

Nutation effect in a system of coherent excitons, photons, and biexcitons in the region of the M band

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(Submitted 29 December 1979)

Pis'ma Zh. Eksp. Teor. Fiz. **29**, No. 4, 223-227 (20 February 1979)

The mechanism coherent conversion of excitons and photons into biexcitons and back to excitons and photons in the region of the M luminescence band of the semiconductors is investigated. It is shown that the conversion rate depends greatly on the initial state of the system.

PACS numbers: 71.35. + z

The action of a strong monochromatic wave on a two-level atomic system causes its population to vary periodically (nutate) with a frequency that depends on the matrix element of the dipole moment and on the field intensity. Burshtein and Pusep^[1] predicted the occurrence of a supernutation effect when the concentration of photons is much smaller than that of the two-level atoms. If the frequency of laser radiation is in resonance with the exciton level, then the excitons in the crystal will be produced faster than the nutation process in the individual atom. This situation imposes basically new constraints on the coherent nonlinear effects in the exciton region of the spectrum. Moskalenko *et al.*^[2] examined the coherent exciton-photon nutation effect, taking into account the exciton-exciton interaction and Davydov and Serikov^[3] studied it in the harmonic approximation in open quantum systems.

In this communication, we propose a method for identifying the biexcitons, which is based on the examined coherent exciton-biexciton nutation effect in the region of the M luminescence band for much shorter times than the relaxation time of the excitons and biexcitons. The picosecond pulses from the coherent resonance laser radiation can produce large concentrations of coherent biexcitons, which subsequently recombine radiatively and produce an exciton and light quanta in the region of the M luminescence band. We define exciton-biexciton nutation as the process of multiple conversion of coherent biexcitons into coherent excitons and photons and back into coherent biexcitons and the nutation frequency as the frequency of such conversions. The dynamics of this process differ significantly from those of exciton-photon nutation.

The Hamiltonian of the coherent exciton, photon, and biexciton system has the form

$$\frac{1}{\hbar} H = \Omega_{\mathbf{k}+\mathbf{q}} a_{\mathbf{k}}^+ a_{\mathbf{k}+\mathbf{q}} + \omega_{\mathbf{q}} b_{\mathbf{q}}^+ b_{\mathbf{q}} + c k C_{\mathbf{k}}^+ C_{\mathbf{k}} + i \frac{\kappa(\mathbf{k}, \mathbf{q})}{\sqrt{V}} \times (C_{\mathbf{k}}^+ b_{\mathbf{q}}^+ a_{\mathbf{k}+\mathbf{q}} - a_{\mathbf{k}+\mathbf{q}}^+ b_{\mathbf{q}} C_{\mathbf{k}}), \quad (1)$$

where a_k , b_k , and C_k are the annihilation operators of the biexciton, exciton, and photon, respectively, with the wave vector k ; $\hbar\Omega_k$ and $\hbar\omega_k$ are the energies of the biexciton and the exciton production, $\hbar ck$ is the photon energy, $g(k, q)$ is the matrix element of the optical conversion of an exciton into a biexciton,^[4,5] and V is the volume of the system. Since the oscillator power of the exciton-biexciton conversion is much greater than that of the exciton transition,^[4,5] we shall not examine the transitions of a crystal from the ground state to the excitonic state. We assume that the binding energy of the biexciton is sufficiently large, as in the case of the CuCl and CuBr crystals and that the M and A bands are separated sufficiently well. Since the states of the excitons, photons, and biexcitons are assumed to be macrofilled, we replace the operators by the time functions.

On the basis of the Heisenberg equations of motion for the operators, we obtain two independent first integrals of motion, which connect the concentrations of the biexcitons n_a , excitons n_b , and photons n_c :

$$n_a + n_b = \text{const}, \quad n_a + n_c = \text{const}. \quad (2)$$

Introducing the amplitudes and phases of the corresponding functions ($a = Ae^{i\phi}$, etc), we obtain a set of second-order, nonlinear differential equations for the amplitude A and the phase ϕ of the biexcitons (henceforth, for simplicity we drop the wave-vector indices of the operators and the values associated with them):

$$\begin{aligned} \ddot{A} - A\dot{\phi}^2 - (\Omega + \omega + ck)A\dot{\phi} - \Omega(\omega + ck)A + \frac{g^2}{V}(B^2 + C^2)A &= 0, \\ A\ddot{\phi} + 2\dot{A}\dot{\phi} + (\Omega + \omega + ck)\dot{A} &= 0, \end{aligned} \quad (3)$$

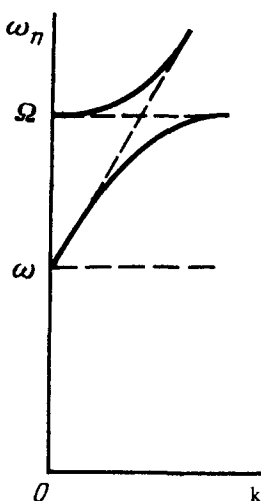


FIG. 1. Polariton dispersion curves for optical transitions in the region of the M band of biexciton luminescence as a function of the wave vector k of a photon.

where the dot denotes differentiation with respect to time.

