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Many-photon coherence of Bose-condensed excitons: Luminescence and related nonlinear optical phenomena

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We consider many-photon coherent emission governed by coherent recombination of many excitons from Bose condensate. Momentum conservation makes photons simultaneously created in the coherent recombination of several excitons from the condensate have zero sum momentum. Many-photon correlations in the processes of simultaneous many-photon production (photons squeezing) could be detected by photon counting experiments with several detectors spatially arranged in an appropriate way, i.e., by Hanbury-Brown-Twiss-like experiments. We analyze the stimulated processes of N-exciton coherent recombination from the condensate resonantly induced by $N-1$ external laser beams with momenta $\mathbf{k}_1, \mathbf{k}_2, \ldots, \mathbf{k}_{N-1}$. Such stimulated processes must reveal themselves through a unidirectional beam with recoil momentum $\mathbf{k}_N = -\sum_{i=1}^{N-1}\mathbf{k}_i$. In particular, two-exciton coherent recombination from the condensate stimulated by a laser beam effectively acts in a three-dimensional (3D) exciton system as a laser beam backscattering from exciton Bose condensate. For 2D coherent exciton systems besides the backscattering stimulated two-exciton coherent recombination has an additional manifestation—anomalous beam transmission with the only change in the sign of in-plane momentum component of inducing laser beam. We estimate the rates of many-photon coherent emission from a 3D system of Cu_2O excitons and from a 2D exciton system in GaAs/AlGaAs coupled quantum wells. The estimations show that these nonlinear optical effects are experimentally observable.

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I. INTRODUCTION

The detection of exciton Bose-Einstein condensation[1,2] is one of the most desirable goals in modern solid state physics. Promising experimental results[3–10] have been obtained in the study of coherent phase of two-dimensional (2D) excitons in coupled quantum wells.[11–26] As to the other recently studied exciton system, 3D excitons in Cu_2O, interesting achievements[1,2,27–34] have been extensively discussed in the literature (see Ref. 35, and references therein), which makes the search for new qualitative effects that can serve as clear and undoubtable evidence of exciton Bose-condensate formation, a vital problem.

Any exciton system inherently has a finite lifetime since excitons can recombine producing photons. During its lifetime exciton system luminesce and therefore the natural way to explore the system is to study exciton luminescence. It is also understood that the photons emitted by exciton system should reflect the properties of their emitter. Specifically, the coherence of recombining excitons must be in some way transmitted to the photons.

With the appearance of the condensate a system of bosons acquires many-particle coherent correlations. Exciton luminescence reflecting the coherence of Bose-condensed exciton system should possess many-photon correlations as well. Many-photon correlations cannot be revealed in intensity measurements with one photon detector, which are usually used to study exciton systems, as they measure only one-photon properties of exciton luminescence (correspondingly one-exciton properties) such as spectral shape or spatial coherence.[33]

We argue that many-exciton correlations can be transmitted to exciton luminescence by coherent many-exciton recombination processes where several excitons simultaneously recombine producing several correlated photons. In diagramatic language such processes can be represented by connected diagrams with several entering excitons and with the same number of photons leaving the diagram (see Fig. 1). Coherent many-exciton recombination processes determine nonlinear optical properties of an exciton system. In this paper we show that an exciton system with appearance of Bose condensate qualitatively changes its nonlinear optical properties in such a way that there arise a family of nonlinear optical effects with a mechanism of laser beam backscattering from condensed excitons. The detection of these effects could serve as a signature of exciton condensation.

Before we proceed with the presentation of the material let us confine ourselves on general remarks at this stage. The most popular semiconductors for studying a coherent phase of exciton systems are Cu_2O and GaAs. In bulk GaAs

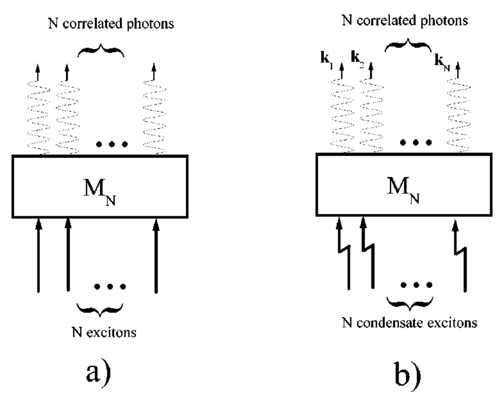

FIG. 1. (a) A generic diagram of many-exciton coherent recombination (b) a diagram of many-exciton coherent recombination from the condensate.

samples, 3D excitons do not form Bose condensate but, rather, gather in electron-hole droplets. Recently, GaAs excitons and their coherent properties are mainly studied in two dimensions—in coupled quantum wells (CQW) (see Ref. 3–26, and references therein). In CQW spatially indirect excitons have electric dipoles and their interaction has strongly repulsive character, so that they can form stable Bose-condensate at $T=0$ (or superfluid phase with quasi-long-range nondiagonal order and local quasicondensate at temperatures smaller than the temperature of Kosterlitz-Thouless transition).[11]

In this paper we are going to consider both the system of 3D exciton in Cu_2O and the system of quasi-2D excitons in GaAs/AlGaAs CQW. The idea of the paper in described in Sec. II. Section III is devoted to the system of 3D Bose-condensed Cu_2O excitons, which model is outlined in Sec.III A. Coherent N-exciton recombination from the condensate in this system is discussed in Secs. III B, III C, and III D for the cases $N=2$, 3, and 4, respectively. The process of two-exciton coherent recombination, laser beam backscattering, and anomalous laser beam transmission in the system of quasi-2D excitons in GaAs/AlGaAs CQW are studied in Sec. IV. Section V concludes with several notes to the results of the paper.

II. COHERENT MANY-EXCITON RECOMBINATION FROM BOSE CONDENSATE

To describe the idea of the paper let us consider the system of 3D excitons. The main process of exciton recombination is the one-exciton recombination where excitons independently recombine. This process determines such (one-photon) properties of exciton luminescence as a spectrum shape[33] or spatial coherence. Among the other recombination processes this process has the greatest rate since it is of the lowest order on relatively weak exciton-photon interaction. Consequently, exciton lifetime is mainly defined by one-exciton recombination. Let us denote the rate of this process as W_1. One exciton recombination is of the first order on exciton density so that $W_1 \propto \rho$, where ρ is the spatial density of excitons.

