Dynamical phase transitions and pattern formation induced by a pulse pumping of excitons to a system near a thermodynamic instability

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We suggest a phenomenological theory of dynamical phase transitions and the subsequent spaciotemporal evolution induced by a short optical pulse in a system which is already prone to a thermodynamic instability. We address the case of pumping to excitons whose density contributes additively to the thermodynamic order parameter like for charge-transfer excitons in electronic charge-ordering transitions. To describe both thermodynamic and dynamical effects on equal footing, we adopt for the phase transition a view of the "excitonic insulator" (EI) and suggest a formation of the macroscopic quantum state for the pumped excitons. The double nature of the ensemble of excitons leads to an intricate time evolution: the dynamical transition between number-preserved and phase-locked regimes, macroscopic quantum oscillations from interference between the Bose condensate of excitons, and the ground state of the EI. Modeling for an extended sample shows also stratification in domains of low and high densities which evolve through local dynamical phase transitions and a sequence of domain merges.

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I. INTRODUCTION

Phase transformations induced by short optical pulses is a new mainstream in studies of cooperative electronic states, see, e.g. [1–3]. In experiments on pump-induced phase transitions (PIPTs) in electronic systems, usually the initial pumping proceeds among depths of filled and empty electronic bands, i.e. via excitations at the photon energy E_{ph} well above the electron-hole (e-h) gap E_g. After an immeasurably rapid cooling down to an energy bottleneck, the next observable stage follows when a distribution is formed of electrons and holes near the upper and lower rims of the gap interval E_g. At the further stage, the inverted population of fermions initiates the very well-observed evolution of the electronic spectra and of their population together with collective degrees of freedom. The latter usually are related to the symmetry breaking in the virgin state which is responsible for the origin of the gap E_g from interactions among electrons or with the lattice. Such most commonly studied states are high- T_c superconductors, Mott and Peierls insulators, charge density waves, charge-ordered states, etc. This is the most explored PIPT scenario described in many publications, e.g. in collections [1–3].

There is a less common case when the transformation is provoked by subgap optical excitations—the excitons—which can be viewed as bound e-h pairs whose energy $E_{\rm ex}$ lies below E_g. These excitons can be produced by absorption of photons whose energy $E_{\rm ph}$ lies in between: $E_{\rm g} > E_{\rm ph} > E_{\rm ex}$. The common case of pumping to high-energy unbound electrons and holes, when $E_{\rm ph} > E_{\rm g}$, is not excluded from this consideration, provided the early cooling leads promptly to binding of e-h pairs with formation of excitons. Experimental concentration of excitons can reach a very high value of 10% per unit cell [4–8]. This brings new opportunities to study the mixed dynamics of the PIPT and of the ensemble of excitons with opening to coherent effects, namely to formation of the quasi-condensate of excitons.

By now, experiments with the ultrafast pumping to excitons have been restricted to the so-called neutral-ionic transitions, see [4–8] and references therein, but actually the range of realizations is unlimited since all nonmetallic systems prone to phase transitions possess also excitons available for pumping.

Photons can be tuned to generate excitons of different origin—intramolecular excitations (Frenkel excitons) or intersite ones (called charge-transfer of Wannier-Mott excitons). A very interesting situation emerges when the transition order parameter and the intensity of pumping excitations are of the same origin, e.g. for the low-energy charge transfer excitons [8] in media with the neutral-ionic transition, both the optical excitation and the thermodynamic long-range ordering are built from processes of electronic transfer between donor and acceptor molecules. That will bring about the duality of the Bose-Einstein condensate (BEC) of excitons [9] and of the excitonic insulator (EI) ground state.

In this paper, we exploit the concept of the EI to resolve a challenging question, taking into account simultaneously and upon the unique bases the two interweaved natures of excitons, as the reservoir of pumped excitons in a coherent state and as the stable condensate forming the thermodynamic ground state.

