Formation of dissipative polariton structures in semiconductors

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Using the Keldysh equations, which describe weakly inhomogeneous (in space and time) excitons and photons, we predict the possibility of formation of dissipative structures in a system of coherent polaritons in semiconductors. In the slowly varying envelope approximation we derive a system of equations that describe the evolution of coherent quasiparticles and allow for external pumping and damping. We obtain uniform time-independent solutions and study their stability. We establish the conditions in which the uniform solutions for coherent excitons and photons become unstable and, as aresult of which a superlattice of coherent polaritons is formed. Finally, we give estimates for the CdS crystal.

1. INTRODUCTION

The existence of giant nonlinearities and short relaxation times for excitons in semiconductors has recently stimulated research in the field of coherent nonlinear effects at the long-wave intrinsic-absorption edge of a crystal in connection with resonant exciton excitation.

Coherent nonlinear phenomena in the excitonic range of the spectrum have their own specific features and differ significantly from similar phenomena in two-level systems.

The fact is that when the exciton number density is not very high, i.e., when we can think of excitons as bosons, the Hamiltonian representing the exciton–photon interaction is quadratic while the dependence of the electromagnetic field amplitude E on the exciton wave amplitude a is linear: $E \propto a$. This clearly distinguishes the exciton problem from the model of two-level atoms, where the Hamiltonian representing the interaction of light with a two-level medium is cubic and the system exhibits natural nonlinearity.

With excitons the nonlinearity is caused by the dynamic and kinematic exciton–exciton interactions. Here the space–time evolution of dipole-active excitons in crystals is described by the well-known system of Keldysh equations,[1] which play the role of the Maxwell–Bloch equations for two-level systems. The system consists of equations of the Ginzburg–Landau type and describes the coherent exciton and photon states that vary slowly in space and time. The Keldysh equations served as a basis for studying many aspects of coherent nonlinear propagation of light in dense condensed media in the excitonic region of the spectrum. For one thing, by employing these equations we constructed[2–11] a theory of self-induced transparency (SIT) in the excitonic range of the spectrum, a theory of optical bistability (OB) and optical switching, and a theory of periodic and stochastic self-pulsations. The SIT and OB phenomena in the excitonic range of the spectrum have been observed in experiments.[12,13]

This paper is devoted to a new cooperative nonlinear phenomenon in condensed media, the occurrence of dissipative structures in a system of coherent excitons and photons (polaritons). The occurrence of spontaneous dissipative structures is under extensive study in various fields of physics, chemistry, and biology.[14,15]

The possibility of a dissipative structure appearing as a superlattice of the exciton number density in a system of mechanical incoherent high-number-density excitons was first demonstrated by Sugakov,[16] who established the formation thresholds, the properties, and conditions for stability of the one-dimensional superlattice that develops owing to the dynamical attractive interaction of excitons.

In contrast to Ref. 16, here we study dissipative structures in a more complex system, a system of dipole-active coherent excitons and photons, and the formation of polariton superlattices.

2. STATEMENT OF THE PROBLEM AND BASIC EQUATIONS

Suppose that a plane monochromatic wave is incident on a cavity whose mirrors are the polished faces of the crystal under investigation. The wave excites the cavity mode coupled to the excitons. The interaction of the active substance with the coherent external pumping radiation and the heat sink, which ensures that relaxation processes take place, will be taken into account at a certain stage phenomenologically.

The system of equations that describe the slowly varying (in space and time) coherent excitons and photons without allowing for external pumping and dissipation effects was derived by Keldysh.[1] For waves propagating parallel to the x-axis this system has the form

$$i\frac{\partial a}{\partial t}=\left(\Omega_\perp-\frac{\hbar}{2m}\frac{\partial^2}{\partial x^2}+\frac{g|a|^2}{\hbar V}\right)a-\frac{d}{\hbar}E^+, \tag{1}$$

$$c^2\frac{\partial^2E^+}{\partial x^2}-\frac{\partial^2E^+}{\partial t^2}=\frac{4\pi d}{v_0}\frac{\partial^2 a}{\partial t^2}, \tag{2}$$

where $a(x,t)$ is the coherent-exciton amplitude, $E^+(x,t)$ is the positive-frequency part of the alternating electromagnetic field, g is the exciton–exciton coupling constant, d is the dipole moment of the transition from the ground state of the crystal to the excitonic state, m is the translational electron mass, v_0 is the volume of a unit cell, V is the volume of the crystal, and $\Omega_\perp$ is the cutoff transverse-exciton frequency.