One-exciton recombination is not the only process in which excitons transform into photons. Consider the following possibility (see Fig. 1): several (say N) excitons interacting with each other simultaneously recombine and produce several (N) photons (and no other particles are produced). These are the processes of our interest and throughout the paper we will refer them to as coherent many (N-) exciton recombination. The processes are of higher order on exciton-photon interaction and consequently their rates, which we denote as W_N, are smaller than the rate of one-exciton recombination, $W_N \ll W_1$ and $W_N \propto \rho^N$.

When the exciton system is Bose condensed a macroscopic number of particles populate lowest one-particle quantum state of zero momentum $\mathbf{p}=\mathbf{0}$ with energy equal to exciton chemical potential μ. Chemical potential of excitons μ reckoned from the upper edge of valence band can be given as $E_g - E_{ex} + \tilde{\mu}$, where E_g is the gap, E_{ex} is the exciton binding energy, and $\tilde{\mu}$ is the chemical potential of excitons, arising from interexciton repulsion. In fact, $\tilde{\mu}, E_{ex} \ll E_g$, so that $\mu \approx E_g$.

In the coherent phase of exciton systems there arises a considerable possibility (dependent on the density of the condensate) that in the processes of coherent many-exciton recombination all the recombining excitons belong to Bose condensate. To picture the diagram representing such a process we simply "sink" entering exciton lines in the diagram given in Fig. 1(a) into the condensate [Fig. 1(b)]. Each condensate exciton line brings into the matrix element of this process a Bose factor $\sqrt{N_0}$, where N_0 is the number of condensate excitons, so that the rate of this process $W_N \propto \rho_{\mathrm{cond}}^N$, where ρ_{cond} is the condensate spatial density. From now on we will consider only the part of luminescence which comes from the condensate excitons. This can also be viewed as a low exciton density and low temperature limit where almost all the excitons are in the condensate $\rho \approx \rho_{\mathrm{cond}}$.

In the process of N-exciton recombination from the condensate momentum and energy conservation laws dictate the following requirements

$$\sum_{i=1}^{N} \mathbf{k}_i = 0, \quad \sum_{i=1}^{N} ck_i = N\mu, \tag{1}$$

where $\mathbf{k}_i$ is the momentum of ith created photon[36] and c is the speed of light in the medium. For the sake of brevity, we will measure photon energies from chemical potential μ so that the photon dispersion law is $\omega_{\mathbf{k}} = ck - \mu \equiv c(k-k_0)$, where k_0 is momentum of a photon with energy μ. This photon energy scale is used throughout the paper unless otherwise specified. Such form of photon dispersion fixes the energy scales for both photons and excitons, so that the excitons in the condensate have "zero" energy. The requirement for photon energies (1) now reads $\Sigma \omega_i = 0$, $\omega_i \equiv \omega_{\mathbf{k}_i}$.

For $N=2$, the requirement (1) is sufficient to fix relative orientation of momenta of two photons created in coherent two-exciton recombination. Namely, the photons have opposite momenta $\mathbf{k}_1 = -\mathbf{k}_2$ and the same energies equal exciton chemical potential. For $N>2$ cases, this requirement itself is insufficient to fix relative orientation of N created photons. Nevertheless, as it will be shown below the rate of the process is maximal when photon energies equal some values around chemical potential μ so that the magnitude of photon momenta with a great accuracy equal k_0. This fact along with requirement (1) fixes relative orientation of photon momenta in case of $N=3$ so that the angles between them equal $2\pi/3$.

The photons created in the process of N-exciton recombination are "squeezed" among different modes of photon field (N-mode squeezing). Such many-photon coherence can be studied by a classic method—photon counting experiments with N detectors (Hanbury-Brown-Twiss-like experiments). The specificity of our case is that the detectors and the exciton system must have such a relative position that the momentum conservation requirement (1) fulfills. Such experiments are of a distinct interest but in the present paper we focus on new nonlinear optical effects, which are governed by N-exciton coherent recombination from the condensate.

Imagine one induces the process of coherent N-exciton recombination from the condensate by $N-1$ laser beams with momenta $\mathbf{k}_1, \ldots, \mathbf{k}_{N-1}$, respectively. Provided that the energy constraint (1) is fulfilled, the stimulated process must manifest itself as a beam coming out of exciton system with recoil momentum and energy

$$\mathbf{k}_N = -\sum_{i=1}^{N-1} \mathbf{k}_i, \quad \omega_N = -\sum_{i=1}^{N-1} \omega_i. \tag{2}$$

Let us refer to this effect as *stimulated* N-exciton coherent recombination and to the arising beam (2) as a recoil beam. For instance, in case of $N=2$, the process of coherent two-exciton recombination stimulated by a laser beam with momentum $\mathbf{k}$ must result in appearance of a recoil beam with momentum $-\mathbf{k}$. In other words stimulated two-exciton coherent recombination acts as a laser beam backscattering from exciton condensate.

Being stimulated, the rate of the N-exciton coherent recombination in direction (2) increases (compared with spontaneous N-exciton coherent recombination rate in this direction) by a factor of

$$\prod_{i=1}^{N-1} (N_i+1),$$

where N_i is the average number of quanta per mode in inducing laser beam with momentum $\mathbf{k}_i$. Contrary to N-exciton recombination, one-exciton recombination in the direction $\mathbf{k}_N$ [see Eq. (2)] is not stimulated by laser beams, as the photons created in stimulated one-exciton recombination processes belong to laser beams modes only, i.e., such photons have momenta $\mathbf{k}_i \neq \mathbf{k}_N$. Therefore, spontaneous one-exciton recombination can be viewed as a background on which stimulated many-exciton coherent recombination is to be recognized.

A laser beam has a great number of quanta per mode. For concreteness, let this parameter be equal 10^3. The recoil beam intensity can exceed the background intensity (one-exciton spontaneous recombination intensity in the direction $\mathbf{k}_N$) if the following inequality is fulfilled:

$$10^{3(N-1)} W_N > W_1 \tag{3}$$

This inequality is a condition of that the process of stimulated N-exciton coherent recombination is experimentally detectable. Thus what we are to do now is to estimate the spontaneous rates W_N in comparison with W_1.

III. THE SYSTEM OF BOSE-CONDENSED CUPROUS OXIDE EXCITONS

A. The model

We consider 3D Bose-condensed exciton system in Cu_2O at zero temperature. In Cu_2O, there are two types of excitons: paraexcitons ($S=1$) and orthoexcitons ($S=0$) (interconversion rate between the branches being very low, see, e.g., Ref. 29). The type of the excitons, however, does not play any role for the effects to be discussed, so that when we refer to excitons we deal with a specific exciton branch.

