

# QUANTUM OPTICS AND SOLID STATE SPECTROSCOPY

ALEXANDER S. SHUMOVSKY

*Department of Physics, Bilkent University  
Bilkent 06533, Ankara, Turkey*

**Abstract.** Possibility of formation of strongly correlated states of the bosonic excitations in solids similar to the squeezed states of photons is considered. The possibility of observation of these states in optical spectroscopy is discussed.

## 1. Introduction

By definition, the Quantum Optics is considered as a branch of physics studying the quantum statistical properties of the optical fields, in other words, the quantum state of light and the picture of its quantum fluctuations. Since the experimental information of the quantum statistical properties of light is provided by the correlation measurements, the quantum correlation functions are the principal objects of this branch of modern physics. Some specific effects with no classical analogies such as antibunching, sub-Poissonian statistics of photons and squeezing of quantum fluctuations are stipulated by the nonlinear processes.

We note that within the framework of quantum field theory the photons are described by the Bose-operators which obey the commutation relations of the Weyl-Heisenberg algebra. In solid state physics we know a number of quasi-particles or collective excitations such as phonons, polaritons, excitons, Cooper pairs (approximately) which are also bosons. In many physically important cases they take part in various kinds of nonlinear processes or interactions. Therefore, in analogy to the photons, it is quite natural to expect some unconventional quantum statistical properties of the Bose-type excitations in solids which could be important for deeper understanding of a number of physical phenomena.

Unfortunately, we usually cannot measure directly the high-order correlation functions for the collective excitations in solids. However, in many important cases the light can be scattered by these excitations. In this case, the photon states are correlated with the degrees of freedom of the excitations inside the solid. Therefore, the scattered light is naturally expected to reflect the quantum statistical properties of the excitations. In turn, the correlation measurements for the scattered light could give information about the quantum statistics of the excitations.

Therefore, there are two important questions. 1) What kind of the statistical properties we can expect for the bosonic excitations in solids? 2) How to observe these properties with the aid of optical correlation measurements? For all that, it is usually necessary to

take into account that boson excitations in condensed matter ought to be considered at thermal equilibrium.

The main purpose of this paper is to discuss briefly the present state of the problems mentioned above.

## 2. Squeezed States

The definition of the squeezed states of bosons [1, 2, 3] is based on the Bogoliubov's canonical transformation. This transformation has been introduced in connection with the problem of superfluidity of liquid  ${}^4\text{He}$  [4]. The original form of this transformation is

$$U^\dagger a_k U = u_k a_k + v_k a_{-k}^\dagger \equiv \mathbf{a}_k, \quad |u_k|^2 - |v_k|^2 = 1 \quad (1)$$

where the operators  $a_{\pm k}$ ,  $a_{\pm k}^\dagger$  describe the over-condensate excitations ( $k \neq 0$ ) at low temperatures when almost all atoms of the  ${}^4\text{He}$  are in the Bose-Einstein condensate state. The unitary operator  $U$  has the form

$$U = \prod_k U_k, \quad U_k = \exp(\xi_k a_k^\dagger a_{-k}^\dagger / 2 - \xi_k^* a_{-k} a_k / 2) \quad (2)$$

with

$$|\xi_k|^2 = \cosh^{-1} u_k, \quad \arg \xi_k = \arg v_k - \pi$$

and diagonalizes the Hamiltonian of over-condensate excitations

$$H = \frac{1}{2} \sum_{k \neq 0} [\epsilon_k (a_k^\dagger a_k + a_{-k}^\dagger a_{-k}) + \lambda_k a_k^\dagger a_{-k}^\dagger + \lambda_k^* a_{-k} a_k]. \quad (3)$$

Hence, the operators  $\mathbf{a}_k, \mathbf{a}_k^\dagger$  describe the simultaneous creation and annihilation of the bosons with opposite momenta. Here

$$u_k = \frac{|\lambda_k|}{\sqrt{2\epsilon_k(\epsilon_k - E_k)}}, \quad v_k = \frac{\epsilon_k - E_k}{\sqrt{2\epsilon_k(\epsilon_k - E_k)}} e^{i \arg \lambda_k}$$

and  $E_k = \sqrt{\epsilon_k^2 - |\lambda_k|^2}$  represents the spectrum of pair over-condensate excitations

$$H = \frac{1}{2} \sum_{k \neq 0} E_k (\mathbf{a}_k^\dagger \mathbf{a}_k + \mathbf{a}_{-k}^\dagger \mathbf{a}_{-k}) \quad (4)$$

