Pis'ma v ZhETF, vol. 74, iss. 5, pp. 318 – 322

Stimulated light backscattering from exciton Bose-condensate

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Submitted 8 August 2001

The new effect – light backstattering from exciton Bose-condensate is considered. The effect is connected with photoinduced coherent recombination of two excitons in the condensate with production of two photons with opposite momenta. The effect of two-exciton coherent recombination leads also to the appearence of the second order coherence in exciton luminescence connected with squeezing between photon states with opposite momenta. The estimations given for Cu_2O and GaAs excitons show that the effect of stimulated light backscattering can be detected experimentally. Moreover, in the system of 2D excitons in coupled quantum wells the effect of *stimulated anomalous light transmission* must also take place. Analogous effects can take place also in systems of Bose-condensed atoms in excited (but metastable) states.

PACS: 71.35.Lk, 78.45.+h

The great progress has been recently made in the study of exciton Bose-condensation in 3D and 2D exciton systems (see [1 – 8] and references therein). The study of various ways which exciton condensate should unambiguously reveal itself by its optical properties is vital [9, 10].

In the present Letter we analyze the coherent coupling of photons with opposite momenta, originating from coherent recombination of two excitons from the condensate. We reveal that this process leads to new effect – stimulated light *backscattering* from exciton condensate. This effect can be viewed as a photoinduced coherent recombination of two *condensate* excitons with production of two photons with opposite momenta.

1. Let us first consider 3D excitons in Cu_2O. Direct recombination of electron and hole in Cu_2O is a forbidden process and an exciton decays mainly with production of a photon as well as of an optical phonon. The interaction Hamiltonian is:

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

where $\hat{a}(\hat{a}^{\dagger})$, $\hat{b}(\hat{b}^{\dagger})$ and $\hat{c}(\hat{c}^{\dagger})$ are exciton, phonon and photon destruction (creation) operators, respectively; $\omega_{phn,\mathbf{q}}$ is the optical phonon dispersion; V is volume of the system; L is effective interaction constant; $\omega_{\mathbf{k}} = ck$ is the frequency of photons with wave vector $\pm\mathbf{k}$, and $c = c_0/n$ is the speed of light in Cu_2O ($n \approx 3$).

In *normal* phase of the exciton system the main process of exciton recombination is determined by

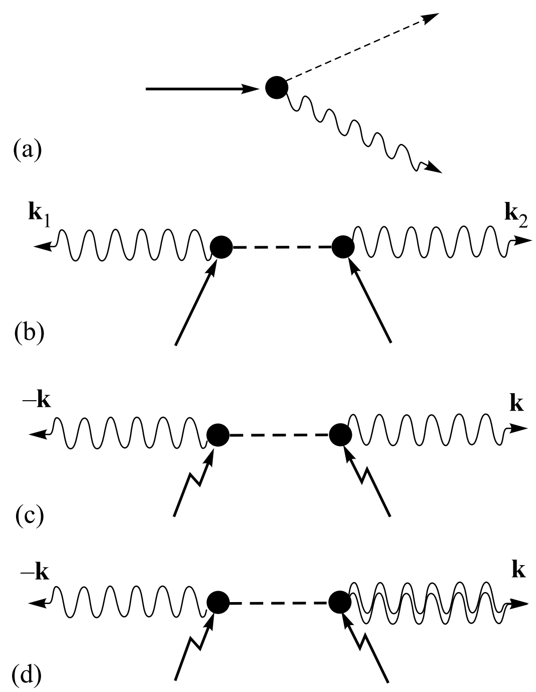

Fig.1. (a) The diagram of one-exciton recombination in Cu_2O; dashed line represents phonon, straight line represents exciton and wiggly line represents photon. (b) The diagram of two-exciton recombination process where phonon is virtual. (c) The diagram of recombination of two excitons from the condensate. Due to momentum conservation the photons have opposite wave vectors. (d) The process of two-exciton recombination stimulated by external beam with wave vector k; double wiggly line represents classical laser field

the diagram in Fig.1a. The spectrum of the exciton luminescence has broadened peak at the frequency

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$\approx \omega_{ex}(\mathbf{0}) - \omega_{phn}(\mathbf{0})$ [9]. The radiative lifetime of isolated exciton with wave vector $\mathbf{k} = 0$ can be given as:

$$\frac{1}{\tau_{\mathrm{Cu_2O}}} = \frac{\left|\mathrm{L}\left(k_0, k_0\right)\right|^2 k_0^2}{\pi c} \tag{2}$$

where L is the matrix element in Eq.(1); k_0 is the absolute value of photon wave vector $\approx E_g/c\hbar$, E_g is the energy gap in Cu_2O, $E_g \approx 2\,\mathrm{eV}$. The process of two exciton recombination (Fig.1b) has extra vertex of exciton-photon interaction and, therefore, is substantially weaker.