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We write the macroscopic amplitudes of the excitons and the field in the form of modulated plane waves with a carrier frequency ω and a wave vector k:

$$a(x,t)=\sqrt{V}\tilde{A}\exp(ikx-i\omega t), \tag{3}$$

$$E^{+}(x,t)=\sqrt{V/v_0}\tilde{\varepsilon}\exp(ikx-i\omega t), \tag{4}$$

where the slowly varying functions $\tilde{A}$ and $\tilde{\varepsilon}$ are the envelopes of the respective wave packets.

Further discussion will be conducted in the framework of the slowly varying envelope approximation, which holds if

$$\left|\frac{\partial\tilde{A}}{\partial t}\right|\ll\omega|\tilde{A}|, \quad \left|\frac{\partial\tilde{A}}{\partial x}\right|\ll k|\tilde{A}|, \tag{5}$$

etc. This means that the wave packet envelopes are fairly smooth functions in comparison to the rapidly oscillating part. The envelopes change little over one wavelength and during one time period of the light impinging on the crystal.

Substituting (3) and (4) into Eqs. (1) and (2), we arrive in the slowly varying amplitude approximation at the following system of equations:

$$\frac{\partial\tilde{A}}{\partial t}=i\left(\omega-\Omega_{\perp}-\frac{g}{\hbar}|\tilde{A}|^2\right)\tilde{A}-\gamma_{ex}\tilde{A}+i\frac{d}{\hbar\sqrt{v_0}}\tilde{\varepsilon}-\frac{\hbar k}{m}\frac{\partial\tilde{A}}{\partial x}, \tag{6}$$

$$\frac{\partial\tilde{\varepsilon}}{\partial t}=i\frac{2\pi\omega d}{\sqrt{v_0}}\tilde{A}+i\frac{\omega^2-c^2k^2}{2\omega}\tilde{\varepsilon}-'\gamma\tilde{\varepsilon}+\tilde{\varepsilon}_0-\frac{kc^2}{\omega}\frac{\partial\tilde{\varepsilon}}{\partial x}, \tag{7}$$

where γ_{ex}, γ, and $\tilde{\varepsilon}_0$ are the exciton and photon damping factors and the amplitude of the external coherent pumping field (all three quantities are introduced into Eqs. (6) and (7) phenomenologically). These equations provide a complete description of the space–time evolution of coherent excitons and photons in the presence of external pumping and damping. In the spatially homogeneous case they coincide with the equations for the exciton and photon amplitudes in Ref. 17. These were derived strictly within the quantum theory of fluctuations and damping from the flux part of the corresponding Fokker–Planck equation without the fluctuation terms.

In what follows it is more convenient to deal with dimensionless quantities. Allowing for the fact that in the most general case the amplitudes $\tilde{A}$ and $\tilde{\varepsilon}$ are complex-valued quantities and introducing

$$z=\frac{\tilde{A}}{A_0}=z_3+iz_4, \quad y=\frac{\tilde{\varepsilon}}{\varepsilon_0}=z_1+iz_2,$$

$$\delta=\frac{\omega-\Omega_{\perp}}{\gamma_{ex}}, \quad \Delta=\frac{\omega^2-c^2k^2}{2\pi\gamma_{ex}\omega}, \quad A_0=\sqrt{\frac{\hbar\gamma_{ex}}{|g|}},$$

$$\varepsilon_0=\sqrt{2\pi\hbar\omega}\,A_0, \quad \tau=\gamma_{ex}t, \quad \Omega_0=\frac{4\pi d^2}{\hbar v_0}, \tag{8}$$

$$\sigma=\frac{\gamma}{\gamma_{ex}}, \quad \nu=\frac{g}{|g|}=\pm1, \quad \alpha=\sqrt{\frac{\omega\Omega_0}{2\gamma_{ex}^2}},$$