The exciton system is assumed to be in a quasiequilibrium state with respect to exciton-exciton interactions. The interactions of the excitons with electromagnetic and phonon fields are weak and considered as perturbations. The normal and anomalous exciton Green functions are taken in ladder (Beliaev) approximation

$$G_{\mathbf{k}}(\omega) = \frac{\omega + \sqrt{\varepsilon_{\mathbf{k}}^2 + \tilde{\mu}^2}}{[\omega - (\varepsilon_{\mathbf{k}} - i\eta_{\mathbf{k}})][\omega + (\varepsilon_{\mathbf{k}} - i\eta_{\mathbf{k}})]},$$

$$F_{\mathbf{k}}(\omega) = -\frac{\tilde{\mu}}{[\omega - (\varepsilon_{\mathbf{k}} - i\eta_{\mathbf{k}})][\omega + (\varepsilon_{\mathbf{k}} - i\eta_{\mathbf{k}})]}, \tag{4}$$

$$\tilde{\mu} = \rho_{\text{cond}} U_0, \quad \varepsilon_{\mathbf{k}} = \sqrt{\frac{\tilde{\mu}k^2}{m} + \left(\frac{k^2}{2m}\right)^2}.$$

Here U_0 is zero Fourier-component of exciton-exciton interaction potential and m is exciton mass. In the following, we use for Cu_2O the estimates $m \approx 2.7 m_e$, $\tilde{\mu} \approx 0.5$ meV (0.8×10^{12} s^{-1}), $\rho_{\text{cond}} = 10^{19}$ cm^{-3}, and $\eta \approx 0.1\tilde{\mu}$. In Cu_2O, dielectric susceptibility $\epsilon \approx 9$ and $c = c_0/\sqrt{\epsilon} \approx 10^{10}$ cm/s, $E_g \approx 2$ eV so that $k_0 \approx 3 \times 10^5$ cm^{-1}.

In Cu_2O, direct recombination of an electron and a hole is very weak and an exciton decays mainly with production of a photon as well as of an optical phonon. The operator taking into account radiative decay of excitons is

$$\hat{V}^{(1)}(t) = \sum_{\mathbf{p}-\mathbf{k}-\mathbf{q}=0} \frac{L_{\mathbf{k},\mathbf{q}}}{\sqrt{V}} \hat{c}^{\dagger}_{\mathbf{p}+\mathbf{q}} e^{-i\omega_{\mathbf{p}+\mathbf{q}}t} \hat{a}_{\mathbf{p}}(t) \hat{\phi}_{\mathbf{q}}(t) + \text{H.c.},$$

$$\hat{\phi}_{\mathbf{q}}(t) = (\hat{b}_{\mathbf{q}} e^{-i\Omega t} + \hat{b}^{\dagger}_{-\mathbf{q}} e^{i\Omega t}),$$

where a's, b's, and c's are exciton, optical phonon, and photon destruction operators, respectively; Ω is the optical phonon energy (for simplicity we assume energy of optical phonon being momentum independent); L is effective interaction constant and V is the volume of the system.

Phonon Green function has the following form:

$$\mathcal{G}_{\mathbf{q}}(\omega) \equiv \mathcal{G}(\omega) = \frac{2\Omega}{[\omega - (\Omega - i\gamma)][(\omega + (\Omega - i\gamma)]}.$$

In Cu_2O, energy of the optical phonon $\Omega \approx 10^{-2}$ eV. For the estimates we take $\gamma = 10^9$ s^{-1} which corresponds to that the optical phonon lifetime is approximately 10^{-9} s.

As it will be seen later, the process of three-exciton coherent recombination requires the vertex of exciton-phonon interaction as well. Operator of exciton-phonon interaction can be taken in the following form:

$$\hat{V}^{(2)}(t) = \sum_{\mathbf{p}-\mathbf{k}-\mathbf{q}=0} \frac{g_{\mathbf{k},\mathbf{q}}}{\sqrt{V}} \hat{a}^{\dagger}_{\mathbf{p}+\mathbf{q}}(t) \hat{a}_{\mathbf{p}}(t) \hat{\phi}_{\mathbf{q}}(t) + \text{H.c.}$$

For simplicity, we consider both interaction constants g and L being independent of momenta $\mathbf{k},\mathbf{q}$, i.e., $g_{\mathbf{k},\mathbf{q}} \equiv g$ and $L_{\mathbf{k},\mathbf{q}} \equiv L$. We assume $g = 10^2 L$ for the following estimates.[37]

The radiative lifetime of an exciton τ, which is determined mainly by one-exciton recombination, equals

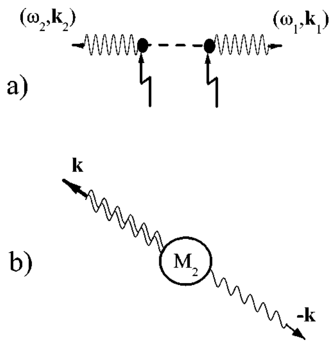

FIG. 2. (a) The diagram of two-exciton coherent recombination from the condensate in the lowest order on interparticle interaction. The broken arrows represent condensate excitons and the dashed line is a virtual optical phonon. (b) Stimulated two-exciton coherent recombination or laser beam backscattering, double wiggly line represents inducing laser beam.

$$\tau^{-1}=\frac{L^2k_0^2}{\pi c} \tag{5}$$

In Cu_2O, exciton radiative lifetime $\tau\approx 10$ μs, that corresponds to $L\approx 2{,}5\times 10^2$ s^{-1} $cm^{3/2}$. The spontaneous rate of one-photon recombination per unit volume is

$$W_1=\frac{\rho_{\rm cond}}{\tau}.$$

One-exciton recombination spectrum has a peak at the frequency $\mu-\Omega$, since in the process of one-exciton recombination exciton energy is distributed between a photon and an optical phonon.

The rate of the photon emission in spontaneous N-exciton coherent recombination per unit volume can be given as

$$W_N=N\int 2\pi\delta\left(\sum_{i=1}^{N}\omega_i\right)|\mathcal{M}_N|^2V^N\prod_{i=1}^{N-1}\frac{d^3\mathbf{k}_i}{(2\pi)^3}, \tag{6}$$

where $\mathcal{M}_N$ is the matrix element of the process and the factor N accounts for the fact that at any elementary act of N-exciton coherent recombination N photons are created. In fact, the matrix element $\mathcal{M}_N$ implicitly has N factors $V^{-1/2}$ but the volume of the system V drops out of the final result for W_N, so that one can set $V=1$.