In the *Bose-condensed* phase of exciton system, the exciton lines on Fig.1b can belong to excitons from the condensate (see Fig.1c). One-exciton recombination from the condensate (Fig.1a) has the only great factor $\sqrt{N_0}$, where N_0 is the number of quanta in macroscopically populated lowest quantum exciton state, while *two-exciton recombination has two such factors.* Additional factor can compensate the weakness of the process in comparison with the process given in Fig.1a and thus make the rate of two-exciton recombination be of order of one-exciton recombination rate. In pure samples due to momentum and energy conservation laws two created photons have the same energy and opposite momenta. The luminescence originating from this recombination process has a peak at the chemical potential of exciton system measured form the valence band $\hbar\mu$ ($\hbar\mu \approx E_g$). Due to that in two-exciton recombination no energy in transformed to phonons the frequency of two-exciton recombination luminescence is greater than the that of one-exciton recombination by the magnitude $\omega_{phn} \ll E_g/\hbar$ (see Fig.2).

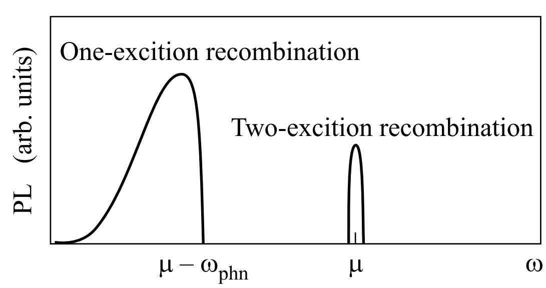

Fig.2. The schematic photoluminescence (PL) spectrum of Cu_2O. One exciton recombination results in the spectrum on the left side of the figure. This part of spectrum was studied, e.g., in Ref.[9]. The part of the spectrum on the right side of the figure corresponds to two-exciton recombination. It has higher energy since no energy is transformed to phonons

The rate of two-exciton recombination with production of two photons with wave vectors $\pm\mathbf{k}$ is:

$$W^{\mathrm{spon}}_{\mathrm{Cu_2O}}(\mathbf{k}) = 2\pi\delta(2\omega_{\mathbf{k}} - 2\mu)\left|M(\mathbf{k})\right|^2 \tag{3}$$

where M is the matrix element of the process. The overall rate of the spontaneous photon emission in two-exciton recombination process per unit volume obtains by integrating the $W^{\mathrm{spon}}_{\mathrm{Cu_2O}}(\mathbf{k})$ over all photon wave vectors $\mathbf{k}$ and by multiplying the result by 2 (at any elementary act of the process two photons are created):

$$W^{\mathrm{spon}}_{\mathrm{Cu_2O}} = \frac{k_0^2}{\pi c}\left|M(k_0)\right|^2. \tag{4}$$

Matrix element $M(\mathbf{k})$ has the form:

$$M_{\mathrm{Cu_2O}}(k_0) = G_{phn}\left(0, k_0\right)\mathrm{L}\left(k_0, k_0\right)^2 \rho_{\mathrm{cond}} \tag{5}$$

where ρ_{cond} is the spatial density of the condensate and

$$G_{phn}(\omega, \mathbf{q}) = \left(\omega - \omega_{\mathbf{q}} + i\delta\right)^{-1} - \left(\omega + \omega_{\mathbf{q}} - i\delta\right)^{-1} \tag{6}$$

is the phonon Green's function. Combining Eqs.(4), (5) and (2), one obtains:

$$W^{\mathrm{spon}}_{\mathrm{Cu_2O}} = \frac{1}{\tau_{\mathrm{Cu_2O}}}\frac{4\pi c\rho^2_{\mathrm{cond}}}{k_0^2\omega^2_{phn}\tau_{\mathrm{Cu_2O}}}. \tag{7}$$