$$P=\frac{\tilde{\varepsilon}_0}{\gamma_{ex}\varepsilon_0}, \quad \beta=\frac{\hbar k}{m\gamma_{ex}}, \quad \chi=\frac{kc^2}{\omega\gamma_{ex}},$$

we obtain from Eqs. (6) and (7) the following:

$$\frac{\partial z_1}{\partial\tau}=-\sigma z_1-\Delta z_2+\alpha z_4+P-\chi\frac{\partial z_1}{\partial x}, \tag{9}$$

$$\frac{\partial z_2}{\partial\tau}=\Delta z_1-\sigma z_2+\alpha z_3-\chi\frac{\partial z_2}{\partial x}, \tag{10}$$

$$\frac{\partial z_3}{\partial\tau}=-\alpha z_2-z_3-[\delta-(z_3^2+z_4^2)]z_4-\beta\frac{\partial z_3}{\partial x}, \tag{11}$$

$$\frac{\partial z_4}{\partial\tau}=\alpha z_1+[\delta-(z_3^2+z_4^2)]z_3-z_4-\beta\frac{\partial z_4}{\partial x}, \tag{12}$$

For uniform time-independent solutions we have

$$z_1=\frac{[\sigma(\delta-n)+\Delta](\delta-n)+[\alpha^2+\sigma-\Delta(\delta-n)]}{[\sigma(\delta-n)+\Delta]^2+[\alpha^2+\sigma-\Delta(\delta-n)]^2}P, \tag{13}$$

$$z_2=\frac{[\sigma(\delta-n)+\Delta]-(\delta-n)[\alpha^2+\sigma-\Delta(\delta-n)]}{[\sigma(\delta-n)+\Delta]^2+[\alpha^2+\sigma-\Delta(\delta-n)]^2}P, \tag{14}$$

$$z_3=-\frac{\alpha P[\sigma(\delta-n)+\Delta]}{[\sigma(\delta-n)+\Delta]^2+[\alpha^2+\sigma-\Delta(\delta-n)]^2}, \tag{15}$$

$$z_4=\frac{\alpha P[\alpha^2+\sigma-\Delta(\delta-n)]}{[\sigma(\delta-n)+\Delta]^2+[\alpha^2+\sigma-\Delta(\delta-n)]^2}, \tag{16}$$

where $n=|z|^2=z_3^2+z_4^2$ is the dimensional exciton number density.

3. STABILITY OF THE UNIFORM TIME-INDEPENDENT DISTRIBUTION OF QUASIPARTICLES IN THE CRYSTAL

To study the stability of the uniform time-independent solution with respect to small perturbations we put

$$\delta y\propto\exp(\lambda t+iqx), \quad \delta z\propto\exp(\lambda t+iqx). \tag{17}$$

Then the characteristic equation for the uniform time–independent states of the system has the form

$$D=\begin{vmatrix} \lambda+\sigma & -(q\chi+\Delta) & 0 & \alpha \\ q\chi-\Delta & \lambda+\sigma & -\alpha & 0 \\ 0 & \alpha & \lambda+1-2z_3z_4 & A-2z_4^2-\beta q \\ -\alpha & 0 & -A+2z_3^2+\beta q & \lambda+1+2z_3z_4 \end{vmatrix}=0. \tag{18}$$

For simplicity we assume that the frequency ω of the external electromagnetic field coincides with the natural frequency ck of the cavity mode, i.e., $\Delta=0$. For the time-independent solutions to be stable, all roots of Eq. (18) must have negative real parts. This requires that the following inequalities hold:

