Photon correlation in inelastic scattering of light

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Intensity correlations in the scattering of light by acoustic phonons or polaritons in the case of weak absorption are considered. The relationship between the bunching parameter of the scattered light and the bunching parameter of the scattering quasiparticles is calculated perturbatively in the interaction representation. The cases of thermal and coherent statistics of the phonons (respectively, polaritons) are considered separately. For the case of arbitrary statistics of the scattering quasiparticles, it is shown that the bunching parameter for them can be measured by means of a Brown–Twiss experiment for light scattered into the anti-Stokes region. Scattering by two uncoupled phonons is considered separately.

1. INTRODUCTION

In quantum optics, the statistics of light fields radiated in various linear and nonlinear optical processes has been well studied. In particular, there has been study of the photon statistics[1)] for different forms of scattering: elastic,[1–3] spontaneous and stimulated Raman[4–6] and parametric and hyperparametric[5,6] scattering. In our view, a question particularly worthy of attention is that of the relationship between the statistics of the scattered radiation and the statistics of the scattering phonons, polaritons, or other quasiparticles. Indeed, if such a relationship does exist, then by investigating the statistics of the scattered light it will be possible to learn significantly more about the phonon-like excitations than can be done simply by measuring the scattering intensity. In Ref. 7, a study is made of the relationship between the phonon and photon statistics for the case of Stokes Raman scattering—the Mandel factor of the scattered light is calculated for the case when the photons are in an energy, or squeezed state. From the point of view of studying the statistics of the scattering excitations, it would appear to be more informative to investigate the bunching parameter of the Stokes and anti-Stokes radiation and also the corresponding cross correlation moments. The fact is that in a number of cases the bunching parameter of the scattered light (measured in Brown–Twiss experiments) is simply equal to the bunching parameter of the scattering quasiparticles. This is indicated by the result obtained in Ref. 6 for scattering by thermal phonons. In this case, the bunching parameter of the scattered light is equal to the bunching parameter of the phonons ($g=2$). However, is this the case for arbitrary statistics of the scattering quasiparticles? Indeed, is it possible at all to speak of phonon-like excitations with nonthermal statistics?

In this paper, we attempt to answer the first question. For the simplest case of scattering by particles with negligibly small absorption (acoustic phonons, polaritons), we calculate the bunching parameter of the scattered light in terms of the bunching parameter of the scattering quasiparticles. We show that unambiguous conclusions about the bunching of phonons or polaritons can be deduced from the fourth moments of the field of the scattered light.

As an answer to the second question—of whether phonon-like excitations with nontrivial statistics are possible—we give two examples. In the case of coherent pumping of the phonon or polariton modes of a medium, the corresponding excitations will obviously not be in a thermal state but in a nearly coherent state. Even more interesting should be the statistics of phonon-like excitations in the presence of lattice anharmonicity. For example, the decay of coupled two-phonon states (biphonons) into single phonons will increase the bunching parameter for the corresponding single-phonon mode. In the equilibrium case, this increase will probably not be significant. However, if the biphonon is artificially populated, then the binary phonon bunching in the first-order spectrum may be appreciable. At the same time, the bunching parameter must depend on the anharmonicity constant responsible for the formation and decay of the biphonons. We note also that, as is shown in Ref. 8, phonon polaritons may exhibit nonclassical statistics, but only in experiments that permit detection of only the phonon (or only the photon) "component" of the polaritons.

Thus, in this paper we use perturbation theory to calculate the fourth moments of the fields (the intensity correlation functions) of inelastically scattered light in terms of the moments of the field of the scattering quasiparticles. The problem is solved in the approximation of a fairly large mean free path of the scattering excitations, and so this approach is valid only for scattering of light by acoustic phonons and polaritons with not too strong absorption. We also consider separately the case when measurements are made of the intensity correlation between anti-Stokes scattering by a phonon-like excitation and Stokes scattering by two uncoupled such excitations. The bunching parameter in this case is found to be 3.

