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Radiative coupling and weak lasing of exciton-polariton condensates

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In spite of having finite life-time exciton-polaritons in microcavities are known to condense at strong enough pumping of the reservoir. We present an analytical theory of such Bose-condensates on a set of localized one-particle states: condensation centers. To understand physics of these arrays one has to supplement the Josephson coupling by the radiative coupling caused by the interference of the light emitted by different centers. Combination of these couplings with the one-site interaction between the bosons leads to a rich nonlinear dynamics. In particular, a new regime of radiation appears. We call it weak lasing: the centers have macroscopic occupations and radiate coherently, but the coupling alone is sufficient for stabilization. The system can have several stable states and switch between them. Moreover, the time reversal symmetry in this regime is, as a rule, broken.

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Introduction.—Condensation of exciton-polaritons (EP) in semiconductor microcavities formed by two Bragg mirrors with a quantum well between them was recently discovered^{1–5}. As experimental implementation of the Bose-Einstein condensation (BEC) these systems have some advantages as compared with cold gases, e.g., the vortex dynamics and the superfluid motion can be accessed by optical methods^{6–10}. At the same time, in contrast with the atomic BEC the EP-condensates are not in the thermodynamic equilibrium. To reach a macroscopic number of EPs, which life-time is finite, one needs an outside pumping. Being driven, the EP-condensates differ fundamentally from the conventional BEC-systems. In particular, EPs can condense into one-particle excited state^{3,11} or even into several excited states^{12,13}.

It is not unusual for a one-particle bosonic state to be localized. Such states either formed by disorder or intentionally prepared^{2,3} can serve as condensation centers (CC). The bosons (photons, excitons, polaritons, etc.) arrive to each CC from an incoherent reservoir created by the pumping and escape in the form of light radiation. At low enough pumping one should expect a system of disconnected BEC droplets emitting light of different frequencies. As the pumping increases these sources of radiation tend to become coherent and to shine as a laser. This effect resembles the Insulator-Superfluid transition, but the similarity is limited by the dissipative nature of the system. In this paper we develop the theory of lasing in non-equilibrium BEC, which allows one to understand qualitatively and describe quantitatively a number of unusual and surprising experimental observations.

Existing theoretical approaches to non-equilibrium EP condensates^{14,15} are mainly based on the Gross-Pitaevskii equation (GPE) modified to account for the finite life-time of the bosons, the feed of the condensate from the reservoir, and effects of reservoir depletion¹⁶. Numerical analysis seems to agree qualitatively with some experiments. However, the nonlinear GPE can have several distinct solutions and the choice is ambiguous.

Our starting idea is that the sensitivity of the bosonic

life-time to the symmetry of the wave-function dominates this choice. If two CCs are close to the resonance and not too far apart, the interference of the light emitted from different CCs is constructive for symmetric (bonding) state and destructive for antisymmetric (anti-bonding) state, i.e., the bosons live longer in the anti-bonding case. Existence of several states with different life-times leads to a crossover range of pumping strengths (rather than a single threshold) where the income and outcome rates match. Within this range a coherent condensed state is formed. In this respect, the non-equilibrium EP condensation in disordered cavities resembles the random lasing phenomena: the lasing states also possess the longest life-times. Important difference is the nonlinearity: interaction between the EPs is able to synchronize the frequencies of CCs^{17,18}, which have no reason to be in resonance at low occupancies. One can call this regime “weak lasing”: the occupations of the CCs are macroscopic and the radiation is coherent, but the state is stabilized by the coupling between CCs rather than, e.g., by the depletion of the reservoir.

Below we show that CCs indeed become coherent and form a particular long-living condensate. In certain range of the parameters the system can have several linearly stable states, and switch between them. We argue that the properties of these states and the switches naturally explain otherwise mysterious experimental observations.

