Pattern Formation as a Signature of Quantum Degeneracy in a Cold Exciton System

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The development of a Turing instability to a spatially modulated state in a photoexcited electron-hole system is proposed as a novel signature of exciton Bose statistics. We show that such an instability, driven by kinetics of exciton formation, can result from stimulated processes that build up near quantum degeneracy. The stability of an electron-hole interface which describes recently observed exciton rings is analyzed. Interface instability occurs below a critical temperature, with a periodic 1D pattern developing via a continuous (type II) transition, in a qualitative agreement with observations.

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Recently, striking spatial photoluminescence (PL) patterns [1–5] have been observed in photoexcited quantum well (QW) structures [Fig. 1(a)]. In addition to macroscopic concentric rings and "bright spots," the electron-hole system exhibits an abrupt transition at ca. 2 K in which the outermost ring "fragments" into regularly spaced beads of high PL intensity [1,3]. While the gross features of PL have been explained within the framework of classical transport, attributing the internal rings to nonradiative exciton transport and cooling [1] and the outermost rings and "bright spots" to macroscopic charge separation [3–6], the origin of the instability remains unidentified.

Pattern formation in electron-hole systems has long been known in connection with electron-hole droplet formation [7]. However, the coupled quantum well geometry [1,3] is engineered so that droplet formation is energetically unfavorable, the ground state is excitonic, and the interaction between excitons is repulsive: Indirect excitons, formed from electrons and holes confined to different QWs by a potential barrier, behave as dipoles oriented perpendicular to the plane, and an exciton or electron-hole density increase causes an enhancement of energy [8,9]. The repulsive character of the interaction is evidenced in experiment as a positive and monotonic line shift with increasing density [10–13]. Crucially, the same behavior is observed in the regime of modulational instability. Here the spectral shift observed in PL varies along the circumference of the ring in concert with the intensity (see Fig. 2f in Ref. [1]), with the highest energy found in the brightest regions. Noting that the repulsive interaction eliminates the standard mechanism of droplet formation [7,14], one is led to conclude that the paradigm of electron-hole droplets is too narrow to account for bead formation and a new mechanism must be sought. Further evidence for the integrity of the excitonic state is provided by the narrow line shape observed in PL which is some 4 times smaller than that achievable for an electron-hole plasma.

The observed spatial modulation calls for an explanation involving a symmetry breaking instability of a homogeneous to a patterned state. However, droplet formation in a system with a repulsive interaction seems hard to explain on the basis of purely energetic arguments. This conundrum can be resolved if kinetic effects play an essential role. In this work we propose a novel mechanism, based on the kinetics of exciton formation from optically excited electrons and holes, that can lead to an instability in the exciton system. Interestingly, these kinetic effects become especially strong in the regime near *exciton quantum degeneracy,* due to the stimulated enhancement of the electron-hole binding rate. The transition to a state with a spatially modulated exciton density presents a directly observable signature of degeneracy.

Pattern formation possesses universal features often making it insensitive to underlying microscopic physics. Thus, while the above mechanism produces phenomenology consistent with the observations [1,3], we caution the reader not to take it as unambiguous evidence for exciton degeneracy. Nevertheless, an observation of macroscopic instability may complement other manifestations of degeneracy discussed in the literature, such as changes in the exciton recombination [15,16] and scattering [17] rates, transport [15], and the PL spectrum [18].

As a pattern formation problem, the observed behavior is reminiscent of a Turing instability in reaction-diffusion systems [19]. However, in contrast to the spatially uniform

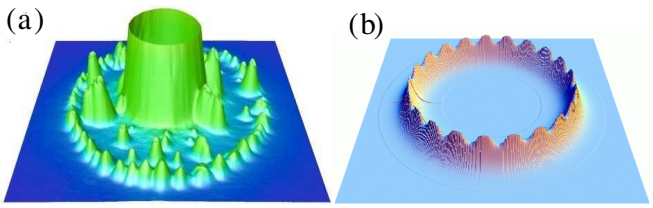

FIG. 1 (color online). (a) Modulational instability of an exciton ring as revealed in the 2D PL profile (from Ref. [3]). (b) Spatial instability as recorded in g_x obtained from a numerical solution of Eq. (11) in a circular geometry with $u/g_x^* = 10$.

