Stratification of a coherent state of excitons sustained by optical pumping

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An instability of a coherent state of Wannier–Mott excitons sustained by a resonant pump wave is analyzed. The instability involves a spatially periodic stratification with a period $L \sim L_{ex} = (\hbar/m\gamma)^{1/2}$, where m is the effective exciton mass, and γ is the reciprocal exciton lifetime.

1. A coherent state of Wannier–Mott excitons sustained by a resonant electromagnetic wave has been studied in several papers. A basic system of equations for the system consisting of coherent excitons and an electromagnetic field was constructed by Keldysh.[1] Elesin and Kopaev[2] analyzed a concentration bistability of coherent excitons for the case of a given field, with recombination processes. Zalozh *et al.*[3] and Parkanskiĭ and Potaru[4] studied spatially uniform temporal instabilities and corresponding high-frequency autopulsations of the concentration of excitons and of the transmission of light in a bistable optical resonator filled with a medium with coherent excitons.

Our purpose in the present letter is to analyze spatially nonuniform instabilities in a system of coherent Wannier–Mott excitons in the cases with and without a resonator.

2. In the given field of a uniform monochromatic pump wave, we have the following equation for the wave function Φ, which describes the macroscopic quantum state of the system of excitons (recombination processes are taken into account in accordance with Ref. 1):

$$i\frac{\partial\Phi}{\partial t}+i\frac{\gamma}{2}\Phi-[(\omega_0-\omega)\Phi-\varkappa\,|\Phi|^2\,]\Phi+\frac{\hbar}{2m}\nabla^2\Phi=\frac{1}{\hbar}d\varepsilon_0. \tag{1}$$

Here ω is the frequency of the pump wave, $\omega_0-\omega\equiv\Delta$ is the frequency detuning from the frequency of the exciton transition, γ is the reciprocal lifetime of an exciton, $\varkappa$ is the coefficient of the nonlinear interaction of excitons, m is the effective mass of an exciton, d is the dipole matrix element for the exciton transition, ε_0 is the amplitude of the pump field (Φ and ε_0 are normalized in such a way that $|\Phi|^2$ and $|\varepsilon_0|^2$ give us, respectively, the densities of coherent excitons and of photons in the pump wave), and $\hbar$ is Planck's constant. Equation (1) holds under the condition $Nr_0^3\ll 1$ (r_0 is the radius of the exciton, and N is the concentration of excitons) at time scales $\delta t\gg\omega^{-1}$ and spatial scales $\delta r\gg N^{1/3}$. The length scale $L_{ex}=(\hbar/m\gamma)^{1/2}$ can be distinguished in Eq. (1). We assume that we are dealing with a thin semiconducting film of thickness $l< L_{ex}$ which is pumped by a wave which is uniform in the (X,Y) plane and which is propagating along the normal to the surface of the film (along the Z axis). We assume a uniform distribution of excitons along Z; we are ignoring the absorption of the wave over the thickness of the film ($\alpha l < 1$, where α is the absorption coefficient for the pump wave).

As of yet there are no reliable experimental data on the parameter $\varkappa$ (Ref. 5). In principle, the coefficient of the nonlinear interaction of excitons could have either sign. The instability in which we are interested here occurs for either sign of $\varkappa$.

Uniform steady-state solutions Φ_0 of Eq. (1) are given by

$$\Phi_0 = \frac{d\varepsilon_0}{(i\gamma/2 + \Delta - \varkappa|\Phi_0|^2)\hbar} \text{ or } |\Phi_0|^2 = \frac{|d\varepsilon_0|^2}{[(\gamma/2)^2 + (\Delta - \varkappa|\Phi_0|2)^2]\hbar^2}. \quad (2)$$

The equation for $|\Phi_0|^2$ can have from one to three solutions, depending on the parameters of the medium and on the pump intensity. In the three-solution case, which prevails under the conditions $|\Delta| > \gamma\sqrt{3}/2$ and $\varkappa\Delta > 0$, the plot of $|\Phi_0|^2$ versus $|\varepsilon_0|^2$ is S-shaped,[2] and an average of the three values cannot be taken. We can thus say that the system is bistable.

Let us examine the stability of a uniform, steady, and otherwise arbitrary state of the system, Φ_0, with respect to small space–time fluctuations. We write Φ in the form

$$\Phi = \mathscr{F} e^{i\Psi}, \quad \mathscr{F} = \Phi_0 + f, \quad \Psi = \Psi_0 + \psi.$$

Linearizing Eq. (1) with respect to f and ψ, and choosing $f \sim \psi \sim \exp(\Omega t + kX)$, we find a dispersion relation for the fluctuations along the X axis:

$$\Omega = -\frac{\gamma}{2} \pm i\frac{\gamma}{2}\left[\left(\frac{2\Delta}{\gamma} - \frac{6\varkappa|\Phi_0|^2}{\gamma} + L_{\text{ex}}^2 k^2\right)\left(\frac{2\Delta}{\gamma} - \frac{2\varkappa|\Phi_0|^2}{\gamma} + L_{\text{ex}}^2 k^2\right)\right]^{1/2}. \quad (3)$$

