MULTIPLE QUANTUM TERMS IN THE FRÖHLICH KINETIC EQUATION

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Energy stored in a coherent state may shift the vibration system into a highly nonlinear region where strong interaction without the help of the heat bath may be significant. Multiple quantum processes of this type are analyzed.

Fröhlich derived the kinetic equation (FKE) determining the excitation of coherent states by the energy supply to a band of polarization waves consisting of normal modes [1,2]. Energy condensation occurs in the lowest frequency mode. Under convenient conditions the energy may condense even in higher frequency modes [3]. The FKE contains the fundamental terms based on one- and two-quantum processes representing energy supply, energy losses and energy channeling between the modes with the help of the heat bath (S, ϕ and X terms, respectively). Livshitz and Yushina derived the additional two-quantum terms [4–6]. If the transition probabilities of the additional terms are conveniently chosen these terms may compensate the Fröhlich condensation X term and a coherent state cannot be reached. In a normal state the aforementioned condition is very likely rather an artificial one [7–10]. Nevertheless, under extraordinary conditions the additional terms might cause the quenching of coherent states. On the other hand the FKE includes only the basic principle of the coherent state excitation process.

The energy stored in the lowest frequency mode as a function of energy supply steeply increases in the vicinity of the critical energy supplied to the system in question as follows from a computer simulation [11]. The whole system of oscillations is, therefore, shifted from the original state where the FKE may be a good approximation to a highly nonlinear region. Multiple quantum processes become significant and the behavior of the system changes.

We assume that besides the Fröhlich terms the direct interaction terms determined by a strong interaction between polarization waves without the help of the heat bath are significant in the highly nonlinear region. The possibility of a strong interaction without the heat bath help was discussed in ref. [2]. The hamiltonian of the biosystem can be given as:

$$H = H_0 + H_1 + H_2 , \tag{1}$$

where H_0 and H_1 satisfy the relations [10,12,13]

$$H_0 = \sum_i \hbar\omega_i a_i^+ a_i + \sum_k \hbar\Omega_k c_k^+ c_k + \sum_p \hbar\bar{\Omega}_p P_p^+ P_p , \tag{2}$$

$$H_1 = \sum_{i,k} (\varphi c_k^+ a_i + \varphi^* c_k a_i^+) + \sum_{i,p} (\xi P_p a_i^+ + \xi^* P_p^+ a_i) + \sum_{i,l,k} (\chi a_l^+ a_i c_k^+ + \chi^* a_l a_i^+ c_k) . \tag{3}$$

In eq. (1) we introduce the multiple quantum term H_2 given by

$$H_2 = \sum_{i,l,j} \sum_{m,n} [\kappa a_l^+ (a_i)^m (a_j)^n + \kappa^* a_l (a_i^+)^m (a_j^+)^n + \lambda a_l^+ (a_i^+)^m (a_j)^n + \lambda^* a_l (a_i)^m (a_j^+)^n + \mu a_l^+ (a_i)^m (a_j^+)^n + \mu^* a_l (a_i^+)^m (a_j)^n] . \tag{4}$$

Here $\omega_i, a_i^+, a_i; \Omega_k, c_k^+, c_k; \bar{\Omega}_p, P_p^+, P_p$ are the circular frequencies, creation operators and annihilation operators of the ith normal vibration mode of the system, of the kth normal vibration mode of the environmental medium and of the pth normal vibration mode

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of the source, respectively. φ, ξ, χ, κ, λ and μ in eqs. (2)–(4) are the coupling coefficients of particular processes. $(a_i)^m$ stands for $a_i a_i a_i \ldots a_i$ (m times). In eq. (2) the environmental medium (the heat bath) is represented only by the oscillator terms, which very likely is not satisfied in real systems but that has no influence on the heat bath independent terms.