2. THEORETICAL MODEL

We consider scattering of light by phonons or polaritons. As a result of scattering, a pumping photon can be transformed into a Stokes photon with production of a phonon (or polariton) or into an anti-Stokes photon with absorption of a phonon (or polariton). In the interaction representation, these processes are described by an effective phenomenological Hamiltonian of the form

$$\mathcal{H}=\mathcal{H}_a+\mathcal{H}_{si} \tag{2.1}$$

where

$$\mathcal{H}_s=-\frac{1}{2}\sum_{i,j}\int_V d^3\mathbf{r}\mathcal{E}_0^{(+)}(\mathbf{r},t)\chi_s(\omega_0,\omega_{si},\omega_{pj})E_{si}^{(-)}(\mathbf{r},t)E_{pj}^{(-)}(\mathbf{r},t)+\text{h.c.},$$

$$\mathcal{H}_a=-\frac{1}{2}\sum_{i,j}\int_V d^3\mathbf{r}\mathcal{E}_0^{(+)}(\mathbf{r},t)\chi_a(\omega_0,\omega_{ai},\omega_{pj})E_{ai}^{(-)}(\mathbf{r},t)E_{pj}^{(+)}(\mathbf{r},t)+\text{h.c.} \tag{2.2}$$

Here V is the volume of the sample in which the scattering takes place; $E_{\alpha i}^{(+)}$ and $E_{\alpha i}^{(-)}$ are the operators of the positive- and negative-frequency parts of the field; more specifically,

$$E_{\alpha i}^{(+)}(\mathbf{r},t)=\frac{i}{2\pi}\sqrt{\hbar\omega_{\alpha i}\left(\frac{2\pi}{L}\right)^3}a_{\alpha i}\exp[i(\mathbf{k}_{\alpha i}\mathbf{r}-\omega_{\alpha i}t)] \tag{2.3}$$

is the positive-frequency operator of the field that includes only frequencies $\omega_{\alpha i}$ less than the pumping frequency ω_0 for the Stokes component ($\alpha=s$) and greater than ω_0 for the anti-Stokes component ($\alpha=a$); a_α is the annihilation operator of a boson: a photon ($\alpha=s,a$) or phonon-like excitation ($\alpha=p$) with wave vector $\mathbf{k}_\alpha$ and frequency ω_α. The index i labels the different modes for all three fields.

The phenomenological coupling constants χ_α are related to the quadratic susceptibility for the case of scattering by polaritons and to the elasto–optical tensor for the case of scattering by acoustic phonons.

We consider the case when the scattering efficiency is not too great, so that the pumping can be regarded as given. In addition, we assume that it is classical, specifying its field in the form of a plane monochromatic wave propagating along the z axis:

$$\mathcal{E}_0^{(+)}=\mathcal{E}_{00}^{(+)}e^{i(k_0z-\omega_0t)}. \tag{2.4}$$

We do not take into account four-photon processes quadratic in the pumping, making the assumption that the pumping is weak. We ignore the damping of the acoustic phonons (or polaritons) not associated with the scattering of the light. We also ignore the effect of scattering of the light by the boundaries of the sample.

We write the solution of the Schrödinger equation

$$i\hbar\frac{d|\psi\rangle}{dt}=\mathcal{H}|\psi\rangle,$$

which determines the evolution of the quantum state $|\psi\rangle$ of the system, using second-order perturbation theory:[4]

$$|\psi(t)\rangle=U|\psi(t_0)\rangle,$$

with the evolution operator

$$U\approx 1+\frac{1}{i\hbar}\int_{t_0}^t dt_1\mathcal{H}(t_1)+\frac{1}{(i\hbar)^2}\int_{t_0}^t dt_1\int_{t_0}^{t_1}dt_2\mathcal{H}(t_1)\mathcal{H}(t_2). \tag{2.5}$$

