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LETTER TO THE EDITOR

Stratification and switching fronts in a coherent state of excitons sustained by optical pumping

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Abstract. Instabilities of a coherent state of Wannier–Mott excitons sustained by a resonant pumping wave in cavity and cavityless cases are analysed. In particular, instability with respect to spatially periodic stratification can exist. The period of such stratification is $L \sim L_{ex} = [\hbar/m\gamma]^{1/2}$, where m is the effective exciton mass, and γ is the reciprocal exciton lifetime. The results of numerical modelling of corresponding non-linear structures are presented. For 'exciton-pumping' bistability, non-monotonic switching fronts are demonstrated numerically.

A coherent state of Wannier–Mott excitons sustained by a resonant electromagnetic wave has been discussed in several papers. Equations for the system consisting of coherent excitons and an electromagnetic field were constructed first in [1]. A concentration bistability of coherent excitons for the case of a given field was analysed in [2]. Spatially uniform temporal instabilities and corresponding high-frequency self-pulsations of exciton concentration and of the transmission of light in a bistable optical resonator filled with a medium with coherent excitons were studied in [3]. A detailed description of exciton condensate and an analysis of its linear response were also given in [4] and [5]. Delicate experiments with a coherent exciton state in quantum well structures and ultra-short light pulses are described in [6] and [7].

Our goal in the present letter is to analyse spatial instabilities in a system of coherent Wannier–Mott excitons with and without a resonator and to demonstrate several corresponding non-linear effects.

In the given field of a uniform monochromatic pumping wave $E = E_0 \exp(\mathrm{i}\omega t)$, we have the following equation for the wave function Φ, which describes the macroscopic quantum state of excitons (recombination processes are taken into account):

$$\mathrm{i}\partial\Phi/\partial t + \mathrm{i}(\gamma/2)\Phi - [\Delta_{ex} - \theta|\Phi|^2]\Phi + (\hbar/2m)\nabla_\perp^2\Phi = (1/\hbar)dE_0. \qquad (1)$$

Here $\Delta_{ex} \equiv \omega_{ex} - \omega$ is the field frequency detuning from the frequency of exciton transition; γ is the reciprocal lifetime of an exciton; θ is the coefficient of non-linear interaction of excitons; m is the effective mass of an exciton; d is the dipole matrix element for the exciton optical transition; $\hbar$ is Planck's constant. Functions Φ and E_0 are normalized in such a way that $|\Phi|^2$ and $|E_0|^2$ are, respectively, the densities of coherent excitons and of photons in the pumping wave. Laplacian $\nabla_\perp^2$ operates in the system plane (X, Y), which is transverse to the direction of pumping wave propagation.

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Equation (1) is complemented by the boundary conditions (one-dimensional case)

$$\partial\Phi/\partial X|_{0,L} = 0 \tag{2}$$

which corresponds to the impermeability of boundaries ($X = 0, X = L$) for excitons.

Equation (1) holds under the condition $N_0 r_0^3 \ll 1$ (r is the radius of the exciton, N_0 is the averaged concentration of excitons) at time scales $\delta t \gg \omega^{-1}$ and spatial scales $\delta r \gg N_0^{1/3}$. The characteristic length $L_{ex} = (\hbar/m\gamma)^{1/2}$ can be distinguished in (1). We assume that we are dealing with a thin semiconductor film of thickness $l < L_{ex}$; we ignore the absorption of the wave over the thickness of the film, therefore the distribution of excitons along the film thickness is uniform.

In principle, the coefficient of the non-linear interaction of excitons, θ, could have either sign. Non-trivial behaviour of exciton condensate is possible with either sign.

The concentration of excitons in an homogeneous steady state, Φ_s, from (1) is:

$$|\Phi_s|^2 = |dE_0|^2/\{\hbar^2[(\gamma/2)^2 + (\Delta_{ex} - \theta|\Phi_s|^2)^2]\}. \tag{3}$$

Equation (3) can have from one to three solutions, depending on parameters of the medium and on pumping intensity. Three solutions exist if $|\Delta| > \gamma\sqrt{3}/2$ and $\theta\Delta > 0$. The plot of $|\Phi_s|^2$ versus $|E_0|^2$ is S shaped [2], and the middle of three roots cannot be taken, so the system is bistable.