The quantity Re $\Omega(k)$ is a measure of the stability of the system. Specifically, the system is stable if Re $\Omega(k) < 0$ for all k. If both expressions instead in the radical in (3) are positive, we have Re $\Omega(k) = -\gamma/2$; i.e., the system is stable. If these expressions in parentheses differ in sign in some interval of k, two real branches of Re $\Omega(k)$ appear. At a certain value of the adjustable parameters $|\varepsilon_0|^2$ and Δ, an instability occurs: Re $\Omega(k) > 0$ (Fig. 1). We wish to stress that we have Im $\Omega(k) = 0$ for these values of k. If we choose $|\varkappa|\,|\Phi_0|^2 = \gamma/2$, then the curve of Re $\Omega(k)$ just touches the abscissa, and at this threshold point we have the characteristic value $k_{\text{th}} = (2 \text{ sign } \varkappa - 2\Delta/\gamma)^{1/2} L_{\text{ex}}^{-1}$ (in the case $\varkappa < 0$ we have $\Delta < 0$). Working from (3), we can also, and easily, find the threshold pump value ε_{th}, but we will not reproduce that expression here. Just above the critical value, a quasiharmonic static stratification of the concentration of excitons and of the phase of the coherent state, with a characteristic period on the order of L_{ex}, should arise in the semiconductor as the result of the instability. With $m \sim 0.1 m_0$ (m_0 is the mass of a free electron) and $\gamma \sim 10^9$ s^{-1}, we have $L_{\text{ex}} \sim 10^{-4}$ cm. If $r_0 \approx 0.5 \times 10^{-6}$ cm, and if we choose the pump and the other parameters in such a way that the average exciton concentration is $N_0 \sim 10^{17}$ cm^{-3}, then the conditions for the applicability of this analysis, $r_0 \ll N^{1/3} \ll L_{\text{ex}}$, are satisfied. As the inequality Re $\Omega(k) > 0$ becomes stronger with increasing pump intensity, the instability interval along the k scale expands, and the structure which arises should

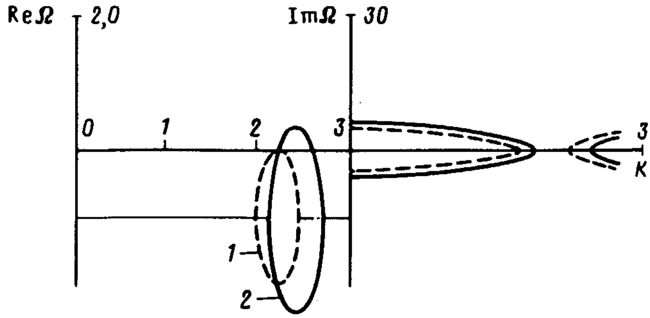

FIG. 1. Solution of the dispersion relation for model (1). *1*—$\Delta/\gamma=-1.5$, $\varkappa|\Phi_0|^2=\gamma/2$; *2*—$\Delta/\gamma=-1.5$, $\varkappa|\Phi_0|^2=1.4\gamma/2$. Here and in Fig. 1, Ω is in units of $\gamma/2$, and k in units of $L_{\rm ex}^{-1}$.

deviate from a quasiharmonic status and should take the form of a system of exciton clusters. The nonlinear form of the one- and two-dimensional structures requires a special analysis.

At $\varkappa>0$, the instability occurs in both the monostable and bistable cases. In the latter case, with $\Delta>\gamma$, this instability corresponds to the upper branch of the plot of $|\Phi_0|^2$ versus $|\varepsilon_0|^2$, while in the case $\sqrt{3}/2<\Delta/\gamma<1$ it corresponds to both branches. If $\varkappa$ has the opposite sign, the instability occurs on the lower branch of the S-shaped curve.

Lugiato and Lefever[6] have studied a similar instability. This was an instability of the electromagnetic field in a Fabry–Perot interferometer with an instantaneous-response nonlinear Kerr medium. That instability was transverse with respect to the propagation direction of the pump wave. The onset of the instability results in the formation of a system of self-focusing filaments. In our case we can speak in terms of a self-focusing instability of the exciton polarization field. The circumstance that in both Ref. 6 and our own case the instability occurs for either sign of the nonlinearity is a consequence of the presence of the additional parameter Δ, which can be given either sign. A fluctuational appearance of focusing or defocusing inhomogeneities, a diffractive spreading of the field, and a local change in the detuning (and thus in the pumping of excitons) as result of the nonlinearity are involved in the formation of the structure.