II. COHERENT ENSEMBLES OF EXCITONS: FROM THE EI TO THE BEC

The EI has been suggested as a hypothetical phase of a semiconductor or a semimetal, and this term became the common nickname for a state formed by appearance of the e-h condensate, see [10–14]. This notion has been revived nowadays as a convenient interpretation of phase transitions

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in various electronic materials [15–17]. We also recall the old suggestions and attempts to reach the EI state by means of extreme conditions such as high magnetic field and pressure (see [18] for experiments and [19,20] for peculiarities in theory).

Semantically, it would be most logical to treat the EI as a result of a metal-insulator transition. At the origin of these theories, that was called also the Keldysh-Kopaev state [11,13] when the gap is formed on the small Fermi surface of a semimetal. In a similar sense, this term is used in contemporary experiments by means of time-resolved angle-resolved photoemission spectroscopy (ARPES) in layered systems [21–23]. Here, the EI is associated with a gap-opening state which resembles the charge density wave (CDW) but is formed primarily by a self-consistent undulation of the electronic density rather than by a more conventional lattice periodicity, which is commonly called the Peierls insulator. Nevertheless, in the origin of this notion and in most basic theoretical studies [10,13,24], the term EI has been used in another sense: a transition among two insulating phases which can happen if the energy of the elementary exciton drops to zero $E_{\rm ex} \to 0$. In this sense, the notion of the EI has been employed in more recent, and relevant to us, studies of semiconductors under a strong stationary electromagnetic field [25]. This is the terminology which we shall follow in this article: the EI appears if the binding energy $E_{\rm b}$ exceeds the single-particle gap $E_{\rm g}$, hence the total energy of the exciton $E_{\rm ex} = E_{\rm g} - E_{\rm b}$ becomes negative. It has been well established since [13,14] that these two limiting definitions of the EI are actually the two corners of the single-phase diagram which starts at negative $E_{\rm g}$ (the semimetal) and ends at the positive $E_{\rm g} > 0$ (the semiconductor) while at still negative $E_{\rm ex} < 0$.

The theories of the thermodynamic EI phase just below the transition and of the BEC of optically pumped excitons (neglecting their decay) are closely related at the microscopic level. They differ by the respective monitoring parameters: either the chemical potential of excitons for the EI or their density for the BEC. For a system under the stationary optical pumping, this duality has been noticed in theory of optical Stark effect in semiconductors, see [25] for a review and references. The similarity among states of the densely pumped excitons and the thermodynamically stable phase of the EI appeared already in earlier microscopic theories, see, e.g. [26]; a short but very clear review and complete references can be found in [27].

In this paper, we deal with an intriguing situation where the excitons are pumped to the system which itself is already prone to formation of the excitonic condensate as the thermodynamic ground state. To describe both thermodynamic and dynamical effects on the same root, we adopt for the phase transition the view of the EI. With only the main ingredient—vanishing of the excitation energy—the EI concept is too broad, as just a general view of quantum phase transitions. The focused concept of the EI is distinguished when the number of excitons, both in the ground state and out of equilibrium, is approximately conserved as we shall specify in the next section. Under pumping conditions, the dualism of the exciton density and of the thermodynamic order parameter looks not to be quite compatible: the thermodynamic charge transfer is expected to be given by a single real nonconserved field q, both in equilibrium and in evolution. The charge transfer density from the pumped ensemble of excitons is given by the exciton number density $q = |\Psi|^2$, which is also a real field, but its evolution is given by the complex wave function $\Psi = q^{1/2} \exp(i\varphi)$ of the BEC, then the hidden degree of freedom—the phase φ comes to the sight. This phase can be traced directly since its time derivative gives the observable instantaneous energy of the exciton $E_{\rm ex}(t) = -\hbar\partial_t\varphi$. We shall see that there are effects of anomalous interband scattering of electrons and holes which can enforce the phase locking, making the clear dynamical boundary between the regimes of the BEC and the EI states.