Now we examine the stability of an homogeneous steady state Φ_s with respect to small space–time exponential ($\sim \exp(\Omega t + \mathrm{i}kX)$) fluctuations. Linearizing (1) we have the following dispersion equation:

$$\Omega = -\frac{\gamma}{2} \pm \mathrm{i}\frac{\gamma}{2}\left[\left(\frac{2\Delta_{ex}}{\gamma} - \frac{6\theta|\Phi_s|^2}{\gamma} + L_{ex}^2k^2\right)\left(\frac{2\Delta_{ex}}{\gamma} - \frac{2\theta|\Phi_s|^2}{\gamma} + L_{ex}^2k^2\right)\right]^{1/2}. \tag{4}$$

The function $\mathrm{Re}\,\Omega(k)$ characterizes the stability of the system. The system is stable if $\mathrm{Re}\,\Omega(k) < 0$ for all wave numbers k. If both expressions in brackets in (4) are positive, $\mathrm{Re}\,\Omega(k) = -\gamma/2$, i.e. the system is stable. If these expressions differ in sign in some interval of k, two real branches of $\mathrm{Re}\,\Omega(k)$ appear. At certain values of the adjustable parameters $|E_0|^2$ and Δ_{ex} an instability occurs; $\mathrm{Re}\,\Omega(k) > 0$ (figure 1), and $\mathrm{Im}\,\Omega(k) = 0$ for these values of k. The curve of $\mathrm{Re}\,\Omega(k)$ just touches the abscissa axis if $|\theta||\Phi_s|^2 = \gamma/2$. Using (3) we can find the threshold pumping value E_{th}. At this threshold point the characteristic value $L_{ex}k_{th} = (\mathrm{sign}\,\theta - 2\Delta_{ex}/\gamma)^{1/2}$ (if $\theta < 0$ for instability we have to choose $\Delta_{ex} < 0$). As the result of the instability just above E_{th} a quasiharmonic static stratification of the concentration of excitons and of the phase of the coherent state with a characteristic spatial period of the order of L_{ex} should arise in the semiconductor. If $\gamma \sim 10^9\ \mathrm{s}^{-1}$ and $m \sim 0.1m_0$ (m_0 is the mass of a free electron), then $L_{ex} \sim 10^{-4}$ cm. If $r_0 \simeq 0.5 \times 10^{-6}$ cm, and the pumping and other parameters are chosen in such a way that the average exciton concentration $N_0 \sim 10^{17}\ \mathrm{cm}^{-3}$, then our analysis is valid ($r_0 \ll N_0^{1/3} \ll L_{ex}$). When the unequality $\mathrm{Re}\,\Omega(k) > 0$ becomes stronger and the instability interval along the k axis expands, the structure takes the form of a system of exciton clusters and non-stationary regimes can exist.

At $\theta > 0$, the instability occurs in both monostable and bistable cases. In the latter case, with $\Delta_{ex} > \gamma$, the instability corresponds to the upper branch of the S-shaped plot of $|\Phi_s|^2$ versus $|E_0|^2$; if $\sqrt{3}/2 < \Delta_{ex}/\gamma < 1$ it corresponds to both branches. If $\theta < 0$, the instability occurs on the lower branch of the S-shaped curve.

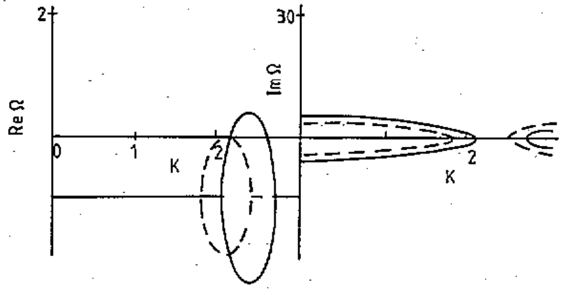


Figure 1. Solution of the dispersion equation (4). Parameters: dashed line, $\Delta_{ex}/\gamma = -1.5, \theta|\Phi_s|^2 = \gamma/2$; solid line, $\Delta_{ex}/\gamma = -1.5, \theta|\Phi_s|^2 = 1.4\gamma/2$. Here and in figure 5 Ω is in units of $\gamma/2$, and k is in units of L_{ex}^{-1}.

