman scattering.

## § 2. Adiabatic elimination from the Fokker-Planck equation

Let us consider the following model equations:

$$dA_i(t)/dt = v_i(A(t)) + \alpha_{ij}^{(1)}(A(t))B_j(t) + \alpha_{ijk}^{(2)}(A(t))B_j(t)B_k(t) + r_i^A(t), \quad (2.1)$$

$$dB_j(t)dt = \beta_j(A(t)) - \gamma_{jk}(A(t))B_k(t) + r_j^B(t), \quad (2.2)$$

where  $r_i^A(t)$  and  $r_j^B(t)$  are Gaussian white noises with mean values zero and

correlations

$$\langle r_i^A(t) r_i^A(t'); a_0 b_0 \rangle = 2L_{ii}^{AA} \delta(t-t'), \quad L_{ii}^{AA} = L_{ii}^{AA}, \quad (2.3)$$

$$\langle r_i^A(t) r_j^B(t'); a_0 b_0 \rangle = 2L_{ij}^{AB} \delta(t-t'), \quad L_{ij}^{AB} = L_{ji}^{BA}, \quad (2.4)$$

$$\langle r_j^B(t) r_k^B(t'); a_0 b_0 \rangle = 2L_{jk}^{BB} \delta(t-t'), \quad L_{jk}^{BB} = L_{kj}^{BB}, \quad (2.5)$$

Here  $\langle \dots; a_0 b_0 \rangle$  denotes the conditional averages with the initial values  $A(0) = a_0$  and  $B(0) = b_0$ . We assume that  $A(t)$  and  $B(t)$  are relevant and irrelevant variables, respectively, and the time scale  $\tau_A$  of  $A(t)$  is distinctly larger than the time scale  $\tau_B$  of  $B(t)$ .

The Fokker-Planck equation corresponding to the Langevin equations (2.1) and (2.2) is given by<sup>6)</sup>

$$\partial P(a, b, t) / \partial t = \Gamma^+(a, b) P(a, b, t), \quad (2.6)$$

where

$$\begin{aligned} \Gamma^+(a, b) = & -(\partial/\partial a_i)[v_i(a) + \alpha_{ij}^{(1)} b_j + \alpha_{ijk}^{(2)} b_j b_k] - (\partial/\partial b_j)[\beta_j - \gamma_{jk} b_k] \\ & + (\partial^2/\partial a_i \partial a_l) L_{il}^{AA} + 2(\partial^2/\partial a_i \partial b_j) L_{ij}^{AB} + (\partial^2/\partial b_j \partial b_k) L_{jk}^{BB} \end{aligned} \quad (2.7)$$

The probability density  $P(a, b, t)$  is given by

$$P(a, b) = \int db P(a, b, t). \quad (2.8)$$

Let us put

$$P(a, b, t) = P(a, t) q(b|a, t), \quad (2.9)$$

$$\int db q(b|a, t) = 1, \quad (2.10)$$

and define, for an arbitrary function  $G(b)$ ,

$$\langle G(b)|a, t \rangle \equiv \int db G(b) q(b|a, t). \quad (2.11)$$

Integrating (2.6) over  $b$  we obtain

$$\begin{aligned} \frac{\partial}{\partial t} P(a, t) = & \left[ -\frac{\partial}{\partial a_j} \{v_j(a) + \alpha_{jk}^{(1)}(a) \langle b_k|a, t \rangle + \alpha_{jkl}^{(2)}(a) \langle b_k b_l|a, t \rangle\} \right. \\ & \left. + \left( \frac{\partial^2}{\partial a_j \partial a_k} \right) \langle L_{jk}^{AA}|a, t \rangle \right] P(a, t). \end{aligned} \quad (2.12)$$