The final, and most complicated, term in (2.5) can be expressed in terms of the previous term by defining

$$\langle\psi(t)|\psi(t)\rangle\approx\langle\psi(t_0)|\left[1+\frac{1}{\hbar^2}\left[\int_{t_0}^t et_1\mathcal{H}(t_1)\right]^2-\frac{2}{\hbar^2}\int_{t_0}^t dt_1\mathcal{H}(t_1)\int_{t_0}^{t_1}dt_2\mathcal{H}(t_2)\right]|\psi(t_0)\rangle. \tag{2.6}$$

Then in accordance with the normalization condition

$$\langle\psi(t)|\psi(t)\rangle\equiv\langle\psi(t_0)|\psi(t_0)\rangle, \tag{2.7}$$

we have

$$2\int_{t_0}^t\mathcal{H}(t_1)\int_{t_0}^{t_1}\mathcal{H}(t_2)dt_1dt_2=\left[\int_{t_0}^t\mathcal{H}(t_1)dt_1\right]^2. \tag{2.8}$$

The sign of approximate equality has been used in (2.6) because terms of higher than second order in the small quantity $\chi_\alpha\mathcal{E}_{00}$ have been omitted.

The limits of integration t_0 and t determine the times at which the pumping is switched on and off. For continuous pumping, we have $t_0\to-\infty$ and $t\to\infty$, and integration over the time gives

$$\int_{-\infty}^{\infty}e^{i(\omega_0-\omega_{si}-\omega_{pj})t}dt\approx 2\pi\delta(\omega_0-\omega_{si}-\omega_{pj}),$$

$$\int_{-\infty}^{\infty}e^{i(\omega_0-\omega_{ai}+\omega_{pj})t}dt\approx 2\pi\delta(\omega_0-\omega_{ai}+\omega_{pj}). \tag{2.9}$$

These relations ensure energy conservation in the stationary case in each scattering event. The modes si and pj for the Stokes process, and also the modes ai and pj for the anti-Stokes process, are related to each other pairwise.

We now go over to integration of the Hamiltonian (2.1) over the volume V. Let the nonlinear medium be unbounded in the transverse directions of its thickness l, the entrance and exit faces of which are perpendicular to the z axis. Then in the diffractionless approximation integration over the transverse coordinates gives

$$\iint_{-\infty}^{\infty}\exp[i\mathbf{r}_\perp(\mathbf{k}_{si\perp}+\mathbf{k}_{pj\perp})]dxdy=4\pi^2\delta^{(2)}(\mathbf{k}_{si\perp}+\mathbf{k}_{pj\perp}),$$

$$\iint_{-\infty}^{\infty}\exp[i\mathbf{r}_\perp(\mathbf{k}_{ai\perp}-\mathbf{k}_{pj\perp})]dxdy=4\pi^2\delta^{(2)}(\mathbf{k}_{ai\perp}-\mathbf{k}_{pj\perp}), \tag{2.10}$$

and this ensures fulfillment of the conservation law for the transverse momentum at the microscopic level in each elementary scattering event.

Integration with respect to the longitudinal coordinate gives

$$\int_{-l}^{0} dz e^{iz\Delta_{\alpha ij}} = \frac{1-e^{-il\Delta_{\alpha ij}}}{il\Delta_{\alpha ij}} l = l e^{-il\Delta_{\alpha ij}/2} \mathrm{sinc}\left(\frac{l\Delta_{\alpha ij}}{2}\right), \tag{2.11}$$

where the longitudinal wave detunings are

$$\Delta_{sij} = k_0 - k_{siz} - k_{pjz}, \quad \Delta_{aij} = k_0 - k_{aiz} + k_{pjz}. \tag{2.12}$$

We specify the initial state of the system in the form

$$|\psi(t_0)\rangle \equiv |\psi_0\rangle = \prod_{si,ai,pi} |0\rangle_{si}|0\rangle_{ai}|\rangle_{pi}, \tag{2.13}$$

where the product is over the vacuum Stokes and anti-Stokes modes and also over the polariton modes in an arbitrary state. For simplicity, we consider only one triplet of modes ai, si, pi (assuming, for example, that the moments of the fields are measured by detectors with high spatial and frequency resolutions). We shall omit the index i throughout.