B. Stimulated coherent two-photon emission and light backscattering from exciton Bose condensate

In the lowest order of exciton-photon interaction the matrix element of coherent two-exciton recombination from the condensate is given in Fig. 2(a). Two condensate excitons coherently recombine through interchange of a virtual optical phonon. Two photons created in the process have opposite momenta $\mathbf{k}_1=-\mathbf{k}_2$ and consequently they have the same energies equal exciton chemical potential [see Eq. (1)]. Analytical expression for the matrix element is

$$\mathcal{M}_2(\mathbf{k}_1,-\mathbf{k}_1)=\mathcal{G}(\omega_1)L^2\rho_{\rm cond}. \tag{7}$$

Using Eqs. (7), (6), and (5), one obtains the total rate of the photon emission in spontaneous two-exciton recombination process in the following form:

$$W_2=\frac{4k_0^2L^4\rho_{\rm cond}^2}{\pi c\Omega^2}=\frac{4L^2\rho_{\rm cond}}{\Omega^2}\times W_1.$$

For the parameters adopted (see previous section), the rate of spontaneous photon emission in the process of interest is estimated as

$$W_2\approx 0.6\times 10^{-2}\times W_1.$$

According to the condition (3), the process is experimentally detectable.

Stimulated two-exciton coherent recombination from the condensate effectively acts as a laser beam backscattering. Indeed, at any elementary act of the stimulated process, there is created not only the photon propagating along inducing beam direction but also the photon propagating in the opposite direction [see Fig. 2(b)].

The position of the spectral peak of two-exciton coherent recombination is higher by an optical phonon energy Ω than the one-exciton recombination peak, since in the processes of two-exciton coherent recombination no phonons are produced. This fact is an additional advantage allowing to distinguish between laser beam backscattering and the background radiation (spontaneous one-exciton recombination).

C. Stimulated coherent three-photon emission

In the lowest order on interparticle interactions the matrix element of three-exciton coherent recombination from the condensate is the sum over photon permutations in the matrix element given in the following:

$$\mathcal{M}_3(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)=gL^3\rho_{\rm cond}^{3/2}\sum_{i\neq j}\mathcal{G}(\omega_i)\mathcal{G}(\omega_j)G_{-\mathbf{k}_j}(\omega_j), \tag{8}$$

where the subscripts i and j, enumerating the photons, take on the values 1,2,3. The main contribution to the rate of the process comes from the (resonant) regions where the Green functions in the matrix element are mostly close to their poles. In these regions, energies of photons differ from the chemical potential of excitons relatively slightly. Indeed, the difference has the order of phonon energy or the energy of elementary excitation in exciton system [see Eq. (10)]. These differences, in turn, are negligibly small in comparison to the semiconductor gap and/or the chemical potential of exciton system μ. Consequently, photon wave vectors magnitudes differ from k_0 negligibly. In Eq. (8), the momentum $-\mathbf{k}_j$ can be substituted by k_0 so that the matrix element (8) is the function of photon energies only. Such approximation can be called fixed-photon-momenta-magnitude approximation.

Assuming $\gamma\ll\Omega$ and $\eta\ll\varepsilon_{k_0}$, it is possible to use the resonant approximation (pole approximation) and in $|\mathcal{M}|^2$ leave only six resonant terms which after integration over photon energies give the same contributions to the rate of the process. Now we can write

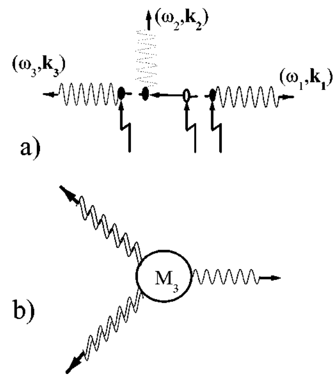

FIG. 3. (a) The diagram of three-exciton coherent recombination from the condensate in the lowest order on interparticle interaction. Straight line represents normal exciton Green function. (b) The system is exposed to two laser beams (double wiggly lines) directed to each other at the angle $2\pi/3$. Stimulated three-exciton coherent recombination should reveal itself by a recoil beam (single wiggly line).

$$|\mathcal{M}_3(\omega_1,\omega_2,\omega_3)|^2=6g^2L^6\rho_{\mathrm{cond}}^3|\mathcal{G}(\omega_2)\mathcal{G}(\omega_1)G_{k_0}(\omega_1)|^2.$$

The fixed-photon-momenta-magnitude approximation makes it possible to reduce the expression (6) for the rate of spontaneous photon emission in case of $N=3$ to the following (see Appendix 1):

$$W_3=18\frac{g^2L^6\rho_{\mathrm{cond}}^3k_0^3}{\pi c^3}\int|\mathcal{G}(\omega_1)G_{k_0}(\omega_1)|^2\times\frac{d\omega_1}{2\pi}\int|\mathcal{G}(\omega_2)|^2\frac{d\omega_2}{2\pi}.$$

The energy of optical phonon $\Omega\approx10^{-2}$ eV is much greater than the energy of elementary excitation of exciton system $\varepsilon_{k_0}\approx10^{-4}$ eV and the result simplifies as

$$W_3=18\frac{g^2L^6\rho_{\mathrm{cond}}^3k_0^3}{\pi c^3}\frac{1}{\Omega^2\gamma}\left[\frac{1}{\gamma}+\frac{1}{\eta}\left(1+\frac{\tilde{\mu}^2}{2\varepsilon_{k_0}^2}\right)\right]$$
$$=18\frac{g^2L^4\rho_{\mathrm{cond}}^2k_0}{c^2\Omega^2\gamma}\left[\frac{1}{\gamma}+\frac{1}{\eta}\left(1+\frac{\tilde{\mu}^2}{2\varepsilon_{k_0}^2}\right)\right]W_1. \quad (9)$$

For the parameters adopted (see Sec. III) the rate at which the photons are emitted due to the three-exciton coherent recombination can be estimated as

$$W_3\approx10^{-2}\times W_1,$$

i.e., every hundredth exciton decays in such a way.

