A new method for detection of exciton Bose condensation using stimulated two-photon emission

Yu. E. Lozovik and A V. Poushnov*

Institute of Spectroscopy, Russian Academy of Sciences, 142092 Troitsk, Moscow Region, Russia

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Abstract

Stimulated two-photon emission by Bose-condensed excitons accompanied by a coherent two-exciton recombination, i.e., by simultaneous recombination of two excitons with opposite momenta leaving unchanged the occupation numbers of excitonic states with momenta $p \neq 0$, is investigated. Raman light scattering (RLS) accompanied by a similar two-exciton recombination (or generation of two excitons) is also analyzed. The processes under consideration can occur only if a system contains Bose condensate, therefore, their detection can be used as a new method to reveal Bose condensation of excitons. The recoil momentum, which corresponds to a change in the momentum of the electromagnetic field in the processes, is transferred to phonons or impurities. If the recoil momentum is transmitted to optical phonons with frequency $\omega_0^s$, whose occupation numbers are negligible, and the incident light frequency $\omega < 2\Omega_-$, where $\Omega_- = \Omega - \omega_0^s$ and $\Omega$ is the light frequency corresponding to the recombination of an exciton with zero momentum, the stimulated two-photon emission and RLS with the coherent two-exciton recombination lead to the appearance of a line at $2\Omega_- - \omega$ and an anti-Stokes component at $\omega + 2\Omega_-$, respectively. At $\omega > 2\Omega_-$ the RLS spectrum contains Stokes and anti-Stokes components at frequencies $\omega \pm 2\Omega_-$, whereas the stimulated two-photon emission is impossible. Formulas for the cross sections at finite temperatures are obtained for the processes under consideration. Our estimates indicate that a spectral line at $2\Omega_- - \omega$, corresponding to the stimulated two-photon emission accompanied by the coherent optical phonon-assisted two-exciton recombination can be experimentally detected in $\text{Cu}_2\text{O}$.

*E-mail: poushnov@isan.troitsk.ru
1. Introduction

The most interesting collective effects in systems of excitons are the anticipated exciton Bose condensation and superfluidity (see Refs. 1-7 and references therein). Recently a number of publications reported on the detection of Bose condensation and superfluidity of excitons in Cu$_2$O based on observations of changes in exciton luminescence spectrum and ballistic transport of excitons, which have been discussed in literature. Observations of condensation of indirect excitons in coupled quantum wells under strong magnetic fields have also been reported (see Ref. 15, a theoretical discussion in Refs. 16-18, and references therein). In this connection, the detailed investigation of coherent exciton properties, whose detection could be used to reveal exciton Bose condensation, seems to be important.

If a system of excitons is in a Bose condensed state, the mean values of the annihilation (creation) operator of the exciton with zero momentum in the ground state are not zero:

$$\langle N - 1 | Q_0 | N \rangle = \langle N + 1 | Q_0^+ | N \rangle = \sqrt{N_0}. \quad (1)$$

Here $| N \rangle$ is the ground state of the excitonic system with the average number of excitons $N$, $Q_0$ is the annihilation operator of an exciton with zero momentum, and $N_0$ is the number of excitons in the condensate.

Equation (1) clearly shows that, as a result of the recombination (generation) of an exciton with zero momentum, a system of Bose-condensed excitons transfers to the ground state, that differs from the initial one in the average number of excitons with momentum $p = 0$. The recombination of excitons with zero momentum leads to the appearance of a peak (the so-called condensate peak) in the excitonic luminescence spectrum at frequency

$$\Omega = [E_0(N) - E_0(N - 1)]/\hbar,$$

where $E_0(N)$ is the energy of the ground state of the excitonic system.

If the exciton-exciton interaction is nonvanishing, then, in addition to the mean values defined by Eq. (1), products of two annihilation (creation) operators of excitons with opposite momenta averaged over the ground state of the Bose-condensed excitonic system (the so-called anomalous averages) are not zero:

$$\langle N - 2 | Q_{-p} Q_p | N \rangle \neq 0, \quad \langle N + 2 | Q_{-p}^+ Q_{p}^+ | N \rangle \neq 0. \quad (2)$$

The unusual optical properties inherent in Bose-condensed state of interacting excitons due to nonvanishing anomalous means (2) are considered in this paper. It will be shown that due to the interaction with the electromagnetic field, the coherent recombination (or generation), i.e., the simultaneous recombination (or creation) of two excitons with opposite momenta, corresponding to anomalous averages (2) is possible. In such processes, the
occupation numbers of excitons with \( p \neq 0 \) are unchanged, and the final state of excitons differs from the initial one only in the average number of excitons with zero momentum. In particular, after the two-exciton recombination, the average number of condensate excitons is reduced by two.

The coherent two-exciton recombination can contribute, for example, to the stimulated two-photon emission or to Raman light scattering (RLS) by Bose-condensed excitons. RLS can also be accompanied by the coherent generation of two excitons. In these processes, the momentum of the exciton-photon system is not conserved: the recoil momentum, equal to the change in the momentum of the electromagnetic field, is transferred to phonons or impurities. In this paper we consider the processes in which the recoil momentum is transferred to two optical phonons. Such processes seem to be most probable in the excitonic system in Cu\(_2\)O crystal, which is one of the most interesting crystals in view of the observation of exciton Bose condensation. In fact, the radiative recombination accompanied by the transmission of the recoil momentum to one optical phonon is typical for excitons in Cu\(_2\)O. Using the energy and momentum conservation laws, one can prove that, in a defect-free crystal, the coherent recombination of two excitons is possible only if the recoil momentum is transferred to two phonons.

At low temperatures, the occupation numbers of optical phonons are small, therefore, it is most probable that the recoil momentum is transferred to two phonons generated in the process. If the phonon dispersion is negligible and the incident light frequency \( \omega < 2\Omega_\omega \), a line in the spectrum of the stimulated two-phonon emission at \( 2\Omega_\omega - \omega \) and an anti-Stokes component in the RLS spectrum at \( \omega + 2\Omega_\omega \) should appear. Here \( \Omega_\omega = \Omega - \omega_0^s \) and \( \omega_0^s \) is the optical phonon frequency. Both these lines correspond to the coherent two-exciton recombination: the energy of the initial state of the system is higher than the energy of its final state by \( 2\hbar\Omega \), where \( \Omega \) is the frequency corresponding to the recombination of an exciton with zero momentum. If \( \omega > 2\Omega_\omega \), the RLS spectrum should contain the anti-Stokes component at \( \omega + 2\Omega_\omega \), which corresponds to the coherent two-exciton recombination, and the Stokes component at \( \omega - 2\Omega_\omega \) due to the coherent generation of two excitons. The stimulated emission of two photons is impossible in this case. The appearance of the lines at frequencies \( |\omega \pm 2\Omega_\omega| \) is possible only if the excitons are in the Bose-condensed state, and after a transition to the normal state these lines should disappear.

The paper is organized as follows. In Section 2 the stimulated two-photon emission with the coherent two-exciton recombination accompanied by the transmission of the recoil momentum to phonons is considered. The diagram technique is used to obtain the cross sections of two-photon processes involving the coherent two-exciton recombination (or generation)
at finite temperatures. This approach allows one to express the appropriate elements of the S-matrix in a natural manner in terms of anomalous Green’s functions of Bose-condensed excitons. The cross section of the stimulated two-photon emission with the coherent phonon-assisted two-exciton recombination is obtained, and its temperature dependence is studied. It turns out that this dependence can be nonmonotonic under certain conditions. Namely, in a certain temperature interval below $T_c$ the cross section of the stimulated two-photon emission can increase with the growth of temperature and can become even higher than it is at $T = 0$. The causes of this unusual temperature dependence is investigated.

Section 3 is dedicated to RLS accompanied by the coherent processes of two-exciton recombination or generation. In Section 4 the possibility of the experimental observation of the lines at frequencies $|\omega \pm 2\Omega -|$ corresponding to the stimulated two-photon emission and RLS is analyzed. Our numerical estimates for excitons in Cu$_2$O indicate that a spectral line at $2\Omega - \omega$ corresponding to the stimulated optical phonon-assisted two-exciton recombination can be detected and, therefore, can be used to reveal exciton Bose condensation.

2. Stimulated two-photon emission accompanied by coherent two-exciton recombination

The effective Hamiltonian describing phonon-assisted radiative recombination (generation) of excitons can be expressed as follows (see Ref. 20 and Appendix A):

$$\hat{H}_L = \sum_{pq} \left[ L_{pq}^> e^{-i\Omega t} Q_p(t) c_q^+(t) b_{p-q}^+(t) + L_{pq}^< e^{-i\Omega t} Q_p(t) c_q^+(t) b_{q-p}^+(t) \right. \right.$$  
$$\left. + L_{pq}^> e^{-i\Omega t} Q_p(t) c_q^+(t) b_{p+q}^+(t) + L_{pq}^< e^{-i\Omega t} Q_p(t) c_q^+(t) b_{-p-q}^+(t) + \text{H.c.} \right], \tag{3}$$

where

$$L_{pq}^>(<) = i\sqrt{2\pi\omega_q} \epsilon^f_{pq}^<(>)$$

$\Omega$ is the frequency corresponding to the recombination of an exciton with zero momentum. Hamiltonian (3) is written in the Heisenberg representation. Here $Q_p(t) = Q_p \exp[-i\epsilon(p)t]$ and $b_p(t) = b_p \exp(-i\omega_p t)$ are the annihilation operators of an exciton and a phonon with momentum $p$, respectively, $c_q(t) = c_q \exp(-i\omega_q t)$ is the annihilation operator of a photon with momentum $q$ ($\omega_q$ and $\epsilon$ are the photon frequency and its polarization unit vector). The exciton energy is measured with respect to the bottom of the exciton band: $\epsilon(0) = 0$. The effective matrix elements $f_{pq}^>(<)$ are responsible for the recombination of an exciton with momentum $p$, which includes, in addition to the emission (absorption) of
a photon with momentum \( \mathbf{q} \), the simultaneous emission or absorption of a phonon\(^1\) (see Ref. 20 and Appendix A).