One can see that  $U_k$  in the Eq. (2) formally coincides with the operator corresponding to the so-called two-mode coherent state in quantum optics [5, 6, 7]. The vacuum state of the system described by the Hamiltonian (4) is

$$|\Phi\rangle = \prod_{k \neq 0} |\psi_k\rangle, \quad |\psi_k\rangle = U_k |0\rangle_k |0\rangle_{-k} \quad (5)$$

where  $|0\rangle_{\pm k}$  denotes the vacuum state defined by the ground state stability condition  $a_{\pm k} |0\rangle_{\pm k} = 0$ . Then

$$\begin{aligned} \langle a_k^\dagger a_k \rangle_\Phi &\equiv \bar{n}_k = |v_k|^2, \\ \langle (a_k^\dagger a_k)^2 \rangle_\Phi &= |v_k|^2 (u_k^2 + |v_k|^2) = \bar{n}_k (2\bar{n}_k + 1) \end{aligned}$$

where  $\langle \dots \rangle_{\Phi}$  denotes the expectation value with respect to the state (5). The number variance is

$$V_{\Phi}(a_k^{\dagger} a_k) \equiv \langle (a_k^{\dagger} a_k)^2 \rangle_{\Phi} - \langle a_k^{\dagger} a_k \rangle_{\Phi}^2 = \bar{n}_k^2 + \bar{n}_k \neq 0 \quad (6)$$

while in the case of ordinary vacuum state  $V(a_k^{\dagger} a_k) = 0$ . It means that there are very strong quantum fluctuations of the number of  ${}^4\text{He}$  atoms in the over-condensate state of a superfluid system [8, 9]. It should be stressed that this result is valid also for the total number of particles since

$$N = \sum_{k \neq 0} a_k^{\dagger} a_k, \quad \bar{N} = \sum_{k \neq 0} |v_k|^2, \quad V_{\Phi}(N) = \sum_{k \neq 0} u_k^2 |v_k|^2.$$

Let us note that using the terminology of quantum optics (see, e.g. [10]) one can say now that the vacuum state (5) of the system (3) and (4) corresponds to a super-Poissonian number distribution as far as

$$V_{\Phi}(N) > \bar{N}.$$

Although the investigation of the *squeezing phenomenon* is not the aim of this paper, let us note that there is no squeezing in the mode  $k$  since

$$V_{\Phi}([a_k + a_k^{\dagger}]/2) = (2u_k^2 - 1)/4 > 1/4$$

although at

$$\frac{\pi}{6} + l\pi < \arg \lambda_k < \frac{5\pi}{6} + l\pi, \quad l = 0, 1, \dots$$

squeezing is present in the mixture of two modes with the opposite  $k$ .

The investigation of the quantum number fluctuations in the superfluid liquid  ${}^4\text{He}$  is not a subject of the optical spectroscopy and can be done with the aid of neutron scattering [8, 9]. Therefore, let us consider now a different example of manifestation of squeezed states in condensed matter physics which is the polariton in an ionic crystal. This system is usually described by the Hamiltonian [11, 12]

$$H = \sum_{k \geq 0} [\omega_k (a_k^{\dagger} a_k + a_{-k}^{\dagger} a_{-k}) + \Omega_L (b_k^{\dagger} b_k + b_{-k}^{\dagger} b_{-k})] - \sum_{k \geq 0} D_k [(a_k^{\dagger} - a_{-k})(b_k + b_{-k}^{\dagger}) + (a_{-k}^{\dagger} - a_k)(b_{-k} + b_k^{\dagger})]. \quad (7)$$

Here the operators  $a$  describe photons with the frequency  $\omega_k = kc$ , the operators  $b$  describe the transversal optical (TO) phonons,  $\Omega_L = \Omega_T \sqrt{\epsilon_0} \epsilon_{\infty}$  is the frequency of longitudinal vibrations of ions and

$$D_k = -\frac{i}{2} \sqrt{\Omega_L \omega_k \eta}, \quad \eta = \frac{\epsilon_0 - \epsilon_{\infty}}{\epsilon_0} > 0.$$