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system studied by Turing and subsequently by others, in the present geometry, the mechanism of exciton formation (as resulting from an ambipolar diffusion of electrons and holes) presents the instability as an interface phenomenon more closely allied with the instabilities recorded in boundary-fed chemical reaction cells [20–23].

The kinetics of exciton formation is complex [24]: Besides stimulated processes, the exciton energy distribution may depend on intraband Auger processes [25]. While, in our model, the former lead to instability and the latter do not, linking the observed patterns with degeneracy is probably somewhat premature. Instead, our main aim is to present the Turing instability from a broader phenomenological perspective, as a general effect of quantum kinetics.

Following Ref. [3], we consider a transport theory involving electron, hole, and exciton densities $n_{\mathrm{e,h,x}}$ obeying a system of coupled nonlinear diffusion equations:

$$\begin{aligned} &\text{(e)}\ \partial_t n_{\mathrm{e}} = D_{\mathrm{e}}\nabla^2 n_{\mathrm{e}} - w n_{\mathrm{e}} n_{\mathrm{h}} + J_{\mathrm{e}}, \\ &\text{(h)}\ \partial_t n_{\mathrm{h}} = D_{\mathrm{h}}\nabla^2 n_{\mathrm{h}} - w n_{\mathrm{e}} n_{\mathrm{h}} + J_{\mathrm{h}}, \\ &\text{(x)}\ \partial_t n_{\mathrm{x}} = D_{\mathrm{x}}\nabla^2 n_{\mathrm{x}} + w n_{\mathrm{e}} n_{\mathrm{h}} - \gamma n_{\mathrm{x}}, \end{aligned} \tag{1}$$

where $J_{\mathrm{e,h}}$ describe the carrier photoproduction and the leakage current in the QW structure. The nonlinear couplings in Eq. (1) account for free electron and hole binding to form an exciton. We adopt a phenomenological approach, justified in Ref. [24], which assumes that the system can be described by an effective exciton temperature, leaving out the effects of nonequilibrium energy distribution and cooling due to phonon emission [17].

We argue that the dipolar exciton repulsion, not accounted for in the model (1), cannot suppress the instability. To assess the role of repulsion in the dynamics, one must first keep in mind that, in $d = 2$, the $1/r^3$ interaction is effectively short ranged. In the hydrodynamical regime, at low exciton density, the associated contribution to transport deriving from the contact interaction is small as compared to diffusion. Moreover, our model implies an exponential growth of the scattering rate with density (2), which should supersede a power law growth of the interaction. Therefore, the latter may add only quantitative corrections without changing the origin of the instability.

The proposed mechanism of instability relies on a strong dependence of the electron-hole binding rate w on local exciton density n_{x} and temperature. The latter naturally arises from stimulated electron-hole binding processes mediated by phonons which enhance the binding rate by the occupation number factor $f = 1 + N_E^{\mathrm{eq}}$. (At low temperatures, one can ignore the reverse processes of exciton dissociation.) For the dominant process involving scattering into the ground state with $N_{E=0} > 1$, the ideal Bose gas result gives

$$w \sim f = e^u, \qquad u \equiv \frac{n_{\mathrm{x}}}{n_0(T)}, \qquad n_0(T) = \frac{2g m_{\mathrm{x}} k_{\mathrm{B}} T}{\pi\hbar^2}, \tag{2}$$

with g the degeneracy, and m_{x} the exciton mass [26]. Equivalently, $u \equiv T_0/T$ where $T_0 = (\pi\hbar^2/2g m_{\mathrm{x}} k_{\mathrm{B}}) n_{\mathrm{x}}$ is the degeneracy temperature. At the crossover from classical to quantum statistics, $T \sim T_0$ ($n_{\mathrm{x}} \sim n_0$), the enhancement factor f increases sharply.