In Cu_2O, the radiative lifetime of the exciton $\tau_{\mathrm{Cu_2O}}$ (see Eq.(2)) is approximately $10\mu s$; energy of optical phonon ω_{phn} is $10^{-2}\,\mathrm{eV}$. Taking for estimation $\rho_{\mathrm{cond}} = 10^{19}\,\mathrm{cm}^{-3}$ we have that the rate of photon creation in coherent *two-exciton* recombination process is:

$$W^{\mathrm{spon}}_{\mathrm{Cu_2O}} \approx \frac{1.5\cdot 10^{-2}}{\tau_{\mathrm{Cu_2O}}}\rho_{\mathrm{cond}}, \tag{8}$$

while the rate of photon creation in *one-exciton* recombination from the condensate process is $\tau^{-1}_{\mathrm{Cu_2O}}\rho_{\mathrm{cond}}$. This result shows that approximately every hundredth exciton in the condensate decays due to the process under consideration. Therefore, the spontaneous two-exciton recombination process is still weak in spite of extra bosonic (excitonic) factor $\sqrt{N_0}$ in the matrix element of the process (Fig.1c). But the process can be induced by resonant external radiation (with frequency $\omega = \mu$) propagating through exciton Bose-condensed system (Fig.1d). At any act of such excitons recombination there is created not only the photon propagating along inducing radiation direction but also the photon propagating in the opposite direction. Consequently, it emerges that the process effectively acts as a light *backscattering* from the condensate. The rate at which photons are emitted in the opposite direction is:

$$W_{-\mathbf{k}} = (N_{\mathbf{k}} + 1)W^{\mathrm{spon}}_{\mathbf{k}} \tag{9}$$

where N_k is the average number of quanta in the quantum state of inducing photon beam with the wave vector

k; the unity takes into account the spontaneous two-exciton recombination.

Consequently, inducing beam at the frequency $\omega = \mu$ which has approximately hundred quanta per mode makes two-exciton recombination luminescence in opposite direction be of the same intensity with one-exciton recombination luminescence (see Eq.(8)). This comment implies that the effect of stimulated backscattering of laser beam on exciton condensate can be detected experimentally.

In the absence of inducing beam two-exciton recombination luminescence is squeezed between the photon states with opposite momenta (two-mode squeezing). Luminescence at a given direction does not possess statistical coherence. The only statistical correlation, which luminescence has, is the correlation between opposite direction luminescence intensities. This correlation can be detected by Hunburry-Brown-Twiss measurements with two detectors arranged diagonally with respect to the exciton system.

2. Now we consider direct gap semiconductor with allowed interband transition such as GaAs. However, in GaAs, 3D excitons do not form Bose-condensate but, rather, gather in metallic electron-hole drops. Recently, GaAs excitons and their coherent properties are mainly studied in two dimensions – in coupled quantum wells (CQW). 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 quasi-long nondiagonal order superfluid phase with local quasi-condensate at temperatures smaller than the temperature of Kosterlitz-Thouless transition). CQW is a quasi-2D system where only *2D in-plane momentum is conserved.* The operator of exciton-photon interaction in interaction picture can be given as:

$$\hat{V}(t) = \hbar \sum_{\mathbf{k}} \frac{\mathrm{g}(\omega_{\mathbf{k}})}{\sqrt{L}} \left(\hat{a}_{\mathbf{k}_{\parallel}}(t) \hat{c}^{\dagger}_{\mathbf{k}} e^{-i\omega_{\mathbf{k}} t} + \mathrm{H.c.} \right) \tag{10}$$

where $\hat{a}(\hat{a}^{\dagger})$ and $\hat{c}(\hat{c}^{\dagger})$ are exciton and photon destruction (creation) operators, respectively, and $\mathbf{g}(\omega)$ is the interaction constant, vectors $\mathbf{k}_{\parallel}$ are two-dimentional; L is the width of the system in normal to CQW direction.

To the lowest order on exciton-photon interaction the spontaneous rate of the process with production of two photons with in-plane wave vector components $\pm\mathbf{k}_{\parallel}$ (see Fig.3a) is:

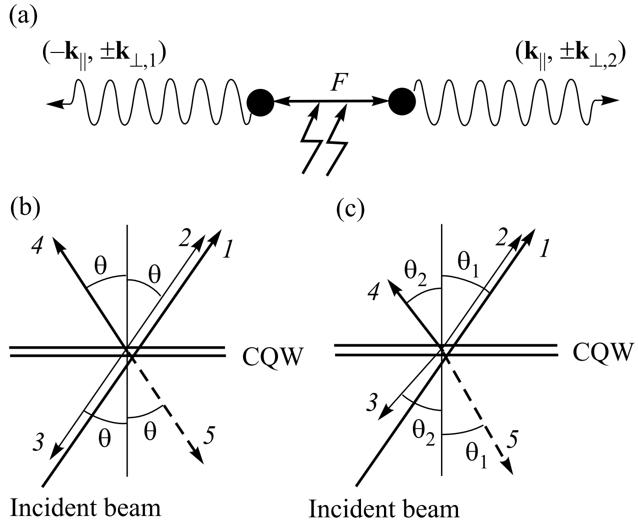

Fig.3. (a) The diagram of two-exciton recombination from 2D exciton condensate in QW or CQW. F is the anomalous Green function; wiggly lines represent photons with opposite in-plane wave vector components. (b) Laser beam induces two processes of two-exciton recombination which are different in the sign of normal wave vector components of photons with in-plane components $-k_{\parallel}$. Light also undergoes normal reflection on CQW planes. 1) Normally transmitted laser beam; 2) stimulated emission; 3) stimulated backscattering; 4) stimulated anomalous transmission; 5) normal reflection on CQW. (c) Nonresonant case, i.e. stimulating beam has energy substantially different from chemical potential of excitons $\hbar\mu$; the photons with in-plane components $-k_{\parallel}$ (c,d) have energies $\hbar\omega_2$ different from the energy of incident light ω_1. Normal components are also different. The angles θ_1, θ_2 obey the relation $\sin(\theta_1)/\sin(\theta_2) = \omega_2/\omega_1$. 1, 2, 3, 4, 5 – denote the same as in the part (b) of the Figure

$$W^{\mathrm{spon}}_{\mathrm{GaAs}}(\mathbf{k}_{\parallel}) = \int (2\pi)\delta(\omega_{\mathbf{k}_1} + \omega_{\mathbf{k}_1} - 2\mu) \times \\ \times |\mathbf{g}(\omega_{\mathbf{k}_1})\mathbf{g}(\omega_{\mathbf{k}_2})|^2 \left|F(\omega - \mu, \mathbf{k}_{\parallel})\right|^2 \times \\ \times dk_{\perp,1}(2\pi)^{-1} dk_{\perp,2}(2\pi)^{-1} \tag{11}$$

where F is anomalous Green function of exciton subsystem. For estimations we take anomalous Green function in the form:

$$F(\omega, \mathbf{k}_{\parallel}) = -\beta\left(\omega - \mu - \omega_{ex}(\mathbf{k}_{\parallel}) + i\eta\right)^{-1} \times \\ \times \left(\omega - \mu + \omega_{ex}(\mathbf{k}_{\parallel}) - i\eta\right)^{-1},$$

$$\omega_{ex}(\mathbf{k}_{\parallel}) = \sqrt{\beta\hbar k_{\parallel}^2 m_{ex}^{-1} + \left(\hbar k_{\parallel}^2 m_{ex}^{-1}\right)^2}, \quad \hbar\beta = \rho_{\mathrm{cond}} V_0,$$

where m_{ex} is the exciton mass (in GaAs, $m_{ex} \approx 0.22 m_0$, m_0 is the mass of free electron); ρ_{cond} is 2D density of excitons in the condensate; V_0 is the zero 2D Fourier component of exciton-exciton interaction potential; ω_{ex} is the dispersion of elementary excitation in exciton subsystem; η is the rate of decay of elementary excitation in exciton subsystem, for simplicity, we take this parameter be frequency independent. Energy of elementary excitation in exciton system is negligibly small in comparison with the gap between valence and conduction bands. Therefore, in Eq.(11) we can make the substitution:

$$\left|F(\omega-\mu,\mathbf{k}_{\|})\right|^2 \to$$
$$\to \frac{\pi(\delta(\omega-\mu-\omega_{ex}(\mathbf{k}_{\|}))+\delta(\omega-\mu+\omega_{ex}(\mathbf{k}_{\|})))\beta^2}{((2\omega_{ex}(\mathbf{k}_{\|}))^2+\eta^2)\eta} \approx$$
$$\approx \frac{2\pi\delta(\omega-\mu)\beta^2}{((2\omega_{ex}(\mathbf{k}_{\|}))^2+\eta^2)\eta}. \tag{12}$$

This approximation can be viewed as a resonant approximation. In resonant processes the photons are created with frequencies $\mu \pm \omega_{ex}(\mathbf{k}_{\|}) \approx \mu$. These magnitudes differ from μ negligibly. Therefore, the absolute values of photon wave vectors with frequencies $\mu+\omega_{ex}(\mathbf{k}_{\|})$ and $\mu-\omega_{ex}(\mathbf{k}_{\|})$ differ negligibly as well. Consequently, since they have the same absolute magnitude of in-plane wave vector components the angles of their propagation θ (see Fig.3b) are almost the same.