By expanding the evolution operator

\[
\hat{S}(t) = T_t \exp \left[ -i \int_{-\infty}^{t} \hat{H}_L(t') dt' \right]
\]

in powers of \( \hat{H}_L \) and retaining terms of up to the second order, we obtain an expression for the elements of the \( S \)-matrix corresponding to phonon-assisted two-photon processes:

\[
S_{n'n} = \frac{(-i)^2}{2!} \int_{-\infty}^{\infty} dt' \int_{-\infty}^{\infty} dt'' \langle n'|T_t \hat{H}_L(t') \hat{H}_L(t'')|n\rangle dt' dt'',
\]

where \( n \) and \( n' \) label the initial and final states of the system composed of excitons and phonons + electromagnetic field.

Let us consider the two-photon emission by excitons in the Bose condensed state due to the coherent two-exciton recombination, i.e., a transition of the excitonic system from state \( |n\rangle_{\text{exc}} = |n, N\rangle_{\text{exc}} \) to the state \( |m\rangle_{\text{exc}} = |n, N-2\rangle_{\text{exc}} \), which differs from the initial state in the average number of excitons with momentum \( \mathbf{p} = 0 \). The change in the electromagnetic field momentum is \( \mathbf{k}' + \mathbf{k} \) in this process, where \( \mathbf{k} \) and \( \mathbf{k}' \) are the momenta of emitted photons. The recoil momentum \( \delta \mathbf{k} = -(\mathbf{k}' + \mathbf{k}) \) is entirely transferred to phonons since the momentum of the excitonic system is zero in both the initial and final states.

For the element of the \( S \)-matrix corresponding to the coherent phonon-assisted two-exciton recombination, we have

\[
(S_p)_{mn} = -\frac{1}{2} \int_{-\infty}^{\infty} dt' \int_{-\infty}^{\infty} dt'' \exp [-i\Omega(t' + t'')] \times \left\{ \left[ L^{b}_{pk} L^{b}_{-pk'} \langle m|T_t Q_p(t') Q_{-p}(t'')|n\rangle_{\text{exc}} \langle f|T_t b^+_{p-k} (t') b^+_{-p-k'} (t'')|i\rangle_{\text{phon}} \\
+ L^{b}_{q-p,k} L^{b}_{-q-k'} \langle m|T_t Q_{q-p}(t') Q_{-q}(t'')|n\rangle_{\text{exc}} \langle f|T_t b^+_{-p-k'} (t') b^+_{p-k} (t'')|i\rangle_{\text{phon}} \right] \times \langle f|T_t c^+_k (t') c^+_k (t'')|i\rangle_{\text{phot}} \right. \\
+ \left. \left[ L^{b}_{-pk'} L^{b}_{pk} \langle m|T_t Q_{-p}(t') Q_p(t'')|n\rangle_{\text{exc}} \langle f|T_t b^+_{p-k} (t') b^+_{-p-k'} (t'')|i\rangle_{\text{phon}} \\
+ L^{b}_{q-k'} L^{b}_{-q-k} \langle m|T_t Q_{q-k}(t') Q_{-q-k}(t'')|n\rangle_{\text{exc}} \langle f|T_t b^+_{-p-k'} (t') b^+_{p-k} (t'')|i\rangle_{\text{phon}} \right] \times \langle f|T_t c^+_k (t') c^+_k (t'')|i\rangle_{\text{phot}} \right\},
\]

\(^1\)In a general case, the radiative recombination of an exciton can result in emission (absorption) of an arbitrary number of phonons. When using Hamiltonian (3), we limit our analysis for simplicity to the case of excitonic recombination with emission (absorption) of one phonon.
where $\mathbf{q} = \mathbf{k} - \mathbf{k}'$. Here $|i\rangle_{\text{phot}} = |0\rangle_{\text{phot}}$ and $|f\rangle_{\text{phot}} = |1_{k}, 1_{k'}\rangle_{\text{phot}}$ are the initial and final states of the electromagnetic field, respectively. Assuming that the phonons are optical and the lattice temperature $T_{\text{lat}}$, which is, generally speaking, different from the excitonic temperature $T$, is sufficiently small ($T_{\text{lat}} \ll \omega_0^s$, where $\omega_0^s$ is the characteristic energy of optical phonons), we suppose that $|i\rangle_{\text{phon}} = |0\rangle_{\text{phon}}$ and $|f\rangle = |1_{p-k}, 1_{-p-k'}\rangle_{\text{phon}}$.

By performing averaging over the Gibbs distribution for the excitonic system, we obtain the element of the $S$-matrix responsible for the two-photon emission that transforms the system from its state of thermodynamic equilibrium $|i\rangle_{\text{exc}} = \sum_n \exp[(F - E_n(N) + \mu N)/T]|n, N\rangle_{\text{exc}}$ to $|f\rangle_{\text{exc}} = Q^2_0|i\rangle/N_0$:

$$(S_p)_{fi} = \sum_n \exp[(F - E_n(N) + \mu N)/T](S_p)_{mn}. \tag{6}$$

By expressing the $S$-matrix element (6) in terms of the anomalous Green’s function of excitons, we obtain

$$(S_p)_{fi} = -\frac{1}{2} \int_{-\infty}^{\infty} dt' dt'' \exp[-i\Omega(t' + t'')] \times \left\{ \left[ L_{pk}^p L_{qk}^{q'} \left( n_0(T) \delta_p + i \hat{G}_{p-q}(t') \right) \right]|T|_{p-k}^+ (t') b_{p-k'}^+ (t'') |i\rangle_{\text{phon}} \\
+ L_{q-p,k}^p \left( n_0(T) \delta_{p-q} + i \hat{G}_{p-q}(t') \right) |f| T_{b_{p-k}^+ (t')} b_{p-k}^+ (t'') |i\rangle_{\text{phon}} \\
+ \left[ L_{pk}^p L_{qk}^{q'} \left( n_0(T) \delta_p + i \hat{G}_{p-q}(t') \right) \right]|T|_{b_{p-k}^+ (t')} b_{p-k}^+ (t'') |i\rangle_{\text{phon}} \\
+ L_{q-p,k}^p \left( n_0(T) \delta_{p-q} + i \hat{G}_{p-q}(t') \right) |f| T_{b_{p-k}^+ (t')} b_{p-k}^+ (t'') |i\rangle_{\text{phon}} \right\} , \tag{7}$$

where $\delta_p = 1$ at $p = 0$ and $\delta_p = 0$ at $p \neq 0$. Here $\hat{G}_p(t'-t'')$ is the causal Green’s function of Bose-condensed excitons at temperature $T$:

$$\hat{G}_p(t'-t'') = -i(1-\delta_p) \times \sum_n \exp[(F - E_n(N) + \mu N)/T] |n, N-2|T|Q_{p-q}(t')Q_{p}(t'')|n, N\rangle_{\text{exc}}, \tag{8}$$

and function $n_0(T)$ is the density of excitons in the condensate at this temperature.

The resulting element (7) of the $S$-matrix is expressed by the sum of diagrams shown in Fig. 1. The lines with oppositely directed arrows denote the causal anomalous Green’s function of excitons in the Bose-condensed state at $T > 0$ (if the momenta next to this line vanish, it corresponds to function $n_0(T)$). The wavy lines correspond to photon creation operators, and the dashed lines indicate photon creation operators. The vertices on these
diagrams correspond to matrix elements $L_{pk}^> > p_k$, where $p$ and $k$ are the momenta of the exciton and photon lines originating at the vertex.

Integration with respect to $t' - t''$ and $t''$ yields

$$
\langle S_p \rangle_{fi} = 2\pi i T_{k'kk} (p) \left[ (\sqrt{2} - 1) \delta(p - q/2) + 1 \right] \delta(\omega' + \omega + \omega_{p-k}^s + \omega_{-p-k'}^s - 2\Omega),
$$

where

$$
T_{k'kk} (p) = i \left\{ L_{pk}^> L_{pqk'}^> \left[ 2\pi n_0(T) \delta_p \delta(\omega + \omega_{p-k}^s - \Omega) + i \hat{G}_p(\omega + \omega_{p-k}^s - \Omega) \right]
+ L_{q-p,k}^> L_{q-p,k'}^> \left[ 2\pi n_0(T) \delta_{p-q} \delta(\omega + \omega_{p-k'}^s - \Omega) + i \hat{G}_p(\omega + \omega_{p-k'}^s - \Omega) \right] \right\}
$$

is the matrix element of the two-photon emission due to the coherent phonon-assisted two-exciton recombination, which is similar to the scattering amplitude in the collision problem.

In deriving this equation, we have taken into account the fact that the anomalous Green’s function is an even function of frequency and does not depend on the momentum direction. The sum in the brackets in Eq. (9) takes into account the fact that the momenta of emitted phonons are equal at $p = q/2$.

Let us limit our discussion to the stimulated phonon-assisted two-photon emission with a negligible dispersion of phonons ($\omega_0^s = \omega_0^s$). It follows from Eq. (9) that the stimulated two-photon emission of this kind leads to the appearance of a line at frequency $2\Omega - \omega$, where $\Omega_0 = \Omega - \omega_0^s$ and $\omega$ is the incident light frequency.

The differential cross section of the stimulated two-photon emission corresponding to the coherent phonon-assisted two-exciton recombination is given by

$$
d\sigma = \frac{2\pi}{c} \left[ \frac{1}{2} \sum_{\neq q/2} \left| T_{k'kk} (p) \right|^2 + 2 \left| T_{k'kk} (q/2) \right|^2 \right] \left( \frac{(2\Omega_0 - \omega)^2}{(2\pi c)^3} \right) d\omega,
$$

where

2Calculations concerning the two-photon emission and RLS under discussion could be performed using Keldysh’s elegant diagrammatic technique (see, e.g., Ref. 21, which is devoted to a problem that requires a similar technique). In our opinion, however, our approach used in this specific case is more visual.