$$2(1+\sigma)>0,$$

$$\sigma Q^2+C+B+(1+\sigma)^3+(1+\sigma)(\sigma+\alpha^2)>0,$$

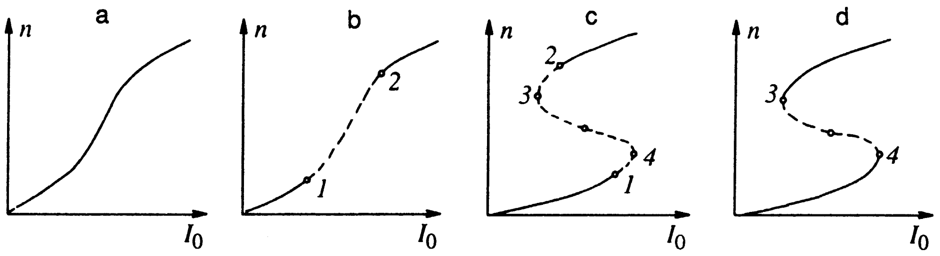

FIG. 1. Exciton concentration vs the intensity of external coherent pumping for $\alpha>\sigma$, $\delta<\delta_1$ (a), $\alpha>\sigma$, $\delta_1<\delta<\delta_2$ (b), $\alpha>\sigma$, $\delta>\delta_2$, (c) and $\alpha<\sigma$, $\delta>\delta_2$ (d).

$$(\sigma+\alpha^2)^2+\sigma^2(B+C)+Q^2(1+B+C)-2\alpha^2QF>0, \tag{19}$$

$$\sigma Q^2+(1+\sigma)^2(2\sigma+\alpha^2)(Q^2+B+C)$$
$$+\sigma(B+C)(B+C-2Q^2)+2\alpha^2QF(1+\sigma)^2$$
$$+(1+\sigma)^4(\sigma+\alpha^2)>0,$$

where

$$A=\delta-n,\ B=(\delta-n)(\delta-3n),$$
$$C=\beta q(4n-2\delta+\beta q),\ F=2n-\delta+\beta q.$$

Further investigations require establishing the values of the reduced wave vector $Q=\chi q$ at which the stationary states determined by the external pumping become unstable.

To determine the stability region we performed a computer experiment. For a given pair of values of certain parameters, the dimensionless intensity $I_0=|P|^2$ of the field incident on the crystal and the pair of parameters I_0,Q or n,Q, the validity of the inequalities (19) was checked. If at least one inequality was found to be violated, the respective point was marked on the $I-Q$ diagram. The shaded regions in Figs. 2–5 below mark the ranges of the parameter values at which the uniform time-independent states become unstable. On the boundaries of such regions $\mathrm{Re}(\lambda_i)=0$. Outside the shaded regions the system is stable.

In the region of unstable parameters, any small deviation from uniform time-independent values leads to formation of a distinctly periodic spatial structure of quasiparticles with a period $d=2\pi/q$, i.e., formation of a polariton superlattice.

4. DISCUSSION AND CONCLUSIONS

If spatial dispersion is ignored and the field is spatially homogeneous, there can be two critical resonance detuning values, δ_1 and δ_2, determining the dependence of the stationary exciton number density on external pumping:

$$\delta_1=\sqrt{3}(\sigma+1),\quad \delta_2=\sqrt{3}\left(\frac{\alpha^2}{\sigma}+1\right). \tag{20}$$

For $\alpha>\sigma$ and $\delta<\delta_1$ all stationary points are stable, and the $n(I_0)$ dependence is one-to-one [Fig. 1(a)]. If $\delta_1<\delta<\delta_2$, on the stationary $n(I_0)$ curve [Fig. 1(b)] a range of values $I_1<I_0<I_2$ occurs for which the stationary points lose their stability, with I_1 and I_2 given by the following expressions:

$$I_{1,2}=\frac{2\delta^3\sigma^2}{27\alpha^2}\left[1+3v\mp\left(1+\frac{\bar{\omega}-3v}{2}\sqrt{1-\bar{\omega}}\right)\right],$$
$$\bar{\omega}=3\left(\frac{\sigma+1}{\delta}\right)^2,\quad v=3\left(\frac{\sigma+\alpha^2}{\sigma\delta}\right)^2. \tag{21}$$

For $\alpha\sigma$ and $\delta>\delta_2$, the system of coherent excitons and photons becomes optically bistable, with the turning point I_3 and I_4 being [Fig. 1(c)]

$$I_{3,4}=\frac{2\delta^3\sigma^2}{27\alpha^2}[1+3v\mp(1-v)^{3/2}]. \tag{22}$$

In this case, instability windows appear on both the lower branch of optical bistability and the upper branch. The lower

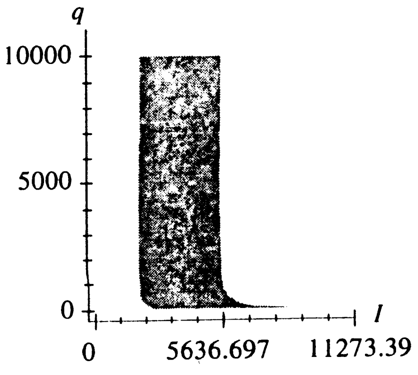

FIG. 2. Wave vector vs the intensity $I(n)$ of external pumping at $\alpha=22.9$, $\delta=10$, $\sigma=10$, and $\beta/\chi=0$.