In accordance with (2.1)–(2.3), (2.5), (2.6), and (2.10)–(2.13), the first approximation of perturbation theory gives

$$\frac{1}{i\hbar}\int_{-\infty}^{\infty} dt_1 \mathscr{H}(t_1)|\psi_0\rangle$$

$$= C_1 \mathscr{E}_{00}^{(+)} \Big[\chi_s(\omega_0,\omega_s,\omega_p)\sqrt{\omega_s\omega_p}$$

$$\times \delta(\omega_0-\omega_s-\omega_p)\delta^{(2)}(\mathbf{k}_{s\perp}+\mathbf{k}_{p\perp})e^{-il\Delta_s/2}$$

$$\times \mathrm{sinc}\left(\frac{l\Delta_s}{2}\right) a_p^+|1\rangle_s|0\rangle_a|\rangle_p + \chi_a(\omega_0,\omega_a,\omega_p)\sqrt{\omega_a\omega_p}$$

$$\times \delta(\omega_0-\omega_a+\omega_p)\delta^{(2)}(\mathbf{k}_{a\perp}-\mathbf{k}_{p\perp})e^{-il\Delta_a/2}$$

$$\times \mathrm{sinc}\left(\frac{l\Delta_a}{2}\right) a_p|0\rangle_s|1\rangle_a|\rangle_p \Big], \tag{2.14}$$

where

$$C_1 = -\frac{i8\pi^4 l}{L^3}. \tag{2.15}$$

We find the mean number of photons per mode in the Stokes and anti-Stokes beams:

$$N_s \equiv \langle n_s\rangle \equiv \langle\psi(t)|a_s^+a_s|\psi(t)\rangle$$

$$= |C_1\mathscr{E}_{00}^{(+)}|^2|\chi_s|^2\omega_s\omega_p\delta^{(2)}(\omega_0-\omega_s-\omega_p)$$

$$\times \delta^{(4)}(\mathbf{k}_{s\perp}+\mathbf{k}_{p\perp})\mathrm{sinc}^2\left(\frac{l\Delta_s}{2}\right)_p \langle|a_pa_p^+|\rangle_p, \tag{2.16}$$

$$N_a \equiv \langle n_a\rangle \equiv \langle\psi(t)|a_a^+a_a|\psi(t)\rangle$$

$$= |C_1\mathscr{E}_{00}^{(+)}|^2|\chi_a|^2\omega_a\omega_p\delta^{(2)}(\omega_0-\omega_a+\omega_p)\delta^{(4)}(\mathbf{k}_{a\perp}$$

$$-\mathbf{k}_{p\perp})\mathrm{sinc}^2\left(\frac{l\Delta_a}{2}\right)_p \langle|a_p^+a_p|\rangle_p. \tag{2.17}$$

We now go to the second approximation of perturbation theory, which is described by double application of the Hamiltonian (2.1) to the state (2.13). Because it is lengthy we shall not write out fully the result of the calculation of the last term in (2.5). We merely note that it contains not only vacuum states but also the following two-phonon states in which we are interested:

$$\sqrt{2}\chi_s^2(a_p^+)^2|2\rangle_s|0\rangle_a|\rangle_p, \tag{2.18}$$

$$\sqrt{2}\chi_a^2(a_p)^2|0\rangle_s|2\rangle_a|\rangle_p, \tag{2.19}$$

$$\chi_s\chi_a(a_p^+a_p+a_pa_p^+)|1\rangle_s|1\rangle_a|\rangle_p. \tag{2.20}$$

It is these states that determine nonvanishing second moments of the intensity correlation functions, the calculation of which will be made below.