Formalism.—In the familiar Glauber-Sudarshan formalism of coherent states^{19,20} the occupation n and the phase ϕ of each CC are encoded in the complex number $z = \sqrt{n} e^{i\phi}$. N CCs are thus characterized by N -component complex vector $\mathbf{Z} = \{z_1, z_2, \dots, z_N\}$, where z_μ corresponds to the CC number μ . Each state of this system is fully described by the density matrix $\rho(\mathbf{Z}, \mathbf{Z}^*, t)$, which evolves in time according to ($\hbar = 1$)

$$\dot{\rho} = \sum_{\mu} [W_{\mu} \partial_{\mu}^* \partial_{\mu} \rho + 2\Re \{ \partial_{\mu} (\rho \partial_{\mu}^* \mathcal{H}) \}]. \quad (1)$$

Here W_{μ} is the incoming rate of the bosons to the μ -th CC, $\partial_{\mu} = \partial/\partial z_{\mu}$, dot indicates the time derivative, and

\mathcal{H} is the complex Hamiltonian function

$$\mathcal{H}(\mathbf{Z}, \mathbf{Z}^*) = \sum_{\mu} \mathcal{H}^{(1)}(|z_{\mu}|^2) + \frac{1}{2} \sum_{\mu \neq \nu} V_{\mu\nu} z_{\mu}^* z_{\nu}. \quad (2)$$

The Hamiltonian function of an isolated CC is

$$\mathcal{H}^{(1)}(|z_{\mu}|^2) = \frac{1}{2} (\Gamma_{\mu} - W_{\mu}) |z_{\mu}|^2 + iH_{\mu}(|z_{\mu}|^2), \quad (3)$$

where Γ_{μ} is the escape rate, $H_{\mu}(n_{\mu})$ is the energy of bosons in the μ -th CC, which alone emits light with the frequency $\Omega_{\mu} = dH_{\mu}(n_{\mu})/dn_{\mu}$. For weak interaction

$$H_{\mu}(n_{\mu}) = \omega_{\mu} n_{\mu} + \frac{1}{4} \alpha_{\mu} n_{\mu}^2, \quad \Omega_{\mu} = \omega_{\mu} + \frac{1}{2} \alpha_{\mu} n_{\mu}, \quad (4)$$

with one-particle energy ω_{μ} and the coupling constant $\alpha_{\mu} > 0$. The coupling is weak, $\alpha_{\mu} \ll \omega_{\mu}$, and Ω_{μ} slightly increases with the pumping. In general, the income rates W_{μ} are dynamical variables determined by the occupations of the reservoir states. However, in (1-3) W_{μ} enter as \mathbf{Z} -independent external parameters. We adopt this simplifying assumption, which is valid when the life-time of the reservoir excitations is not limited by the inelastic relaxation into CCs. It is quite straightforward to generalize our approach to include the dynamics of W_{μ} .

The coupling $V_{\mu\nu}$ between CCs in (2) is bilinear, i.e., the bosons from different CCs do not interact, and it consists of radiative (dissipative) and Josephson (non-dissipative) parts, $V_{\mu\nu} = \gamma_{\mu\nu} - iJ_{\mu\nu}$. The Josephson coupling favors symmetric ground state, i.e., $J \geq 0$. The radiative coupling $\gamma_{\mu\nu}$ is due to the interference of the light from different CCs. The matrices $\gamma_{\mu\nu}$ and $J_{\mu\nu}$ are Hermitian and have zero diagonal matrix elements, which are absorbed by the first term in (2). Besides, we assume no explicit time reversal symmetry violation and thus $\gamma_{\mu\nu}$ and $J_{\mu\nu}$ are real. Note that the Josephson coupling caused by the tunneling is exponentially small, when CCs are spatially separated, while $\gamma_{\mu\nu}$ can be still substantial.

The time evolution of the density matrix without Josephson coupling should reflect the following physical picture. According to (2) and (3) both $\Re z_{\mu}$ and $\Im z_{\mu}$ oscillate with the high frequency Ω_{μ} . As long as $\Omega_{\mu} \neq \Omega_{\nu}$, coupling between the CCs μ and ν averages out as time exceeds $|\Omega_{\mu} - \Omega_{\nu}|^{-1}$. However the interaction between the bosons synchronizes the frequencies. As soon as this happens, the interference suppresses the escape if the phases of CCs are opposite. This suppression in turn stabilizes this nontrivial stationary state (NTSS) with high occupations and coherent radiation of the CCs. NTSS differs drastically from the trivial stationary state (TSS) with small n_{μ} and different emission frequencies. We emphasize that NTSSs appear due to the off-diagonal radiative coupling even at $J_{\mu\nu} = 0$.