3In a general case, the number of phonons involved in the process can be arbitrary. Moreover, the recoil momentum (the whole or a fraction of it) can be transferred to impurities. Thus, the stimulated two-photon emission can result in the appearance of the spectral lines at frequencies $2(\Omega - n\omega_0^s)$, where $n$ is an arbitrary integer.
\[ T_{k'k}(p) = i \left\{ L_{pk}^> L_{p'k'}^> \left[ 2\pi n_0(T) \delta_p \delta(\omega - \Omega_-) + i \hat{G}_p(\omega - \Omega_-) \right] ight. \\
+ \left. L_{q-p,k}^> L_{p-q,k'}^> \left[ 2\pi n_0(T) \delta_{p-q} \delta(\omega - \Omega_-) + i \hat{G}_{p-q}(\omega - \Omega_-) \right] \right\}. \tag{12} \]

The factor 1/2 in front of the sum over \( p \) in Eq. (11) is introduced because the sum over all possible \( p \) includes the emission of two phonons with momenta \( p - k \) and \( -p - k' \) two times: \( T_{k'k}(p) = T_{k'k}(-p + q) \).

It is clear that at \( \omega \neq \Omega_- \) the summands proportional to \( n_0(T) \) do not contribute to the cross section (11). In this case, it is proportional to anomalous Green’s functions, which are determined, as is well known, not only by the presence of Bose condensate, but also by the interaction among particles. Thus, the stimulated two-photon emission corresponding to the coherent phonon-assisted two-exciton recombination at \( \omega \neq \Omega_- \) can take place only in a non-ideal gas of excitons with Bose condensate.

Assuming that the condition \( \omega \neq \Omega_- \) is fulfilled, let us express cross section (11) as follows:

\[ d\sigma^L = \frac{\omega(2\Omega_- - \omega)^3}{e^4} \left[ \frac{1}{2} \sum_{p \neq q/2} |(s_p)_{nm} e_n^* e_m^*|^2 + 2|(s_{q/2})_{nm} e_n^* e_m^*|^2 \right] d\omega', \tag{13} \]

where

\[ (s_p)_{nm} = \hat{G}_p(\omega - \Omega_-)(f_{pk}^>)_n (f_{pk}^>)_m + \hat{G}_{p-q}(\omega - \Omega_-)(f_{p-q,k}^>)_n (f_{p-q,k}^>)_m \tag{14} \]

is the tensor of the two-photon emission corresponding to the coherent phonon-assisted two-exciton recombination.

The causal Green’s function \( \hat{G}_p(\omega) \) is related to the advanced and retarded Green’s functions by the following formula:

\[ \hat{G}_p(\omega) = \frac{1}{2} \left( 1 + \coth \frac{\omega}{2T} \right) \hat{G}_p^R(\omega) + \frac{1}{2} \left( 1 - \coth \frac{\omega}{2T} \right) \hat{G}_p^A(\omega). \tag{15} \]

Substituting it in the tensor (14) yields

\[ (s_p)_{nm} = \frac{1}{2} \left\{ \left[ \left( 1 + \coth \frac{\Delta\omega}{2T} \right) \hat{G}_p^R(\Delta\omega) + \left( 1 - \coth \frac{\Delta\omega}{2T} \right) \hat{G}_p^A(\Delta\omega) \right] (f_{pk}^>)_n (f_{pk}^>)_m \\
+ \left[ \left( 1 + \coth \frac{\Delta\omega}{2T} \right) \hat{G}_{p-q}^R(\Delta\omega) + \left( 1 - \coth \frac{\Delta\omega}{2T} \right) \hat{G}_{p-q}^A(\Delta\omega) \right] (f_{p-q,k}^>)_n (f_{p-q,k}^>)_m \right\}, \tag{16} \]

where \( \Delta\omega = \omega - \Omega_- \).

Using this expression, we calculate the sum over \( p \) in Eq. (13) for the cross section of the stimulated two-photon emission:
\[
\sum_{p \neq q/2} |(s_p)_{nn} e_n^* e_{m}^*|^2 \\
= \frac{1}{2} \sum_{p \neq q/2} \left\{ 2 \left[ \left( 1 + \coth \frac{\Delta \omega}{2T} \right) |\hat{G}_p^R(\Delta \omega)|^2 + \left( 1 - \coth \frac{\Delta \omega}{2T} \right) \text{Re} \left[ \hat{G}_p^R(\Delta \omega) \right]^2 \right] \cdot \right. \\
\left. \times \left( |(e^* f^>_{-pk})(e f^>_{pk})|^2 \right)^2 \right. \\
+ \left. \left[ \left( 1 + \coth \frac{\Delta \omega}{2T} \right) \hat{G}_p^R(\Delta \omega) + \left( 1 - \coth \frac{\Delta \omega}{2T} \right) \hat{G}_p^{R*}(\Delta \omega) \right] \cdot \\
\left. \times \left[ \left( 1 + \coth \frac{\Delta \omega}{2T} \right) \hat{G}_{p-q}^R(\Delta \omega) + \left( 1 - \coth \frac{\Delta \omega}{2T} \right) \hat{G}_{p-q}^{R*}(\Delta \omega) \right] \cdot \\
\left. \times (e^* f^>_{-pk})(e f^>_{pk})(e f^>_{-p\prime k})(e f^>_{p\prime k}) \right\} \cdot (17)
\]

In deriving this formula, we have taken into account the relation between the advanced and retarded Green’s functions on the real axis of \( \omega \): \( G_p^A(\omega) = G_p^{R*}(\omega) \).

Further calculation of the stimulated two-photon emission cross section (13) requires an expression for the retarded anomalous Green’s function of excitons at a finite temperature. It can be obtained through the analytical continuation of the anomalous Green’s function in the Matsubara representation to the upper half-plane of \( \omega \).

The anomalous Green’s function of a Bose system in the Matsubara representation is given by the following expression.\(^2\)

\[
\hat{G}_p(\omega) = -\frac{(1 - \delta_p) \Sigma_{\omega_p}^{02}}{(i \omega_s - \epsilon_0(p) + \mu - \Sigma_{\omega_s}^{11}) (i \omega_s + \epsilon_0(p) - \mu + \Sigma_{\omega_s}^{11}) + \Sigma_{\omega_s}^{20} \Sigma_{\omega_s}^{02}},
\]

where \( \omega_s = 2\pi sT \) and \( s \) is integer. Here \( \epsilon_0(p) = p^2/2m \) and \( \mu \) is the system chemical potential defined by the formula \( \mu = [\Sigma_{\omega_s}^{11} - \Sigma_{\omega_s}^{02}]|_{\omega_s=p=0} \).

At \( T \sim T_c \), where \( T_c \) is the Bose condensation temperature of an ideal Bose gas, the self-energy parts of a dilute Bose system with interparticle interaction can be expressed as follows.\(^2\)

\[
\Sigma_{\omega_s}^{11} = \frac{8\pi}{m} n \alpha, \quad \Sigma_{\omega_s}^{20} = \Sigma_{\omega_s}^{02} = \frac{4\pi}{m} n_0(T) \alpha,
\]

where \( n \) is the total density of particles, \( \alpha \) is the amplitude of their mutual scattering, \( n_0(T) \) is the total density of particles in the Bose condensate, which can be approximately calculated by the formula \( n_0(T) = n[1 - (T/T_c)^{3/2}] \).

Thus, the anomalous Green’s function for a dilute excitonic gas can be expressed as

\[
\hat{G}_p(\omega) = (1 - \delta_p) \frac{\zeta(T)}{\omega_s^2 + \epsilon_p^2},
\]

where
\[ \epsilon_p = \sqrt{\frac{\xi^2}{m} - \zeta^2(T)}, \quad \xi_p = \frac{\beta^2}{2m} + \zeta(T), \quad \zeta(T) = \mu(0) \left[ 1 - \left( \frac{T}{T_c} \right)^{3/2} \right], \quad \mu(0) = \frac{4\pi n a}{m}, \]

\( n \) is the density of excitons and \( m \) is the exciton mass. The parameter \( \mu(0) \) is the chemical potential of excitons at \( T = 0 \).

The analytical continuation of \( \hat{G}(\omega_n) \) to the upper half-plane yields the expression for the retarded anomalous Green’s function:

\[ \hat{G}^R_p(\omega) = -(1 - \delta_p) \frac{\zeta(T)}{(\omega - \epsilon_p + i\Gamma_p/2)(\omega + \epsilon_p + i\Gamma_p/2)}. \]  

(21)

Here \( \Gamma_p = \tau_p^{-1}, \tau_p \) is the lifetime of a quasiparticle with momentum \( p \) in the excitonic system.

By substituting Eq. (21) in (17), one can easily find that the main contribution to the cross section of the stimulated two-photon emission (13) at \( |\Delta \omega| \gg \Gamma_p \) is due to the summands in which \( \epsilon_p \sim |\Delta \omega| \). Therefore, matrix elements \( f_{p, q}^{>\alpha} \) and \( f_{p, q}^{<\alpha} \) can be replaced by their values corresponding to the momentum \( p_L \) that satisfies the condition \( \epsilon(p_L) = \Delta \omega \) and carried out of the integrand. Moreover, if the \( p_L \gg q \) is fulfilled, one can set \( q = 0 \) in the sum over \( p \) in Eq. (17). Thus, we have

\[ \sum_p |(s_p)_{nm} \epsilon^*_n \epsilon_m^*|^2 = 2 \left[ (1 + \coth^2 \frac{\Delta \omega}{2T}) \sum_p |\hat{G}^R_p(\Delta \omega)|^2 
+ (1 - \coth^2 \frac{\Delta \omega}{2T}) \sum_p \text{Re}[\hat{G}^R_p(\Delta \omega)]^2 \right] |f_n(\omega'_L)f_m(\omega_L)\epsilon^*_n \epsilon_m^*|^2; \]  

(22)

where

\[ f(\omega_L) = \frac{1}{4\pi} \int f^{>}(p_L, k)d\omega_L, \quad f(\omega'_L) = \frac{1}{4\pi} \int f^{>}(p_L, k')d\omega_L \]

are the matrix elements averaged over the directions of vector \( p_L \).