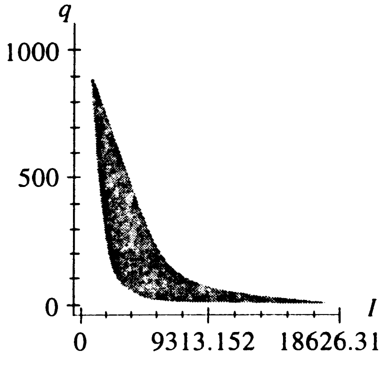

FIG. 3. Wave vector vs the intensity $I(n)$ of external pumping at $\alpha=22.9$, $\delta=10$, $\sigma=10$, and $\beta/\chi=0.01$.

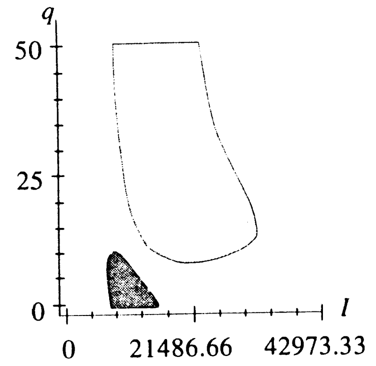

FIG. 4. Wave vector vs the intensity $I(n)$ of external pumping at $\alpha=22.9$, $\delta=30$, $\sigma=10$, and $\beta/\chi=0.01$.

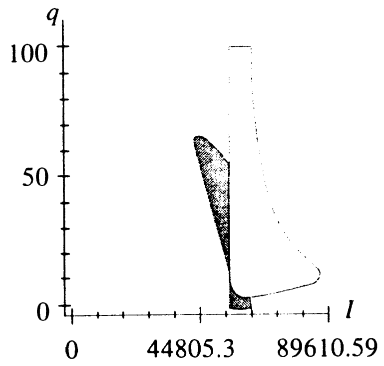

FIG. 5. Wave vector vs the intensity $I(n)$ of external pumping at $\alpha=22.9$, $\delta=110$, $\sigma=10$, and $\beta/\chi=0.01$.

branch is stable for $0 < I_0 < I_1$ and unstable in interval $I_1 < I_0 < I_4$. The upper branch is stable for $I_0 > I_2$ and unstable for $I_3 < I_0 < I_2$.

For $a<\sigma$ at $\delta<\delta_2$ there is no bistability in the system, and the entire $n(I_0)$ curve is stable. When the detuning satisfies $\delta > \delta_2$, the upper and lower optical bistability branches are stable [Fig. 1(d)].

Allowing for spatial dispersion and field nonuniformity leads to the following results. Figures 2 and 3 depict the wave vector values as functions of external pumping at $\sigma=10$, $\delta=10$, and $\alpha=22.9$ (the case where $\alpha > \sigma$ and $\delta < \delta_1$). Clearly, allowing for spatial dispersion and field nonuniformity leads to spatial instability and formation of a superlattice for parameter values at which the $n(I_0)$ was formerly stable.

Figure 4 depicts the $q(I)$ dependence for $\delta=30$, $\alpha=22.9$, and $\sigma=10$. We see that for such values of the parameters there are two regions of spatial instability.

Figure 5 is the diagram for the case where optical bistability occurs in the system of coherent excitons and photons for $\sigma=10$, $\delta=110$, and $\alpha=22.9(\alpha>\sigma,\delta>\delta_2$. In this case two superlattices with periods $d_h\sim10^{-4}-10^{-5}$ m and $d_l\sim 2(10^{-4}-10^{-5})$ m for the upper and lower optical bistability branches, respectively, appear in the system. One superlattice is formed from a low-density exciton domain, and the other from a high-density exciton domain.