The mechanism for hydrodynamic instability mirrors that discussed in general reaction-diffusion systems: A local fluctuation in exciton density leads to an increase in the stimulated electron-hole binding rate. The depletion of local carrier concentration causes neighboring carriers to stream towards the point of fluctuation presenting a mechanism of positive feedback. The wavelength, determined by the most unstable harmonic of the density, characterizes the length scale of spatial modulation in the nonuniform state. The transport has implicit constraints imposed by particle number *and* electric charge conservation. These are obtained by considering the linear combinations (e) − (h), (e) + (h) + 2(x) of Eqs. (1). In both cases, the nonlinear term drops out and one has

$$\hat{L}_{\mathrm{e}} n_{\mathrm{e}} - \hat{L}_{\mathrm{h}} n_{\mathrm{h}} = J_{\mathrm{e}} - J_{\mathrm{h}}, \tag{3}$$

$$\hat{L}_{\mathrm{e}} n_{\mathrm{e}} + \hat{L}_{\mathrm{h}} n_{\mathrm{h}} + 2\hat{L}_{\mathrm{x}} n_{\mathrm{x}} + 2\gamma n_{\mathrm{x}} = J_{\mathrm{e}} + J_{\mathrm{h}} \tag{4}$$

with $\hat{L}_{\mathrm{e(h,x)}} = \partial_t - D_{\mathrm{e(h,x)}}\nabla^2$. Since the origins of (3) and (4) are rooted in conservation laws, they are robust and insensitive to the form of the electron-hole binding term.

Although our main aim is to address the experimental geometry, it is instructive to consider first the uniform system, $J_{\mathrm{e}}(\mathbf{r}) = J_{\mathrm{h}}(\mathbf{r}) \equiv J$, realized by an extended photoexcitation, wherein $\bar{n}_{\mathrm{x}} = J/\gamma$, $\bar{n}_{\mathrm{e,h}} = [J/w(\bar{n}_{\mathrm{x}})]^{1/2}$. Applying a linear stability analysis to (1), an expansion around the uniform state with a harmonic modulation $\delta n_{\mathrm{e,h,x}} \propto e^{\lambda t} e^{i\mathbf{k}\cdot\mathbf{r}}$ obtains the stability condition

$$\frac{d\ln w}{d\ln \bar{n}_{\mathrm{x}}} \equiv u \geq u_c = (1 + r^{1/2})^2, \tag{5}$$

where $r = (D_{\mathrm{e}}^{-1} + D_{\mathrm{h}}^{-1}) D_{\mathrm{x}} \bar{n}_{\mathrm{x}}/\bar{n}_{\mathrm{e}}$. Since the binding rate $w(n_x)$ builds up near the BEC transition due to the growth of stimulated processes, while other transport coefficients are not directly sensitive to the degree of exciton degeneracy, the system becomes unstable at temperatures approaching T_{BEC}. At threshold $u = u_c$ all modes with $|\mathbf{k}| = k_* \equiv r^{1/4}/\ell_{\mathrm{x}}$, where $\ell_{\mathrm{x}} = \sqrt{D_{\mathrm{x}}/\gamma}$ denotes the exciton diffusion length, become unstable simultaneously. The resulting 2D density distribution is determined by the effect of harmonics mixing due to higher order terms in (1) expanded in $\delta n_{\mathrm{e,x}}$ about the uniform state. Since these equations contain quadratic terms, the favored combination of harmonics is threefold symmetric, $\mathbf{k}_j = k_*(\cos(2\pi j/3 + \theta), \sin(2\pi j/3 + \theta))$, $j = 1, 2, 3$, with the parameter θ describing the degeneracy with respect to 2D rotations. This leads to a density distribution $\delta n \propto \sum_j e^{\pm \mathbf{k}_j\cdot\mathbf{r}}$ with maxima arranged in a triangular lattice. On symmetry grounds, since the triangular pattern is stabilized by quadratic terms, the mean field analysis predicts

that the transition to the modulated state in this case is abrupt, of a type I kind. Such behavior mirrors closely Turing instabilities explored long ago in the context of Bénard convection [27] and, more recently, in chemical reaction cells [28].