III. THE PHENOMENOLOGICAL MODEL AND EVOLUTION EQUATIONS

The theory of PIPTs faces very high challenges when it is started *ab initio* at the microscopic level (see, e.g. [3,28–32]), but over longer timescales, the evolution should be governed by collective variables allowing for a phenomenological description at macroscopic scales. The phenomenology becomes indispensable to recover inhomogeneous states which are inherent to large enough samples [33,34], as we shall demonstrate below. The efficiency of such an approach has been demonstrated in successful detailed modeling of experiments on dynamics of other PIPTs [35–37]. The prerequisite for this approach is the rapidity of the initial relaxation whose origin was not always fully understood, and we shall take it here for granted as experimentally confirmed fact.

The optical pumping gives rise to a high density of excitons which, as bosons, may be described by the common wave function $\Psi(x,t)$. Our main assumption is that the quasicondensate of optically pumped excitons appears sufficiently early as the macroscopic quantum state. It is plausible because of the very high initial pumping (0.1 per site, i.e. about 1 per the exciton core length). Still, for a not well-tuned pumping, when the light is absorbed to hot excitonic states via phonon-assistant processes, an intermediate two-fluid stage appears where the particle conversion from the normal bath has to be taken into account. A general conclusion drawn from studies of basic Bose-gas models (see, e.g. [38] and references therein) is that the condensate part can still be described by a macroscopic wave function which needs to be treated stochastically rather than deterministically.

While in real systems several fields (e.g. both the onsite charges and the bonds deformations) can be involved simultaneously, here, we shall concentrate on a generic case where the thermodynamic transition is governed by only one field: the charge transfer density q. This field is not symmetry breaking, similar to the liquid-gas or to the Mott transitions; hence, a first-order phase transition is expected in accordance with a typical experimental situation for most charge-transfer systems.

The basic phenomenological energy density can be chosen in the simplest polynomial form

$$W(q) = E_{\rm ex}^0 q + \frac{a}{2}q^2 + \frac{b}{3}q^3; \quad q = |\Psi|^2. \tag{1a}$$

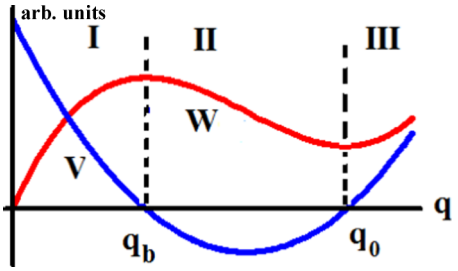

FIG. 1. Ground state energy density $W(q)$ and the thermodynamic energy of exciton $V(q)$ for the weakly first-order phase transition.

Here, $E^0_{\rm ex}$ is the bare exciton energy and the term $\sim q^2$ gives the interaction of excitons, similar to the term $\sim|\Psi|^4$ in the standard Gross-Pitaevskii theory of Bose ensembles.

In the standard Ginzburg-Landau theory for the second-order phase transition of both BEC of real excitons and the virtual condensate of the EI theory, the only term $aq^2/2$ (with the positive a) would be necessary. Below the second-order transition, i.e. at $E^0_{\rm ex} < 0$, this term stabilizes the density at a ground state value q_0. The first-order transition requires for negative $a < a_{cr} = -2(bE^0_{ex})^{1/2}$ at yet positive $E^0_{\rm ex} > 0$. The whole energy is stabilized thanks to the higher-order nonlinearity $\sim bq^3$ with $b > 0$. The negative $a < 0$ means the attraction of excitons at their low density. The effects of exciton attraction and its subsequent stabilization (or forming a biexciton gas as an alternative) has been studied microscopically (for conventional semiconductors) in [39].

The thermodynamic definition of the exciton energy is generalized for their interacting ensemble as

$$E_{\rm ex}(q) = V(q) = \frac{dW}{dq} = E^0_{\rm ex} + aq + bq^2. \qquad (1b)$$

Its dynamical definition is given by the time derivative of the wave function phase: $E_{\rm ex}(t) = -\hbar\partial_t\varphi$.