At this point we adopt the Langevin approach, which remains valid for a complex Hamiltonian function. We start with the formal solution of Eq. (1) for $\rho(\mathbf{Z}, \mathbf{Z}^*, t)$ with the initial condition $\rho_0(\mathbf{Z}, \mathbf{Z}^*)$ in the form of Feynman path integral over the ‘‘trajectories’’ $\mathbf{Z}(t)$

$$\rho(\mathbf{Z}, \mathbf{Z}^*, t) = \int \rho_0(\mathbf{Z}_0, \mathbf{Z}_0^*) d^{2N} \mathbf{Z}_0 \int_{\mathbf{Z}(0)=\mathbf{Z}_0}^{\mathbf{Z}(t)=\mathbf{Z}} \mathcal{D}\mathbf{Z}(\tau) e^{-\mathbb{S}}, \quad (5)$$

$$\mathbb{S} = \sum_{\mu} \int_0^t \frac{1}{W_{\mu}} |\dot{z}_{\mu}(\tau) + \partial_{\mu}^* \mathcal{H}|^2 d\tau, \quad (6)$$

Note that the pumping intensity W_{μ} of the μ -th CC plays the role of its effective temperature.

One can rewrite (5) as the average $\langle \delta(\mathbf{Z} - \mathbf{Z}^{(f)}(t)) \rangle$, where $z_{\mu}^{(f)}(t)$ are the solutions of equations

$$\dot{z}_{\mu} + \partial_{\mu}^* \mathcal{H} = f_{\mu}(t), \quad (7)$$

with $f_{\mu}(t)$ being a realization of the Gaussian random processes with zero mean $\langle f_{\mu}(t) \rangle = 0$ and δ -like two-point correlation function $\langle f_{\mu}(t) f_{\nu}^*(t') \rangle = W_{\mu} \delta_{\mu\nu} \delta(t - t')$.

Reduction of the description of the system to the classical Fokker-Plank-Langevin scheme, Eqs. (1) and (7), is justifiable because the CCs are coupled to the systems with the continuous spectrum. The noise term in (7) reflects the randomness in the particle influx from the incoherent reservoir. The radiative escape of the particles from CCs is a noiseless decay as it occurs into the vacuum state of photons.

Our analysis of (7) consists of the following steps. First we omit noise to find the stationary points. Next we analyze the stability of these points and thus identify the long-living stationary states. If such states coexist, the time the system spends in each basin of attraction is finite due to the noise $f_{\mu}(t)$. The set of these times determines the behavior of the system. Below we evaluate these times for the simplest case. The final step—the analysis of the fluctuations of \mathbf{Z} in the vicinity of the stationary points, which determine the line-shapes of the radiation and its statistics—lies beyond the scope of this letter and will be discussed elsewhere.

Substitution of (2,3) into (7) at $f_{\mu} = 0$ leads to

$$\dot{z}_{\mu} = -(\Gamma_{\mu} - W_{\mu} + 2i\Omega_{\mu}) \frac{z_{\mu}}{2} - \sum_{\mu \neq \nu} (\gamma_{\mu\nu} - iJ_{\mu\nu}) \frac{z_{\nu}}{2}. \quad (8)$$

Being similar to the equations used in¹⁷ Eqs. (8) differ in two aspects. First one is the radiative coupling. The second difference is the absence of the reservoir dynamics. We believe that the depletion of reservoir is not qualitatively important in the weak lasing regime.

Two Condensation Centers.—Most of the important physics of strongly disordered systems is determined by rare configurations. In our case these are pairs of coupled CC. From now on we restrict ourselves with such a pair. The density matrix $\rho^{(2)}(z_1, z_2)$ depends on four real variables, however, the total phase, is irrelevant to what follows. We choose to parameterize the remaining degrees of freedom by three components of a 3D classical spin $\mathbf{S} = (S_x, S_y, S_z)$ using the Pauli matrices $\sigma_{\mu\nu}$,

$$\mathbf{S} = \frac{1}{2} \sum_{\mu, \nu=1,2} z_{\mu}^* \sigma_{\mu\nu} z_{\nu}, \quad (9)$$