One can see that the operator structure of the Hamiltonian (7) is somewhat different in comparison with (3) since it contains the supplementary terms  $a_{\pm k}^{\dagger} b_{\pm k}$  and  $b_{\pm k}^{\dagger} a_{\pm k}$ . The Bogoliubov's canonical transformations in this case have the form

$$c_{kj} = u_{1j}(k) a_k + u_{2j}(k) b_k - v_{1j}^*(k) a_{-k}^{\dagger} - v_{2j}^*(k) b_{-k}^{\dagger}, \quad j = 1, 2 \quad (8)$$

and obey the conditions

$$\sum_l (u_{lj} u_{lj'}^* - v_{lj}^* v_{lj'}) = \delta_{jj'}.$$

The explicit form of the coefficients in (8) is

$$\begin{aligned} u_{1j}(k) &= (i)^{j-1} \frac{(E_{kj} + \omega_k) |D_k| \sqrt{\Omega_L}}{\sqrt{\Phi_{kj}}}, \\ v_{1j}(k) &= (-i)^{j-1} \frac{(E_{kj} - \omega_k) |D_k| \sqrt{\Omega_L}}{\sqrt{\Phi_{kj}}}, \\ u_{2j}(k) &= i^j \frac{(E_{kj}^2 - \omega_k^2)(E_{kj} + \Omega_L)}{2\sqrt{\Omega_L \Phi_{kj}}}, \\ v_{2j}(k) &= (-1)^{j-1} i^j \frac{(E_{kj}^2 - \omega_k^2)(E_{kj} - \Omega_L)}{2\sqrt{\Omega_L \Phi_{kj}}} \end{aligned} \quad (9)$$

where

$$\Phi_{kj} \equiv E_{kj} [(E_{kj}^2 - \omega_k^2)^2 + 4\omega_k \Omega_L |D_k|^2],$$

and

$$E_{kj}^2 = \frac{\omega_k^2 + \Omega_L^2}{2} \pm \frac{1}{2} \sqrt{(\omega_k^2 - \Omega_L^2)^2 + 16\omega_k \Omega_L |D_k|^2} \quad (10)$$

are the two branches of the polariton spectrum [11, 12] (see Fig. 1). In the polariton

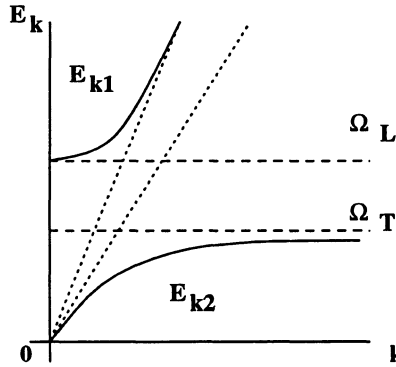


Figure 1. Two branches of the polariton excitations versus  $k$ .

(diagonal) representation, the Hamiltonian (7) has the following form

$$H_{pol} = \sum_{k \geq 0} \sum_j E_{kj} (c_{kj}^+ c_{kj} + c_{-kj}^+ c_{-kj}) + E_0 \times \mathbf{1} \quad (11)$$

where

$$E_0 = - \sum_k \sum_{l,j} E_{klj} |v_{lj}|^2.$$

The eigenstates of the Hamiltonian (11) which are the Fock number states of polaritons are squeezed states in terms of the photon and phonon degrees of freedom. It means, that its structure corresponds to the simultaneous creation of pairs of photons and phonons and also of a photon and phonon simultaneously. The corresponding squeezing operator has the form [9]

$$S_k = \exp(\xi_k a_k^{+2} + \eta_k b_k^{+2} + \zeta_k a_k^+ b_{-k}^+ - h.c.)$$

where  $\xi_k$ ,  $\eta_k$ , and  $\zeta_k$  are known but quite complicated functions of the coefficients  $u$  and  $v$ . One can see that  $S_k$  represents some generalization of the two-mode squeezing operator (2).