Functions $W(q)$ and $V(q)$ are plotted in Fig. 1 for parameters above the phase transition: the metastable state at $q = q_0$ coexists with the stable state at $q = 0$. The two states are separated by the barrier at $q = q_b$. Two vertical lines in Fig. 1 separate three intervals of q corresponding to different behavior of excitons. In the region I, $0 < q < q_b$, the excitons are attractive: being still positive, $E_{\rm ex}(q)$ decreases with q. In the region II, $q_b < q < q_0$, the thermodynamically defined exciton energy $E_{\rm ex}(q)$ is negative, allowing for their production, thus drawing the order parameter value towards the energy minimum at $q = q_0$. Finally, in the region III, $q > q_0$, the exciton energy $E_{\rm ex}(q)$ becomes again positive, and the excitons are repulsive.

Two types of Coulomb interactions give rise to the microscopic formation of the exciton as a quantum bound state of the electron and the hole. The major long-range attractive scattering conserves the band numbers of particles (Fig. 2, left panel). In addition, there is also the anomalous scattering (Fig. 2, right panel), coming from matrix elements of the Coulomb interaction which transfer two electrons across the gap, between filled and empty bands; that corresponds to the simultaneous creation or annihilation of two e-h pairs [11,27].

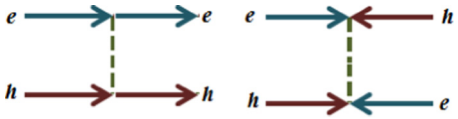

FIG. 2. Two types of the e-h scattering: normal (left) and with annihilation (right).

For e-h bound states, that means the creation or annihilation of pairs of excitons from/to the vacuum.

Normally, this is a virtual process (the energy deficit is $2E_g$ for free particles or $2E^0_{\rm ex}$ for bound pairs), which can contribute only to the second-order perturbation theory, but for the correlated state of a large number of excitons, we arrive at the amplitudes S, S^* of the simultaneous creation/annihilation of two excitons, then the free energy (1a) acquires a phase-locking term

$$\Delta W = \tfrac{1}{2}(S^*\Psi^2 + S\Psi^{*2}) = |S|q\cos 2(\varphi - \alpha);$$
$$S = |S|\exp(i\alpha), \qquad (2)$$

whose phase dependence violates the particle conservation law. We shall assume the given scattering phase value as $\alpha = \pi/2$ which always can be done by shifting the origin of the variable phase φ.

Variation over Ψ of the energy functional $W + \Delta W + W_{\rm kin}$ given by expressions in Eqs. (1a) and (2), augmented by the kinetic energy of excitons, yields the generalized Gross-Pitaevskii equation

$$i\hbar\partial_t\Psi = -\frac{\hbar^2}{2M}\partial_x^2\Psi + (V - i\hbar\Gamma)\Psi - S\Psi^*, \qquad (3)$$

where M is the excitonic mass and Γ is the introduced relaxation rate of excitons. The perturbations related with Γ and S in Eq. (3) describe the relaxation of the amplitude and the locking of the phase correspondingly.

The function $\Gamma(q)$ might have a complicated behavior passing through different regimes depending on q (see the intervals I–III in Fig. 1). In the most dilute limit of isolated excitons, apparently $\Gamma(q) \to 1/\tau_{\rm ex}$, which is the inverse lifetime $\tau_{\rm ex}$ for the single-exciton recombination. For traditional experiments attempting the BEC of excitons under the stationary pumping (see references in [27,40]), this time was too short—nanoseconds—to maintain the necessary concentration (whose obstacle has been overcome in space-separated bilayers [41] with τ reaching microseconds), but for the PIPTs, this time is oppositely too long to be considered. Moreover, our vanishing q still assumes a macroscopic concentration when the radiative recombination is dominated by stimulated emission; then $\Gamma(q)$ decreases as $\Gamma \propto q$. This assumption corroborates with firm observations in semiconductors, particularly well elaborated for Cu_2O, where the law $\Gamma \propto q$ has been firmly observed and associated with Auger processes [40,42].

In the region II, $q_b < q < q_0$, the thermodynamically defined exciton energy $E_{\rm ex}(q)$ is negative. That assumes the production of excitons which indeed is necessary to draw the order parameter towards the high-q energy minimum. As a consequence, here $\Gamma(q) < 0$ at $E_{\rm ex}(q) < 0$, with a precaution on a definition of $E_{\rm ex}$ at presence of S terms, as we shall specify below.