with $S^2 = S_x^2 + S_y^2 + S_z^2 = (|z_1|^2 + |z_2|^2)^2/4$. For simplicity we assume that $(\Gamma_1 - W_1) = (\Gamma_2 - W_2) = g$; a small

difference between $(\Gamma_1 - W_1)$ and $(\Gamma_2 - W_2)$ would not affect the results qualitatively. From (8) and (9) we have

$$\dot{\mathbf{S}} = -S\nabla(\Re\mathcal{H}) - [\mathbf{S} \times \nabla(\Im\mathcal{H})], \quad (10)$$

where $\nabla = \partial/\partial\mathbf{S}$. Calling $\omega = \omega_1 - \omega_2$, $\alpha = (\alpha_1 + \alpha_2)/2$, and $\beta = (\alpha_2 - \alpha_1)/2$, we rewrite (10) explicitly

$$\dot{S}_x = -gS_x - \gamma S - [\omega + \alpha S_z - \beta S] S_y, \quad (11a)$$

$$\dot{S}_y = -gS_y + JS_z + [\omega + \alpha S_z - \beta S] S_x, \quad (11b)$$

$$\dot{S}_z = -gS_z - JS_y, \quad (11c)$$

and accordingly $\dot{S} = -gS - \gamma S_x$. We used $\gamma_{\mu\nu} = \gamma\sigma_{\mu\nu}^x$ and $J_{\mu\nu} = J\sigma_{\mu\nu}^x$.

The stationary states are solutions of (11) at $\dot{\mathbf{S}} = 0$. It turns out that, apart from TSS $\mathbf{S} = 0$, as long as $0 < g < \gamma$ there exist NTSSs. In polar coordinates (S, ϑ, φ) their positions are

$$\varphi = \pi - \arctan r, \quad \cos \vartheta = -Jr/\gamma, \quad (12a)$$

$$S = \frac{\gamma[g\omega + (J^2 + g^2)r]}{g[\beta\gamma + \alpha J r]}, \quad r^2 = \frac{\gamma^2 - g^2}{J^2 + g^2}. \quad (12b)$$

Since $r = \pm|r|$, Eq. (12b) for S gives two solutions. Each *positive* solution corresponds to a NTSS. S changes the sign together with either its numerator or its denominator, i.e., g has two bifurcation values, g_0 and g_∞ :

$$\omega^2 g_0^2 = (J^2 + g_0^2)(\gamma^2 - g_0^2), \quad (13a)$$

$$(\beta^2 \gamma^2 + \alpha^2 J^2) g_\infty^2 = \gamma^2 J^2 (\alpha^2 - \beta^2). \quad (13b)$$

At $g = g_0$ one of the NTSSs merges with the TSS. At $g = g_\infty$ and $r < 0$ the value of S diverges.

To manifest itself as an attractor the NTSS has to be linearly stable. The stability diagram Fig. 1 follows from the analysis of the Lyapunov exponents. The TSS loses its stability at g_0 . The stability of NTSS with a given S (12b) changes at $g = g_c$ defined by the equation

$$[(J^2 - g^2)\beta\gamma r + (\gamma^2 + g^2)\alpha J] gS = 2\gamma(g^4 + \gamma^2 J^2). \quad (14)$$

Here we are not aiming to describe the nonlinear dynamics of the two CCs system in all details. In particular, we discussed only the stationary states. However, there are numerical evidences for the existence of the limiting cycle (LC) in the regions IV and V in Fig. 1. Moreover, within the region V all of the stationary states are unstable, i.e., there should be at least one stable LC. Analysis of stable *time-dependent* solutions and their experimental manifestations is a subject for further studies.

Kramers transitions.—In the regions II and VII in Fig.1 two stable stationary states coexist, which leads to an additional problem: the noise term in (7) causes switching between these states. The properties of the radiation from the two states are very different. If both escape rates τ^{-1} are small enough, this switching can be observed directly. Otherwise we expect a combination of the two signals, the weights being determined by the relation between the escape rates.