Substituting (12) into (11), one obtains:

$$W^{\rm spon}_{\rm GaAs}(\mathbf{k}_{\|}) = \frac{\beta^2}{((2\omega_{ex}(\mathbf{k}_{\|}))^2+\eta^2)\eta} \times$$
$$\times \left(\int 2\pi\delta(\omega_{\mathbf{k}_{\|},k_\perp}-\mu)|\mathrm{g}(\mu)|^2\frac{dk_\perp}{2\pi}\right)^2. \tag{13}$$

The magnitude in the parenthesis in nothing but radiative lifetime reciprocal $\tau^{-1}_{\rm GaAs}$ of isolated exciton with in-plane wave vector $\mathbf{k}_{\|}$. For spatially indirect exciton in GaAs CQW radiative lifetime $\tau_{\rm GaAs}$ approximately equals 10^{-8} sec. Letting $\tau_{\rm GaAs}$ be independent on $k_{\|}$ and assuming that entire radiative zone ($k_{\|} < k_0$) corresponds to linear part of elementary excitation spectrum in exciton subsystem, one can integrate $W^{\rm spon}_{\rm GaAs}(k_{\|})$ over radiative zone what yields total spontaneous rate of the process per unit area of CQW in the form:

$$W^{\rm spon}_{\rm GaAs} = \frac{\beta m_{ex}}{16\pi\hbar\eta\tau^2_{\rm GaAs}}\mathrm{Log}\left(1+\frac{4\beta\hbar k_0^2}{m_{ex}\eta^2}\right). \tag{14}$$

Taking for estimation exciton density $\rho_{cond} = 10^{10}\,\mathrm{cm}^{-2}$, $\hbar\beta = 0.5\,\mathrm{meV}$, $\eta = 10^{7\div10}\,\mathrm{sec}^{-1}$, $k_0 = 3\cdot10^5\,\mathrm{cm}^{-1}$, we have:

$$W^{\rm spon}_{\rm GaAs} \approx \frac{10^{2\div-2}}{\tau_{GaAs}}\rho_{\rm cond}.$$

This result shows (compare to Eq.(8)) that the effect of interest can be detected in the system of indirect excitons in GaAs/AlGaAs CQW.

Since in two dimensions the third component of photon wave vector is not fixed and is set only by energy conservation law, the rate Eq.(11) corresponds to *four processes* in which the photons with wave vectors $(\mathbf{k}_{\|}, \pm k_\perp)$ and $(-\mathbf{k}_{\|}, \pm k_\perp)$ are created. Therefore, stimulating laser light with in-plane wave vector component $\mathbf{k}_{\|}$ induces *two processes* in which the photons with in-plane components $\mathbf{k}_{\|}$ are emitted in two directions $(-\mathbf{k}_{\|}, \pm k_\perp)$, i.e. besides stimulated backscattering there arises *stimulated anomalous light transmission* in which only in-plane wave vector component changes its sign (see Fig.3b).

Furthermore, in the process of interest it is not necessary for the photons to be "resonant", i.e. the photons can have energies ω different from $\mu \pm \omega_{ex}(\mathbf{k}_{\|})$ (see Eq.(12)). However, the rate of this process is weak and for $|\omega-\mu| \gg \omega_{ex}(\mathbf{k}_{\|})$ the rate of the process in proportional to $(\omega-\mu)^{-4}$. In such processes the photons with different energies ω_1 and ω_2 have different normal wave components as well. The angles of their propagation obey the relation $\sin(\theta_1)/\sin(\theta_2) = \omega_2/\omega_1$ (see Fig.3c).

In conclusion, we predicted and estimated new optical effect of stimulated light backscattering on exciton Bose-condensate. Moreover, being detected this effect is nothing but the signature of exciton Bose-condensate since there is no other possibilities for the light to undergo backscattering from exciton subsystem. Estimations given in the Letter for Cu_2O excitons and excitons in GaAs/AlGaAs CQW show that the effect can be observed experimentally[2)]. The angle distribution in backscattering process is smeared by impurities or interface roughness due to that exciton momentum is not a good quantum number. The characteristic angle at which the photons are emitted can be given as $1/k_0 l$, where l is the mean free path of an exciton.

For the effect described to exist it is essential that exciton condensate be *quasi*-equilibrium, i.e. Bose-condensed system is not a ground state of the total system but it has excess energy which can be transferred to photons. For example, there is no such effect in the problem of Bose-condensate of atoms at their *ground state.* However, analogous effect can exist in the system of metastable Bose-condensed atoms. Studies of Bose-condensation in the systems of metastable atoms have been recently initiated.

[2)]Experimentally the effect of backscattering can be studied employing semitransparent mirror.

The work has been supported by INTAS, Russian Foundation of Basic Research and Program Solid State Nanostructures.

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