To apply these ideas to the experimental geometry, one must first determine the profile of the uniform distribution. The rings represent an interface between regions populated by electrons and holes at which they bind to form excitons, with the steady state maintained by a constant flux of carriers. The parameter regime which is both relevant and simple to analyze is that of long exciton lifetime γ^{-1} where the diffusion length ℓ_x exceeds the range of electron and hole overlap. In this case, approximating the source of excitons by a straight line $c\delta(x)$, where c is the total carrier flux and x is the coordinate normal to the interface, the exciton density profile is given by $(c\ell_x/2D_x)e^{-|x|/\ell_x}$. Accordingly, one can seek the electron and hole profile treating $w(n_x)$ as constant, restoring its dependence on n_x when turning to the instability. The profiles can be inferred from two coupled nonlinear diffusion equations

$$D_{e(h)}\partial_x^2 n_{e(h)} = w n_e n_h, \tag{6}$$

with the boundary condition $D_{\substack{e\\h}}\partial_x n_{\substack{e\\h}}|_{\pm\infty} = \pm c\theta(\pm x)$. From Eq. (3) one obtains $D_e n_e - D_h n_h = cx$ which allows the elimination of n_h. Applying the rescaling $n_{e(h)} = c\ell g_{e(h)}/D_{e(h)}$, where $\ell = (D_e D_h/wc)^{1/3}$, one obtains

$$\partial_{\tilde{x}}^2 g_e = g_e(g_e - \tilde{x}), \qquad \tilde{x} \equiv x/\ell. \tag{7}$$

From the rescaling one can infer that the electron and hole profiles overlap in a range of width $\ell \sim c^{-1/3}$ while $g_e(|x| \gg \ell) = \tilde{x}\theta(\tilde{x}) + \mathcal{O}(|\tilde{x}|^{-1/4}e^{-2|\tilde{x}|^{3/2}/3})$.

Although Eqs. (6) are nonlinear, they do not admit a spatial instability. However, if one restores the dependence of the binding rate on exciton density, the same mechanism of positive feedback which triggers the instability in the uniform system becomes active. To explore the instability, one may again expand linearly in fluctuations around the spatially uniform solution, $g_e(x, y) = \bar{g}_e(x) + \delta g_e(x)e^{iky}$ (similarly g_h and g_x), where y is the coordinate along the interface, $\bar{g}_e(x)$ denotes the uniform profile obtained from (7), and $\bar{g}_x = D_x\bar{n}_x/c\ell_x$. With $\ell_x \gg \ell$, the exciton density remains roughly uniform over the electron-hole interface. Denoting this value by $n_x^* \equiv \bar{n}_x(0)$, in the vicinity of the interface, one may again develop the linear expansion $w[n_x] \simeq w[n_x^*](1 + u\delta n_x/n_x^*)$ where, as before, $u = d\ln w/d\ln n_x$. Noting that Eq. (3) enforces the steady-state condition $\delta g_e = \delta g_h$, a linearization of Eqs. (1) obtains the Schrödinger-like equation

$$[-\partial_{\tilde{x}}^2 + (\ell k)^2 + \bar{g}_e + \bar{g}_h]\delta g_e + \frac{u}{g_x^*}\bar{g}_e\bar{g}_h\delta g_x = 0, \tag{8}$$

together with the condition on the Fourier components $\delta g_{x(e,h)}(q) = \int e^{-iqx}\delta g_{x(e,h)}(x)dx$,

$$\delta g_x(q) = -\delta g_e(q)\left(1 - \frac{1}{Q^2 + (q\ell_x)^2}\right), \tag{9}$$