We will use the time dependent perturbation theory to determine the perturbed state vector $|\psi(t)\rangle$ as was described by Wu and Austin in refs. [12,13]. The commutation relations enable us to evaluate the rate of change of the number of the qth mode quanta from the equation $\dot{N}_q = -(\mathrm{i}/\hbar)[N_q, H]$. From the equations for the expectation values $\langle \dot{N}_q \rangle = \langle \psi(t)|\dot{N}_q|\psi(t)\rangle$ we derive the general relations

$$\sum_{m=-\infty}^{\infty} \sum_{n=0}^{\infty} \frac{n\langle \dot{N}_q \rangle' \omega_q}{m\omega_i + n\omega_j} = 0 ,$$

$$\sum_{n=-\infty}^{\infty} \sum_{m=0}^{\infty} \frac{m\langle \dot{N}_q \rangle' \omega_q}{m\omega_i + n\omega_j} = 0 , \qquad (5)$$

where $N_q = a_q^+ a_q$, $\omega_q = |m\omega_i + n\omega_j|$ and $\langle \dot{N}_q \rangle'$ represents a part of the total expectation value connected with the interaction between modes without the help of the heat bath. The relations given by eq. (5) are identical with the famous Manley–Rowe power relations derived for nonlinear electrical elements of the classical type [14]. The total expectation values $\langle \dot{N}_q \rangle$ are subjected to the conditions

$$\sum_{m=-\infty}^{\infty} \sum_{n=0}^{\infty} \frac{n\langle \dot{N}_q \rangle \omega_q}{m\omega_i + n\omega_j} = \sum_{m=-\infty}^{\infty} \sum_{n=0}^{\infty} \frac{nF_q \omega_q}{m\omega_i + n\omega_j} ,$$

$$\sum_{n=-\infty}^{\infty} \sum_{m=0}^{\infty} \frac{m\langle \dot{N}_q \rangle \omega_q}{m\omega_i + n\omega_j} = \sum_{n=-\infty}^{\infty} \sum_{m=0}^{\infty} \frac{mF_q \omega_q}{m\omega_i + n\omega_j} , \qquad (6)$$

where F_q stands for the original FKE terms [1,2], i.e.

$$F_q = S_q - \phi[\langle N_q \rangle \exp(\beta\omega_q) - (1 + \langle N_q \rangle)]$$

$$- \sum_i X\{\langle N_q \rangle (1 + \langle N_i \rangle) \exp[\beta(\omega_q - \omega_i)]$$

$$- \langle N_i \rangle (1 + \langle N_q \rangle)\} . \qquad (7)$$

The modified kinetic equation may be written in the form

$$\langle \dot{N}_q \rangle = F_q + A_q , \qquad (8)$$

where A_q is derived from eq. (5).

We assume a simple case – the direct interaction without the heat bath help between three modes with the frequencies ω_1 ($m_1 = 1, n_1 = 0$), ω_2 ($m_2 = 0, n_2 = 1$) and ω_3 ($m_3 = m, n_3 = n$). The corresponding A_q values in eq. (8) are given by

$$A_1 = \langle \dot{N}_1 \rangle' , \quad A_2 = (n/m)\langle \dot{N}_1 \rangle' , \quad A_3 = -(1/m)\langle \dot{N}_1 \rangle' , \qquad (9)$$

respectively. We assume the energy stored in the coherently excited mode to be high enough. For $n < 0$ and $m > 0$ the energy is channeled from the ω_1 mode to the remaining two modes. The interaction process without the heat bath help may channel the energy to the higher frequency modes (upwards) if the mode frequencies are in convenient relationships. The Fröhlich X process transports the energy from the higher frequency modes to the lowest frequency one (downwards) with energy losses. Energy circulation occurs.

The interaction processes without the heat bath help lead to the energy saturation in the coherently excited mode, contribute to the coherent excitation of higher frequency modes [3] and cause the higher energy losses (the downward step of the energy circulation is connected with the losses). The mode frequency changes may disturb the nonlinear energy transfer without the heat bath help. If the energy supply decreases, transition from the highly nonlinear region into a region with the less pronounced nonlinearity takes place and the role of the multiple quantum processes is diminished.

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