The results of numerical modelling of a non-linear process of structure forming is shown in figure 2. If the size of a sample along the X axis is finite, due to boundary conditions (2) the allowed values of transverse wave number k become discrete, and different wave numbers k can be in the instability interval. In particular, for a small enough sample there is no wave number in the instability interval, and the system is stable. Figure 2, where the coordinate dependence of function $|\Phi|^2$ is shown for different moments in time, corresponds to one unstable transverse mode.

A similar field instability in a Fabry–Perot interferometer with an inertialess medium was studied in [8]. It leads to the formation of self-focusing filaments, so we can speak in terms of a self-focusing instability of the exciton polarization field. In both [8] and our work the instability occurs for either sign of the non-linearity, and it is a consequence of the presence of the additional parameter Δ_{ex}, which can be chosen of either sign.

Due to inhomogeneities in the medium, structures can arise even when the stratification instability is not achieved in or out the region of inhomogeneity. The amplitude of the structure decreases on going away from inhomogeneity. Such a structure, connected with the local change of parameter ω_{ex}, is shown in figure 3. In real systems with a great number of defects (which lead to the inhomogeneities of ω_{ex} and γ) complicated structure can exist in the pre-stratification case. However, if the stratification instability is realized, numerical simulation shows that regular structures arise only with small defects near the inhomogeneities.

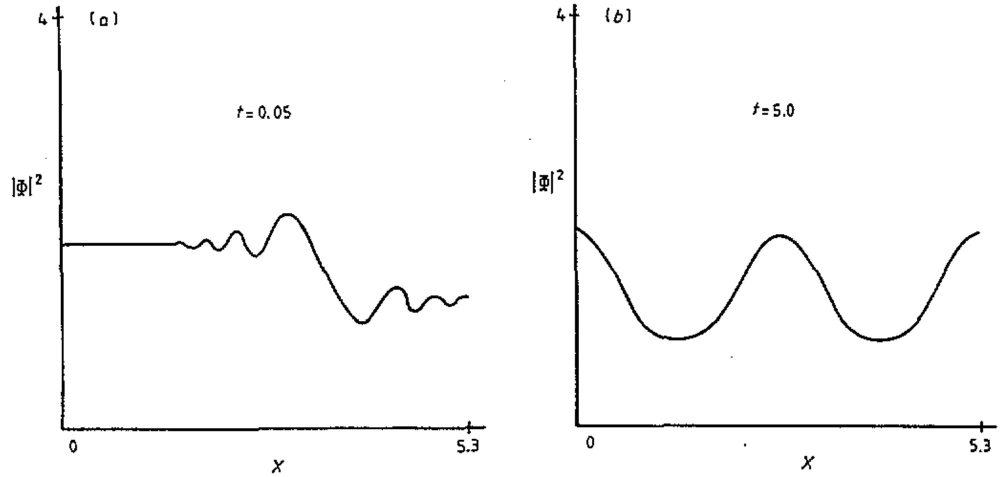
In the bistable case switching fronts exist between the upper and lower branches of the S-shaped curve of $|\Phi_s|^2$ versus $|E_0|^2$. A characteristic feature of the front is the spatial oscillations near the front. This is shown in figure 4 for the case when both upper and lower states are stable. In bistable systems with diffusion (for example, the heat reversible breakdown of a semiconductor) the switching fronts are usually monotonic [9]. The coherent exciton state is described by an equation of wave type and it alone determines the non-monotonic shape of the front.

For the 'coherent excitons in a Fabry–Perot resonator' system the following model is used:

$$\mathrm{i}\partial\Phi/\partial t + \mathrm{i}(\gamma/2)\Phi - [\Delta_{ex} - \theta|\Phi|^2]\Phi + (\hbar/2m)\nabla_\perp^2\Phi = -(1/\hbar)d\mathcal{E} \tag{5}$$

$$\mathrm{i}\partial\mathcal{E}/\partial t + \mathrm{i}(\Gamma/2)\mathcal{E} - \Delta_f\mathcal{E} + (c/2K)\nabla_\perp^2\mathcal{E} = \nu E_0 - (2\pi\omega/c)d\Phi. \tag{6}$$

Here $\mathcal{E}$ is the field in the interferometer, which is described in the approximation of one longitudinal mode and which is averaged over distances of the order of the optical