Replacing the summation in Eq. (22) by the integration on \( p \), we obtain

\[ \sum_p |\hat{G}^R_p(\Delta \omega)|^2 = \int_0^\infty \frac{d^3 p}{(2\pi)^3} \frac{\zeta^2(T)}{|(\Delta \omega + i\Gamma_p/2) - \epsilon_p^2|^2}. \]  

(23)

This integral diverges as \( \Gamma_p \to 0 \). Replacing it as a sum of two integrals each of which converges at \( \Gamma_p \to 0 \) and replacing \( p \) integration by the integration on \( t = \xi_p/\zeta(T) \) yields

\[ \sum_p |\hat{G}^R_p(\Delta \omega)|^2 = \sqrt{\frac{2m^3}{\zeta^2(T)}} \left[ \int_1^\infty \frac{dt\sqrt{t - 1}}{t^2 - \beta_+^2} - \int_1^\infty \frac{dt\sqrt{t - 1}}{t^2 - \beta_-^2} \right], \]  

(24)

where

\[ \beta_\pm = (\alpha_L \pm i\gamma_L)^2 + 1, \quad \alpha_L = \frac{|\Delta \omega|}{\zeta(T)}, \quad \gamma_L = \frac{\Gamma_L}{2\zeta(T)}, \quad \Gamma_L = \Gamma_{pL}. \]
Thus, in calculating the integrals on the right of the resulting equation, one can set \( \beta^2 = \beta^2 \pm i\delta \). As a result, we obtain

\[
\sum_{p} |\hat{G}^R_p(\Delta \omega)|^2 = \frac{\sqrt{2m^3\zeta(T)(\sqrt{\alpha_L^2 + 1} - 1)}}{4\pi\alpha_L \Gamma_L \sqrt{\alpha_L^2 + 1}}.
\] (25)

The second sum over \( p \) in Eq. (22) converges even as \( \Gamma_p \to 0 \). Therefore, if \( |\Delta \omega| \gg \Gamma_p \), we can set in this sum \( \Gamma_p = 0+ \). In this case, we have

\[
\text{Re} \sum_{p} [\hat{G}^R_p(\Delta \omega)]^2 = -\frac{\sqrt{2m^3/\zeta(T)}}{16\pi} \frac{\sqrt{\alpha_L^2 + 1} - 1}{\alpha_L \sqrt{\alpha_L^2 + 1}^3} \left(1 + \coth^2 \frac{\Delta \omega}{2T}\right)
\times |f_n(\omega'_L)f_m(\omega_L)e_{m'}^*e_{n'}^*|^2.
\] (26)

It is clear that for \( |\Delta \omega| \gg \Gamma_p \) the following relation takes place:

\[
\left| \sum_{p} \text{Re} [\hat{G}^R_p(\Delta \omega)]^2 \right| \ll \sum_{p} |\hat{G}^R_p(\Delta \omega)|^2.
\]

Thus,

\[
\sum_{p} |(s_p)_{nn'}e_{n'}^*e_{m'}|^2 = \frac{\sqrt{2m^3\zeta(T)}}{2\pi\alpha_L \Gamma_L \sqrt{\alpha_L^2 + 1}} \left(1 + \coth^2 \frac{\Delta \omega}{2T}\right)
\times |f_n(\omega'_L)f_m(\omega_L)e_{m'}^*e_{n'}|^2.
\] (27)

By substituting this expression in the formula for the differential cross section (13), we obtain

\[
d\sigma^L = \frac{\omega(2\Omega_+ - \omega)^3}{4\pi c^4} \frac{\sqrt{2m^3\zeta(T)}}{\sqrt{\alpha_L^2 + 1} - 1} \left(1 + \coth^2 \frac{\Delta \omega}{2T}\right)
\times |f_n(\omega'_L)f_m(\omega_L)e_{m'}^*e_{n'}|^2 d\omega'.
\] (28)

If the exciton-phonon system is isotropic, and the incident light is monochromatic and has a linear polarization, one has \( |e_{m'}^*f_m(\omega_L)|^2 = f^2(\omega_L)/3 \). Summing over the polarizations of photon \( \omega' \) and integrating over the directions of its momentum (note that in the case of stimulated two-photon emission the photon \( \omega \) is identical to the incident one), we obtain the total cross section of the stimulated two-photon emission corresponding to the coherent phonon-assisted two-exciton recombination:

\[
\sigma^L(\omega, T) = \frac{\omega(2\Omega_+ - \omega)^3}{c^4} \frac{\sqrt{8m^3\zeta(T)}}{9\alpha_L \Gamma_L \sqrt{\alpha_L^2 + 1}} \left(1 + \coth^2 \frac{\Delta \omega}{2T}\right) f^2(\omega_L)f^2(\omega'_L).
\] (29)
It should be noted that, if the conditions $\Delta \omega \ll \Omega_\perp$, $\mu(0) \ll \Omega$, and $\tau^L = \text{const}$ are fulfilled, then, at a given ratio between the exciton chemical potential $\mu(0)$ at zero temperature and twice the temperature of their Bose condensation, $\gamma = \mu(0)/2T_c$, the parameter $\sigma^L(\Delta \omega, T)/\sigma^L(0, 0)$ is uniquely determined by two quantities, $x = \Delta \omega/2T_c$ and $y = T/T_c$:

$$\frac{\sigma^L(\Delta \omega, T)}{\sigma^L(0, 0)} = \frac{z^2 \sqrt{x^2 + z^2} - z}{|x| \sqrt{2\gamma(x^2 + z^2)}} \left(1 + \coth^2\frac{x}{y}\right),$$  

(30)

where $z = \gamma(1 - y^{3/2})$.

The dependence of the cross section (29) on frequency (strictly speaking, on the difference between the incident light frequency $\omega$ and $\Omega_\perp$) is shown in Fig. 2a for different temperatures of the excitonic subsystem. This cross section as a function of temperature at different fixed values of the difference $\Delta \omega = \omega - \Omega_\perp$ is shown in Fig. 2b. All the curves in Fig. 2 correspond to $\gamma = 0.3$, and it is assumed that $\tau^L = \text{const}$. It is clear that at $|\Delta \omega| \ll T_c$ and $T < T_c$ there is a temperature interval where the cross section (29) of the stimulated two-photon emission is a nonmonotonic function of temperature: $\sigma^L$ increases with the growth of temperature and can even become larger than it is at $T = 0$.

The reason for this unusual temperature dependence is the following. The cross section (29) of the stimulated two-photon emission is determined by two quantities that depend on the temperature differently, namely, by $\zeta(T)$, proportional to the number of excitons in the condensate, and by occupation numbers of quasiparticle levels of the excitonic system with the quasiparticle energy $\epsilon(p_L) = |\Delta \omega|$. Really, the density of condensate and, hence, $\zeta(T)$ decreases as the temperature increases. It leads, in turn, to a decrease in the cross section (29). On the other hand, using Bogoliubov’s $u - v$ transforms, one can easily show that the coherent two-exciton recombination, which is a second-order process with respect to Hamiltonian (3), proceeds via intermediate states of the excitonic system containing one particle more (less) than the state of thermodynamic equilibrium (see also Refs. 19 and 20). The cross section of the stimulated two-photon emission corresponding to the coherent two-exciton recombination is proportional to

$$(n_{p_L} + 1)^2 + n_{p_L}^2 = \frac{1}{2} \left(1 + \coth^2\frac{\Delta \omega}{2T}\right),$$

where $n_{p_L} = [\exp(\epsilon_{p_L}/T) - 1]^{-1}$ is the occupation number of the quasiparticle state with energy $\epsilon(p_L) = |\Delta \omega|$ in the excitonic system. As the temperature increases, $n_p$ also rises, which leads to a larger cross section (29). If this tendency dominates, the cross section of the stimulated two-photon emission corresponding to the coherent two-exciton recombination
should increase with the temperature. Of course, the tendency to decrease the cross section should overcome sooner or later as $T \to T_c$, since it must turn to zero at $T = T_c$.

Note that the temperature dependence of the cross section (29) of the stimulated two-photon emission accompanied by the coherent two-exciton recombination has been calculated in the approximation (19), which is correct only in a narrow temperature interval about the Bose condensation temperature $T_c$, which is considered to be equal to Bose condensation temperature in an ideal Bose gas. Although this approximation allows one to reproduce formally our results$^{19}$ for $T = 0$, in the intermediate temperature interval the curve of the cross section $\sigma^L(\Delta\omega, T)$ versus temperature should be different from that plotted in Fig. 2. Nonetheless, the conclusion about the nonmonotonic temperature dependence of the cross section of the stimulated two-photon emission is valid. For example, at $\Delta\omega/2T_c = 0.2$ we have $\sigma^L(\Delta\omega, T) > \sigma^L(\Delta\omega, 0)$ even for $T_c - T \ll T_c$ (Fig. 2b), where the approximation (19) is correct.

3. Raman light scattering

The coherent two-exciton recombination can be revealed not only in the stimulated two-photon emission but also in the Raman light scattering (RLS). Abrikosov and Falkovsky$^{25}$ analyzed RLS in a superconductor, whose analogue in a semiconductor was RLS by a dense electron-hole plasma with coupling between electrons and holes (a phase transition in this system was studied by Keldysh and Kopaev$^{26}$, see also the review by Kopaev$^{27}$ and references therein). But we are discussing the case of a low density of electrons and holes (excitonic gas). Moreover, it is essential for the case of RLS under consideration that the electron-hole system is not in equilibrium, because this is the situation when the coherent two-exciton recombination (generation) leading to a transition of an exciton system to a state with a lower (higher) energy is possible. In the case of such RLS with the transfer of the recoil momentum to two generated optical phonons the energy conservation is described by the formula

$$\omega + 2\Omega_0 = \omega'$$

(31)

The case considered here corresponds to the appearance of an anti-Stokes RLS component at frequency $\omega'$ defined by this formula.

In addition, an RLS process with the coherent two-exciton generation is also possible,
and the energy conservation in this case is described by the equation\(^\text{4}\)

\[
\omega - 2\Omega_\omega = \omega'.
\]  

(32)

This formula determines the frequency \(\omega'\) of the Stokes component corresponding to this Raman scattering. It is clear that RLS with the coherent phonon-assisted two-exciton generation is possible only when \(\omega > 2\Omega_\omega\). The stimulated two-photon emission corresponding to the coherent phonon-assisted two-exciton recombination is impossible in this case.