To conclude this paper, we list the numerical estimates for a CdS crystal: $g=2.4\times10^{-32}$ erg·cm^{-3}, $\gamma_{ex}\sim3\times10^{11}$ s^{-1}, $\gamma\sim3\times10^{12}$ s^{-1}, and $\Omega_0=10^{-4}\Omega_\perp=4\times10^{11}$ s^{-1}. For these parameter values the period of the superlattice lies in the range 10–100 μm for exciton densities $n\sim10^{17}$ cm^{-3}. In this case the intensity of external pumping at which polariton superlattices can form in the crystal is in the 10–100 mW/cm^2 range, respectively.

Thus, allowing for the spatial dispersion of excitons and the spatial variation of the field gives rise to spatial instability and to the appearance of a polariton superlattice formed by coherent excitons and photons.

[1] L. V. Keldysh, in *Problems of Theoretical Physics: In Memory of I. E. Tamm*, Nauka, Moscow (1972), p. 433 [in Russian].
[2] S. A. Moscalenco, Kh. Rotaru, and P. I. Khadzhi, Opt. Commun. **23**, 367 (1977).
[3] S. N. Belkin, P. I. Khadzhi, S. A. Moscalenco, and A. H. Rotaru, J. Phys. C **14**, 4109 (1981).
[4] A. Kh. Rotaru, P. I. Khadzhi, M. I. Baznat, and G. D. Shibarshina, Fiz. Tverd. Tela (Leningrad) **29**, 535 (1987) [Sov. Phys. Solid State **29**, 304 (1987)].
[5] S. A. Moscalenco, A. H. Rotaru, Yu. M. Shvera, and V. A. Zaloj, Phys. Status Solidi B **149**, 187 (1988).
[6] S. A. Moscalenco, A. H. Rotaru, and V. A. Zaloj, Phys. Status Solidi B **150**, 401 (1988).
[7] V. A. Zalozh, S. A. Moskalenko, and A. Kh. Rotaru, Zh. Eksp. Teor. Fiz. **95**, 601 (1989) [Sov. Phys. JETP **68**, 338 (1989)].
[8] S. A. Moskalenko, V. A. Zalozh, and A. Kh. Rotaru, Fiz. Tverd. Tela (Leningrad) **31**, 265 (1989) [Sov. Phys. Solid State **31**, 500 (1989)].
[9] B. Sh. Parkanskiı and A. Kh. Rotaru, Zh. Eksp. Teor. Fiz. **99**, 899 (1991) [Sov. Phys. JETP **72**, 499 (1991)].
[10] B. Sh. Parkanskiı and A. Kh. Rotaru, Fiz. Tverd. Tela (Leningrad) **33**, 3378 (1991) [Sov. Phys. Solid State **33**, 1905 (1991)].
[11] B. Sh. Parkanskiı and A. Kh. Rotaru, Fiz. Tverd. Tela (Leningrad) **33**, 2250 (1991) [Sov. Phys. Solid State **33**, 1268 (1991)].
[12] F. Bryukner, Ya. T. Vasil'ev, V. S. Dneprovskiı, D. G. Koshchug, and E. K. Silina, Zh. Eksp. Teor. Fiz. **67**, 2219 (1974) [Sov. Phys. JETP **40**, 1101 (1975)].
[13] G. P. Golubev, V. S. Dneprovskiı, E. A. Kiselev, Z. D. Kovalyuk, and V. A. Stadnik, Dokl. Akad. Nauk SSSR **280**, 591 (1985) [Sov. Phys. Dokl. **30**, 71 (1985)].
[14] M. I. Rabinovich and D. I. Trubetskov, *Oscillations and Waves in Linear and Nonlinear Systems*, Kluwer Academic, Dordrecht (1989).
[15] Yu. M. Romanovskiı, N. V. Stepanova, and R. S. Chernavskiı, *Mathematical Modeling in Biophysics*, Nauka, Moscow (1975) [in Russian].
[16] V. I. Sugakov, Fiz. Tverd. Tela (Leningrad) **28**, 2441 (1986) [Sov. Phys. Solid State **28**, 1365 (1986)].
[17] P. D. Drummond and D. F. Walls, J. Phys. A **13**, 725 (1980).

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