3. CORRELATION OF THE INTENSITIES IN THE SCATTERED LIGHT AND ITS RELATIONSHIP TO THE STATE OF THE SCATTERING QUASIPARTICLES

Using the wave function obtained in the previous section, we find the intensity correlation functions for the scattered light. We begin by defining the normalized correlation coefficient of the Stokes and anti-Stokes beams:

$$K_{sa} \equiv \frac{\langle n_s n_a\rangle}{\langle n_s\rangle\langle n_a\rangle}. \tag{3.1}$$

A nonvanishing contribution to this correlation function is made by the component (2.20) of the state (2.5). With allowance for (2.16) and (2.17), we have

$$K_{sa} = \frac{\langle(2n_p+1)^2\rangle}{4\langle n_p\rangle(\langle n_p\rangle+1)} = \frac{4\langle:n_p^2:\rangle+8\langle n_p\rangle+1}{4\langle n_p\rangle(\langle n_p\rangle+1)}. \tag{3.2}$$

Here the averaging is over the initial state of the phonon (or polariton) mode $|\ \rangle_p$, and the colon denotes normal ordering.

If the phonons (polaritons) possess thermal statistics, then

$$\langle:n_p^m:\rangle = m!\langle n_p\rangle^m$$

and

$$K_{sa}^{(T)} = \frac{8\langle n_p\rangle^2+8\langle n_p\rangle+1}{4\langle n_p\rangle(\langle n_p\rangle+1)}. \tag{3.3}$$

The graph of the dependence of $K_{sa}^{(T)}$ on $\langle n_p\rangle$ is shown in Fig. 1. For $\langle n_p\rangle \gg 1$, we have $K_{sa}^{(T)} \to 2$, in agreement with the results of Ref. 6.

For phonons or polaritons in a coherent state

$$\langle:n_{p0}^m:\rangle = \langle n_{p0}\rangle^m,$$

and

$$K_{as}^{(C)} = \frac{4\langle n_{p0}\rangle^2+8\langle n_{p0}\rangle+1}{4\langle n_{p0}\rangle(\langle n_{p0}\rangle+1)}. \tag{3.4}$$

The corresponding graph is also shown in Fig. 1. For $\langle n_{p0}\rangle \gg 1$, we have $K_{as}^{(C)} \approx 1$, i.e., $K_{as}^{(C)}$ is half the value for thermal phonons.

In the case of an arbitrary bunching parameter

$$K_{pp} \equiv \frac{\langle:n_{p0}^2:\rangle}{\langle n_{p0}\rangle^2}$$

of the phonon system, we can write

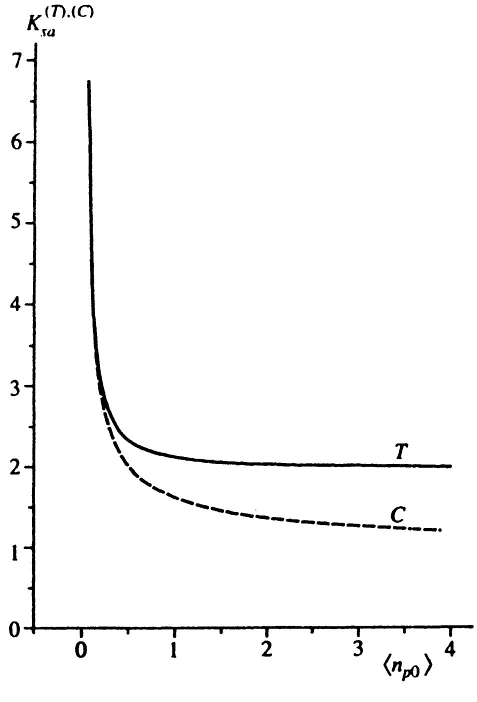

FIG. 1. Normalized correlation coefficient of Stokes and anti-Stokes scattering as function of the mean number of polaritons (phonons) in the mode for thermal (T) and coherent (C) statistics of the scattering excitations.

$$K_{as}=\frac{4K_{pp}\langle n_{p0}\rangle^2+8\langle n_{p0}\rangle+1}{4\langle n_{p0}\rangle(\langle n_{p0}\rangle+1)}. \tag{3.5}$$

Thus, for a large number of phonons (polaritons) in the mode, the correlation coefficient K_{sa} is equal to the coefficient K_{pp}, the bunching parameter of the scattering quasiparticles.