with $Q^2 \equiv (k\ell_x)^2 + 1$. Now, since the product $\bar{g}_e\bar{g}_h$ is strongly peaked around the interface, the main contribution from the last term in (8) arises from Fourier elements $q\ell \sim 1$. Then, with $\ell_x \gg \ell$, the second contribution to $\delta g_x(q)$ can be treated as a small perturbation of the first and, to leading order, neglected, i.e., $\delta g_x \simeq -\delta g_e$. In this approximation, the most unstable mode occurs at $k = 0$. Qualitatively, an increase in u triggers an instability at a critical value u_c when the linear equation first admits a nonzero solution, $\delta g_e^{(c)}$. Numerically, one finds that $u_c/g_x^* = (2\ell/\ell_x)u_c \equiv a_0 \simeq 6.516$, while the corresponding solution $\delta g_e^{(c)}(x)$ is shown in Fig. 2.

While the approximation above identifies an instability, a perturbative analysis of the k-dependent corrections implied by (9) reveals that the most unstable mode is spatially modulated. To the leading order of perturbation theory, an estimate of the shift of u_c obtains

$$\frac{\delta u_c}{u_c} = \frac{\ell}{a_0 a_1}\left((k\ell)^2 + \frac{a_0 a_2(Q)}{\ell_x^2 Q}\right),$$

where $a_1 = \int_{-\infty}^{\infty} d\tilde{x}\bar{g}_e(\tilde{x})\bar{g}_h(\tilde{x})g_e^{(c)}(\tilde{x}) \simeq 0.254$, and

$$a_2(Q) = \frac{1}{2}\int_{-\infty}^{\infty} d\tilde{x}d\tilde{x}'\bar{g}_e(\tilde{x})\bar{g}_h(\tilde{x})g_e^{(c)}(\tilde{x})e^{-Q|\tilde{x}-\tilde{x}'|\ell/\ell_x}g_e^{(c)}(\tilde{x}').$$

With $\ell_x \gg \ell$, it follows that $Q \ll \ell_x/\ell$, and the latter takes the constant value $a_2 \simeq 0.461$, independent of k. Finally, minimizing δu_c with respect to k, one finds that the instability occurs with a wave vector set by

$$k_c\ell_x \simeq (a_0 a_2 \ell_x/2\ell)^{1/3} \tag{10}$$

implying a shift of u_c by $\delta u_c/u_c \sim (\ell/\ell_x)^{4/3}$. As a result, one can infer that the wavelength of spatial modulation $\lambda_c \sim \ell^{1/3}\ell_x^{2/3}$ is larger than the electron-hole overlap ℓ, but smaller than ℓ_x. Finally, an expansion to higher order in

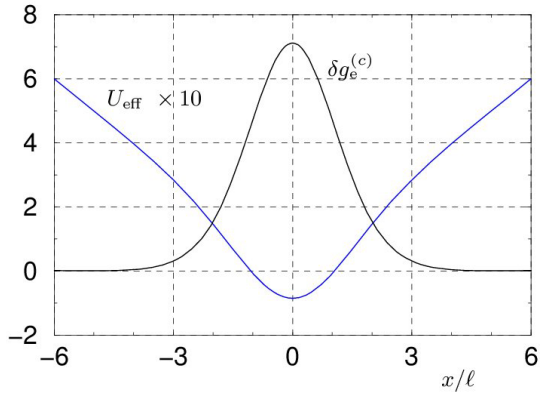

FIG. 2 (color online). Numerical solution of the (normalized) bound state solution $\delta g_e^{(c)}(\tilde{x})$ at instability threshold together with the effective potential $U_{\rm eff} = \bar{g}_e + \bar{g}_h - a_0\bar{g}_e\bar{g}_h$ of the Schrödinger-like Eq. (8).

