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Stochastic Processes in Raman Scattering

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Abstruct

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.[1-2] We have studied the ordered structures in non-linear chemical reactions and in non-linear optics.[3-5] 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 develope 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

$$< r_i^A(t) r_l^A(t');\ a_0 b_0 > = 2L_{il}^{AA}\delta(t-t'),\quad L_{il}^{AA} = L_{li}^{AA}, \tag{2.3}$$

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

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

Here $<\cdots; a_0 b_0>$ 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 τ_A of $A(t)$ is distinctly larger than the time scale τ_B of $B(t)$.

The Fokker-Planck equation corresponding to the Langevin equations (2.1) and (2.2) is given by[6]

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

where

$$\Gamma^+(a,b) = -(\partial/\partial a_i)[v_i(a) + \alpha_{ij}^{(1)} b_j + \alpha_{ijk}^{(2)} b_j b_k] - (\partial/\partial b_j)[\beta_j - \gamma_{jk} b_k] \tag{2.7}$$

$$+ (\partial^2/\partial a_i \partial a_l) L_{il}^{AA} + 2(\partial^2/\partial a_i \partial b_j) L_{ij}^{AB} + (\partial^2/\partial b_j \partial b_k) L_{jk}^{BB}$$

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

$$P(a,b) = \int db\ P(a,b,t). \tag{2.8}$$

Let us put

$$P(a,b,t) = P(a,t) q(b|a,t), \tag{2.9}$$

$$\int db\ q(b|a,t) = 1, \tag{2.10}$$

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

$$<G(b)|a,\ t> \equiv \int db\ G(b) q(b|a,t). \tag{2.11}$$

Integrating (2.6) over b we obtain

$$\frac{\partial}{\partial t} P(a,\ t) = [-\frac{\partial}{\partial a_j}\{v_j(a) + \alpha_{jk}^{(1)}(a) <b_k|a,t> + \alpha_{jkl}^{(2)}(a) <b_k b_l|a,t>\}$$

$$+ \left(\frac{\partial^2}{\partial a_j \partial a_k}\right) <L_{jk}^{AA}|a\ t>] P(a,t). \tag{2.12}$$

From (2. 6) (2.9) and (2.11) we obtain

$$\frac{\partial q(b|a\ t)}{\partial t} = \left\{-\left(\frac{\partial}{\partial b_j}[\beta_j - \gamma_{jk} b_k] + \left(\frac{\partial^2}{\partial b_j \partial b_k}\right) L_{jk}^{BB}\right\} q(b|a,t)$$

$$- \frac{1}{P(a,t)}\left\{\left(\frac{\partial}{\partial a_j}\right)[\alpha_{jk}^{(1)}\widehat{b_k} + \alpha_{jkl}^{(2)}\widehat{b_k b_l}] - 2\left(\frac{\partial^2}{\partial a_j \partial b_k}\right) L_{jk}^{AB}\right\} P(a,t) q(b|a,t)$$

$$-\left\{v_j + \alpha_{jk}^{(1)} <b_k|a,t> + \alpha_{jkl}^{(2)} <b_k b_l|a,t> - \frac{1}{P(a,t)}\frac{\partial}{\partial a_k} <L_{jk}^{AA}|a,t> P(a,t)\right\}$$

$$\times \frac{\partial}{\partial a_j} q(b|a,t) - \frac{q(b|a,t)}{P(a\ t)}\frac{\partial^2}{\partial a_j \partial a_k} <L_{jk}^{AA}|a,t> P(a\ t), \tag{2.13}$$