The analysis of RLS accompanied by the coherent two-exciton recombination (or generation) is similar to that of the stimulated two-photon emission with the coherent two-exciton recombination. Since the formulas for the cross section of RLS with the coherent two-exciton recombination or generation are cumbersome, here we only indicate how these formulas can be derived from Eq. (29) using appropriate substitutions.

1. The cross section of RLS accompanied by the coherent phonon-assisted two-exciton recombination is obtained by replacing some variables in Eq. (29):

\[ f(\omega_L) \rightarrow f'(\tilde{\omega}_L), \quad f(\omega'_L) \rightarrow f'(\tilde{\omega}'_L), \quad \omega \rightarrow -\omega, \quad \Delta \omega \rightarrow \omega + \Omega_\omega, \quad \alpha_L \rightarrow \tilde{\alpha}_L, \quad \Gamma_L \rightarrow \tilde{\Gamma}_L.\]

Here \(\tilde{\alpha}_L = (\Omega_\omega + \omega)/\zeta(T)\), \(\tilde{\Gamma}_L\) is the reciprocal lifetime of a quasiparticle with energy \(\epsilon(\tilde{\rho}_L) = \Omega_\omega + \omega\) in the excitonic system,

\[ f'(\tilde{\omega}_L) = \frac{1}{4\pi} \int f'^>(\tilde{\rho}_L, k) d\tilde{\rho}_L, \quad f'(\tilde{\omega}'_L) = \frac{1}{4\pi} \int f'^>(\tilde{\rho}_L, k') d\tilde{\rho}_L.\]

2. The cross section of RLS due to the coherent phonon-assisted two-exciton generation \((\omega > 2\Omega_\omega)\) is derived from Eq. (29) by substituting

\[ f(\omega_L) \rightarrow f'(\omega_L), \quad 2\Omega_\omega - \omega \rightarrow \omega - 2\Omega_\omega, \]

where

\[ f'(\omega_L) = \frac{1}{4\pi} \int f'^>(\rho_L, k) d\rho_L.\]

\(^4\)In a general case, RLS, like the two-photon emission, can involve an arbitrary number of phonons. Moreover, the recoil momentum (entirely or partially) can be transferred to impurities. Thus, RLS accompanied by the coherent two-exciton generation or recombination can lead to the appearance of anti-Stokes and Stokes components at frequencies \(\omega + (2\Omega - n\omega_0) = \omega'\) and \(\omega - (2\Omega - n\omega_0) = \omega'\), respectively, where \(n\) is an arbitrary integer (see also Ref. 20).
4. Possibility of experimental detection of two-photon processes accompanied by coherent two-exciton recombination

Let us analyze the possibility of the experimental detection of the stimulated two-photon emission and RLS accompanied by the coherent phonon-assisted two-exciton recombination. First we consider the stimulated two-photon emission.

The light intensity \( I_L^{(\omega')} \) at frequency \( \omega' = 2\Omega_\omega - \omega \) resulting from the stimulated two-photon emission with transfer of the recoil momentum to generated optical phonons is given by the expression

\[
I_L^{(\omega')} = \frac{\omega'}{\omega} \sigma_L^{(\omega)} I(\omega),
\]

where \( \sigma_L^{(\omega)} \) is the cross section of this process (Eq. (29)), \( I(\omega) \) (W/cm\(^2\)) is the intensity of incident light of frequency \( \omega \).

The intensity (33) can be expressed as a sum of two terms:

\[
I_L^{(\omega')} = \Delta I_L^{(\omega')} + \bar{I}_L^{(\omega')},
\]

where \( \bar{I}_L^{(\omega')} \) is the intensity of the two-photon emission resulting from two consecutive processes: the spontaneous emission at frequency \( \omega' = 2\Omega_\omega - \omega \) and the subsequent stimulated emission at frequency \( \omega \), each of which satisfies the energy conservation law.

If the incident light frequency \( \omega > \Omega_- \), then \( \omega' < \Omega_- \). In this case, the spontaneous emission at frequency \( \omega' = 2\Omega_\omega - \omega \) is due to the excitonic recombination with generation of a Bogoliubov quasiparticle with momentum \( \mathbf{p}_L \) that satisfies the condition \( \epsilon(p_L) = \Delta\omega \) (see Appendix B, \( \Delta\omega = -\Delta\omega' \)). The spontaneous recombination of excitons generates in the excitonic system \( I_s^{(\omega')} \) quasiparticles with energy \( \epsilon(p_L) = \Delta\omega \) per unit time, where \( I_s^{(\omega')} \) is the luminescence intensity (57) (see Appendix B and Ref. 28). These quasiparticles disappear in the time of order of \( \tau^L \). The disappearance of some of these quasiparticles is accompanied by the stimulated recombination of excitons and the induced emission of light at frequency \( \omega \). Thus, at \( \omega > \Omega_- \), the intensity \( \bar{I}(\omega') \) is given by the relation

\[
\bar{I}_L^{(\omega')} = \frac{\tau_{r}^L}{\tau_{r'}^L} I_s^{(\omega')},
\]

where \( \tau_{r}^L \) is the lifetime of the quasiparticle with energy \( \epsilon(p_L) \) due to its recombination, which leads to the stimulated emission at frequency \( \omega \), provided that the excitonic system contains one quasiparticle with momentum \( \mathbf{p}_L \) more than it does in the state of thermodynamic equilibrium. The time \( \tau_{r}^L \) can be easily calculated using Fermi’s ‘golden rule’.
\[
\frac{1}{\tau^L_r} = \frac{(2\pi)^2}{3c} f^2(\omega_L) u^2_{pL} (n_{pL} + 1) I(\omega),
\]
\[
u^2_{pL} = \frac{1}{2} \left( \frac{\sqrt{\alpha^2_L + 1}}{\alpha_L} + 1 \right), \quad n_{pL} = \frac{1}{e^{\Delta \omega/T} - 1},
\]
where \(u_{pL}\) is Bogoliubov’s coefficient and \(n_{pL}\) is the distribution function of quasiparticles with energy \(\epsilon(p_L) = \Delta \omega\) at temperature \(T\).

If the incident light frequency \(\omega < \Omega_−\) and, hence, \(\omega' > \Omega_−\), the situation is similar to that discussed above. In this case, the spontaneous emission at frequency \(\omega'\) is due to the recombination of an exciton accompanied by the disappearance of one Bogoliubov quasiparticle with energy \(\epsilon(p_L) = -\Delta \omega\) in the excitonic system. At \(\omega' > \Omega_−\), the number of quasiparticles of energy \(\epsilon(p_L) = -\Delta \omega\) that disappear per unit time as a result of spontaneous recombination of excitons is \(I^L_s(\omega')/\omega'\). In the time of order of \(\tau^L\), the disappeared quasiparticles are replaced by new ones, and the appearance of some of them is accompanied by the stimulated radiation at frequency \(\omega\). Thus, at \(\omega < \Omega_−\) we have
\[
\tilde{I}^L(\omega') = \frac{\tau^L}{\tau^L_c} I^L_s(\omega'),
\]
where \(\tau^L_c\) is the lifetime of an exciton with momentum \(p_L\) with respect to the stimulated recombination, which results in both a stimulated emission at frequency \(\omega\) and generation of a quasiparticle of energy \(\epsilon(p_L) = -\Delta \omega\), provided that the excitonic system contains one quasiparticle with momentum \(p_L\) less than it does in the state of thermodynamic equilibrium.

Using Fermi’s ‘golden rule’ yields the following expression for \(\tau^L_c\):
\[
\frac{1}{\tau^L_c} = \frac{(2\pi)^2}{3c} f^2(\omega_L) u^2_{pL} n_{pL} I(\omega),
\]
\[
u^2_{pL} = \frac{1}{2} \left( \frac{\sqrt{\alpha^2_L + 1}}{\alpha_L} - 1 \right).
\]

Using Eqs. (35)-(38) and (57) from Appendix B, we obtain the intensity \(\tilde{I}^L(\omega')\) in a general case:
\[
\tilde{I}^L(2\Omega_− - \omega) = \frac{2\Omega_− - \omega}{\omega} \tilde{\sigma}^L(\omega) I(\omega),
\]
\[
\tilde{\sigma}^L(\omega) = \tau^L \frac{\omega(2\Omega_− - \omega)^3}{c^4} \sqrt{\frac{2m^3}{\zeta(T)}} \left( \sqrt{\frac{\alpha^2_L + 1}{\alpha_L^2} - 1} \right) f^2(\omega_L) f^2(\omega'_L)
\]
\[
\times \left[ \text{sign}(\Delta \omega) + \coth \frac{\Delta \omega}{2T} \right]^2.
\]
The spectral line at frequency $\omega' = 2\Omega_+ - \omega$ due to the stimulated two-photon emission accompanied by the coherent phonon-assisted two-exciton recombination will be observed against the background of the luminescence spectrum of Bose-condensed excitons. It clearly follows from Eqs. (35) and (37) that $\tilde{I}^L(\omega')$ determines a fraction of the spontaneous emission intensity $I_s^L(\omega')$. Thus, the total intensity of the emission at frequency $\omega'$ can be expressed as follows:

$$I_{\text{tot}}^L(\omega') = \Delta I^L(\omega') + I_s^L(\omega'),$$

(40)

where $I_{\text{tot}}^L(\omega')$ is the luminescence intensity at frequency $\omega'$ in the absence of the incident light of frequency $\omega$, $\Delta I^L(\omega')$ is the observed light intensity at frequency $\omega'$ due to the stimulated two-photon emission with the coherent two-exciton recombination. By substituting the cross section (29) in Eq. (33) and using Eqs. (34) and (39), we obtain the observed light intensity $\Delta I^L(\omega')$:

$$\Delta I^L(2\Omega_+ - \omega) = \frac{2\Omega_+ - \omega}{\omega} \Delta \sigma^L(\omega) I(\omega),$$

$$\Delta \sigma^L(\omega) = \tau L \frac{\omega(2\Omega_+ - \omega)^3}{c^4} \sqrt{8m^3\zeta(T)\left(\sqrt{\alpha_L^2 + 1} - 1\right)} \frac{f^2(\omega_L)f^2(\omega'_L)}{9\alpha_L\sqrt{\alpha_L^2 + 1}}$$

$$\times \left[1 + \coth^2 \frac{\Delta \omega}{2T} - \frac{1}{4} \left(\text{sign}(\Delta \omega) + \coth \frac{|\Delta \omega|}{2T}\right)^2\right].$$

(41)

One can easily prove that $1/2 \leq \Delta \sigma^L(\omega)/\sigma^L \leq 1$. In particular, at $T = 0$ we have $\Delta \sigma^L(\omega) = \sigma^L(\omega)$ at $\omega < \Omega_+$ and $\Delta \sigma^L(\omega) = \sigma^L(\omega)/2$ at $\omega > \Omega_+$.