Let us examine statistical properties of phonons in the system (7). As in standard quantum optics, these properties can be characterized by the degree of coherence [10]

$$G_{\Theta}^{(2)} = \frac{\langle b_k^{+2} b_k^2 \rangle}{\langle b_k^+ b_k \rangle^2} \quad (12)$$

Without going into straightforward calculations let us examine the temperature dependence of (12) given in Fig. 2 (also see [13]). Taking into account that in the case of Gaussian distribution of phonons  $G^{(2)} = 2$  at  $\Theta = 0$ , one can see from the Fig. 2 that there is a *super-Gaussian* distribution of phonons in the polariton system. It means that at low

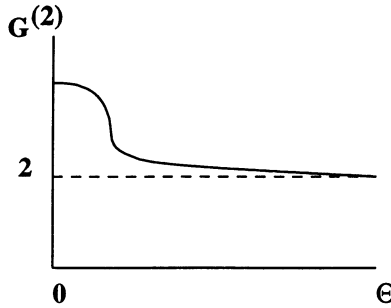


Figure 2. Temperature dependence of the degree of coherence (12) at fixed  $k$ .

temperature, there are quite strong quantum number fluctuations of TO phonons in an ionic crystal. Increase of temperature amplifies the thermal number fluctuations and make the quantum fluctuations unobservable. The estimation of the threshold of observation of the quantum number fluctuations gives for a typical ionic crystal  $T_{th} \leq 30K$  [13] which is not too low temperature for the experimental study. A similar behavior can be observed also in the photon sub-system [14].

Thus, the above considered examples show that realization of a squeezed state in material systems (quantum liquid, solid state etc.) leads to a quite strong quantum number fluctuations of bosons which can be observed at relatively low temperatures.

Let us discuss briefly some other sources of squeezed states in solids. Since the origin of the emergence of squeezed states is the simultaneous creation or annihilation of two (or more) bosons, the simplest process in a real crystal which leads to a squeezed state is connected with the third-order phonon anharmonicity. In this case, we have for example the scattering of two phonons into one or decay of one phonon into two phonons (see Fig. 3). These processes are realized under the condition of the conservation of energy but it is not necessary to have conservation of quasi-momentum [11].

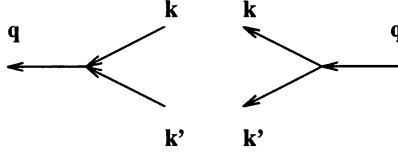


Figure 3. Scattering of two phonons into one and decay of a phonon into two phonons. The processes obey the energy conservation  $\Omega(q) = \Omega(k) + \Omega(k')$  and one of the relations  $\vec{q} = \vec{k} + \vec{k}'$  or  $\vec{q} = \vec{k} + \vec{k}' + \vec{g}$  where  $\vec{g}$  is the vector of reciprocal lattice.

Another example is provided by the photon-phonon interaction responsible for the processes of emission and absorption of electromagnetic waves by a crystal. The energy of interaction in the dipole approximation in this case has the form

$$H_{int} = - \sum_f \vec{d}_{f\alpha} \cdot \vec{E}(f) \quad (13)$$

where

$$\vec{d}_{f\alpha} = \sum_k \sqrt{\frac{\hbar}{2\Omega_\alpha(k)N}} \vec{d}_\alpha(k) e^{ikf} (b_{k\alpha} + b_{-k\alpha}^+)$$

is the electric dipole moment of the primitive cell of the crystal, index  $\alpha$  marks the branch of the phonon oscillations of the frequency  $\Omega_\alpha$ ,  $b$  and  $b^+$  are the phonon operators, and

$$\vec{E}(f) = i \sqrt{\frac{2\pi\hbar}{N\epsilon}} \sum_{q,\sigma} \vec{e}_\sigma(q) \sqrt{\omega_q} (a_{q\sigma} - a_{-q\sigma}^+) e^{iqf}$$

is the electric field on the site  $f$ . Here  $\epsilon$  is the permittivity of the crystal,  $\vec{e}$  describes the photon polarization, and the operators  $a$  and  $a^+$  describe the photons with the frequency  $\omega_q$  corresponding to the region of transparency of the crystal. One can see that the Hamiltonian (13) formally coincides with the polariton model (7).