From (2.6) (2.9) and (2.11) we obtain

$$\begin{aligned} \frac{\partial q(b|a, t)}{\partial t} = & \left\{ -\left( \frac{\partial}{\partial b_j} [\beta_j - \gamma_{jk} b_k] + \left( \frac{\partial^2}{\partial b_j \partial b_k} \right) L_{jk}^{BB} \right) q(b|a, t) \right. \\ & - \frac{1}{P(a, t)} \left\{ \left( \frac{\partial}{\partial a_j} \right) [\alpha_{jk}^{(1)} \widehat{b}_k + \alpha_{jkl}^{(2)} \widehat{b}_k \widehat{b}_l] - 2 \left( \frac{\partial^2}{\partial a_j \partial b_k} \right) L_{jk}^{AB} \right\} P(a, t) q(b|a, t) \\ & - \left\{ v_j + \alpha_{jk}^{(1)} \langle b_k|a, t \rangle + \alpha_{jkl}^{(2)} \langle b_k b_l|a, t \rangle - \frac{1}{P(a, t)} \frac{\partial}{\partial a_k} \langle L_{jk}^{AA}|a, t \rangle P(a, t) \right\} \\ & \left. \times \frac{\partial}{\partial a_j} q(b|a, t) - \frac{q(b|a, t)}{P(a, t)} \frac{\partial^2}{\partial a_j \partial a_k} \langle L_{jk}^{AA}|a, t \rangle P(a, t) \right\}, \end{aligned} \quad (2.13)$$

$$\text{where } \widehat{b}_j = b_j - \langle b_j | a, t \rangle, \quad (2.14)$$

$$\langle \widehat{b}_j \widehat{b}_k \rangle = b_j b_k - \langle b_j b_k | a, t \rangle. \quad (2.15)$$

Thus we obtain from (2.11) and (2.13)

$$\begin{aligned} \partial \langle b_j | a, t \rangle / \partial t &= \beta_j - \gamma_{jk} \langle b_k | a, t \rangle - \left\{ v_j + \alpha_{jk}^{(1)} \langle b_k | a, t \rangle + \alpha_{jkl}^{(2)} \langle b_k b_l | a, t \rangle \right. \\ &\quad \left. - \frac{1}{P(a, t)} \frac{\partial}{\partial a_k} \langle L_{jk}^{AA} | a, t \rangle P(a, t) \right\} \times \left\{ \frac{\partial}{\partial a_j} \langle b_j | a, t \rangle \right\} - \frac{1}{P(a, t)} \\ &\quad \frac{\partial}{\partial a_j} \left\{ \alpha_{jk}^{(1)} \chi_{jk}(a, t) + \alpha_{jkl}^{(2)} \chi_{jkl}^{(3)}(a, t) + 2 \langle L_{jj}^{AB} | a, t \rangle \right\} P(a, t) - \frac{\langle b_j | a, t \rangle}{P(a, t)} \\ &\quad \frac{\partial^2}{\partial a_j \partial a_k} \langle L_{jk}^{AA} | a, t \rangle P(a, t) \end{aligned} \quad (2.16)$$

where

$$\chi_{jk}(a, t) = \langle \widehat{b}_j \widehat{b}_k | a, t \rangle, \quad (2.17)$$

$$\chi_{jkl}^{(3)}(a, t) = \langle b_j \widehat{b}_k \widehat{b}_l | a, t \rangle = \langle \widehat{b}_j \widehat{b}_k \widehat{b}_l | a, t \rangle. \quad (2.18)$$