Approaching the high-q equilibrium phase, where the excitons constitute the ground state, $\Gamma(q)$ should vanish, i.e. $\Gamma(q) \to 0$ since, at the energy minimum, there is no decay channel. Namely, in the course of increasing q, the excitons gradually change their nature from the reservoir of excitations (even if Bose condensed) to the order parameter of the ground state of the EI. The qualitative boundary, as we shall demonstrate below, is the phase lock-in transition which happens approaching the thermodynamically (meta)stable ground sate at q_0. In their last incarnation, the excitons cannot decay without increasing of the system energy.

In view of the phase dependence, the equilibrium state is determined by both q and φ, approaching the energy minimum [$\varphi = 0$ (modulo π), $q \approx q_0$] in some complicated way as we shall demonstrate below. Instead of guessing $\Gamma(q,\varphi)$ as a function of the two variables, it is more instructive and basic to realize that the energy relaxation terminates when the dynamical energy of the exciton becomes frozen at zero, i.e. at $\partial_t \varphi = 0$. These expectations for behavior of $\Gamma(q,\varphi)$ at all limits are qualitatively satisfied if we write

$$\Gamma\ (q) = -\frac{G(q)}{2i}\ (\Psi^* \partial_t \Psi - \Psi \partial_t \Psi^*), \tag{4}$$

where $G(q)$ is a smooth interpolation function which we shall take as a constant in the numerical modeling.

For the spaciohomogeneous regime $\partial_x \Psi \equiv 0$, it is instructive to rewrite Eqs. (3) and (4) for Ψ in variables (q,φ)

$$\hbar \partial_t \varphi = -V + |S| \cos(2\varphi), \tag{5}$$

$$\partial_t q = -\frac{G}{\hbar} q^2 [V - |S| \cos(2\varphi)] + |S| q \sin(2\varphi). \tag{6}$$

Approaching the bare state $q \to 0$, we get from Eq. (3) the isolated excitons with the energy $E_S = [(E^0_{\rm ex})^2 - |S|^2]^{1/2}$ shifted by the S term in Eq. (2). Their wave function oscillates as

$$\Psi \sim \sqrt{E^0_{\rm ex} - |S|} \cos\left(\frac{t E_S}{\hbar}\right) - i\sqrt{E^0_{\rm ex} + |S|}\ \sin\left(\frac{t E_S}{\hbar}\right). \tag{7}$$

The phase is not locked, but instead the excitonic level is down-shifted, and both positive and negative energies are present in the eigenmode. This is a consequence of pair creation (annihilation) from (to) the vacuum. The limit $E^0_{\rm ex} = |S|$ corresponds to the absolute instability of the normal state if it is supercooled with respect to the EI state.

In the opposite limit of high q, the truly static state $\partial_t \Psi = 0$ of the EI is reached as $\varphi = \pi n$ with the equilibrium value of q displaced from q_0 to q_1 such that $V(q_1) = |S|$.

IV. THE NUMERICAL MODELING

A. Choice of parameters

In this section, we present the results of numerical modeling based on the above equations.

The energies are supposed to be measured in electronvolts and the time in femtoseconds. For our modeling, we normalize the expression (1a) in such a way that $E^0_{\rm ex} = 1$ and $q_0 = 1$. The choice of coefficients in (1a,b) as $a = -3.5$ and $b = 2.5$ gives rise to the plots of Fig. 1(a) with $q_0 = 1$ and $q_b = 0.4$. With these parameters, we are above the thermodynamic phase transition to the EI state which nevertheless can exist as a metastable one. The known experimental values are, e.g. $E^0_{\rm ex} = 0.6$ and $q_0 = 0.3$ which allows interpreting, within the order of magnitude, our rescaled parameters as physical ones. The less clear parameter is the energy S which must be much smaller than 1; typically, we shall use $S = 0.01$. For the coefficient G in the expression in Eq. (4), we adopt the experimental values known for conventional semiconductors. The law for the concentration $c(t)$ decay $dc/dt = -Ac^2$ with $A \sim (10^{-20}-10^{-16})\,{\rm cm}^3 \cdot {\rm ns}^{-1}$ [40,42] transfers to systems of our interest as $G \sim (10^{-4}-10^{0})/({\rm site} \cdot {\rm fs})$, and we shall employ an intermediate $G = 0.01$. The distance x will be measured in intermolecular spacing d; the experimentally known [6–8] width of absorption lines for the charge transfer exciton gives the estimation of the effective mass M as $\hbar^2/(Md^2) \sim 0.2\,{\rm eV}$.