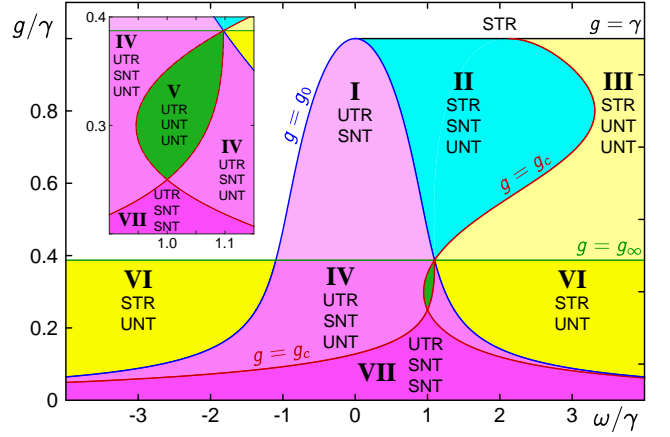


FIG. 1. Bifurcation and stability curves, according to Eqs. (13) and (14), for $J/\gamma = 0.25$ and $\alpha = 2\beta$. Stable(unstable) TSS and NTSS are indicated by STR(UTR) and SNT(UNT) in each domain. Zoomed domain V is shown in the insert.

One can evaluate τ^{-1} analytically when $J = 0$ and according to (13b) $g_\infty = 0$. In this case the region III disappears and the bistable region II is defined by the inequalities $\omega > \gamma > g > 0$. At $g = 0$ the system is conservative and integrable, i.e., (11) has two integrals of motion: S_z [see (11c)] and $A = 2\beta(\gamma S_y + \omega S) - \beta^2 S^2$. Stationary states correspond to $S_z = 0$ for all $g > 0$. For TSS $A = 0$, while for NTSSs $A = (\omega \pm \gamma)^2 = A_\pm$ (the stable and unstable NTSS correspond to + and -, respectively). Other values of A at $g = 0$ label periodic trajectories (see Fig. 2). Slow time evolution of A at small $g > 0$ is governed by

$$\dot{A} = \frac{2gE}{K_{-1}} + F(t), \quad \langle F(t)F(t') \rangle = \beta T \delta(t - t'), \quad (15)$$

$$T = \frac{4W}{K_{-1}} \left[2\omega(E + K_{-1}A) + (\gamma^2 - \omega^2 - A) \frac{\partial E}{\partial \omega} \right], \quad (16)$$

where $E = K_{+1} - 2\pi\omega\theta(A_- - A)$,

$$K_{\pm 1} = \int_0^{2\pi} R^{\pm 1/2} \theta(R) d\varphi, \quad R = (\omega + \gamma \sin \varphi)^2 - A, \quad (17)$$

with $\theta(x > 0) = 1$ and $\theta(x < 0) = 0$.

Equation (15) is a typical case of the one-dimensional Kramers problem—classical transitions between two potential minima due to the thermal noise—slightly modified by the fact that “effective temperature” (16) depends on the “coordinate” A . Using the familiar solution^{21–23} we estimate the escape rates from the stable TSS and NTSS as

$$\frac{\beta}{W\omega} \exp \left\{ -\frac{2g}{\beta} \int_{A_u}^{A_s} \frac{E dA}{K_{-1}T} \right\}, \quad (18)$$

where $A_u = A_- = (\omega - \gamma)^2$, $A_s = 0$ and $A_s = A_+ = (\omega + \gamma)^2$ for TSS and NTSS, respectively. This approach