Equation (3) allows steady-state solutions of the form

$$A = \text{const}, \quad B = \text{const}, \quad C = \text{const}, \quad \dot{\phi} = -\omega_p = \text{const}, \quad (4)$$

where

$$\omega_p = \frac{1}{2}(\Omega + \omega + ck) \pm \frac{1}{2}\sqrt{(\Omega - \omega - ck)^2 + 4g^2(n_b + n_c)}. \quad (5)$$

In the case of stationary excitation, Eq. (5) can be considered a polariton-type dispersion law for optical exciton-biexciton transitions in the region of the M band, which has two branches whose location and shape depend on the equilibrium concentrations of the excitons n_b and photons n_c , i.e., on the excitation level of the crystal (Fig. 1). It is assumed that in Fig. 1 the wave vector of the exciton q is given, the wave vector of the photon k varies, and the wave vector of the biexciton is equal to the sum of the wave vectors of the exciton and photon. The upper branch of the dispersion curves represents the quasi energy of the biexciton and the lower branch denotes the quasi energy of the photon plus exciton in the region $ck + \omega < \Omega$. The maximum splitting of the branches occurs in the actual region of the k space, where $\Omega - \omega = ck$, and its magnitude depends on the equilibrium concentrations of the excitons and photons.

Equation (3) also has nonsteady-state solutions. Assuming that the resonance detuning is equal to zero ($\Delta = \Omega - \omega - ck = 0$) and the initial concentrations of the excitons, photons, and biexcitons, respectively, are n_{b0} , n_{c0} , and n_{a0} , we obtain the following expression for the nutation frequency ω_H :

$$\omega_H = \frac{\pi g \sqrt{n_{max}}}{K(k)}, \quad k^2 = \frac{n_{min}}{n_{max}}, \quad (6)$$

$$n_{min}^{max} = \frac{min}{max}(n_{a0} + n_{b0}, n_{a0} + n_{c0}),$$

where $K(k)$ is the (total) first-order elliptic integral^[6] with the modulus k and n_{min} and n_{max} , respectively, are the minimum and maximum of the two expressions in the parentheses; $0 \leq n_a \leq n_{min}$, $n_{a0} + n_{b0} - n_{min} \leq n_b \leq n_{a0} + n_{b0}$, and $n_{a0} + n_{c0} - n_{min} \leq n_c \leq n_{a0} + n_{c0}$. The concentrations of the excitons, photons, and biexcitons vary periodically with the frequency ω_H . One of the values, n_b or n_c , decreases to zero due to the oscillations, whereas the other oscillates above the constant background. This is attributable to the fact that at $n_{b0} \neq n_{c0}$ only the smaller value determines the additional number of the produced biexcitons. The nutation frequency ω_H depends in a complicated way on the initial concentration of the system. At a constant concentration of the biexcitons, the nutation frequency is determined by the modulus of the difference in concentrations of the excitons and photons $\Delta n = |n_{b0} - n_{c0}|$. In the neighborhood of $\Delta n = 0$ we have $\omega_H = \pi g (n_{max})^{1/2} / \ln [4(n_{max} / \Delta n)^{1/2}]$. As $\Delta n \rightarrow 0$, the nutation frequency decreases and vanishes and when Δn increases it increases mono-

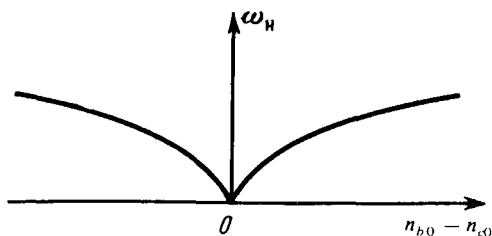


FIG. 2. Dependence of the nutation frequency of the coherent excitons, photons, and biexcitons on the difference in the initial concentrations of excitons and photons at a specified initial concentration of biexcitons.

tonically (Fig. 2). At $n_b = n_c$, the system is devoid of oscillations and the concentration increases monotonically from n_{a0} to $n_{a0} + n_{b0}$ and then remains constant. If we set $n_{a0} \approx n_{b0} \ll n_{c0}$ or $n_{a0} \approx n_{c0} \ll n_{b0}$, then we can easily obtain $\omega_H = 2g(n_{a0} + n_{c0})^{1/2}$ or $\omega_H = 2g(n_{a0} + n_{b0})^{1/2}$, respectively, i.e., the nutation frequency varies as a function of the larger concentration according to the square-root law. At $n_{a0} = 0$, Eq. (3) has a simple solution

$$n_a = n_{a \min} \operatorname{sn}^2(\sqrt{n_{a \max}} g t), \quad \omega_H = \frac{\pi g \sqrt{n_{a \max}}}{K(k)},$$

$$k^2 = \frac{n_{a \min}}{n_{a \max}}, \quad n_{a \min} = \min(n_{b0}, n_{c0}),$$

where $\operatorname{sn}(x)$ is an elliptic function^[6] with the modulus k . The nutation frequency, as before, is determined by the larger concentration and the variation amplitude n_a is determined by the smaller concentration. The dependence of ω_H on Δn is shown in Fig. 2. As in Ref. 1, we define super-nutation in the system of coherent excitons, photons, and biexcitons as a continuous increase of the nutation frequency with increasing concentration of photons (or excitons).

At nonzero resonance detuning, the nutation frequency depends on the magnitude of detuning. This, however, does not produce fundamentally new effects as compared with the case $\Delta = 0$.

It would be of particular interest to prove experimentally the predicted variation of the nutation frequency as a function of the excitation level of the crystal. An increase of the exciton-biexciton nutation frequency (super-nutation) increases the induced polariton transparency effect in the region of the M band of biexciton luminescence. In conclusion, we note that the coherent exciton-biexciton nutation effect is much more abundant than the exciton-photon nutation effect or the nutation in the two-level atoms.

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