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

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

Thus we obtain from (2.11) and (2.13)

$$\partial \langle b_j | a,t \rangle / \partial t = \beta_j - \gamma_{jk} \langle b_k | a,t \rangle - \Big\{ v_f + \alpha^{(1)}_{fk} \langle b_k | a,t \rangle + \alpha^{(2)}_{fkl} \langle b_k b_l | a,t \rangle - \frac{1}{P(a,t)} \frac{\partial}{\partial a_k} \langle L^{AA}_{fk} | a,t \rangle P(a,t) \Big\} \times \Big\{ \frac{\partial}{\partial a_f} \langle b_j | a,t \rangle \Big\} - \frac{1}{P(a,t)} \frac{\partial}{\partial a_f} \Big\{ \alpha^{(1)}_{fk} \chi_{jk}(a,t) + \alpha^{(2)}_{fkl} \chi^{(3)}_{jkl}(a,t) + 2 \langle L^{AB}_{fj} | a,t \rangle \Big\} P(a,t) - \frac{\langle b_j | a,t \rangle}{P(a\ t)} \frac{\partial^2}{\partial a_f \partial a_k} \langle L^{AA}_{fk} | a,t \rangle P(a,t) \tag{2.16}$$

where

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

$$\chi^{(3)}_{jkl}(a,t) = \langle b_j \widehat{b_k b_l} | a,t \rangle = \langle \hat{b}_j \widehat{b_k b_l} | a,t \rangle. \tag{2.18}$$

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

$$\langle b_j | a,t \rangle \cong [\gamma^{-1}(a)]_{jk} \cdot \Big[\beta_k(a) - \Big\{ v_f(a) + \alpha^{(1)}_{fm}(a) \gamma^{-1}_{ml} \beta_l + \alpha^{(2)}_{fmn} \{ \gamma^{-1}_{mp} \beta_p \gamma^{-1}_{nq} \beta_q + \chi_{mn} \Big\} - \frac{1}{P(a,t)} \frac{\partial}{\partial a_m} \langle L^{AA}_{fm} | a,t) \rangle P(a,t) \Big\{ \frac{\partial}{\partial a_f} \gamma^{-1}_{kl} \beta_l \Big\} - \frac{1}{P(a,t)} \frac{\partial}{\partial a_f} \Big\{ \alpha^{(1)}_{fm} \chi_{km}(a,t) + \alpha^{(2)}_{fmn} \chi^{(3)}_{kmn}(a,t) + 2 \langle L^{BB}_{fk} | a,t \rangle \Big\} P(a,t) - \frac{\gamma^{-1}_{jr} \beta_r}{P(a,t)} \frac{\partial^2}{\partial a_f \partial a_m} \langle L^{AA}_{fm} | a,t \rangle P(a,t) \Big]. \tag{2.19}$$

We also obtain from (2.13)

$$\frac{\partial \langle b_m b_n | a,t \rangle}{\partial t} = \int b_m b_r \frac{\partial q(b|a,t)}{\partial t} db$$

$$= \beta_m \langle b_n | a,t \rangle + \beta_n \langle b_m | a,t \rangle - \gamma_{nk} \langle b_m b_k | a,t \rangle - \gamma_{mk} \langle b_n b_k | a,t \rangle + 2 \langle L^{BB}_{mn} | a\ t \rangle - \frac{1}{P(a,t)} \Big\{ \frac{\partial}{\partial a_j} \Big[\alpha^{(1)}_{jk} \langle \widehat{b_m b_n} \hat{b}_k | a\ t \rangle + \alpha^{(2)}_{jkl} \langle \widehat{b_m b_n} \widehat{b_k b_l} | a,t \rangle \Big] + 2 \frac{\partial}{\partial a_j} \Big[\langle b_m L^{AB}_{jn} | a,t \rangle + \langle b_n L^{AB}_{jm} | a,t \rangle \Big] \Big\} P(a,t) - \Big\{ v_j + \alpha^{(1)}_{jk} \langle b_k | a,t \rangle + \alpha^{(2)}_{jkl} \langle b_k b_l | a,t \rangle \Big\} \frac{\partial}{\partial a_f} \langle b_m b_n | a,t \rangle - \frac{1}{P(a,t)} \langle b_m b_n | a,t \rangle \frac{\partial^2}{\partial a_f \partial a_k} \langle L^{AA}_{jk} | a\ t \rangle P(a,t). \tag{2.20}$$

