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STATISTICAL PROPERTIES OF PHONONS IN POLARITON-LIKE SYSTEM AT EQUILIBRIUM

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In the present paper we will be concerned with a bilinear model of a polariton-like system — the subject of a great number of studies made so far. However, whereas most other papers have been dealing either with dispersion properties or with influence on optical properties of polar crystals, we will be investigating the quantum statistical characteristics of phonons. This problem has come more into the focus of attention because of interest in quantum fluctuations of the Bose-type excitations in solids [1]. Due to the strong bound between phonons and infrared photons leading to the strong dispersion [2], the stimulated combination scattering must depend on fluctuations of the number of phonons [3].

The simplest model in the theory of polaritons is described by the following Hamiltonian [4]:

$$H = \hbar\omega_a(a^\dagger a + 1/2) + \hbar\omega_b(b^\dagger b + 1/2) + i\hbar g(a + a^\dagger)(b - b^\dagger), \qquad (1)$$

where $a^\dagger$ and a are creation and annihilation operators of infrared photons, $b^\dagger$ and b are creation and annihilation operators of transverse optical phonons, g is a photon – phonon coupling constant, ω_a and ω_b are frequencies of photons and phonons, respectively. This bilinear form is diagonalized by the Bogolubov canonical transformation [5, 6]

$$c_j = u_{1j}a + u_{2j}b + v_{1j}a^\dagger + v_{2j}b^\dagger, \quad j = 1, 2 \qquad (2)$$

where the parameters u and v are defined by the expressions [7]:

$$u_{1j} = \frac{\Omega_j + \omega_a}{2\omega_a}\Phi_{1j}, \quad u_{2j} = \frac{(\Omega_j + \omega_b)(\Omega_j^2 - \omega_a^2) + 4\omega_a g^2}{2\Omega_j(\Omega_j^2 - \omega_a^2)}\Phi_{2j},$$
$$v_{1j} = \frac{\Omega_j - \omega_a}{2\omega_a}\Phi_{1j}, \quad v_{2j} = \frac{(\Omega_j - \omega_b)(\Omega_j^2 - \omega_a^2) + 4\omega_a g^2}{2\Omega_j(\Omega_j^2 - \omega_a^2)}\Phi_{2j}.$$

Here

$$|\Phi_{1j}|^2 = (-1)^{j+1}\frac{\omega_a(\Omega_j^2 - \omega_b^2)}{\Omega_j(\Omega_1^2 - \Omega_2^2)}, \quad |\Phi_{2j}|^2 = (-1)^{j+1}\frac{\omega_b(\Omega_j^2 - \omega_a^2)}{\Omega_j(\Omega_1^2 - \Omega_2^2)}$$

and

$$\Omega_j^2 = \frac{\omega_a^2 + \omega_b^2}{2} - \frac{(-1)^j}{2}\sqrt{(\omega_a^2 - \omega_b^2)^2 + 16\omega_a\omega_b g^2}.$$

As a result of this transformation, one gets the diagonal form of the Hamiltonian (1):

$$H = \sum_{j=1}^{2} \hbar\Omega_j (c_j^\dagger c_j + 1/2). \tag{3}$$

It should be noted that the operators $c_j^\dagger$ and c_j describe the polaritons. Let us now suppose that the system is in the equilibrium state with temperature T. This state is described by the following density matrix:

$$\rho(\beta) = \frac{e^{-\beta H}}{Tr\left\{e^{-\beta H}\right\}}, \tag{4}$$

where $\beta = 1/kT$ and H is defined by (3). Now we examine the quantum number distribution for phonons in the state with the density matrix (4). This state can be interpreted as a two–mode squeezed chaotic state, and it is a generalization of the state introduced in [8]. Using analogy with quantum optics we calculate the second–order degree of coherence [9]:

$$G_{bb} = \frac{\langle b^{\dagger 2} b^2 \rangle}{\langle b^\dagger b \rangle^2}, \tag{5}$$

where $\langle \ldots \rangle$ is termed the expectation value with the density matrix (4). Corresponding averages have the following form:

$$\begin{aligned} \langle b^\dagger b \rangle &= \mu_1 \bar{n_1} + \mu_2 \bar{n_2} + \eta, \\ \langle b^{\dagger 2} b^2 \rangle &= 2\left(\bar{n_1}\,\mu_1 + \bar{n_2}\,\mu_2 + \eta\right)^2 + (2\,\bar{n_1} + 1)^2 \nu_1^2 + (2\,\bar{n_2} + 1)^2 \nu_2^2 + \\ &+ 2(2\,\bar{n_1} + 1)(2\,\bar{n_2} + 1)\gamma \end{aligned} \tag{6}$$

and

$$\mu_1 = |u_{21}|^2 + |v_{21}|^2, \qquad \mu_2 = |u_{22}|^2 + |v_{22}|^2,$$
$$\nu_1 = |u_{21} v_{21}|, \qquad \nu_2 = |u_{22} v_{22}|,$$
$$\eta = |v_{21}|^2 + |v_{22}|^2, \qquad \gamma = Re\left(u_{21} u_{22}^* v_{21}^* v_{22}\right).$$