Using Eq. (29), we can estimate the cross section $\sigma^L$ of the stimulated two-photon emission. In CGS units this expression has the form

$$\sigma^L(\omega, T) = \tau^L V \frac{\omega(2\Omega_+ - \omega)^3}{9c^4\hbar^4} \sqrt{8m^3\zeta(T)\left(\sqrt{\alpha_L^2 + 1} - 1\right)}$$

$$\times \left[1 + \coth^2 \frac{\Delta \omega}{2T}\right] f^2(\omega_L)f^2(\omega'_L),$$

(42)

where $V$ is the volume of the excitonic system exposed to the incident light and $\alpha_L = \hbar|\Delta \omega|/\zeta(T)$.

We shall consider as an example a system of Bose-condensed excitons in Cu$_2$O at zero temperature. The exciton effective mass in this crystal is $m = 2.7m_e$, the characteristic exciton radius is equal to $a = 7 \text{ Å}$, and the photon energy corresponding to the recombination of an exciton with zero momentum is $\hbar\Omega \simeq 2 \text{ eV}$. The optical recombination of an exciton
in Cu$_2$O is typically assisted by generation of an optical phonon of energy $\hbar \omega_0^s \simeq 10$ meV with a negligible dispersion.

Let us estimate the exciton chemical potential at $T = 0$ by the formula

$$\mu(0) = \frac{4\pi \hbar^2}{m} na,$$

where $n$ is the exciton density. Assuming that $n = 10^{19}$ cm$^{-3}$ (this density was achieved in some experiments), we obtain $\mu(0) \simeq 2.5$ meV. An ideal gas of excitons with $n = 10^{19}$ cm$^{-3}$ should transform to the Bose-condensed state at $T_c \sim 50$ K, in this case $\mu(0)/2T_c \simeq 0.3$.

In the experiment excitons were generated by powerful nanosecond laser pulses at a wavelength $\lambda \simeq 500$ nm focused into the spot of diameter $d \simeq 30 \mu$m on the sample surface. The volume of the excitonic system interacting with the incident light stimulating the two-photon emission can be estimated as $V = d^2 l$, where $l \simeq 1 \mu$m is the penetration depth of radiation with wavelength 500 nm.

As $\omega \to \Omega_-$ ($\alpha_L \to 0$), the cross section $\sigma^L$ increases. Let us suppose that $\hbar(\Omega_- - \omega) = \mu(0)$. In this case

$$f(\omega_L) \simeq f(\omega'_L) \simeq F, \quad F = \frac{1}{4\pi} \int F^>(p_L, k) d\rho_{p_L},$$

where $F$ is the matrix element of the radiative phonon-assisted recombination of an isolated exciton. This matrix element can be estimated by the formula

$$\frac{1}{\tau_{exc}} = \frac{4\Omega^3}{3\varepsilon^3 \hbar} F^2,$$

where $\tau_{exc}$ is the lifetime of an isolated exciton due to its spontaneous recombination with the emission of a photon of energy $\hbar \Omega_-$ and an optical phonon with energy $\hbar \omega_0^s$. The lifetime of para-excitons in Cu$_2$O is $\tau_{exc} \sim 100$ $\mu$s.

The relaxation time $\tau^L$ in the system of Bose-condensed excitons is a subject of further investigation. Even at zero temperature, it can be considerably shorter than the radiative lifetime of excitons $\tau_{exc}$, because a quasiparticle can disappear, for example, due to the emission of one or several acoustic phonons. Assuming that the time $\tau^L$ is within the interval of $10^{-11}$–$10^{-5}$ s (the lower bound is defined by the condition $\Gamma_L = 10^{-1}\epsilon(p_L)$, the upper bound is $10^{-1}\tau_{exc}$), we obtain an estimate for the cross section of the stimulated two-photon emission by Bose-condensed para-excitons in Cu$_2$O at $T = 0$: $\sigma^L = 10^{-16}$–$10^{-10}$ cm$^2$.

The radiative lifetime of ortho-excitons in Cu$_2$O is $\tau_{exc} \sim 300$ ns. Assuming the relaxation time $\tau^L$ in a system of ortho-excitons in the Bose-condensed state is within $10^{-11}$–$10^{-9}$ s (in this case the upper bound is determined by the time of transition between the
ortho-exciton and para-exciton states), yields $\sigma^L = 10^{-11} - 10^{-9}$ cm$^2$ at $T = 0$. Thus, the stimulated two-photon emission accompanied by the coherent two-exciton recombination can be experimentally detected in Cu$_2$O.

The cross section of RLS with the coherent two-exciton recombination accompanied by the generation of two optical phonons is determined by the squared product of two matrix elements:

$$f'(\tilde{\omega}_L) = \frac{1}{4\pi} \int f^{\rangle}(\tilde{p}_L, k)d\tilde{p}_L, \quad f(\tilde{\omega}_L') = \frac{1}{4\pi} \int f^{\rangle}(\tilde{p}_L, k')d\tilde{p}_L,$$

where $\tilde{p}_L$ is determined by the condition $\epsilon(\tilde{p}_L) = \omega + \Omega_-$ (see Section 3). The band gap in Cu$_2$O is wide ($\Omega_- \sim 10^2 \omega_0^0$), therefore $\epsilon(\tilde{p}_L) \gg \omega_0^0$. Using the approach suggested in Appendix A, one can prove that in this case $f(\tilde{\omega}_L')$ and $f'(\tilde{\omega}_L)$ are negligible in comparison with the matrix elements $f(\omega_L)$ and $f(\omega'_L)$ in Eq. (42) at $|\Omega_- - \omega| \sim \mu(0)$. Moreover, the cross section of the RLS under consideration is proportional to the lifetime of a quasiparticle with energy $\epsilon(\tilde{p}_L) = \omega + \Omega_-$, which is essentially shorter than the relaxation time $\tau^L$ in the cross section (42) at $|\Omega_- - \omega| \sim \mu(0)$. Thus, unlike the stimulated two-photon emission, one can hardly detect RLS accompanied by the coherent two-exciton recombination in Cu$_2$O. The situation is similar in the case of RLS with the coherent two-exciton generation.

5. Conclusions

In this paper, we have demonstrated that the coherent two-exciton recombination, i.e., the simultaneous recombination of two excitons with opposite momenta corresponding to the existence of nondiagonal long-range order in the system expressed by nonvanishing anomalous averages of the form $\langle N - 2|Q_{-p}Q_{p}|N\rangle$, is possible in Bose-condensed excitonic system interacting with the electromagnetic field. Similarly, the coherent two-exciton generation corresponding to anomalous averages like $\langle N - 2|Q_{-p}^+Q_{p}^+|N\rangle$ is also possible. In these processes, the exciton occupation numbers are unchanged, and the final and state of the excitonic system differs from the initial one only in the average number of excitons with zero momentum. The coherent two-exciton recombination may also lead to Raman light scattering by excitons in Bose-condensed state (RLS can also be accompanied by the coherent two-exciton generation). The recoil momentum corresponding to the change in the momentum of electromagnetic field is transferred to phonons or impurities. Both the stimulated two-photon emission and RLS with the coherent two-exciton recombination (generation) can occur only in the presence of Bose condensate in a system of interacting excitons, therefore, the observation of these effects can be used as a strong experimental evidence of the existence
of Bose condensation in excitonic systems.

Using the diagrammatic methods, we have developed a technique for calculating the cross sections of the stimulated two-photon emission and RLS accompanied by the coherent two-exciton recombination (or generation) at $T > 0$. In this approach, the elements of the scattering matrix corresponding to the processes in question are expressed in a natural manner in terms of Green’s functions of Bose-condensed excitons (see Eqs. (9), (10), and also (49)).

If the incident light frequency $\omega < 2\Omega_-$, where $\Omega_- = \Omega - \omega_0^s$ ($\Omega$ is the frequency of light due to the recombination of an exciton with zero momentum and $\omega_0^s$ is the optical phonon frequency), the stimulated two-photon emission and RLS accompanied by the coherent phonon-assisted two-exciton recombination result in the appearance of a spectral line at frequency $2\Omega_- - \omega$ and the anti-Stokes component at $\omega + 2\Omega_-$, respectively. At $\omega > 2\Omega_-$ the RLS spectrum contains both the anti-Stokes and Stokes components at frequencies $\omega \pm 2\Omega_-$. The anti-Stokes line corresponds due to the coherent two-exciton recombination, whereas the Stokes component is due to the coherent phonon-assisted two-exciton generation. In this case, the stimulated two-photon emission is impossible.

Using approximation (19), we have derived expressions for the cross sections of the processes under consideration at finite temperatures. If $|\omega - \Omega_-| \ll T_c$ ($T_c$ is the temperature of Bose condensation), the cross section of the stimulated two-photon emission is a nonmonotonic function of temperature. It increases in a certain temperature interval below $T_c$ and can even be larger than it is at $T = 0$. The cause of this nonmonotonic behavior is that the cross section of the stimulated two-photon emission is determined not only by the density of excitons in the condensate, which decreases as the temperature increases and vanishes at $T = T_c$, but also by the occupation numbers of quasiparticles with energies $|\omega - \Omega_-|$ in the excitonic system, which increases as the temperature grows.

Our estimates indicate that, at $|\omega - \Omega_-| \sim \mu(0)$, where $\mu(0)$ is the exciton chemical potential measured with respect to the excitonic band bottom, a spectral line at $2\Omega_- - \omega$ due to the stimulated two-photon emission accompanied by the coherent optical phonon-assisted two-exciton recombination can be experimentally observed in Cu$_2$O.