On the other hand, for $\langle n_{p0}\rangle \ll 1$, i.e., in the case of vacuum phonons (polaritons), K_{as} diverges as (1/4) $\langle n_{p0}\rangle$.

We now turn to the determination of the correlation coefficients K_{ss} and K_{aa} (the bunching parameters for the Stokes and anti-Stokes light), which can be measured, for example, using the Brown–Twiss scheme. For the first coefficient, the component (2.18) makes a nonvanishing contribution; for the second, the component (2.19) does. As a result, we obtain

$$K_{ss}\equiv\frac{\langle :n_s^2:\rangle}{\langle n_s\rangle^2}=\frac{\langle :n_{p0}^2:\rangle+4\langle n_{p0}\rangle+2}{(\langle n_{p0}\rangle+1)^2}, \tag{3.6}$$

$$K_{aa}\equiv\frac{\langle :n_a^2:\rangle}{\langle n_a\rangle^2}=\frac{\langle :n_{p0}^2:\rangle}{\langle n_{p0}\rangle^2}\equiv K_{pp}. \tag{3.7}$$

The last result is very striking. The bunching parameter of the anti-Stokes light is equal to the bunching parameter of the scattering quasiparticles irrespective of the mean number per mode. This result is readily understood if one recalls that the production of an anti-Stokes photon is always accompanied by the annihilation of a phonon (or polariton).

4. SCATTERING BY TWO UNCOUPLED PHONONS

The algorithm that we have considered for solving the problem of the scattering of light by phonons (polaritons) enables us to take into account not only four-photon processes quadratic in the pumping but also other, more complicated processes. All that is changed is the actual form of the Hamiltonian (2.1); all the subsequent sequence of operations remains the same. As an example, we consider the process of scattering by two uncoupled phonons. In this case, the effective Hamiltonian is transformed to

$$\begin{aligned}\mathcal{H}=-\frac{1}{2}\sum_{ijl}\int_V d^3\mathbf{r}\mathcal{E}_0^{(+)}(\mathbf{r},t)[\chi_s(\omega_0,\omega_{si},\omega_{pj})E_{si}^{(-)}\\ \times(\mathbf{r},t)E_{pj}^{(-)}(\mathbf{r},t)+\chi_a(\omega_0,\omega_{ai},\omega_{pj})E_{ai}^{(-)}(\mathbf{r},t)E_{pj}^{(+)}\\ \times(\mathbf{r},t)+\chi_{2s}(\omega_0,\omega_{2si},2\omega_{pj})E_{2si}^{(-)}(\mathbf{r},t)(E_{pj}^{(-)}(\mathbf{r},t))^2\\ +\chi_{2a}(\omega_0,\omega_{2ai},2\omega_{pj})E_{2ai}^{(-)}(\mathbf{r},t)(E_{pj}^{(+)}(\mathbf{r},t))^2]+\text{h.c.}\end{aligned} \tag{4.1}$$