Then (2.16) leads to, for  $t \gg \tau_B$ ,

$$\begin{aligned} \langle b_j | a, t \rangle &\cong [\gamma^{-1}(a)]_{jk} \cdot \left[ \beta_k(a) - \left\{ v_f(a) + \alpha_{fm}^{(1)}(a) \gamma_{ml}^{-1} \beta_l + \alpha_{fmn}^{(2)} \{ \gamma_{mp}^{-1} \beta_p \gamma_{nq}^{-1} \beta_q \right. \right. \\ &\quad \left. \left. + \chi_{mn} \right\} - \frac{1}{P(a, t)} \frac{\partial}{\partial a_m} \langle L_{fm}^{AA} | a, t \rangle P(a, t) \right\} \left\{ \frac{\partial}{\partial a_f} \gamma_{kl}^{-1} \beta_l \right\} \\ &\quad - \frac{1}{P(a, t)} \frac{\partial}{\partial a_f} \left\{ \alpha_{fm}^{(1)} \chi_{km}(a, t) + \alpha_{fmn}^{(2)} \chi_{kmn}^{(3)}(a, t) + 2 \langle L_{jk}^{BB} | a, t \rangle \right\} P(a, t) \\ &\quad - \frac{\gamma_{jr}^{-1} \beta_r}{P(a, t)} \frac{\partial^2}{\partial a_f \partial a_m} \langle L_{jm}^{AA} | a, t \rangle P(a, t) \Big]. \end{aligned} \quad (2.19)$$

We also obtain from (2.13)

$$\begin{aligned} \frac{\partial \langle b_m b_n | a, t \rangle}{\partial t} &= \int b_m b_n \frac{\partial q(b | a, t)}{\partial t} db \\ &= \beta_m \langle b_n | a, t \rangle + \beta_n \langle b_m | a, t \rangle - \gamma_{nk} \langle b_m b_k | a, t \rangle \\ &\quad - \gamma_{mk} \langle b_n b_k | a, t \rangle + 2 \langle L_{mn}^{BB} | a, t \rangle \\ &\quad - \frac{1}{P(a, t)} \left\{ \frac{\partial}{\partial a_j} \left[ \alpha_{jk}^{(1)} \langle \widehat{b}_m \widehat{b}_n \widehat{b}_k | a, t \rangle + \alpha_{jkl}^{(2)} \langle \widehat{b}_m \widehat{b}_n \widehat{b}_k \widehat{b}_l | a, t \rangle \right] \right. \\ &\quad \left. + 2 \frac{\partial}{\partial a_j} \left[ \langle b_m L_{jn}^{AB} | a, t \rangle + \langle b_n L_{jm}^{AB} | a, t \rangle \right] \right\} P(a, t) \\ &\quad - \left\{ v_j + \alpha_{jk}^{(1)} \langle b_k | a, t \rangle + \alpha_{jkl}^{(2)} \langle b_k b_l | a, t \rangle \right\} \frac{\partial}{\partial a_j} \langle b_m b_n | a, t \rangle \\ &\quad - \frac{1}{P(a, t)} \langle b_m b_n | a, t \rangle \frac{\partial^2}{\partial a_f \partial a_k} \langle L_{jk}^{AA} | a, t \rangle P(a, t). \end{aligned} \quad (2.20)$$

Using the following approximations:

$$\widehat{b}_m \widehat{b}_n = \widehat{b}_m \widehat{b}_n + \widehat{b}_m \langle b_n \rangle + \widehat{b}_n \langle b_m \rangle - \langle \widehat{b}_m \widehat{b}_n \rangle, \quad (2.21)$$

$$\chi_{kmn}^{(3)} = \langle \widehat{b}_m \widehat{b}_n \widehat{b}_k \rangle = \langle \widehat{b}_m \widehat{b}_n \widehat{b}_k \rangle + \langle \widehat{b}_m \widehat{b}_k \rangle \langle b_n \rangle + \langle \widehat{b}_n \widehat{b}_k \rangle \langle b_m \rangle, \quad (2.22)$$

$$\chi_{mn,kl}^{(4)} = \langle \widehat{b}_m \widehat{b}_n \widehat{b}_k \widehat{b}_l \rangle = \langle (\widehat{b}_m \widehat{b}_n + \widehat{b}_m \langle b_n \rangle + \widehat{b}_n \langle b_m \rangle - \langle \widehat{b}_m \widehat{b}_n \rangle)$$

$$\times (\widehat{b_k b_l} + \widehat{b_k} \langle b_l \rangle + \widehat{b_l} \langle b_k \rangle - \langle \widehat{b_k b_l} \rangle), \quad (2.23)$$