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Appendix A
Effective matrix elements of excitonic recombination

The objective of this Appendix is to prove that the two-photon emission and RLS accompanied by the coherent two-exciton recombination can be analyzed on the base of first principles, without using the effective Hamiltonian (3). Taking as an example the two-photon emission, we will determine conditions when the analysis based on the effective Hamiltonian (3) is correct. In addition, we will show that the effective matrix elements of excitonic recombination used in this paper do not depend on temperature and equal to those calculated previously for \( T = 0 \).

The Hamiltonian describing the interaction among excitons, phonons, and electromagnetic field can be written as

\[
\hat{V}(t) = \hat{W}(t) + \hat{D}(t),
\]

\[
\hat{W}(t) = \sum_{pq} \left[ W_{pq} Q^+_{q}(t) Q_p(t) b_{q-p}(t) + W^*_{pq} Q^+_{p}(t) Q_q(t) b_{p-q}^+(t) \right],
\]

\[
\hat{D}(t) = \sum_q \left[ D_q e^{-i\Omega t} Q_q(t) c_q^+(t) + D_q^* e^{i\Omega t} Q^\ast q(t) c_q(t) + \text{H.c.} \right],
\]

where Hamiltonian \( \hat{W}(t) \) describes the scattering of excitons by phonons, \( \hat{D}(t) \) is responsible for the interaction between excitons and the electromagnetic field, \( D_q = i\sqrt{2\pi\omega_q} e^* d_q, \) \( D_q^* = -i\sqrt{2\pi\omega_q} e d_q \).

It is clear that the two-photon emission accompanied by the coherent phonon-assisted two-exciton recombination is a process of the fourth order in Hamiltonian \( \hat{V}(t) \). For the element of the \( S \)-matrix of the two-photon emission due to the coherent two-exciton recombination and generation of two optical phonons with momenta \( p - k \) and \( -p - k' \) averaged with the Gibbs distribution, we have

\[
(S_p)_{fi} = \frac{(-i)^4}{4!} \int_{-\infty}^{\infty} \ldots \int \langle f | T_t [\hat{W}(t_1)\hat{W}(t_2)\hat{D}(t_3)\hat{D}(t_4) \nonumber
\]
\[
+ \hat{W}(t_1)\hat{D}(t_2)\hat{W}(t_3)\hat{D}(t_4) + \hat{D}(t_1)\hat{W}(t_2)\hat{W}(t_3)\hat{D}(t_4) + \hat{W}(t_1)\hat{D}(t_2)\hat{D}(t_3)\hat{W}(t_4) \nonumber
\]
\[
+ \hat{D}(t_1)\hat{W}(t_2)\hat{D}(t_3)\hat{W}(t_4) + \hat{D}(t_1)\hat{D}(t_2)\hat{W}(t_3)\hat{W}(t_4) ] |i\rangle dt_1 \ldots dt_4. \tag{45}
\]

Here \( \langle f | \ldots | i \rangle = \sum_n \exp[(F - E_n(N) + \mu N)/T] \langle m | \ldots | n \rangle \), where \( |n\rangle = |n, N\rangle_{\text{exc}} |i\rangle_{\text{phon}} |\ell\rangle_{\text{phot}} \) and \( |m\rangle = |n, N-2\rangle_{\text{exc}} |f\rangle_{\text{phon}} |\ell\rangle_{\text{phot}} \), and the other notations are given in Section 2.

By changing the time variables in each summand of Eq. (45) we can transform it to

\[
(S_p)_{fi} = \frac{1}{4!} \int_{-\infty}^{\infty} \ldots \int \langle f | T_t [\hat{W}(t_1)\hat{W}(t_2)\hat{D}(t_3)\hat{D}(t_4) |i\rangle dt_1 \ldots dt_4, \tag{46}
\]
where

\[
\langle f | T_t \hat{W}(t_1) \hat{W}(t_2) \hat{D}(t_3) \hat{D}(t_4) | i \rangle = D_k D_{k'} \exp \left[ -i \Omega(t_3 + t_4) \right] \sum_{p_1 p_2} W_{p_1 p_1 + p + k} W_{p_2 p_2 - p + k}^* \\
\times \left\{ \left[ (T_t Q^+_{p_1 + p + k'}(t_1) Q_{p_1}(t_1) Q^+_{p_2 - p + k}(t_2) Q_{p_2}(t_2) Q_k(t_3) Q_{k'}(t_4)) \right] \times \\
\times \left\{ (f | T_t b_{-p - k'}(t_1) b^+_{-p - k}(t_2) | i \rangle_{\text{phon}} \\
+ \langle T_t Q^+_{p_2 - p + k}(t_1) Q_{p_2}(t_1) Q^+_{p_1 + p + k'}(t_2) Q_{p_1}(t_2) Q_k(t_3) Q_{k'}(t_4) \rangle \\
\times \langle f | T_t b^+_{-p - k}(t_1) b^+_{-p - k'}(t_2) | i \rangle_{\text{phon}} \langle f | T_t c_{k'}^+(t_3) c_k^+(t_4) | i \rangle_{\text{phot}} \\
\times \left\{ (T_t Q^+_{p_1 + p + k'}(t_1) Q_{p_1}(t_1) Q^+_{p_2 - p + k}(t_2) Q_{p_2}(t_2) Q_k(t_3) Q_{k'}(t_4)) \right\} \right\}. \tag{47}
\]

Here \( \langle \ldots \rangle = \sum_n \exp[(F - E_n(N) + \mu N)/T] \langle n, N - 2 | \ldots | n, N \rangle_{\text{exc}}. \)

In the most interesting case \( k \neq k' \), we have

\[
\sum_{p_1 p_2} W^*_{p_1 p_1 + p + k} W^*_{p_2 p_2 - p + k} \left\langle T_t Q^+_{p_1 + p + k'}(t_1) Q_{p_1}(t_1) Q^+_{p_2 - p + k}(t_2) Q_{p_2}(t_2) Q_k(t_3) Q_{k'}(t_4) \right\rangle \\
= \sum_{p_1} W^*_{p_1 p_1} \left[ W^*_{-k'k'} \langle T_t Q^+_{p_1}(t_1) Q_{p_1}(t_1) \rangle \langle T_t Q_{-k'}^+(t_2) Q_k(t_3) \rangle \langle T_t Q_{-k}^-(t_2) Q_{k'}(t_4) \rangle \\
+ W^*_{-k'k} \langle T_t Q^+_{p_1}(t_1) Q_{p_1}(t_1) \rangle \langle T_t Q_{-k'}^+(t_2) Q_{k'}(t_4) \rangle \langle T_t Q_{-k}^-(t_2) Q_k(t_3) \rangle \delta(p + k') \right] \\
+ \sum_{p_2} W^*_{p_2 p_2} \left[ W^*_{-k'k} \langle T_t Q_{-k}^+(t_1) Q_{k}(t_3) \rangle \langle T_t Q_{-k'}^-(t_2) Q_{k'}(t_4) \rangle \langle T_t Q_{p_2}^+(t_2) Q_{p_2}(t_2) \rangle \right] \delta(p - k) \\
+ W^*_{-k,k'-p} W^*_{-k'-q'-p} \langle T_t Q_{p-q}(t_1) Q_{p-q}(t_2) \rangle \langle T_t Q_{-k}^-(t_2) Q_k(t_3) \rangle \langle T_t Q_{-k'}^-(t_2) Q_{k'}(t_4) \rangle \\
+ W^*_{-k,k'-p} W^*_{-k'-q}-p \langle T_t Q_{p-q}^+(t_1) Q_{p-q}^+(t_2) \rangle \langle T_t Q_{-k}^-(t_2) Q_k(t_3) \rangle \langle T_t Q_{-k'}^-(t_2) Q_{k'}(t_4) \rangle \\
+ W^*_{-k,k'-p} W^*_{-k'-q'-p} \langle T_t Q_{p-q}^+(t_1) Q_{p-q}^+(t_2) \rangle \langle T_t Q_{-k}^-(t_2) Q_k(t_3) \rangle \langle T_t Q_{-k'}^-(t_2) Q_{k'}(t_4) \rangle \\
+ W^*_{-k,k'-p} W^*_{-k'-q'-p} \langle T_t Q_{p-q}^+(t_1) Q_{p-q}^+(t_2) \rangle \langle T_t Q_{-k}^-(t_2) Q_k(t_3) \rangle \langle T_t Q_{-k'}^-(t_2) Q_{k'}(t_4) \rangle \\
+ W^*_{-k,k'-p} W^*_{-k'-q'-p} \langle T_t Q_{p-q}^+(t_1) Q_{p-q}^+(t_2) \rangle \langle T_t Q_{-k}^-(t_2) Q_k(t_3) \rangle \langle T_t Q_{-k'}^-(t_2) Q_{k'}(t_4) \rangle \right). \tag{48}
\]

Similar expressions can be derived from the rest of the summands in Eq. (47).