We calculate the correlation coefficient for anti-Stokes scattering by a phonon and Stokes scattering by two uncoupled phonons. We use here the single-mode approximation, i.e., we identify precisely the triplet of modes (ai, $2si$, and pi) that satisfies the conditions of wave and frequency matching. In this case, it is important that both phonons participating in the anti-Stokes processes belong to a single mode. A single application of the Hamiltonian (3.1) to the vector of the original state

$$|\psi_0\rangle=|0\rangle_s|0\rangle_a|0\rangle_{2s}|0\rangle_{2a}|\rangle_p \tag{4.2}$$

gives the following components of interest to us:

$$\begin{aligned}&\chi_a a_p|0\rangle_s|1\rangle_a|0\rangle_{2a}|0\rangle_{2a}|\rangle_p,\\ &\chi_{2s}(a_p^+)^2|0\rangle_s|0\rangle_a|1\rangle_{2s}|0\rangle_{2a}|\rangle_p.\end{aligned} \tag{4.3}$$

If we apply the Hamiltonian twice, then besides other components, we obtain

$$\chi_{2s}\chi_a(a_p(a_p^+)^2+(a_p^+)^2a_p)|0\rangle_s|1\rangle_a|1\rangle_{2s}|0\rangle_{2a}|\rangle_p, \tag{4.4}$$

from which it immediately follows that

$$\begin{aligned}K_{a2s}&\equiv\frac{\langle n_a n_{2s}\rangle}{\langle n_a\rangle\langle n_{2s}\rangle}\\ &=\frac{{}_p\langle|(a_p^2a_p^++a_p^+a_p^2)(a_p(a_p^+)^2+(a_p^+)^2a_p)|\rangle_p}{{}_p\langle|n_p|\rangle_p\times{}_p\langle|a_p^2(a_p^+)^2|\rangle_p}\\ &=\frac{\langle :n_{p0}^3:\rangle+6\langle :n_{p0}^2:\rangle+7\langle n_{p0}\rangle+1}{\langle n_{p0}\rangle(\langle :n_{p0}^2:\rangle+4\langle n_{p0}\rangle+2)}.\end{aligned} \tag{4.5}$$

For "thermal" phonons, we have

$$K_{a2s}^{(T)}=\frac{6\langle n_{p0}\rangle^2+6\langle n_{p0}\rangle+1}{2\langle n_{p0}\rangle(\langle n_{p0}\rangle+1)}=3+\frac{1}{2\langle n_{p0}\rangle(\langle n_{p0}\rangle+1)}. \tag{4.6}$$

The graph of this dependence is shown in Fig. 2. For $\langle n_{p0}\rangle \gg 1$, we have $K_{a2s}^{(T)}\approx 3$ (in contrast to $K_{as}^{(T)}\approx 2$). This cir-

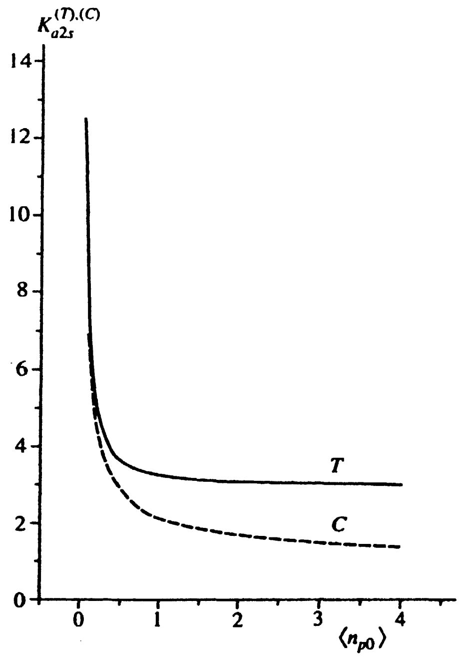

FIG. 2. Normalized coefficient of the correlation between two-particle Stokes scattering and single-particle anti-Stokes scattering as function of the mean number of polaritons (phonons) in the mode for thermal (T) and coherent (C) statistics of the scattering excitations.

cumstance can evidently be used to investigate second-order phonon spectra if it is thereby possible to distinguish coupled two-phonon states from uncoupled states.