we obtain

$$\langle b_j | a, t \rangle \equiv [\gamma^{-1}]_{jk} \{ \beta_k - H_f^{(0)} \frac{\partial}{\partial a_f} (\gamma_{kl}^{-1} \beta_l) - P^{-1} \frac{\partial}{\partial a_f} I_{fk} P \} \quad (2.24)$$

$$\langle \widehat{b_m} \widehat{b_n} | a, t \rangle \equiv \chi_{mn} - H_f^{(0)} ((\partial \chi_{kl} / \partial a_f))_{mn} \quad (2.25)$$

$$- ((I_{fs} \frac{\partial}{\partial a_f} \gamma_{il}^{-1} \beta_l))_{mn} - ((I_{ft} \frac{\partial}{\partial a_f} \gamma_{sl}^{-1} \beta_l))_{mn} - P^{-1} \frac{\partial}{\partial a_f} ((K_{f,kl})_{mn} P),$$

where

$$H_f^{(0)} = v_f + \alpha_{fm}^{(1)} \gamma_{ml}^{-1} \beta_l + \alpha_{fmn}^{(2)} \{ \gamma_{mp}^{-1} \beta_p \gamma_{nq}^{-1} \beta_q + \chi_{mn} \}, \quad (2.26)$$

$$I_{fk} = \alpha_{fm}^{(1)} \chi_{km} + 2 \alpha_{fmn}^{(2)} \chi_{mk} \gamma_{nr}^{-1} \beta_r + 2 \langle L_{fk}^{AB} | a, t \rangle, \quad (2.27)$$

$$K_{f, mn} = \alpha_{fkl}^{(2)} \chi_{mn} \chi_{kl} + 2 \{ \langle \widehat{b_m} L_{fn}^{AB} \rangle + \langle \widehat{b_n} L_{fm}^{AB} \rangle \} \quad (2.28)$$

$$((A_{kl})_{mn}) \equiv \int_0^\infty \tilde{d}s [e^{-\tau s}]_{mk} A_{kl} [e^{-\tau s}]_{nl}, \quad (2.29)$$

$$\chi_{mn} = 2 \langle (L_{kl}^{BB})_{mn} \rangle. \quad (2.30)$$

Then (2.12), (2.24) and (2.25) lead to

$$\frac{\partial}{\partial t} P(a, t) = - \frac{\partial}{\partial a_j} \left\{ H_j^{(0)} + H_j^{(1)} - \frac{\partial}{\partial a_f} E_{jf} \right\} P(a, t), \quad (2.31)$$

where

$$\begin{aligned} H_j^{(1)} = & -H_f^{(0)} \left\{ \left[ \alpha_{jk}^{(1)} + 2 \alpha_{jkl}^{(2)} \gamma_{lm}^{-1} \beta_m \right] \gamma_{kp}^{-1} \frac{\partial}{\partial a_f} \left[ \gamma_{pr}^{-1} \beta_r \right] \right. \\ & + \alpha_{jkl}^{(2)} ((\partial \chi_{mr} / \partial a_f))_{kl} + I_{fp} \frac{\partial}{\partial a_f} \left\{ \left( \alpha_{jk}^{(1)} + 2 \alpha_{jkl}^{(2)} \gamma_{lm}^{-1} \beta_m \right) \gamma_{kp}^{-1} \right\} \\ & \left. - 2 \alpha_{jkl}^{(2)} ((I_{fn} \frac{\partial}{\partial a_f} \gamma_{mt}^{-1} \beta_t))_{kl} + ((K_{f, mn})_{kl}) \frac{\partial}{\partial a_f} \alpha_{jkl}^{(2)}, \right. \end{aligned} \quad (2.32)$$

$$E_{jf} = \langle L_{jf}^{AA} | a, t \rangle + \left[ \alpha_{jk}^{(1)} + 2 \alpha_{jkl}^{(2)} \gamma_{lm}^{-1} \beta_m \right] \gamma_{kp}^{-1} I_{fp} + \alpha_{jkl}^{(2)} ((K_{f, mn})_{kl}). \quad (2.33)$$