The experiment where three-photon coherent emission can be detected is to expose the condensate to two beams directed with respect to each other at angle $(2\pi)/3$ [see Fig. 3(b)]. The process under consideration should result in appearance of a recoil beam with momentum (2) directed at the angle $(2\pi)/3$ with respect to the both inducing laser beams.

The main contribution to the rate of the process (9) comes from the regions on the photon energy scale where the Green functions are mostly close to their resonance. One of the virtual phonons can always be made resonant (real) by choosing $\omega_3=\pm\Omega$ [see Fig. 3(a)]. As to the virtual exciton elementary excitation and the other virtual phonon, they can not be resonant simultaneously since they share the same arguments. Therefore, there are two possibilities (i) the other virtual phonon [the right one in the Fig. 3(a)] is resonant or (ii) the virtual elementary exciton excitation is resonant. In the Fig. 3(a) the cases (i) and (ii) correspond to $\omega_1=\pm\Omega$ and $\omega_1=\pm\varepsilon_{k_0}$, respectively. The possibility (i) is responsible for the first term in the parenthesis in Eq. (9) whereas case (ii) is responsible for the second. Two cases correspond to three sets of energy values of photons:[38]

$$(\mu+\Omega,\mu-\Omega,\mu),\quad(\mu+\Omega,\mu\pm\varepsilon_{k_0},\mu-\Omega\mp\varepsilon_{k_0})$$

and

$$(\mu-\Omega,\mu\mp\varepsilon_{k_0},\mu+\Omega\pm\varepsilon_{k_0}). \quad (10)$$

The first set is case (i) whereas the second and the third sets are case (ii).

To resonantly increase the efficiency of stimulated three-exciton coherent recombination the energies of two stimulating laser beams must be equal to any two values of any of the sets (10). As is seen from Eq. (10) the energies of photons slightly differ from exciton chemical potential $\mu\gg\Omega\gg\varepsilon_{k_0}$. The greatest difference is of order of optical phonon energy Ω. Consequently, photon momenta magnitudes are not equal k_0 exactly. The relative difference is of order of $\Omega/\mu\approx5\times10^{-2}$. This is the order of a value by which actual angles between photon momenta can differ from $2\pi/3$—the magnitude obtained within fixed-photon-momenta-magnitude approximation.

D. Four-exciton coherent recombination

In the lowest order on interparticle interactions the matrix element of four-exciton coherent recombination has the following form:

$$\mathcal{M}_4(\mathbf{k}_1\cdots\mathbf{k}_4)=\rho_{\mathrm{cond}}L^4\sum_{l\neq m,m\neq n,n\neq l}F_{-(\mathbf{k}_l+\mathbf{k}_m)}[-(\omega_l+\omega_m)]\times\mathcal{G}(-\omega_l)\mathcal{G}(-\omega_n),\quad l,m,n=1\cdots4.$$

It consists of 12 diagrams similar to the one given in Fig. 4(a). The diagram explicitly has only two entering lines of condensate excitons. The other two lines are implicitly contained in anomalous exciton Green function.

In resonant approximation (pole approximation) one can leave only resonant terms in $|\mathcal{M}_4|^2$ so that

$$|\mathcal{M}_4|^2=12\rho_{\mathrm{cond}}^2L^8|F_{|\mathbf{k}_1+\mathbf{k}_2|}(\omega_1+\omega_2)\mathcal{G}(\omega_1)\mathcal{G}(\omega_3)|^2. \quad (11)$$

In fixed-photon-momenta-magnitudes approximation (see Appendix 2) the rate of photon emission (6) for $N=4$ can be reduced to

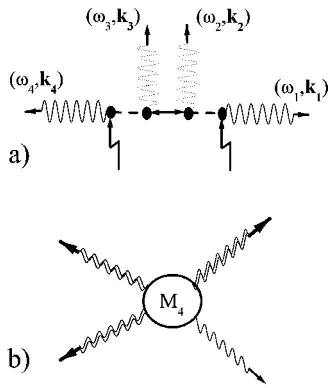

FIG. 4. (a) The diagram of four-exciton coherent recombination from the condensate in the lowest order on interparticle interaction. Double arrowed line represents anomalous exciton Green function. (b) The system is exposed to three laser beams (double wiggly lines) relatively directed in such a way that momentum conservation requirement fulfills [see Eq. (1)]. Stimulated four-exciton coherent recombination should reveal itself by a recoil beam (single wiggly line).

$$W_4=48\rho_{\rm cond}^2L^8\frac{k_0^4}{2\pi^2c^4}\int\frac{du}{(2\pi)}\int_0^{2k_0}dk|F_k(u)|^2 \times\int\frac{d\omega_1}{(2\pi)}|\mathcal{G}(\omega_1)|^2\int\frac{d\omega_3}{(2\pi)}|\mathcal{G}(\omega_3)|^2,$$

where we made the substitutions $u=\omega_1+\omega_2$, $k=|\mathbf{k}_1+\mathbf{k}_2|$. In the approximation of k independent and small η ($\eta \ll \varepsilon_{k_0}$) the first integral simplifies as

$$\int\frac{du}{(2\pi)}\int_0^{2k_0}dk|F_k(u)|^2=\frac{\pi\tilde{\mu}^2}{8\eta^2c_v},$$

where c_v is the speed of exciton system elementary excitation at the linear part of exciton spectrum $c_v=\sqrt{\tilde{\mu}/m}\approx 0.5\times 10^6$ cm/s. Finally, the rate of photon emission in spontaneous four-exciton coherent recombination is

$$W_4=\frac{3\rho_{\rm cond}^2L^8\tilde{\mu}^2k_0^4}{\pi c_v c^4\gamma^2\eta^2}=\frac{3\rho_{\rm cond}L^6\tilde{\mu}^2k_0^2}{c_v c^2\gamma^2\eta^2}\times W_1. \qquad (12)$$

For the parameters adopted (see Sec. III), the corresponding rate can be estimated as

$$W_4=0.5\times 10^{-2}\times W_1.$$

This result shows that approximately every two-hundredth exciton decays due to this process and consequently the stimulated process is experimentally observable.

The momenta of four photons created in the process can be directed as shown in Fig. 4(b). A consequence of the relation (1) is that the sum momenta of any two photons has the same magnitude and opposite direction with that of the other photon pair. The angle between momenta of any two photons equals the angle between momenta of the other two photons. Relative orientation of the two planes where momenta of each pair lie is not fixed however, i.e., the momenta of the photons are generally not complanar.