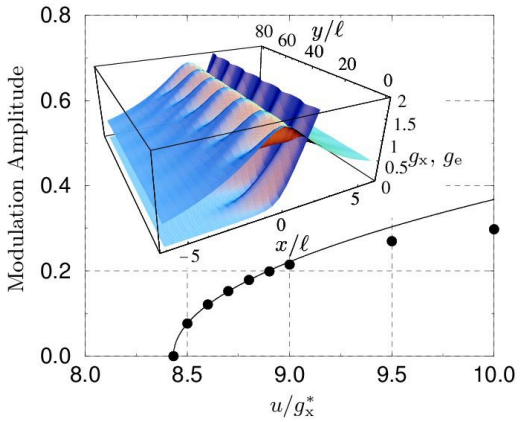

FIG. 3 (color online). Amplitude of the spatial modulation of the exciton density g_x along the electron-hole interface $x = 0$ as a function of u/g_x^* as determined from the numerical solution of the transport Eq. (11) combined with a fit to $(u - u_c)^{1/2}$. Inset: Dimensionless electron g_e and exciton g_x densities for $u/g_x^* = 9.5$ and $\ell_x/\ell = 4$. Here periodic boundary conditions are imposed along the interface. Results for a circular geometry with $u/g_x^* = 10$ and $\ell_x/\ell = 4$ are shown in Fig. 1(b).

fluctuations shows that, for $u > u_c$, the amplitude of the Fourier harmonic k_c grows as $(u - u_c)^{1/2}$.

Once the instability is strongly developed (or when $\ell_x \lesssim \ell$) the linear stability analysis becomes unreliable. Having in mind the mechanism of stimulated scattering, Fig. 3 shows the results of a numerical analysis of the dimensionless nonlinear steady-state equation

$$\nabla_{\tilde{\mathbf{x}}}^2 g_e = \exp\left(\frac{u}{g_x^*}\delta g_x\right) g_e(g_e - \tilde{x}), \qquad (11)$$

where, using Eq. (9), $\delta g_x \equiv g_x - \bar{g}_x$ depends nonlocally on $\delta g_e \equiv g_e - \bar{g}_e$ through the linear relation

$$\delta g_x(\mathbf{x}) = -\delta g_e(\mathbf{x}) + \int \frac{d^2\mathbf{x}'}{2\pi\ell_x^2} K_0\left(\frac{|\mathbf{x} - \mathbf{x}'|}{\ell_x}\right)\delta g_e(\mathbf{x}')$$

with K_0 the modified Bessel function. Although in the ring geometry the modulation is constrained by the periodic boundary conditions imposed along the interface, the critical wave number k_c lies close to that predicted by (10). The constraint leads to a value of u_c/g_x^* a little in excess of that predicted by the linear stability analysis. Otherwise the results for the circular geometry [Fig. 1(b)] and the quasi-1D case are quite similar.

With $\ell_x \gtrsim \ell$, the model predicts both the ring width and modulation wavelength to be of order ℓ_x, which compares favorably with the measured PL profile at the outer ring [1] and $\ell_x \simeq 30\ \mu$m inferred from the internal ring radius [1]. (Also, $\ell_x = \sqrt{D_x/\gamma}$ and $\gamma^{-1} \simeq 100$ ns [12] yield a reasonable value $D_x \simeq 100\ \mathrm{cm^2\,s^{-1}}$.) Moreover, with a degeneracy temperature of $T_0 \simeq 2(n_x/10^{11}\ \mathrm{cm^{-2}})$ K [26], the observed transition at $T_c \equiv T_0/u_c$ translates to an exciton density of ca. $n_x \simeq 10^{10}$–$10^{11}\ \mathrm{cm^{-2}}$, a value consistent with experiment [1]. (However, more recent measurements indicate that the instability can be observed at lower densities which, if confirmed, presents a challenge to theory.) Finally, one may note that enhancement of the exciton scattering rate [17] is observed in the same temperature range as the instability.

To conclude, we have shown that the realization of quantum degeneracy in a cold exciton system is preceded by the development of a spatial density modulation. The mechanism of instability relies on a *strongly* nonlinear dependence of the electron-hole binding rate on the exciton density pointing to the importance of stimulated scattering.

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