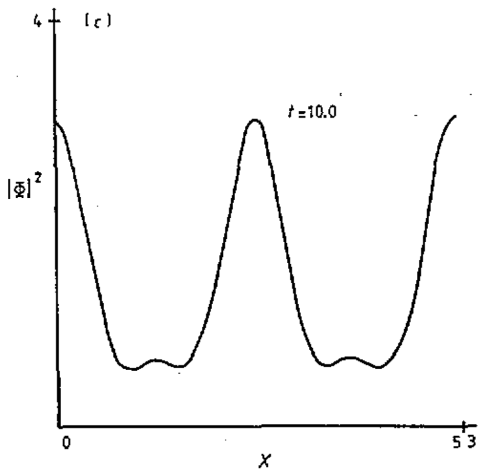


Figure 2. Structure forming in the case of one unstable transverse mode. Parameters: $\Delta_{ex}/\gamma = -1.5$; $\theta|\Phi_s|^2 = 1.3\gamma/2$; $L = 5.3L_{ex}$. In figures 2–4 the time is in units of $2/\gamma$ and $|\Phi|^2$ is in units of $\gamma/(2|\theta|)$.

wavelength λ and over times of the order of ω^{-1}. The field $\mathcal{E}$ has been normalized in such a way that $|\mathcal{E}|^2$ is the density of photons in the resonator. The term with spatial derivatives in (6) describes the diffraction of light in the plane of the interferometer. The parameter Γ characterizes the attenuation of the field in the cavity; $\Delta_f \equiv \omega_f - \omega$ is the frequency detuning of the pumping field with respect to the frequency of the longitudinal mode ω_f; c is the velocity of light in the semiconductor (without the contribution of the exciton state to refractive index); $K = 2\pi/\lambda$ is the wave number of the longitudinal mode; $\nu = c(2L)^{-1}$; L is the resonator length. Other notations are the same as in (1).

The characteristic length of the field in (3) is $L_f = (c/K\Gamma)^{1/2}$. With $\Gamma \sim 10^{11}$ s^{-1} and $\lambda \sim 10^{-4}$ cm we have $L_f \sim 10^{-3}$ cm and $L_f \gg L_{ex}$. Homogeneous steady states, optical bistability and homogeneous linear fluctuations of a system such as (2), (3) were studied in [3].

We have studied the dispersion equation for $\Omega(k)$ for various parameter values. This equation is quite lengthy, and we will not reproduce it here. An example of the dependence of the internal field intensity on pumping power and the solution of the dispersion equation are shown in figure 5. There are two instabilities, and their scales are L_f and L_{ex}. Field peak

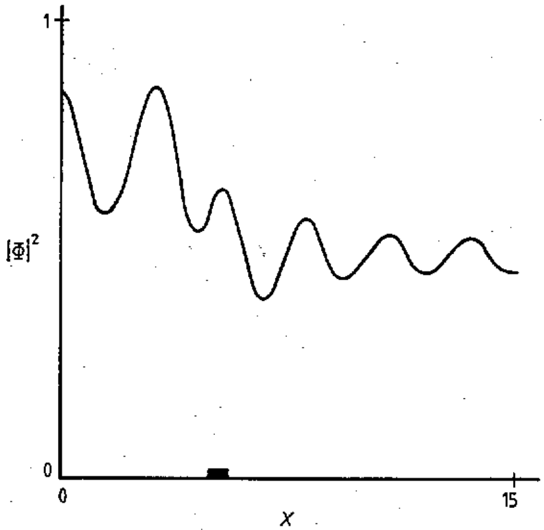


Figure 3. The stationary structure near the inhomogeneity of parameter ω_{ex} in the pre-stratification case. The bold segment of the abscissa axis marks the region of inhomogeneity. Parameters: $\theta|\Phi_s|^2 = 0.7\gamma/2$; $L = 15L_{ex}$. $\Delta_{ex}/\gamma = -2$ in the interval $5L_{ex} < L < 5.6L_{ex}$; otherwise $\Delta_{ex}/\gamma = -1.5$.