Besides the phonons, the processes related to the excitons in semiconductors and molecular crystals can be also considered as the sources of squeezed states. For example, in the case of a molecular crystal the exciton-photon interaction can lead to the formation of the exciton-polaritons [15]. The interaction energy corresponding to the above process is

$$H_{EP} = - \frac{e}{mc} \sum_f \vec{A}(f) \cdot \vec{P}_f + \frac{e^2 S}{2mc^2} \sum_f A^2(f) \quad (14)$$

where  $\vec{A}(f)$  describes the vector potential of electromagnetic field in the site  $f$  and  $\vec{P}_f$  is the total momentum of all  $S$  optically active electrons of a neutral molecule in the lattice site  $f$ . Going over to the quantization in the reciprocal space (the second quantization representation), one can rewrite (14) in the form

$$H_{EP} = \sum_{k,\alpha,f} \mathcal{D}(k, \alpha, f) (B_{kf}^+ - B_{-kf})(a_{k\alpha} + a_{-k\alpha}^+) + \sum_{k,\alpha} \frac{\hbar\Omega_p^2 S}{4c|k|} ((a_{k\alpha}^+ + a_{-k\alpha})(a_{k\alpha} + a_{-k\alpha}^+)) \quad (15)$$

where  $\Omega_p^2$  is the squared plasma frequency and  $\mathcal{D}$  is the coupling constant [15]. Since the exciton operators  $B^+, B$  obey the boson commutation relations we again have the Hamiltonian of the form of (7). The exciton-phonon interaction can be also described in a similar way [15].

Let us mention here an important and interesting problem of the exciton-polaritons in a semiconducting micro-cavity formed by a multiple-quantum-well structure placed between the distributed Bragg reflectors (DBR) [16, 17, 18, 19]. In this case, the two branches of the polariton formed by coupled modes of the radiation field and collective excitations of semiconductor provide a two-level system. Unlike the Jaynes-Cummings model [20, 21, 22], the occupation number of any level of that two-level system can take an arbitrary integer value due to the bosonic nature of the polariton. Nevertheless, the vacuum-field induced Rabi oscillations can be observed in such a system [16, 17]. Let us stress that the study of Rabi oscillations in various physical systems is now an active field of research due to its importance in the investigation of spontaneous emission rate in a cavity and plays a crucial role in the development of new light emitting devices with special properties, as for instance narrow line-width, low-threshold pump-rate, and low noise level [23]. In the case of exciton-polariton system, the properties of radiation are determined by the specific correlation between the processes of simultaneous creation and annihilation of the photons and excitons described in terms of states quite similar to (5) [19].

Another example of great interest is provided by the competition of direct and indirect radiative transitions in the GaAs/AlAs superlattices and multiple-quantum-well structures [24, 25]. The band structure of this system is represented in Fig. 4. In this superlattice, the lowest electronic states of the conducting band are located in the AlAs layers, whereas the lowest electronic states of the valence band are confined in the GaAs layers. After photoexcitation due to the quite short laser pulse close to the resonance with the transition  $|0\rangle \leftrightarrow |1\rangle$  electrons scatter from the  $\Gamma$  states in the GaAs to the lower lying  $X$  states located in the AlAs. This  $\Gamma$ - $X$  transfer is either dominated by the LO-phonon emission [24] or is due to the interface scattering [25]. After that, the level  $|2\rangle$  is populated and indirect (phonon-assisted) transition can occur (type II transition in Fig. 4). This transition leads to an additional peak in the luminescence spectrum [24, 25] which is shifted downwards from the type I peak by the energy  $\sim 0.05eV$  corresponding to the phonon energy. In this case, we again have the simultaneous creation of a photon and phonon. Thus, we can expect also here some kind of squeezed state.

The above examples provide the case when the squeezed states result from the interactions of bosons (phonon-photon, phonon-phonon, exciton-phonon, etc.). However, they can also occur as a result of interaction between bosons and some other particles such as electrons, protons of the hydrogen bonds and so on. An important example is provided by

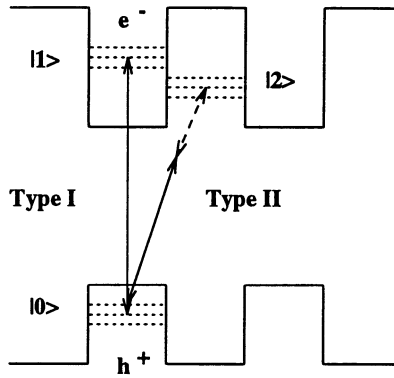


Figure 4. Band structure of a GaAs/AlAs superlattice. Solid lines correspond to the photons while the dotted line corresponds to a phonon.

the phonon squeezing via correlations in the superconducting electron-phonon interaction [26, 27, 28]. Similar process takes place in a polaron system [29, 30].