### § 3. Fokker-Planck equation for the Stokes-scattered light

In this section we consider the generation of Stokes-shifted light in Raman-type process.<sup>7)</sup> If we set  $A = \text{Col}(a_1, a_2) = \text{Col}(a_1, a_1^*)$ ,  $B = \text{Col}(b_1, b_2) = \text{Col}(b_1, b_1^*)$ , then we have

$$\begin{aligned} v_i = & \begin{pmatrix} -\gamma_a a_1 \\ -\gamma_a a_1^* \end{pmatrix}, \quad \beta_j = \begin{pmatrix} E_p \\ E_p^* \end{pmatrix}, \quad \gamma_{jk} = \begin{pmatrix} \gamma & 0 \\ 0 & \gamma \end{pmatrix}, \\ \alpha_{ijk}^{(2)} = & \frac{1}{2} g a_i \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} = \frac{1}{2} g a_i (1 - \delta_{jk}), \end{aligned} \quad (3.1)$$

and  $\alpha_{ij}^{(1)} = 0$  where  $\gamma = \gamma_p + g(a_1 a_1^* + 1)$ .

$A$  describes the Stokes-scattered light amplitude, while  $B$  characterizes the laser mode inside the cavity coupled to the resonant external source  $P$ . The resonant external source  $P$  contains the coherent part  $E$  and the fluctuating part  $F_0$ .  $\gamma_a$  and  $\gamma_b$  are relaxation rates and  $g$  is a coupling constant.

Fluctuating forces  $r_i$  and  $F_0$  are assumed to be Gaussian white noises with  $\langle r_i(t) \rangle = \langle F_0(t) \rangle = 0$  and

$$\begin{aligned} \langle r_j^\mu(t) r_k^\nu(s) \rangle &= 2L_{jk}^{\mu\nu} \delta(t-s), \\ \langle F_0(t) F_0^*(s) \rangle &= 2l_{00} \delta(t-s), \end{aligned} \quad (3.2)$$

$$L_{jk}^{AA} = \begin{pmatrix} 0 & \gamma_a n_a \\ \gamma_a n_a & 0 \end{pmatrix}, \quad L_{jk}^{BB} = \begin{pmatrix} 0 & l_{BB} \\ l_{BB} & 0 \end{pmatrix},$$

and  $L_{jk}^{AB} = 0$ ,

where  $l_{BB} = \gamma_b n_b + (2g)^{-2} l_{00}$ ,  
 $n_a = \langle a_i a_i^* \rangle$ ,  $n_b = \langle b_i b_i^* \rangle$ .

According to § 2, (2.26), (2.32) (3.1) and (3.2) lead to

$$\begin{aligned} H_j^{(0)} &= \{-\gamma_a + g(\gamma^{-2} \beta_1 \beta_2 + \gamma^{-1} l_{BB})\} a_j, \\ H_j^{(1)} &= H_j^{(0)} g^2 a_1 a_2 \gamma^{-3} (l_{BB} + 4\gamma^{-1} \beta_1 \beta_2) \\ &\quad + \frac{1}{2} g^2 \gamma^{-2} a_j [\gamma^{-1} l_{BB}^2 + 4\gamma^{-2} \beta_1 \beta_2 l_{BB}] \\ &\quad - 6g^3 \gamma^{-3} \beta_1 \beta_2 a_1 a_2 a_j l_{BB} \\ E_{ij} &= \gamma_a n_a (1 - \delta_{ij}) + \frac{1}{2} g^2 a_i a_j \gamma^{-3} l_{BB} (l_{BB} + 4\gamma^{-1} \beta_1 \beta_2). \end{aligned} \quad (3.3)$$