For phonons in a coherent state,

$$K_{a2s}^{(C)}=\frac{\langle n_{p0}\rangle^3+6\langle n_{p0}\rangle^2+7\langle n_{p0}\rangle+1}{\langle n_{p0}\rangle(\langle n_{p0}\rangle^2+4\langle n_{p0}\rangle+2)}. \quad (4.7)$$

For $\langle n_{p0}\rangle \gg 1$, the coefficient $K_{a2s}^{(C)}$, like $K_{as}^{(C)}$, is equal to unity.

Finally, for $\langle n_{p0}\rangle \ll 1$ we have $K_{a2s}\approx 1/2\langle n_{p0}\rangle$ irrespective of the state of the phonons, which in this case is always close to the vacuum state (Fig. 2).

We note that in an experiment it is rather difficult to separate scattering by two uncoupled phonons that both belong to one transverse mode (their wave vectors differ little). Indeed, many pairs of phonons with the most varied wave vectors can contribute to the scattering—it is merely necessary that their vector sum correspond to the distinguished anti-Stokes mode. Nevertheless, one may attempt to establish a nonequilibrium occupation of a phonon mode with a distinguished wave vector, for example, as was done in Ref. 9.

5. CONCLUSIONS

Thus measurement of the fourth moments of light scattered by polaritons or acoustic phonons makes it possible to determine the bunching parameter of the scattering quasiparticles. If the number of phonons (polaritons) per mode is much greater than unity, the measurement can be made both with Stokes or anti-Stokes radiation in accordance with the Brown–Twiss scheme or by investigating the cross correlation of the intensities between the Stokes and anti-Stokes radiation. However, in the case when the mode of phonon-like excitations is weakly occupied (and it is this case that is interesting for the investigation of nontrivial phonon statistics), only the intensity correlation function for anti-Stokes scattering (3.8) measured using the Brown–Twiss scheme gives the correct value of the bunching parameter of the phonon (polariton) system.

This paper should be seen as a theoretical addition to the study of Ref. 9, in which the fourth moments were measured experimentally for the scattering of light by acoustic phonons. Moreover, the basic idea of these studies—that bunching of the phonon system can be deduced from bunching of the scattered light—is also due in equal degree to the authors of Ref. 9.

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[1)]Here and in what follows, the expressions "photon statistics" and "photon correlation" mean that correlation functions of the form $K=\langle n_i n_j\rangle \equiv \langle a_i^+ a_i a_j^+ a_j\rangle$, where a_i, a_j are the annihilation operators for modes i and j, are measured or calculated.

[1]B. Crosignani, P. Di Porto, and M. Bertolotti, *Statistical Properties of Scattered Light* (Academic Press, New York, 1975) [Russ. transl., Nauka, Moscow, 1980].

[2]N. I. Lebedev, Vestn. Mosk. Univ. **23**, 41 (1982).

[3]S. P. Kulik, A. N. Penin, and P. V. Prudkovskiĭ, Zh. Éksp. Teor. Fiz. **106**, 993 (1994) [JETP **79**, 543 (1994)].

[4]D. N. Klyshko, *Photons and Nonlinear Optics* [in Russian], (Nauka, Moscow, 1980).

[5]D. N. Klyshko, Zh. Éksp. Teor. Fiz. **64**, 1160 (1973) [Sov. Phys. JETP **37**, 590 (1973)].

[6]D. N. Klyshko, Kvantovaya Elektron. (Moscow) **4**, 1341 (1977) [Sov. J. Quantum Electron. **7**, 755 (1977)].

[7]A. S. Shumovsky and B. Tanatar, Phys. Lett. A **182**, 411 (1993).

[8]M. Artoni and J. F. Birman, Opt. Commun. **104**, 319 (1994).

[9]A. N. Penin, M. V. Chekhova, S. P. Kulik, and P. A. Prudovsky, in *Proc. 15th Int. Conf. on Coherent and Nonlinear Optics* (Technical Digest, Vol. 1 (1995), p. 269).

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