By substituting Eq. (48) in (46) and performing integration over time variables, we obtain

for \( p + k' \neq 0 \) and \( p - k \neq 0 \) (the phonons are supposed to be optical)
(S_p)_{fi} = 2\pi i T_{k'k}(p) \left[ (\sqrt{2} - 1) \delta(p - q/2) + 1 \right] \delta(\omega' + \omega - 2\Omega_-),

T_{k'k}(p) = D_k D_{k'} \left[ W^*_{q-p,k} W^*_{p-q,k'} G_k(\omega - \Omega) G_{k'}(\Omega_- - \omega - \omega_0^s) \tilde{G}_{p-q}(\omega - \Omega_-) \right. \\
+ W^*_{q-p,k} W^*_{p-q,k'} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- + \omega_0^s) \tilde{G}^+_{q-p}(\omega - \Omega_-) \\
+ W^*_{q-p,k} W^*_{p-q,k'} G_k(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- + \omega_0^s) \tilde{G}_{q-p}(\omega - \Omega_-) \\
+ W^*_{q-p,k} W^*_{p-q,k'} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- - \omega) \tilde{G}^+_{q-p}(\omega - \Omega_-) \\
+ W^*_{p-k'} W^*_{k-p} G_k(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- + \omega_0^s) \tilde{G}_{p}(\omega - \Omega_-) \\
+ W^*_{p-k'} W^*_{k-p} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- + \omega_0^s) \tilde{G}^+_{p}(\omega - \Omega_-) \\
+ W^*_{p-k'} W^*_{k-p} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- - \omega) \tilde{G}^+_{p}(\omega - \Omega_-) \\
+ \left. W^*_{p-k'} W^*_{k-p} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_-) \tilde{G}^+_{p}(\Omega_- - \omega) \right], \quad (49)

where \( \tilde{G}_p(\omega) \), \( \tilde{G}^+_p(\omega) \), and \( G_p(\omega) \) are Fourier transforms of the anomalous and normal Green’s functions of excitons in the Bose-condensed state, which are defined as follows:

\[
G_p(t - t') = -i \langle T(t) Q_p(t') \rangle, \\
\tilde{G}^+_p(t - t') = -i \langle T(t) Q^+_p(t') \rangle. \quad (50)
\]

Thus, we have derived the expression for the element of the S-matrix responsible for the two-photon emission due to coherent recombination directly from the Hamiltonian of the interaction between excitons and the electromagnetic field and the Hamiltonian of the exciton-phonon interaction (44). Similarly elements of the S-matrix corresponding to the RLS accompanied by the coherent two-exciton recombination (or generation) can be derived.

In a general case, Eq. (49) cannot be reduced to the corresponding expression (12) derived from the effective Hamiltonian (3). Below we will determine the conditions when this is possible and derive an expression for \( L^p_{pq} \).

By analyzing the stimulated two-photon emission under the condition \(|\omega - \Omega_-| \ll \omega_0^s\), we obtain

\[
T_{k'k}(p) = D_k D_{k'} \left[ W^*_{q-p,k} W^*_{p-q,k'} G_k(\omega - \Omega) G_{k'}(\Omega_- - \omega - \omega_0^s) \tilde{G}_{p-q}(\omega - \Omega_-) \right. \\
+ W^*_{q-p,k} W^*_{p-q,k'} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- + \omega_0^s) \tilde{G}^+_{q-p}(\omega - \Omega_-) \\
+ W^*_{q-p,k} W^*_{p-q,k'} G_k(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- + \omega_0^s) \tilde{G}_{q-p}(\omega - \Omega_-) \\
+ W^*_{q-p,k} W^*_{p-q,k'} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- - \omega) \tilde{G}^+_{q-p}(\omega - \Omega_-) \\
+ W^*_{p-k'} W^*_{k-p} G_k(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- + \omega_0^s) \tilde{G}_{p}(\omega - \Omega_-) \\
+ W^*_{p-k'} W^*_{k-p} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- + \omega_0^s) \tilde{G}^+_{p}(\omega - \Omega_-) \\
+ W^*_{p-k'} W^*_{k-p} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_- - \omega) \tilde{G}^+_{p}(\omega - \Omega_-) \\
+ \left. W^*_{p-k'} W^*_{k-p} \tilde{G}_{-k}(\omega - \Omega) \tilde{G}_{k'}(\omega - \Omega_-) \tilde{G}^+_{p}(\Omega_- - \omega) \right]. \quad (51)
\]
Under the conditions of approximation (19), we have for the retarded Green’s functions

\[ G^R_p(\omega) = -2\pi i n_0(T) \delta_p \delta(\omega) + G^\prime R_p(\omega), \]  

from which an expression for \( G_p(\omega) \) can be obtained. The anomalous Green’s function \( \tilde{G}_p(\omega) \), which is defined by Eq. (50), is related to the Green’s function \( \hat{G}_p(\omega) \) (see definition (8), and Eqs. (15) and (21)) by the formula \( \hat{G}_p(\omega) = -2\pi i n_0(T) \delta_p \delta(\omega) + \tilde{G}_p(\omega) \).

By comparing between the normal and anomalous Green’s functions at \( \omega = \omega^0 \), we obtain \( \tilde{G}_k(\omega^0)/G_k(\omega^0) \ll 1 \) at \( \omega^0 \gg \xi_k \). In this case the element (51) of the scattering matrix is determined by the expression

\[
T^'_{k'k}(p) = D_k D_{k'} \left[ W^*_{q-p,k} W^*_{p-q,k} G_k(\omega^0) G_{k'}(\omega^0) \tilde{G}_{p-q}(\omega - \Omega -) \\
+ W^*_{-pk} W^*_{+qk} G_k(\omega^0) G_{k'}(\omega^0) \tilde{G}_{-p}(\omega - \Omega -) \right],
\]  

where \( G_{k'}(\omega^0) \approx G_k(\omega^0) \approx 1/\omega^0 \).

The comparison between the latter expression and Eq. (12) in Section 2 yields \( L^>_{pq} \) for the effective Hamiltonian of exciton recombination:

\[ L^>_{pq} = -i \frac{D_q W^*_{pq}}{\omega^0}. \]  

The expressions for the other matrix elements of the exciton recombination in effective Hamiltonian (3) can be derived in a similar way.

Thus, the two-photon emission accompanied by the coherent two-exciton recombination can be analyzed with the aid of the effective Hamiltonian (3) with \( L^>_{pq} \) determined by Eq. (54) if the incident light satisfies the conditions \( |\omega - \Omega -| \ll \omega^0 \) and \( \xi_k \ll \omega^0 \). In this case, the matrix element (54) does not depend on temperature and \( f(\omega_L) \) is identical to the effective matrix element \( F \) responsible for the phonon-assisted recombination of an isolated exciton.

Appendix B

Luminescence intensity of Bose-condensed excitons

The luminescence of Bose-condensed excitons at frequency \( \omega' < \Omega_\!\_ \) is due to the optical phonon-assisted excitonic recombination accompanied by the generation of a Bogoliubov quasiparticle with energy \( \epsilon(p'_L) = \Omega_\!\_ - \omega' \) in the excitonic system. The matrix element of this recombination is
\( L = L^{\gamma}_{p' L} v_{p' L} \sqrt{n_{p' L} + 1}, \)  

\( v_{p' L}^2 = \frac{\sqrt{\alpha_L'^2 + 1} - \alpha_L'}{2\alpha_L'}, \quad n_{p' L} = \left[ \exp \left( \frac{\mid \Delta \omega' \mid}{T} \right) - 1 \right]^{-1}, \)  

where \( \Delta \omega' = \omega' - \Omega_- \) and \( \alpha_L' = |\Delta \omega'| / \zeta(T) \).

Using Fermi’s ‘golden rule’, we obtain the optical phonon-assisted luminescence intensity \( I^L_s(\omega') \) of Bose-condensed excitons at frequencies \( \omega' < \Omega_- \):

\[
I^L_s(\omega') = \frac{\omega'^4 \sqrt[4]{2m^3 \zeta(T)} \left( \sqrt{\alpha_L'^2 + 1} - 1 \right) f^2(\omega_L')}{6\pi^2 c^3 \sqrt{\alpha_L'^2 + 1} \left( \sqrt{\alpha_L'^2 + 1} + \alpha_L' \right)} \left[ 1 + \text{coth} \frac{\mid \Delta \omega' \mid}{2T} \right].
\]  

(56)

The luminescence of Bose-condensed excitons at frequency \( \omega' > \Omega_- \) is due to the optical phonon-assisted excitonic recombination accompanied by the disappearance of a quasiparticle with energy \( \epsilon(p'_L) = \omega' - \Omega_- \). The matrix element of this recombination is derived from Eq. (55) by substituting \( v_{p' L} \sqrt{n_{p' L} + 1} \rightarrow \sqrt{1 + v_{p' L}^2 \sqrt{n_{p' L}}} \). The luminescence intensity at frequency \( \omega' > \Omega_- \) is derived from Eq. (56) by substituting \( \alpha_L' \rightarrow -\alpha_L' \) and \( \text{coth}(\mid \Delta \omega' \mid / 2T) + 1 \rightarrow \text{coth}(\mid \Delta \omega' \mid / 2T) - 1 \). Thus, the expression that determines the luminescence spectrum of Bose-condensed excitons at an arbitrary frequency \( \omega' \neq \Omega_- \) has the form

\[
I^L_s(\omega') = \frac{\omega'^4 \sqrt[4]{2m^3 \zeta(T)} \left( \sqrt{\alpha_L'^2 + 1} - 1 \right) f^2(\omega_L')}{6\pi^2 c^3 \sqrt{\alpha_L'^2 + 1} \left( \sqrt{\alpha_L'^2 + 1} - \text{sign}(\Delta \omega') \alpha_L' \right)} \left[ \text{sign}(\Delta \omega') + \text{coth} \frac{\mid \Delta \omega' \mid}{2T} \right].
\]  

(57)

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FIGURES

Fig. 1.

Diagrams corresponding to the two-photon emission accompanied by the coherent phonon-assisted two-exciton recombination (the notations are explained in the text).

Fig. 2.

Cross section of the stimulated two-photon emission accompanied by the coherent optical phonon-assisted two-exciton recombination: (a) as a function of the difference $\Delta \omega = \omega - \Omega_{-}$ between the incident light frequency $\omega$ and $\Omega_{-}$ at different temperatures of the excitonic system: (1) $T/T_{c} = 0.01$; (2) 0.10; (3) 0.60; (4) 0.90; (5) 0.99; (b) as a function of the excitonic system temperature at different $\Delta \omega$: (1) $|\Delta \omega|/2T_{c} = 0.2$; (2) 0.3; (3) 0.9. The curves were plotted using Eq. (30). For all curves $\mu(0)/2T_{c} = 0.3$, and $\tau_{L}$ is assumed to be constant.
Fig. 1.
\[ \frac{\sigma^L(\Delta \omega,T)}{\sigma^L(0,0)} \]

\[ \Delta \omega/(2T_c) \]

Fig. 2a
\[
\frac{\sigma^L(\Delta \omega, T)}{\sigma^L(\Delta \omega, 0)}
\] vs \(T/T_c\) for different curves.