An experiment for detection of the process of four-exciton coherent recombination is to expose the exciton system to three laser beams [see Fig. 4(b)]. The process reveals itself as a recoil beam with momentum (2).

As in the previous section, we note that the main contribution to the rate (12) comes from the regions on photon energies scales where the Green functions are mostly close to their poles. As it can be seen from Fig. 4(a) all the virtual particles can be made ''real'' by appropriate choice of photon energies. In the resonant process the energies of photons must be equal to[39]

$$(\mu+\Omega,\mu-\Omega,\mu-\Omega\pm\varepsilon_{k_0},\mu+\Omega\mp\varepsilon_{k_0}).$$

To increase the efficiency of stimulated four-photon coherent emission the energies of laser beams must be equal any three values of the set.

IV. LASER BEAM BACKSCATTERING AND ANOMALOUS TRANSMISSION IN QUASI-2D EXCITON SYSTEM IN CQW

GaAs is a direct gap semiconductor with allowed dipole interband transition. Excitons in quantum wells are quasi-two-dimensional contrary to the photons which are three-dimensional. In this system only 2D in-plane momentum is conserved. The operator of exciton-photon interaction can be given as

$$\hat{V}(t)=\hbar\sum_{\mathbf{k}}\frac{g}{\sqrt{L}}[\hat{a}_{\mathbf{k}_\parallel}(t)\hat{c}^\dagger_{\mathbf{k}}e^{-i\omega_{\mathbf{k}}t}+\text{H.c.}], \qquad (13)$$

where $\hat{a}$ and $\hat{c}$ are exciton and photon operators, respectively; g is the interaction constant; in-plane vectors $\mathbf{k}_\parallel$ are 2D; L is the width of the system in normal to CQW direction.

In the lowest order on exciton-photon interaction the matrix element of spontaneous two-exciton coherent recombination with production of two photons with in-plane wave vector components $\pm\mathbf{k}_\parallel$ [see Fig. 5(a)] is:

$$\mathcal{M}_2(\mathbf{k}_\parallel)=g^2F(\omega_1,\mathbf{k}_\parallel)$$

so that the rate of the process is

$$W_2(\mathbf{k}_\parallel)=\int(2\pi)\delta(\omega_1+\omega_2)g^4|F(\omega_1,\mathbf{k}_\parallel)|^2\frac{dk_{\perp,1}dk_{\perp,2}}{(2\pi)^2},$$

where F is anomalous Green function of exciton subsystem. The anomalous Green function has the form given in Eq. (4), with different parameters, however, corresponding to GaAs. For simplicity, we consider that elementary excitation decay parameter η is independent on exciton elementary excitation momentum. In the resonant (pole) approximation we make the substitution

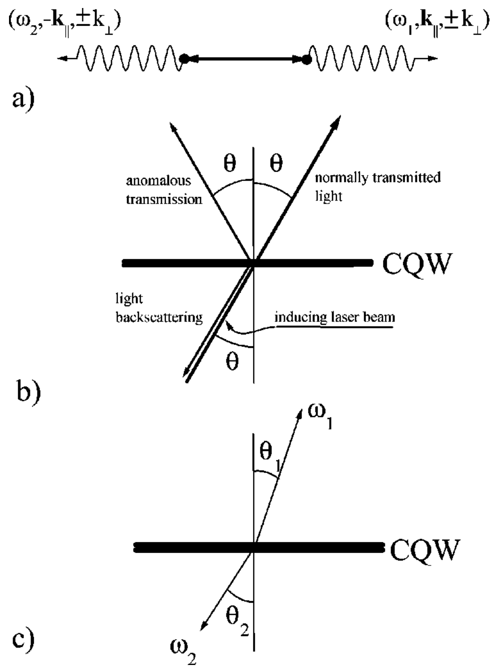

FIG. 5. Light backscattering and anomalous transition in coherent 2D exciton system in GaAs/AlGaAs coupled quantum wells (a) The diagram of two-exciton coherent recombination with production of two photons. (b) Spatial orientation of photon momenta in stimulated two-exciton coherent recombination. Besides the beam with momentum $-\mathbf{k}_\parallel,-\mathbf{k}_\perp$, which corresponds to laser beam backscattering, the beam with momentum $-\mathbf{k}_\parallel,\mathbf{k}_\perp$ arises. The latter corresponds to anomalous light transmission. In resonant processes the angles θ between photon momenta and wells plane are almost the same. (c) In nonresonant processes, where the photons with energies ω_1 and ω_2 (reckoned from exciton chemical potential $\mu=k_0c$) are created, these angles are different and obey the relation $(\omega_2+ck_0)/(\omega_1+ck_0)=\sin\theta_1/\sin\theta_2$.

$$|F(\omega,\mathbf{k}_\parallel)|^2\to\frac{\tilde{\mu}^2\pi[\delta(\omega-\varepsilon_{\mathbf{k}_\parallel})+\delta(\omega+\varepsilon_{\mathbf{k}_\parallel})]}{[(2\varepsilon_{\mathbf{k}_\parallel})^2+\eta^2]\eta}. \tag{14}$$

This substitution corresponds to that we take into consideration only resonant processes where two created photons have the energies $\pm\varepsilon_{\mathbf{k}_\parallel}$ [on the scale accepted, i.e., $c(k-k_0)=\pm\varepsilon_{\mathbf{k}_\parallel}$]. Using Eq. (14) one obtains

$$W_2(\mathbf{k}_\parallel)\approx\frac{\tilde{\mu}^2\prod_{i=1,2}\left\{\int 2\pi g^2\delta[\omega_i+(-1)^i\varepsilon_{\mathbf{k}_\parallel}]\frac{dk_{\perp,i}}{(2\pi)}\right\}}{[(2\varepsilon_{\mathbf{k}_\parallel})^2+\eta^2]\eta}.$$

The energy of elementary excitation $\varepsilon_{\mathbf{k}_\parallel}\ll\mu$ and consequently the approximation of fixed-photon-momenta magnitudes is applicable. In the arguments of δ functions we can omit the terms $\varepsilon_{\mathbf{k}_\parallel}$. In result, the magnitudes in the parenthesis become the radiative lifetime reciprocal of an exciton with momentum $\mathbf{k}_\parallel$:

$$W_2(\mathbf{k}_\parallel)\approx\frac{\tilde{\mu}^2}{[(2\varepsilon_{\mathbf{k}_\parallel})^2+\eta^2]\eta\tau_{\mathbf{k}_\parallel}^2}. \tag{15}$$