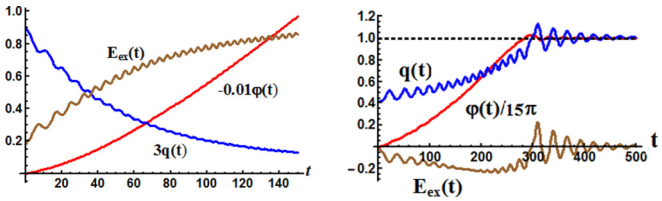

FIG. 3. Plots for $q(t)$ (blue), $\pm\varphi(t)$ (red), and $E_{\rm ex}(t)$ (brown) for space-independent subbarrier (left panel, $q_i = 0.39$) and superbarrier (right panel, $q_i = 0.41$) regimes. The transition at $t \approx 300$ locks the phase in $\varphi = 15\pi$. Notice that, in the left panel, the phase is plotted with the "minus" sign.

B. Space-independent or zero-dimensional regimes

Let us first consider the space-independent regime which can be realized in a small sample or in a heterostructure. Results of the numerical modeling [solutions of Eqs. (3) and (4)] are shown in Fig. 3 as linear plots for $q(t), \pm\varphi(t)$, and $E_{\rm ex}(t)$. The pumping intensity determines the initial value $q_i = |Y(0)|^2$.

For a subbarrier pumping $q_i < q_b$, the system relaxes to the virgin no-exciton state $q = 0$ as shown in Fig. 3 (left panel). The smooth curves are superimposed by oscillations of the macroscopic quantum interference which frequency corresponds to the double energy of the bare exciton, in accordance with the expression in Eq. (7).

Results for the superbarrier pumping are shown in Fig. 3 (right panel) for q_i just above the barrier q_b. Initially and in the whole interval $q_0 > q_i > q_b$, the system is in the regime II of Fig. 1 where $\Gamma < 0$. That allows for creation of excitons from the vacuum, which leads to the further increase of q. For a while, the system demonstrates the unlocked regime where the phase is monotonous in time, but unlike the subbarrier case, the phase increases with t meaning the negative energy of the exciton. That in turn yields the particle production feeding the increase in $q(t)$. In a while, the value of $q(t)$ approaches close enough the energy W minimum at q_0 where the phase gets locked at a definite moment $t \approx 300$.

Just after the lock-in transition, the oscillations rise sharply, but later on, they attenuate, and the system finds a new

equilibrium corresponding to the EI state. In this example, the phase is locked, getting an increment of 15π. Keeping in mind inevitable inhomogeneities in real systems, the number of periods gained before the locking can vary from place to place, giving rise to long-living topological defects—the phase domain walls.

C. Spaciotemporal regimes

Now we release the space-independence restriction to study spaciotemporal regimes with the help of Eqs. (3) and (4). As the initial condition, we have used an almost flat profile of $q(x,t=0)$ dropping to zero at the boundaries: $q(\pm L,t=0)=0$. Its convenient parameterization is given by the Jacobi elliptic function $q(x,t=0)=q_i sn^2[\frac{x+L}{L}K(k);\ k]$ where $K(k)$, here with $k=0.999$, is the complete elliptic integral of the first order.