Using the following approximations:

$$\widehat{b_m b_n} = \hat{b}_m \hat{b}_n + \hat{b}_m \langle b_n \rangle + \hat{b}_n \langle b_m \rangle - \langle \hat{b}_m \hat{b}_n \rangle, \tag{2.21}$$

$$\chi^{(3)}_{kmn} = \langle \widehat{b_m b_n} \hat{b}_k \rangle = \langle \hat{b}_m \hat{b}_n \hat{b}_k \rangle + \langle \hat{b}_m \hat{b}_k \rangle \langle b_n \rangle + \langle \hat{b}_n \hat{b}_k \rangle \langle b_m \rangle, \tag{2.22}$$

$$\chi^{(4)}_{mn,kl} = \langle \widehat{b_m b_n} \widehat{b_k b_l} \rangle = \langle (\hat{b}_m \hat{b}_n + \hat{b}_m \langle b_n \rangle + \hat{b}_n \langle b_m \rangle - \langle \hat{b}_m \hat{b}_n \rangle)$$

$$\times(\widehat{b_k}\widehat{b_l}+\widehat{b_k}<b_l>+\widehat{b_l}<b_k>-<\widehat{b_k}\widehat{b_l}>)>, \tag{2.23}$$

we obtain

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

$$<\widehat{b_m}\widehat{b_n}|a,t>\cong\chi_{mn}-H_f^{(0)}((\partial\chi_{kl}/\partial a_f))_{mn} \tag{2.25}$$

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

where

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

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

$$K_{f,mn}=\alpha_{fkl}^{(2)}\chi_{mn}\chi_{kl}+2\left\{<\widehat{b_m}L_{fn}^{AB}>+<\widehat{b_n}L_{fm}^{AB}>\right\} \tag{2.28}$$

$$((A_{kl}))_{mn}\equiv\int_0^\infty ds\,[e^{-\gamma s}]_{mk}A_{kl}[e^{-\gamma s}]_{nl}, \tag{2.29}$$

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

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

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

where

$$H_j^{(1)}=-H_f^{(0)}\left\{\left[\alpha_{jk}^{(1)}+2\alpha_{jkl}^{(2)}\gamma_{lm}^{-1}\beta_m\right]\gamma_{kp}^{-1}\frac{\partial}{\partial a_f}\left[\gamma_{pr}^{-1}\beta_n\right]\right.$$

$$+\alpha_{jkl}^{(2)}((\partial\chi_{mr}/\partial a_f))_{kl}+I_{fp}\frac{\partial}{\partial a_f}\left\{(\alpha_{jk}^{(1)}+2\alpha_{jkl}^{(2)}\gamma_{lm}^{-1}\beta_m)\gamma_{kp}^{-1}\right\}$$

$$-2\alpha_{jkl}^{(2)}((I_{fn}\frac{\partial}{\partial a_f}\gamma_{mt}^{-1}\beta_t))_{kl}+((K_{f\ mn}))_{kl}\frac{\partial}{\partial a_f}\alpha_{fkl}^{(2)}, \tag{2.32}$$

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

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

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

$$v_i=\begin{pmatrix}-\gamma_a\ a_1\\-\gamma_a\ a_1^*\end{pmatrix},\quad \beta_j=\begin{pmatrix}E_p\\E_p^*\end{pmatrix}\quad \gamma_{jk}=\begin{pmatrix}\gamma&0\\0&\gamma\end{pmatrix},$$

$$\alpha_{ijk}^{(2)}=\frac{1}{2}g\,a_i\begin{pmatrix}0&1\\1&0\end{pmatrix}=\frac{1}{2}g\,a_i\,(1-\delta_{jk}), \tag{3.1}$$

and $\alpha_{ij}^{(1)}=0$ where $\gamma=\gamma_p+g\,(a_1a_1^*+1)$.