Thus, it is shown in this section that many different mechanisms can lead to the formation of squeezed states (or states with correlated processes of creation and annihilation) of the boson-type excitations in solids. It is manifested in the change of quantum statistical properties of these excitations, first of all in the increase of quantum number fluctuations with respect to non-squeezed states.

Let us now turn our attention to the problem of how to measure the quantum statistical properties of the boson excitations in solids.

### 3. Quantum Spectroscopy of Solids

It was shown in the previous section that the type of bose statistics can be qualitatively determined by the normalized correlation function (12). This means that it is necessary to measure the number correlation or, using the terminology of quantum optics, the intensity correlation. For the photons emitted by some source this can be done with the aid of the Hanbury-Brown and Twiss correlation interferometer [10]. In this device, the photon beam is splitted into two equivalent beams which are then detected by two independent photo-detectors. To determine the photon correlations photon counters and digital correlations are used. Unfortunately, it is almost impossible to use the similar scheme of correlation measurements for phonons, excitons and other boson excitations in solids.

At the same time, these excitations can interact with photons in the process of emission or scattering of light by solids. In this case, it is reasonable to think that the quantum statistical properties of photons should contain some information about the quantum statistical properties of bosons in solids.

A simple illustrative example of this process is provided by the photoluminescence in the system shown in Fig. 4 and briefly discussed in the previous section. In fact, the process of photoluminescence in the type II transition is characterized by a correlation between creation of photons and phonons and thus strongly depend on the type of statistics of the initial phonon state.



In many cases, bosonic excitations in solids are Raman active [31]. The process of Raman scattering with generation of the Stokes and anti-Stokes photons is shown in Fig. 5. If we suppose the monochromatic coherent laser field and scattering on an optical

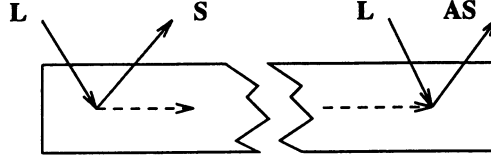


Figure 5. Scheme of the Raman scattering with generation of Stokes (A) and anti-Stokes (AS) components. The Raman active excitations of a medium are shown by the dotted lines.

phonon, the Hamiltonian of the process in question is

$$H = \omega_L a_L^\dagger a_L + \Omega b^\dagger b + \sum_k (\omega_{Sk} a_{Sk}^\dagger a_{Sk} + \omega_{Ak} a_{Ak}^\dagger a_{Ak}) + \sum_k [\gamma_{Sk} a_L^\dagger a_{Sk} b + \gamma_{Ak} b^\dagger a_L^\dagger a_{Ak} + h.c.]. \quad (16)$$

Here the operators  $a_L$  describe the laser (pump) field with the frequency  $\omega_L$ , the phonons of frequency  $\Omega$  are described by the operators  $b$ . Due to the Manley-Rowe relations [31]  $\omega_{Sk} - \omega_{Ak} \sim 2\Omega$ . The operators  $a_{Sk}$  and  $a_{Ak}$  correspond to the Stokes and anti-Stokes photons respectively and  $\gamma_{Sk}$ ,  $\gamma_{Ak}$  are the coupling constants.

The investigation of quantum properties of this Hamiltonian is the subject of a number of works (e.g., see Refs. [32, 33, 34] and references there). However, due to the cubic nonlinearity, the problem is not solved exactly except the case of  $\gamma_{Ak} = 0$  [34, 35, 36].

The Hamiltonian (16) has the following conservation laws

$$\begin{cases} n_L + \sum_k (n_{Sk} + n_{Ak}) \equiv N = \text{const} \times 1 \\ n_b - \sum_k (n_{Sk} - n_{Ak}) \equiv M = \text{const} \times 1 \end{cases} \quad (17)$$

Here  $n_x$  denotes the number operator of the field  $x$ .