3. We also consider a system consisting of coherent excitons and an electromagnetic field in a Fabry–Perot resonator. In contrast with Ref. 6, we consider a dynamic system of coherent excitons with a spatial dispersion of their own, instead of an instantaneous-response Kerr medium. Our model is

$$i\frac{\partial\Phi}{\partial t}+i\frac{\gamma}{2}\Phi-[\Delta-\varkappa|\Phi|^2]\Phi+\frac{\hbar}{2m}\nabla^2\Phi=-\frac{1}{\hbar}d\varepsilon, \tag{4}$$

$$i\frac{\partial\varepsilon}{\partial t}+i\frac{\Gamma}{2}\varepsilon-\tilde{\Delta}\varepsilon+\frac{c}{2K_0}\nabla^2\varepsilon=\nu\epsilon_0-\frac{2\pi\omega}{c}d\Phi. \tag{5}$$

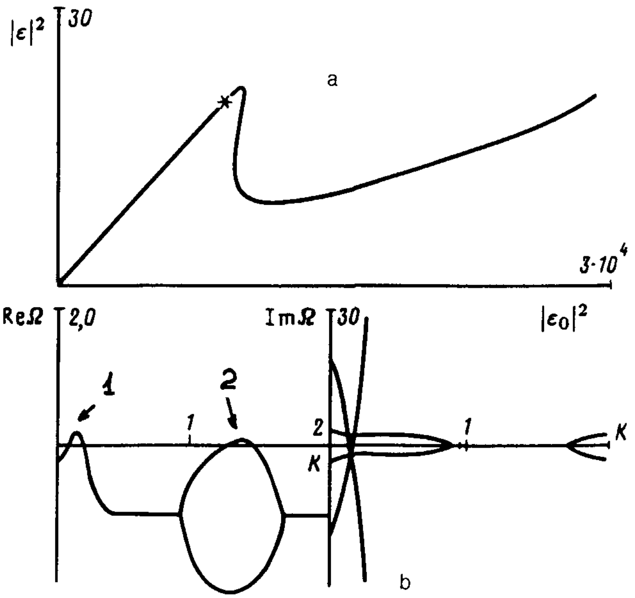

FIG. 2. a: Steady-state uniform intensity of the internal field in the resonator versus the pump power, b: Solution of the dispersion relation for the model in (4), (5). The parameter values are $\Gamma/\gamma=25$, $\Delta/\gamma=-2$, $\tilde{\Delta}/\gamma=-10$, $(8\pi\omega/c\gamma^2\hbar)d^2=30$, and $(L_f/L_{ex})^2=1000$. The intensity of the internal field in the resonator is in units of $\gamma^3\hbar^2/8d^2\varkappa$, and the pump power in units of $\gamma^5\hbar^2/32d^2\varkappa\nu^2$. The asterisk in panel *a* marks the point $(\kappa|\Phi_0|^2=-1.04\gamma/2)$, which corresponds to the dispersion curves in panel *b*.

Here ε is the field in the interferometer, which is described in the single-mode approximation and which is averaged over distances on the order of the optical wavelength λ and over times on the order of ω^{-1}. The field ε has been normalized in such a way that $|\varepsilon|^2$ gives us the density of photons in the resonator. The spatial derivatives of the field equation describe the diffraction of light in the plane of the interferometer. The parameter Γ characterizes the attenuation of the field in the resonator; $\tilde{\Delta}\equiv\tilde{w}_0-w$ is the frequency detuning of the pump field with respect to the frequency $\tilde{\omega}_0$, which is one of the longitudinal modes of the resonator (along the Z axis), c is the velocity of light in the semiconductor (we are ignoring here the contribution of this exciton state to the refractive index), $K_0=2\pi/\lambda$ is the wave number of the selected resonator mode, $\nu=c(2R)^{-1}$, and R is the dimension of the resonator along the Z axis. The notation is otherwise the same as in Eq. (1).

A length scale of field equation (5) is $L_f=(c/K_0\Gamma)^{1/2}$. With $\Gamma\sim10^{11}$ s^{-1} and $\lambda\sim10^{-4}$ cm we have $L_f\sim10^{-3}$ cm and $L_f\gg L_{ex}$. Uniform steady states and uniform fluctuations of a system like (4), (5) were studied in Refs. 3 and 4. We have studied the dispersion relation for $\Omega(k)$ for various parameter values. The equation itself is quite lengthy, and we will not reproduce it here. Part *a* of Fig. 2 shows an example of the behavior of the intensity of the internal field in the resonator as a function of the pump power; part *b* shows a solution of the dispersion relation for model (4), (5) for the case in which the two spatial instabilities occur simultaneously. The length scales of these instabilities are L_f and L_{ex}. Field peak *1* on the Re $\Omega(k)$ curve corresponds to the case Im $\Omega(k)\neq 0$, while near exciton peak *2* we have Im $\Omega(k)=0$. An instability with respect to the excitation of a wave of the field ε and an instability with respect to the stratification of excitons can thus coexist. When the parameters are varied, we also find a case in which the crest of peak *1* in Fig. 2b corresponds to the value $k=0$, and we have Im $\Omega(0)\neq 0$. In this case the system is unstable with respect to uniform oscillations and stratification simultaneously. The dispersion curves become more complex as the parameters γ and Γ move closer together.

In samples whose dimensions along the X and Y axes are comparable to L_f and L_{ex}, the allowed values of the fluctuation wave number k become discrete, and the dimensions may have a strong influence on whether the instabilities occur.

The most suitable systems for an experimental observation of these instabilities would be multilayer systems with quantum wells, in which narrow exciton absorption peaks are observed even at room temperature, and for which the mechanism of the exciton–exciton interaction is the dominant mechanism in the observed optical-bistability effects.

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