A describes the Stokes-scattered light amplitude, while B characterizes the laser mode inside the cavity coupled to the resonant external souse P. The resonant external sourse P contains the coherent part E and the fluctuating part F_0. γ_a and γ_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 $< r_i(t) > = < F_0(t) > = 0$ and

$$< r_f^\mu(t) r_k^\nu(s) > = 2L_{fk}^{\mu\nu}\delta(t-s),$$

$$< F_0(t) F_0^*(s) > = 2l_{00}\delta(t-s), \tag{3 2}$$

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

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

where $\quad l_{BB} = \gamma_b n_b + (2g)^{-2} l_{00},$

$\quad n_a = < a_1 a_1^* >, \; n_b = < b_1 \, b_1^* >.$

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

$$H_j^{(0)} = \{-\gamma_a + g(\gamma^{-2}\beta_1\beta_2 + \gamma^{-1}l_{BB})\} a_j,$$

$$\begin{aligned} H_j^{(1)} &= H_j^{(0)} g^2 a_1 a_2 \gamma^{-3}(l_{BB} + 4\gamma^{-1}\beta_1\beta_2) \\ &+ \frac{1}{2} g^2\gamma^{-2} a_j [\gamma^{-1} l^2_{BB} + 4\gamma^{-2}\beta_1\beta_2 l_{BB}] \\ &- 6g^3\gamma^{-5}\beta_1\beta_2 a_1 a_2 a_j l_{BB} \end{aligned} \tag{3.3}$$

$$E_{ij} = \gamma_a n_a (1-\delta_{ij}) + \frac{1}{2} g^2 a_i a_j \gamma^{-3} l_{BB}(l_{BB} + 4\gamma^{-1}\beta_1\beta_2).$$

Therefore (2.31) leads to

$$\begin{aligned} \frac{\partial P(a_1 a_2, t)}{\partial t} &= -\frac{\partial}{\partial a_1}\Big[H_1^{(0)}\Big\{1 + g^2 a_1 a_2 \gamma^{-3}(l_{BB} + 4\gamma^{-1}\beta_1\beta_2)\Big\} \\ &+ \frac{1}{2} g^2\gamma^{-3} a_1 l_{BB}(l_{BB} + 4\gamma^{-1}\beta_1\beta_2) - 6g^3\gamma^4\beta_1\beta_2 l_{BB} a_1^2 a_2\Big] P \\ &+ \frac{\partial^2}{\partial a_1^2}\Big[\frac{1}{2} g^2 a^2_1 \gamma^{-3} l_{BB}(l_{BB} + 4\gamma^{-1}\beta_1\beta_2)\Big] P \\ &+ \frac{\partial^2}{\partial a_1 \partial a_2}\Big[\gamma_a n_a + \frac{1}{2} g^2 a_1 a_2 \gamma^{-3} l_{BB}(l_{BB} + 4\gamma^{-1}\beta_1\beta_2)\Big] P + comp.\ conj \end{aligned} \tag{3.4}$$

The Langevin equation for a_1 takes the form

$$\begin{aligned} \frac{d a_1}{dt} &= H_1^{(0)}\Big\{1 + g^2\gamma^{-3} a_1 a_2 (l_{BB} + 4\gamma^{-1}\beta_1\beta_2)\Big\} \\ &+ \frac{1}{2} g^? \gamma^{-3} l_{BB} a_1 (l_{BB} + 4\gamma^{-1}\beta_1\beta_2) \\ &- 6g^3\gamma^{-4}\beta_1\beta_2 l_{BB} a^2_1 a_2 \quad + \textit{fluctuating force}. \end{aligned} \tag{3.5}$$

Using

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

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

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

where

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

The Langevin equation for n takes the form

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

where R_n is a fluctuating force.

§4. Summary

An adiabatic elimination from the Fokker-Planck equation correrponding 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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References

1) H. Haken, *Synergetics* (Springer-Verlag, Berlin, 1977).
2) G. Nicolis and I. Prigogine, *Self-Organization in Nonequilibrium Systems* (Wiley, New York, 1977).
3) Y. Fukuyama, Sci. Bull. Fac. Educ., Nagasaki Univ. **30** (1979) , 15.
4) Y. Fukuyama and H. Mori, Prog. Theor. Phys. **63** (1980) ,1181.
5) Y. Fukuyama, Sci. Bull. Fac. Educ., Nagasaki Univ. **32** (1981), 15.
6) T. Morita, H. Mori and K.T. Masiyama. Prog. Theor. Phys. **64** (1980), 500.
7) A. Schenzle and H. Brand, Phys. Rev. **A20** (1979) , 1628.