Therefore (2.31) leads to

$$\begin{aligned} \frac{\partial P(a_1, a_2, t)}{\partial t} &= -\frac{\partial}{\partial a_1} \left[ H_j^{(0)} \left\{ 1 + g^2 a_1 a_2 \gamma^{-3} (l_{BB} + 4\gamma^{-1} \beta_1 \beta_2) \right\} \right. \\ &\quad \left. + \frac{1}{2} g^2 \gamma^{-3} a_1 l_{BB} (l_{BB} + 4\gamma^{-1} \beta_1 \beta_2) - 6g^3 \gamma^4 \beta_1 \beta_2 l_{BB} a_1^2 a_2 \right] P \\ &\quad + \frac{\partial^2}{\partial a_1^2} \left[ \frac{1}{2} g^2 a_1^2 \gamma^{-3} l_{BB} (l_{BB} + 4\gamma^{-1} \beta_1 \beta_2) \right] P \\ &\quad + \frac{\partial^2}{\partial a_1 \partial a_2} \left[ \gamma_a n_a + \frac{1}{2} g^2 a_1 a_2 \gamma^{-3} l_{BB} (l_{BB} + 4\gamma^{-1} \beta_1 \beta_2) \right] P + \text{comp. conj} \end{aligned} \quad (3.4)$$

The Langevin equation for  $a_1$  takes the form

$$\begin{aligned} \frac{d a_1}{d t} &= H_j^{(0)} \left\{ 1 + g^2 \gamma^{-3} a_1 a_2 (l_{BB} + 4\gamma^{-1} \beta_1 \beta_2) \right\} \\ &\quad + \frac{1}{2} g^2 \gamma^{-3} l_{BB} a_1 (l_{BB} + 4\gamma^{-1} \beta_1 \beta_2) \\ &\quad - 6g^3 \gamma^{-4} \beta_1 \beta_2 l_{BB} a_1^2 a_2 \quad + \text{fluctuating force}. \end{aligned} \quad (3.5)$$

Using

$$\begin{aligned} a_1 &= \sqrt{n} e^{i\varphi}, & a_2 &= \sqrt{n} e^{-i\varphi}, \\ \beta_1 &= \beta_0 e^{i\theta}, & \beta_2 &= \beta_0 e^{-i\theta}, \end{aligned} \quad (3.6)$$

we can transform (3.4) into the equation for the photon number  $n$ . Thus we obtain, for the Fokker-Planck equation,

$$\begin{aligned} \frac{\partial p(n, t)}{\partial t} &= -\frac{\partial}{\partial n} \left[ H(n) P(n, t) \right] + \frac{\partial^2}{\partial n^2} \left[ E(n) P(n, t) \right], \\ H(n) &= 2n \{ B(n) + 2M(n) + nN(n) \} + 2A_0, \\ E(n) &= 2n \{ A_0 + 2nM(n) \}, \end{aligned} \quad (3.7)$$

where

$$\begin{aligned} B(n) &= \{ -\gamma_a + g\gamma^{-1}(l_{BB} + \gamma^{-1}\beta_0^2) \} \\ &\quad \times \{ 1 + g^2\gamma^{-3}n(l_{BB} + 4\gamma^{-1}\beta_0^2) \}, \\ M(n) &= \frac{1}{2} g^2\gamma^{-3}l_{BB}(l_{BB} + 4\gamma^{-1}\beta_0^2) \\ N(n) &= -6g^3\gamma^{-5}\beta_0^2l_{BB}, \\ A_0 &= \gamma_a n_a. \end{aligned} \quad (3.8)$$

The Langevin equation for  $n$  takes the form

$$\frac{dn}{dt} = 2n \{ B(n) + 2M(n) + nN(n) \} + R_n(t), \quad (3.9)$$

where  $R_n$  is a fluctuating force.

#### § 4. Summary

An adiabatic elimination from the Fokker-Planck equation corresponding to the Langevin equations contained the non-linear coupling in irrelevant variables has developed. Of particular interest is the behavior of fluctuations in Raman scattering. The Fokker-Planck equation and the Langevin equation for the relevant variable in Raman scattering are obtained. A detailed investigation of the critical phenomena about these equations will be given on another occasion.

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