We shall consider the most interesting situation where the initial pumping intensity is still below the barrier $q_i < q_b$. In spite of that, the early evolution leads to formation of one or several stripes with enhanced values of $q(x,t)$ which is the effect of self-trapping, akin to self-focusing with formation of bright solitons in the nonlinear optics. Indeed, because of the negative curvature $d^2W/dq^2 < 0$ at $q < q_b$ corresponding to the attraction of excitons, at low q, our effective Gross-Pitaevskii equation takes a form of the nonlinear Schrödinger equation with the negative sign of the nonlinearity. A small critical pumping appears whose value $q_c < q_b$ depends on the initial profile. For $q_i < q_c$, the self-focusing starts, but it is not sufficient to overpass the barrier; after a number of oscillations, it all relaxes to the no-exciton state $q(x,t)\to 0$. For $q_i > q_c$, the local enhancement of $q(x,t)$ becomes sufficient to overcome the barrier, and the system is stratified in regions of high and low $q(x,t)$. In a short time $\sim$100, within the high-q stripes, the phase is locked, giving rise to the phase separation into the low-q bulk of the BEC of excitons and the high $q\approx q_0$ stripes of the EI.

Figure 4 shows the modeling results for several values of the subbarrier pumping intensity q_i, always within $q_c < q_i < q_b$. Figure 4 (left panel) shows that for q_i being just slightly above $q_c \approx 0.2$, two domains of the high-q phase are nucleated after fast self-focusing processes. Afterwards, they move convergently toward the sample center where they merge forming one stable central domain.

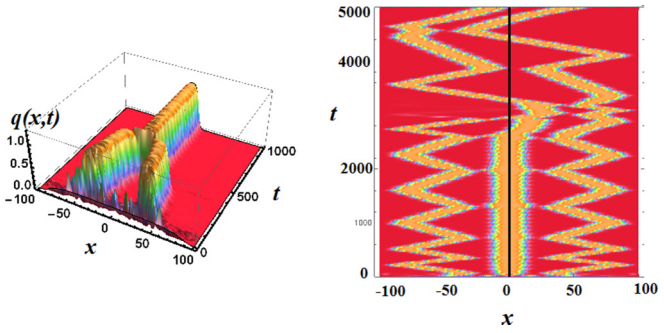

FIG. 4. Spaciotemporal modeling for different (always subbarrier) pumping intensity, showing formation of high-density domains. Three-dimensional (3D) plot of $q(x,t)$ for $q_i=0.19$ (left); density plot of $q(x,t)$ for $q_i=0.25$ (right).

At higher q_i, while still below q_b, three domains are nucleated (Fig. 4, the right panel). The central domain keeps its position in space, while two side ones oscillate in the antiphase manner between the sample boundaries and the central domain, being repelled from both. At some much later moment $t\approx 3500$, the central domain is absorbed by the side ones which keep oscillating, now in phase with the right one. Further on at $t\approx 8500$, the remnant two domains also collide and merge. These events are shown in detail in the panels of Fig. 5 for three selected intervals of time: the initial multiple nucleation which is promptly reduced to three main ones (left panel), the merging of the undulating left domain with the static central one at $t\approx 3500$ (central panel), and the final merging of domains at $t\approx 8500$ (right panel).

To clarify effects of possible inhomogeneities, we have performed also the modeling for a stronger shaped initial distribution (again, for the subbarrier pumping intensity). The results presented in Fig. 6 (left panel) show the two domains emitted from the common nucleation point; afterwards, they diverge with a constant velocity. Then they are reflected from the sample boundaries, move convergently, and finally collide and merge. The earlier stage amplified in the right panel of Fig. 6 clearly demonstrates the self-trapping development followed by the explosive penetration over the barrier.