To examine the problem with the Hamiltonian (16) let us use the method of elimination of Bose variables [37, 38]. In view of our main aim, we follow here the case considered in the Ref. [39]. We should eliminate from the equations of motion the phonon field dynamics of which is described by the following formal expression

$$b(t) = b_f(t) - iB(t), \quad b_f(t) = b(t_0)e^{-i\Omega(t-t_0)}, \\ B(t) = \int_{t_0}^t d\tau e^{-i\Omega(t-\tau)} \sum_k [\gamma_{Sk} a_{Sk}^\dagger(\tau) a_L(\tau) + \gamma_{Ak} a_L^\dagger(\tau) a_{Ak}(\tau)]. \quad (18)$$

Consider now the time evolution of the expectation values  $\langle n_{Sk} \rangle_t$  and  $\langle n_{Ak} \rangle_t$  describing the process of emission of the Stokes and anti-Stokes photons respectively. Here  $\langle \dots \rangle_t$  denotes the average of corresponding operator in the Heisenberg representation with respect to the density matrix of the initial state of the complete system [38]. We have

$$i \frac{d}{dt} \langle n_{Sk} \rangle_t = -\gamma_{Sk} (\langle a_L^\dagger a_{Sk} b \rangle - \langle b^\dagger a_{Sk}^\dagger a_L \rangle), \\ i \frac{d}{dt} \langle n_{Ak} \rangle_t = -\gamma_{Ak} (\langle b^\dagger a_L^\dagger a_{Ak} \rangle - \langle a_{Ak}^\dagger a_L b \rangle).$$

We have to substitute here the expression (18) and conjugated expression for  $b^+$ . It leads to the expectation values containing the mixed averages with participation of  $b(t_0)$  and  $b^+(t_0)$ . Depending on the choice of the initial state, these averages have different values. In the case of thermal harmonic phonons we get [39]

$$\langle \mathcal{A}b_f \rangle_t = i\bar{n}_b \langle [B, \mathcal{A}] \rangle_t \quad (19)$$

where  $\mathcal{A}$  is an arbitrary operator and  $\bar{n}_b$  is the initial number of phonons depending on the temperature, while in the case of squeezed thermal phonons

$$\langle \mathcal{A}b_f \rangle_t = i[|v|^2 + (u^2 + |v|^2)\bar{n}_b] \langle [B, \mathcal{A}] \rangle_t - iuv(2\bar{n}_b + 1) \langle [B^+, \mathcal{A}] \rangle_t. \quad (20)$$

Here  $u$  and  $v$  are the parameters describing the squeezed state. It follows from these exact relations that the mixed correlation functions of thermal harmonic phonons do not contribute into the equations of motion at  $T \sim 0$  when  $\bar{n}_b \sim 0$  while in the case of squeezed phonons, they give a non-vanishing contribution even at  $T = 0$ . Thus, there is a principal difference between these two cases.

Consider, for simplicity, just the case of low temperatures when  $\bar{n}_b \sim 0$  and use the Markov approximation. Then, the equations of motion in the case of harmonic phonons have the form

$$\begin{aligned} \frac{d}{dt} \langle n_{Sk} \rangle_t &= 2\pi\gamma_S^2 \langle a_L^+ a_L \rangle_t \delta_{kk_S} + \pi\gamma_{Sk}\gamma_S (\langle a_L^+ a_L a_S^+ a_{Sk} \rangle_t + \langle a_{Sk}^+ a_S a_L^+ a_L \rangle_t) \\ &\quad + \pi\gamma_{Sk}\gamma_A (\langle a_L^{+2} a_{Sk} a_A \rangle_t + \langle a_A^+ a_{Sk}^+ a_L^2 \rangle_t) \end{aligned} \quad (21)$$

and

$$\begin{aligned} \frac{d}{dt} \langle n_{Ak} \rangle_t &= -\pi\gamma_{Ak}\gamma_A (\langle a_A^+ a_{Ak} \rangle_t + \langle a_{Ak}^+ a_A \rangle_t) - \pi\gamma_{Ak}\gamma_A (\langle a_L^+ a_L a_A^+ a_{Ak} \rangle_t + \langle a_{Ak}^+ a_A a_L^+ a_L \rangle_t) \\ &\quad - \pi\gamma_{Ak}\gamma_S (\langle a_L^{+2} a_S a_{Ak} \rangle_t + \langle a_{Ak}^+ a_S^+ a_L^2 \rangle_t). \end{aligned} \quad (22)$$

Here  $a_S, \gamma_S$  and  $a_A, \gamma_A$  denote the resonance values defined by the conditions

$$\omega_{Sk} = \omega_L - \Omega, \quad \omega_{Ak} = \omega_L + \Omega$$

respectively.