For spatially indirect excitons in GaAs/AlGaAs CQW radiative lifetime τ approximately equals 10^{-8} s. Assuming that $\tau_{\mathbf{k}_\parallel}$ is independent of $\mathbf{k}_\parallel$, i.e., $\tau_{\mathbf{k}_\parallel}\equiv\tau$, and that entire radiative zone corresponds to linear part of elementary excitation spectrum $\varepsilon_{\mathbf{k}_\parallel}\approx\sqrt{\tilde{\mu}/m}k_\parallel$ for $k_\parallel<k_0$, one can integrate $W_2(k_\parallel)$ over radiative zone what yields total spontaneous rate of photon production in the process per unit area of CQW in the following form:

$$W_2=\int_{k_\parallel<k_0}W_2(k_\parallel)\frac{d^2k_\parallel}{(2\pi)^2}=\frac{\tilde{\mu}m}{16\pi\eta\tau^2}\ln\left(1+\frac{4\tilde{\mu}k_0^2}{m\eta^2}\right)$$

In GaAs, $E_g\approx1.5$ eV and $\epsilon\approx12$ so that $k_0\approx2.8\times10^5$ cm^{-1}. Taking for estimation $\rho_{\rm cond}=10^{10}$ cm^{-2}, $\tilde{\mu}=0.5$ meV, $\eta=0.1\tilde{\mu}$, we have

$$W_2\approx2\times10^{-3}W_1,$$

where W_1 is the rate of one-exciton recombination $W_1=\rho_{\rm cond}\tau^{-1}$. This result shows that the effect of stimulated coherent two-exciton recombination can be detected in the system of quasi-2D excitons in GaAs/AlGaAs CQW [see Eq. (3)].

Two created photons have approximately the same magnitudes of their momenta $k_0\pm\varepsilon_{\mathbf{k}_\parallel}/c\approx k_0$ and exactly the same magnitudes of in-plane momenta components. Therefore, the angles of their propagation θ [see Fig. 5(b)] are almost the same.[40]

In two dimensions the third component of photon momentum is not fixed and is set only by energy conservation law. Consequently, the rate $W_2(\mathbf{k}_\parallel)$ corresponds to *four processes* in which the pairs of photons with momenta $(\mathbf{k}_\parallel,\pm k_\perp)$ and $(-\mathbf{k}_\parallel,\pm k_\perp)$ are created. Therefore, stimulating laser beam with in-plane momentum component $\mathbf{k}_\parallel$ induces (contrary to the 3D case) *two processes* in which the recoil beams are emitted in two directions $(-\mathbf{k}_\parallel,\pm k_\perp)$. Thus stimulated two-exciton coherent recombination in 2D coherent exciton system manifests itself via two effects: stimulated laser beam backscattering as well as stimulated anomalous laser beam transmission. In the latter process only in-plane component of laser beam momentum changes its sign [see Fig. 5(b)].

Furthermore, in the process of interest it is not necessary for the photons to be "resonant," i.e., the photons can have energies $\omega_{1,2}$ different from $\pm\varepsilon_{\mathbf{k}_\parallel}$ [on the scale accepted, i.e., $\omega_{1,2}\equiv c(k_{1,2}-k_0)=\pm\varepsilon_{\mathbf{k}_\parallel}$]. However, the rate of such nonresonant process is weak and for $\omega\gg\varepsilon_{\mathbf{k}_\parallel}$ its rate is proportional to ω^{-4}. The photons created in nonresonant processes with different energies ω_1 and ω_2 $(\omega_1+\omega_2=0)$ have different normal wave components as well. The angles of their propagation obey the relation $\sin(\theta_1)/\sin(\theta_2)=(\omega_2+ck_0)/(\omega_1+ck_0)$ [see Fig. 5(c)].

V. CONCLUSION

In conclusion, we considered many-exciton coherent recombination processes in a system of Bose-condensed excitons. It is shown that N-exciton coherent recombination processes, being resonantly induced by $N-1$ external laser beams, must result in that Bose-condensed exciton system irradiates a unidirectional beam with recoil momentum (2).These effects are nonlinear optical properties of exciton Bose-condensed systems and can serve as signatures of exciton Bose condensation.

For the effects under consideration the momentum conservation plays a crucial role. However, the inevitable presence of impurities, interface roughness, etc., leads to the smearing of exciton momentum. This should result in the angular uncertainty of the recoil beam propagation direction of order of $1/(k_0 l)$, where l is a mean free path of an exciton.

The part of spontaneous luminescence corresponding to N-exciton coherent recombination is squeezed among the modes of created photons (N-mode squeezing). Disregard the weakness of these processes, another way to detect them is photon counting experiments with N detectors, e.g., the Hanbury-Brown-Twiss method (see Refs. 41,42) for two-exciton coherent recombination. In such experiments, detectors must be positioned relatively to exciton system in such a way that momentum conservation requirement in N-exciton recombination fulfills, e.g., for two-exciton coherent recombination the two detectors must be placed diagonally with respect to exciton system.

We would also like to emphasize that by now in physics there are two known backscattering mechanisms: the light backscattering from disordered media, connected with weak Anderson localization of light and Andreev reflection of quasiparticles (quasielectrons or quasiholes) on the boundary normal-metal–superconductor. The stimulated two-exciton coherent recombination discussed in the present paper appears to be a mechanism of backscattering.

In a light of the effects discussed in this paper, the presence of normal magnetic field would influence 2D-exciton system in CQW only quantitatively, i.e., by changing exciton mass, condensate density, etc. Therefore, the results of Sec. IV are directly applicable to magnetoexciton problem.[17,26,43]

The method of studying nonlinear properties of exciton system, which is alternative to the proposed in this paper, is four-wave-mixing experiments.[44–46] It must be noted, that four-wave-mixing experiments have essentially different physical background from that of many-exciton coherent recombination. Particularly, in four-wave-mixing experiments exciton matter waves are generated by pumping laser beams whereas in many-exciton coherent recombination it is assumed that there is already a condensed exciton system and laser beams only induce process of their coherent recombination.

We would also like to note that effects described can be generalized to other systems of Bose particles, e.g., for Bose-condensed atoms.[47] At this, however, the Bose particles must be able to disappear producing photons or in other words be metastable.