Here

$$\bar{n_i} = 1/\left(e^{\hbar\beta\Omega_i} - 1\right). \tag{7}$$

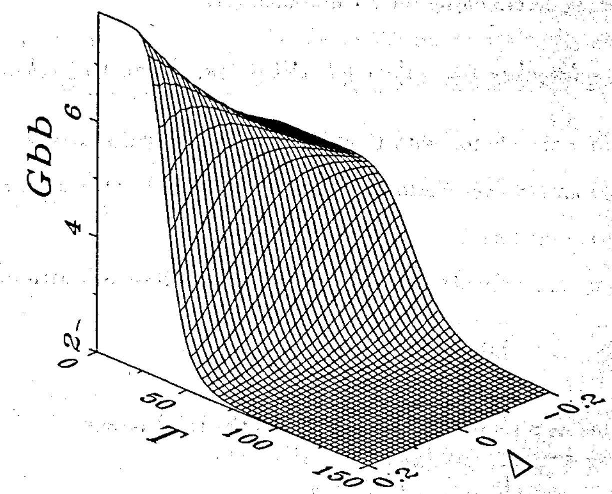

Figure 1: Phonon degree of coherence G_{bb} versus temperature $T\,(in Kelvin)$ and detuning Δ for the photon frequency $\omega_a = 5\cdot 10^{13} sec^{-1}$ and the coupling constant $g = 5\cdot 10^{12} sec^{-1}$.

Evaluating correlation function (5), one can determine the type of statistics. For example, the Poisson probability distribution leads to $G_{bb} = 1$ and the Gaussian probability distribution (chaotic state) at any temperature gives $G_{bb} = 2$. The result of calculations of G_{bb} with the help of expressions (6) and (7) is represented in Fig.1. The degree of coherence is plotted as a function of temperature T and detuning $\Delta = (\omega_a - \omega_b)\,/\,(\omega_a + \omega_b)$. One can see that at any parameters $G_{bb} > 2$ that corresponds to the super–Gaussian probability distribution (i.e., wider than the Gaussian probability distribution). It means that the photon–phonon interaction in an ionic crystal leads to stronger fluctuations of the number of phonons in comparison with the fluctuations in the case of the ordinary thermal lattice vibrations. It is important to note that at low temperatures (when thermal fluctuations are small compared to quantum fluctuations) there exsists a strong enough turning of G_{bb} aside from 2, while enhancement of T (increase of the thermal fluctuations) leads to decrease of that deflection. Therefore, the quantum fluctuation must play an important role only at low enough temperatures.

To observe these fluctuations, one can use the Raman spectroscopy [10]. The Brillouin scattering process is described by the Hamiltonian [11]:

$$H = \hbar\omega_R a_R^\dagger a_R + \hbar\omega_S a_S^\dagger a_S + \hbar\omega_A a_A^\dagger a_A + \hbar\omega_b b^\dagger b - \hbar\left(\kappa_S a_S^\dagger b^\dagger a_R + \kappa_A^* a_A^\dagger b a_R + h.c.\right), \quad (8)$$

where a_R stands for the photon with Rayleigh line frequency ω_R, a_S and a_A correspond to the Stokes and anti–Stokes photons respectively, b describes the phonons and κ_S, κ_A are the coupling parameters.

The short–time approximate solution for spontaneous Raman scattering, when

$$\langle a_S^\dagger(0) a_S(0)\rangle = \langle a_A^\dagger(0) a_A(0)\rangle = 0,$$

can be represented as a series of expansion in t to the third power:

$$\begin{aligned} a_S(t) &= \kappa_S e^{-i\omega_S t}\Big\{ i t b^\dagger a_R + \frac{t^2}{2}\kappa_A^* a_A^\dagger a_R^2 - \\ &\quad - i\frac{t^3}{6} b^\dagger a_R \left[b^\dagger b(|\kappa_A|^2 + |\kappa_S|^2) + a_R^\dagger a_R(|\kappa_A|^2 - |\kappa_S|^2) - |\kappa_A|^2 + 2|\kappa_S|^2\right]\Big\}, \\ a_A(t) &= \kappa_A^* e^{-i\omega_A t}\Big\{ i t b a_R - \frac{t^2}{2}\kappa_S a_S^\dagger a_R^2 - \\ &\quad - i\frac{t^3}{6}\left[b^\dagger b(|\kappa_A|^2 + |\kappa_S|^2) + a_R^\dagger a_R(|\kappa_A|^2 - |\kappa_S|^2) + |\kappa_A|^2 + 2|\kappa_S|^2\right] b a_R\Big\}, \end{aligned} \quad (9)$$

where $b \equiv b(0)$ and $a_j \equiv a_j(0)$, $j = A,\ S,\ R$. Taking into account that the density matrix of the system with the Hamiltonian (8) at $t = 0$ can be written as

$$\rho(0) = \prod_j \rho_j(0),$$

we can calculate expressions for time–dependent intensities:

$$\begin{aligned} I_S(t) &= |\kappa_S|^2 t^2 \Big\{ I_R(1 + \langle n_b\rangle) - \\ &\quad - t^2\Big(\frac{1}{3}\left[(|\kappa_A|^2 - |\kappa_S|^2)(I_R^2 - I_R) + |\kappa_S|^2 I_R\right](1 + \langle n_b\rangle) + \\ &\quad + \frac{1}{3}(|\kappa_A|^2 + |\kappa_S|^2) I_R(\langle n_b\rangle + \langle n_b^2(0)\rangle) - \frac{|\kappa_A|^2}{4}(I_R^2 - I_R)\Big)\Big\}, \\ I_A(t) &= |\kappa_A|^2 t^2\Big\{ I_R\langle n_b\rangle - \\ &\quad - t^2\Big(\frac{1}{3}\left[(|\kappa_A|^2 - |\kappa_S|^2)(I_R^2 - I_R) + |\kappa_S|^2 I_R\right]\langle n_b\rangle + \\ &\quad + \frac{1}{3}(|\kappa_A|^2 + |\kappa_S|^2) I_R\langle n_b^2(0)\rangle - \frac{|\kappa_S|^2}{4}(I_R^2 - I_R)\Big)\Big\}. \end{aligned} \quad (10)$$

where $I_R \equiv I_R(0)$ and $n_b \equiv b^\dagger b$ is the number operator of phonons at $t = 0$. This system of equations (10) gives us a possibility to determine the second–order degree of coherence for phonons at the initial time moment via the intensities of the Stokes and anti–Stokes components of scattered light for $t > 0$ that can be directly measured in experiments. Really, using (10), the degree of coherence (5) can be represented in the following form:

$$G_{bb} = \frac{\langle n_b^2\rangle - \langle n_b\rangle}{\langle n_b\rangle^2}, \quad (11)$$

where

$$\begin{aligned} \langle n_b^2\rangle &= \frac{3}{(|\kappa_A|^2 + |\kappa_S|^2)\, t^2 I_R}\Big\{ -\frac{I_A(t)}{|\kappa_A|^2 t^2} + I_R\langle n_b\rangle - \\ &\quad - t^2\Big(\frac{1}{3}\left[(|\kappa_A|^2 - |\kappa_S|^2)(I_R^2 - I_R) + |\kappa_S|^2 I_R\right]\langle n_b\rangle - \frac{|\kappa_S|^2}{4}(I_R^2 - I_R)\Big)\Big\} \end{aligned}$$

and

$$\begin{aligned} \langle n_b\rangle &= \frac{3}{(|\kappa_A|^2 + |\kappa_S|^2)\, t^2 I_R}\Big\{ \frac{I_A(t)}{|\kappa_A|^2 t^2} - \frac{I_S(t)}{|\kappa_S|^2 t^2} + I_R - \\ &\quad - t^2\Big(\frac{1}{12}(|\kappa_A|^2 - |\kappa_S|^2)(I_R^2 - I_R) + \frac{|\kappa_S|^2}{3} I_R\Big)\Big\}. \end{aligned}$$

Thus, evaluating the degree of coherence (11), one can experimentally verify the super–Gaussian type of phonon statistics in an ionic crystal.

The main result of this paper consists in revealing super–Gaussian character of phonon number probability distribution at thermal equilibrium in a polariton–like system. This is due to the nature of the quantum fluctuations, which turn out to be especially large at low temperatures. The Raman spectroscopy is proposed as a possible way for observation of such fluctuations in an ionic crystal at equilibrium.

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