One can see that unlike the Eq. (21), the right-hand side of (22) does not contain any direct dependence on the mean number of photons in the laser mode  $\langle a_L^+ a_L \rangle_t$ . Using the short-time approximation, it is not hard to see that this property leads to a delay of the anti-Stokes radiation with respect to the Stokes radiation (Fig. 6). In the case of the squeezed phonons, the right-hand side of the Eq. (21) should be added by the terms

$$\begin{aligned} &2\pi|v|^2\gamma_S^2 \langle a_L^+ a_L \rangle_t \delta_{kk_S} - \pi|v|^2\gamma_{Sk}\gamma_S (\langle a_S^+ a_{Sk} \rangle_t + \langle a_{Sk}^+ a_S \rangle_t) \\ &\quad + \pi\gamma_{Sk}\gamma_A (uv \langle a_A^+ a_{Sk} \rangle_t + uv^* \langle a_{Sk}^+ a_A \rangle_t) \end{aligned} \quad (23)$$

which shows the increase of dependence on  $\langle a_L^+ a_L \rangle_t$  and at the same time, the increase of correlation between the Stokes and anti-Stokes photons. Similar additional terms for (22) have the form

$$\begin{aligned} &2\pi|v|^2\gamma_A^2 \langle a_L^+ a_L \rangle_t \delta_{kk_A} - |v|^2\pi\gamma_{Ak}\gamma_A (\langle a_A^+ a_{Ak} \rangle_t + \langle a_{Ak}^+ a_A \rangle_t) \\ &\quad + \pi\gamma_{Ak}\gamma_S (uv \langle a_A^+ a_S \rangle_t + uv^* \langle a_S^+ a_{Ak} \rangle_t). \end{aligned} \quad (24)$$

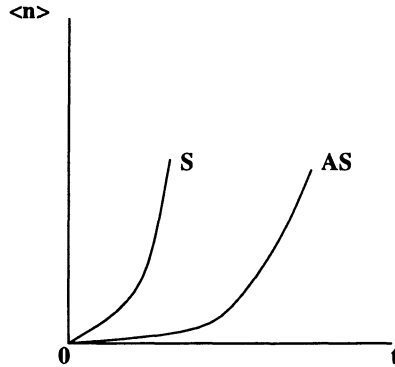


Figure 6. Short-time behavior of the mean photon number for the Stokes and anti-Stokes components described by the Eqs. (21) and (22).

One can see from (24) that now there is a direct dependence of the right-hand side of the equation of motion for the average number of anti-Stokes photons on the number of photons in the laser mode. Therefore, in the case of squeezed phonons there is no delay between the Stokes and anti-Stokes radiation. Thus, the change of state of the phonons leads to some qualitative change of the spectra of scattered light in the Raman scattering process. Some other changes are discussed in the Refs. [9, 33, 34, 35].

Of course, the analysis of the high-order intensity correlations of the Stokes and anti-Stokes components can give richer picture of qualitative and quantitative effects of the dependence of photon statistics of scattered light on the quantum statistical properties of the scatterers [9, 36]. These high-order intensity correlations can be measured by the methods of correlation spectroscopy [40].

#### 4. Summary

It is shown that many different mechanisms of interactions can lead to the formation of strongly correlated (squeezed) state of boson excitations in solids. These correlations are manifested by the quantum number fluctuations which can have a super-Gaussian character at low temperatures. The increase of temperature leads to the increase of thermal number fluctuations exceeding the quantum fluctuations. Therefore, the principal observability of the quantum number fluctuations of bosonic excitations in solids requires the low temperatures.

The quantum number fluctuations of optical-active boson excitations in solids can be observed by the methods of optical spectroscopy, especially by the correlation spectroscopy.

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