In all cases of domains formation, the phase is locked within the domain as it should be for the well-formed EI. Beyond the

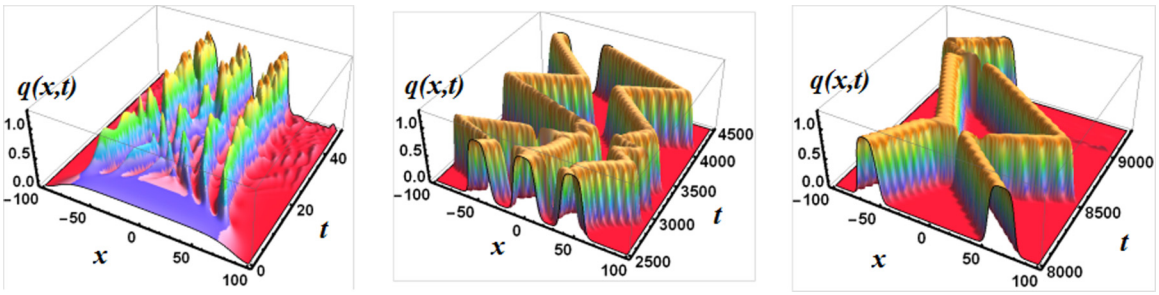

FIG. 5. Details of the evolution [shown in total in Fig. 4 (right panel)] are given here for three characteristic intervals of time. The initial nucleation stage (left), the merging of the undulating left domain with the static central one at $t\approx 3500$ (center), and the final merging of domains at $t\approx 8500$ (right).

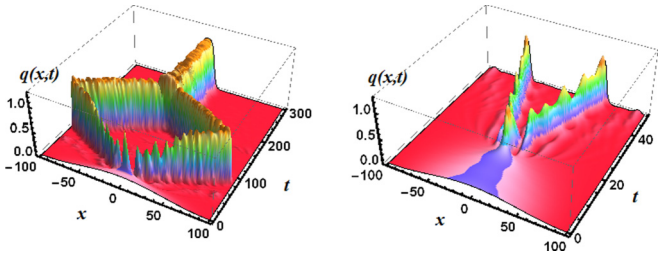

FIG. 6. Modeling for a stronger shaped initial distribution (again, for the subbarrier pumping intensity) showing the common nucleation of two domains, their divergence, then approaching after reflections from the sample boundaries and the final merging.

domain, in regions of small q, the phase evolves linearly in time in accordance with the free exciton energy.

V. DISCUSSION AND CONCLUSIONS

The actual systems can be more complicated than our basic model, particularly involving other degrees of freedom in addition to the charge transfer. Thus, in a well-studied case of the neutral-ionic transitions in donor-acceptor molecular stacks (see the model formulation in [43]), there is also another degree of freedom—the lattice dimerization which appears in the course of the transition. It is quite doable to extend and to study numerically such a model where the energy is a functional of several variables [43], but the supplementary oscillating dynamics related with the lattice complicate the picture, preventing a clear analysis of the fundamental problem of two interfering faces of the excitonic condensate, which is the primary goal of our paper.

Manifestations of phase transitions provoked by pumping to excitons are unlimited in opportunities. Even if we restrain the attention to most relevant cases of charge-transfer transitions, their realizations can be found in very different systems from the inorganic world of compounds based upon transition metals to many realizations as neutral-ionic transitions in organic materials. By now, PIPT experiments have been performed systematically only for this type of ordering in donor-acceptor chains [4–8].

In summary, we have presented results of the phenomenological modeling for a system prone to a weakly first-order phase transition after it is exposed to optical pumping to a high concentration of excitons. We considered the case where the density of the pumped excitations and the variable governing the thermodynamic phase transition present the same entity. Both thermodynamic and dynamical effects can be described on the same root by viewing the ordered state as the EI. The following distinguishing features have been recovered: (i) the oscillations coming from the macroscopic quantum coherence, (ii) the dynamical transition from the particle conserved regime of the BEC of excitons to the phase-locked regime of the EI, and (iii) the inhomogeneous spaciotemporal regime with the self-focusing followed by formation, separation and merging of domains of the high-density phase of the EI embedded to the bulk depleted from the excitons.

We believe that the suggested picture, the approach, and illustrations will encourage a more microscopical theoretical work and will stimulate experimental studies of PIPTs in systems possessing features of the EI and/or allowing for pumping to an excitation mode coupled to a parameter of a nearby phase transition.

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