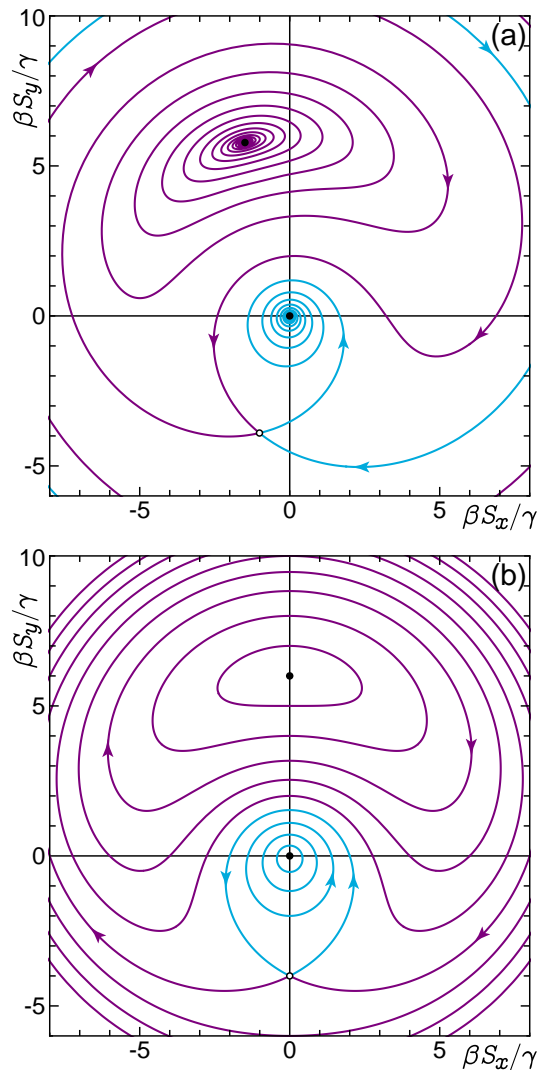


FIG. 2. Flow near NTSSs and TSS for $J = 0$ and $\omega = 5\gamma$. (a) The saddle trajectories for $g = 0.25\gamma$. (b) Several closed trajectories for $g = 0$.

is valid only provided that the exponential factor is small, i.e., when $\omega/\beta \gg 1$ and thus the occupations at NTSS are large.

One can use (18) to compare the steady state occupations of TSS and NTSS. It turns out that for small ω the NTSS prevails. The escape rate from NTSS is approximately independent of ω , while the escape rate from TSS decreases with ω . The rates become equal at $\omega \simeq 4.1\gamma$. For bigger ω TSS dominates.

Comparison to experimental observations.—We believe that a number of recently observed features of the radiation of the disordered exciton-polariton structures can be naturally interpreted in the framework of our theory. Large, $\pi/2 < \varphi < 3\pi/2$, phase differences between the

CCs, which manifests itself in the dip of the radiation intensity $I(\mathbf{k}_\perp)$ at small transverse wave-numbers \mathbf{k}_\perp was reported by several groups^{3,11–13}. A detailed analysis of the radiation from disordered CdTe structures was performed in¹³. It was found that (i) the number of CCs differs from the number of radiated frequencies; (ii) some frequencies are radiated from several CCs, and (iii) its distribution in the \mathbf{k}_\perp -space as a rule has a characteristic annular shape. The last but not the least important observation in¹³ is (iv) the clear absence of the $\mathbf{k}_\perp \rightarrow -\mathbf{k}_\perp$ symmetry, $I(\mathbf{k}_\perp) \neq I(-\mathbf{k}_\perp)$.

Features (i), (ii) is exactly what we should expect: for example, as long as TSS and NTSS in the system of two CCs coexist each CC radiates its own frequency and in addition they together radiate the third one, i.e. two CCs radiate three frequencies and one of them is radiated from both of them. The number of collectively radiated frequencies quickly increases with the number of CCs. (iii) had already been discussed – it reflects the large phase differences favored by the radiative coupling.

(iv) The time inversion symmetry implies that the phase differences between different CCs can be either 0 or π . One can check that under this condition $I(\mathbf{k}_\perp) = I(-\mathbf{k}_\perp)$. However, according to (12a) in the NTSS the phase mismatch $\varphi = \phi_2 - \phi_1$ between the two CCs is neither 0 nor π . We believe that by observing states with broken time inversion symmetry authors of¹³ experimentally proved existence of the radiative coupling.

In conclusion, we considered radiation of coupled centers of condensation of exciton-polaritons in the weak lasing regime, where the occupations n_1 and n_2 of the centers are macroscopically large, while the occupations of the states of the incoherent reservoir remain independent on $n_{1,2}$. Apart from the usual Josephson coupling between the centers we took into account the radiative coupling due to the interference of the two sources of light. The radiative coupling turns out to be responsible for the very existence of the weak lasing regime and crucial for understanding the experiments on coupled condensates. The onsite interaction between the bosons makes the dynamics of the system nonlinear and rich. In particular, depending on the parameters there can be more than one stable stationary states, and some of them are nontrivial: in contrast with the trivial stationary state (independent sources of radiation) the radiation of the two centers is synchronized in frequency. The phase difference is locked thus breaking the time-reversal symmetry. This symmetry breaking was recently observed¹³.

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