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APPENDIX: APPROXIMATION OF FIXED-PHOTON-MOMENTA-MAGNITUDES

The integration over photon momenta in Eq. (6) is performed over 3D momenta of $N-1$ photons whereas in fixed-photon-momentum-magnitudes approximation the matrix elements of three- and four-photon coherent emission depend on less number of variables. It is possible to integrate out the idle variables in the integral (6), i.e., in the expression

$$\int \cdots 2\pi\delta\left(\sum_{i=1}^{N} c(k_i-k_0)\right)\prod_{i=1}^{N-1}\frac{k_i^2 d\omega_i d\cos\theta_i d\phi_i}{c(2\pi)^3}, \tag{A1}$$

where we use spherical coordinates for photon momenta

$$\mathbf{k}_i=k_i(\cos\theta_i;\sin\theta_i\cos\phi_i;\sin\theta_i\sin\phi_i). \tag{A2}$$

The approximation of fixed-photon-momenta-magnitudes is to assume $k_i=k_0, i=1\cdots N-1$. The momentum space differentials take the form

$$\frac{k_0^2 d\omega_i d\cos\theta_i d\phi_i}{c(2\pi)^3}$$

and the energy δ function takes the following form:

$$c^{-1}\delta\left(\left|\sum_{i=1}^{N-1}\mathbf{k}_i\right|-k_0\right). \tag{A3}$$

In the approximation of fixed-photon-momenta magnitudes, the energy conservation law serves to ensure that photon momenta $\mathbf{k}_i, i=1\cdots N-1$ have such a relative spatial orientation that the recoil momentum of the Nth photon, which equals the magnitude (2), has the magnitude k_0.

1. The case of three photons

The matrix element of the three-photon coherent emission depends only on the energies of the photons and we can integrate out the spatial angles variables. With no loss of generality we can choose the azimuth axis of the second photon being directed along the first photon momentum so that $|\mathbf{k}_1+\mathbf{k}_2|=k_0\sqrt{2(1+\cos\theta_2)}$ [see Fig. 6(a)]. The energy δ function (A3) takes the form

$$(ck_0)^{-1}\delta[\sqrt{2(1+\cos\theta_2)}-1]=(ck_0)^{-1}\delta(\cos\theta_2+1/2).$$

Integration over spherical angles of the photons simplify the expression (A1) in the case of three-photon coherent emission as

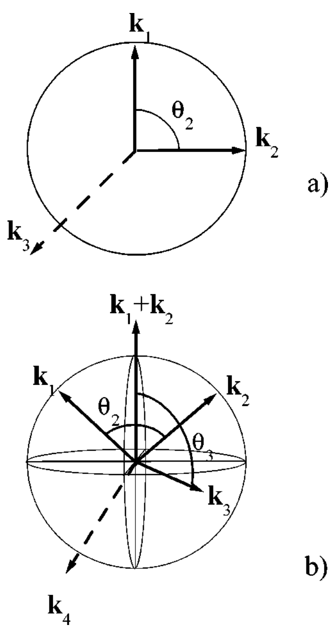

FIG. 6. Photon momenta orientation in fixed-photon-momenta-magnitudes approximation (Appendix). (a) The case of three photons. The magnitudes of momenta $\mathbf{k}_1$ and $\mathbf{k}_2$ equal k_0 whereas the magnitude of the third photon momentum depends on relative orientation of vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. (b) The case of four photons. The magnitudes of momenta $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ equal k_0 whereas the magnitude of the fourth photon momentum depends on relative orientation of vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$.-

$$\frac{k_0^3}{\pi c^3}\int \cdots \prod_{i=1}^{2} \frac{d\omega_i}{(2\pi)}. \tag{A4}$$

The fact that $\cos\theta_2 = -1/2$ implies that the angles between the three created photons momenta equal $2\pi/3$, as they should [see Fig. 3(b)].

2. The case of four photons

For four-photon coherent emission the matrix element $\mathcal{M}_4$ is a function of photon energies and the magnitude $|\mathbf{k}_1+\mathbf{k}_2|$. To reduce the number of variables in Eq. (6) we first choose the azimuth axis of the third photon being directed along the vector $\mathbf{k}_1+\mathbf{k}_2$ so that

$$\left|\sum_{i=1}^{N-1}\mathbf{k}_i\right| = \sqrt{|\mathbf{k}_1+\mathbf{k}_2|^2+k_0^2+2k_0|\mathbf{k}_1+\mathbf{k}_2|\cos\theta_3}.$$

The δ function in Eq. (A3) transforms as

$$c^{-1}\delta\left(\sqrt{|\mathbf{k}_1+\mathbf{k}_2|^2+k_0^2+2k_0|\mathbf{k}_1+\mathbf{k}_2|\cos\theta_3}-k_0\right)$$
$$=(c|\mathbf{k}_1+\mathbf{k}_2|)^{-1}\delta(\cos\theta_3-X),$$
$$X=-(2k_0)^{-1}|\mathbf{k}_1+\mathbf{k}_2|\in[-1,0].$$

Integration over the spherical angles of the third photon simplifies the expression (A1) as

$$\int \cdots \frac{k_0^2}{c^2|\mathbf{k}_1+\mathbf{k}_2|}\frac{d\omega_3}{(2\pi)}\prod_{i=1}^{2}\frac{k_0^2 d\omega_i}{c(2\pi)^3}d\cos\theta_i d\phi_i.$$

Now we choose azimuth axis of the second photon being directed along the first photon momentum so that $|\mathbf{k}_1+\mathbf{k}_2| = k_0\sqrt{2(1+\cos\theta_2)}$ [see Fig. 6(b)]. The integration over $\cos\theta_2$ transforms into integration over $|\mathbf{k}_1+\mathbf{k}_2|$ as

$$\frac{d\cos\theta_2}{|\mathbf{k}_1+\mathbf{k}_2|}=k_0^{-2}d(|\mathbf{k}_1+\mathbf{k}_2|), \quad |\mathbf{k}_1+\mathbf{k}_2|\in(0,2k_0).$$

Finally, for four-photon coherent emission we can rewrite the expression (A1) as

$$\frac{k_0^4}{2\pi^2c^4}\int \cdots d|\mathbf{k}_1+\mathbf{k}_2|\prod_{i=1}^{3}\frac{d\omega_i}{(2\pi)}.$$

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[39] As in Eq. (10), the chemical potential μ is explicitly included in photon energies.
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