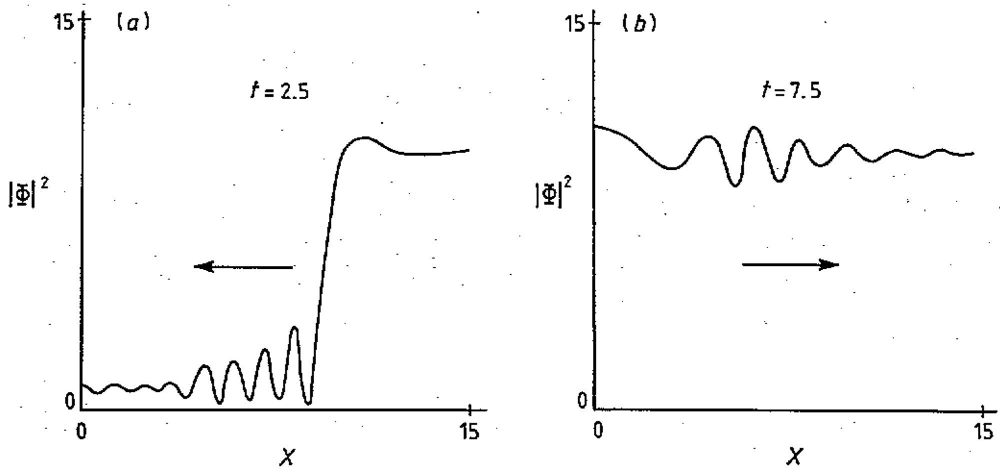


Figure 4. (a) Switching front in the bistable case. (b) The post-front effect (back-propagating wave packet), which is a result of interaction of the front and the boundary. Parameters: $\Delta_{ex}/\gamma = -4$; $\theta|\Phi_s|^2 = -0.8\gamma/2$; $L = 18L_{ex}$. The arrows mark the direction of propagation.

1 on the $\mathrm{Re}\,\Omega(k)$ curve corresponds to $\mathrm{Im}\,\Omega(k) \neq 0$, while near the exciton stratification peak 2 $\mathrm{Im}\,\Omega(k) = 0$. An instability with respect to the excitation of a standing wave of the field $\mathcal{E}$ and an instability with respect to the stratification of excitons can thus coexist. Varying parameters we also find a case in which peak 1 in figure 5(b) corresponds to the instability with $k = 0$, and $\mathrm{Im}\,\Omega(k = 0) \neq 0$. Then the system is unstable with respect to uniform oscillations and stratifications simultaneously.

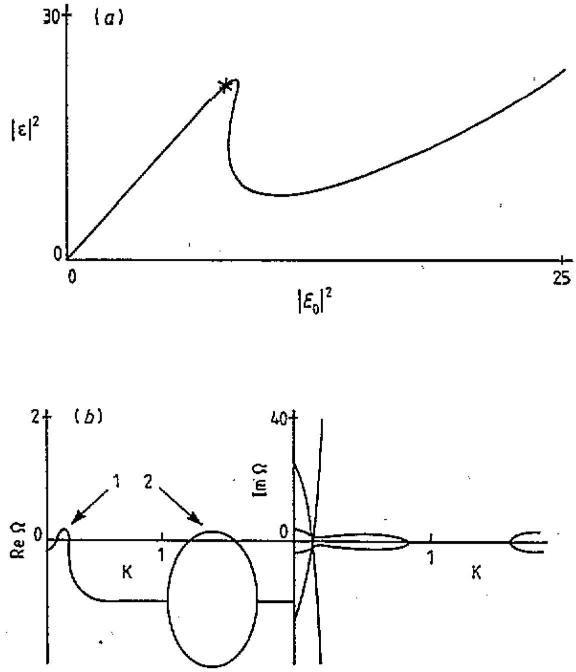


Figure 5. (*a*) Steady-state uniform intensity of the internal field in the resonator versus the pumping; (*b*) solution of the dispersion equation for the model (5), (6). Parameters: $\Gamma/\gamma = 25$, $\Delta_{ex}/\gamma = -2$, $\Delta_f/\gamma = -10$, $(8\pi\omega/c\gamma^2\hbar)d^2 = 30$, $(L_f/L_{ex})^2 = 1000$. The intensity of the internal field in the resonator is in units of $\gamma^3\hbar^2/8d^2|\theta|$, the pumping power is in units of $\gamma^5\hbar^2/32^2 d\theta^2\nu$. The asterisk in (*a*) marks the point $(\theta|\Phi_s|^2 = -1.04\gamma/2)$, which corresponds to the dispersion curve in (*b*).

We suppose that the multilayer systems with quantum wells are suitable for an experimental observation of these instabilities. In such systems narrow exciton absorption peaks are observed even at room temperature, and the exciton–exciton interaction is the dominant mechanism in the non-linear optical effects observed.

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