

Self-Organization of Light in Optical Media with Competing Nonlinearities

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We study the propagation of light beams through optical media with competing nonlocal nonlinearities. We demonstrate that the nonlocality of competing focusing and defocusing nonlinearities gives rise to self-organization and stationary states with stable hexagonal intensity patterns, akin to transverse crystals of light filaments. Signatures of this long-range ordering are shown to be observable in the propagation of light in optical waveguides and even in free space. We consider a specific form of the nonlinear response that arises in atomic vapor upon proper light coupling. Yet, the general phenomenon of self-organization is a generic consequence of competing nonlocal nonlinearities, and may, hence, also be observed in other settings.

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Self-organization constitutes one of the most fascinating phenomena appearing in nonlinear systems. During the process, strong interactions among the system components lead to the formation of spatial structures and long-range ordering. This effect plays a crucial role in a broad context, from biology [1–3], chemistry [4,5] and hydrodynamics [6] to soft-matter physics [7–9]. In optics the spontaneous formation of regular intensity patterns has been observed almost 30 years ago [10], and since been explored in various settings [11–14]. Common to all these experiments is the requirement of an appropriate feedback mechanism, provided, e.g., by an optical cavity or a single mirror that retroreflects traversing light back into the medium, while feedbackless pattern formation in a Kerr medium has been observed [15] from far-field interference of small-scale regular filaments. On the other hand, the formation of spatial structures solely due to the nonlinear propagation of light has attracted great interest over the past years [16,17]. Most prominently, optical solitons emerging from local Kerr-type nonlinearities of various kinds have been actively investigated [18–20] and play an important role for intense light propagation [21] and potential applications to fiber optics communication [22]. Nonlinearities can also cause extended structures to emerge, e.g., from modulation instabilities (MI) that drive a growth of broad-band density modulations and ultimately lead to the formation of randomly arranged filaments [23–26].

In this work, we show that self-organization into spatially ordered patterns [see Fig. 1(a)] of unidirectionally propagating light can occur in media with a spatially nonlocal nonlinearity. Although the absence of any feedback mechanism in our system may be expected to prevent the formation of extended patterns [27], we show that this is not the case and regular patterns can arise from a suitably

designed nonlocality of the medium. This sets it apart from previously studied systems [10–14], and as we will see below, implies profound changes of the underlying physics, including the threshold behavior for optical pattern formation [28–30]. The effect rests upon a sign change of the optical response in Fourier space [31], which in the present case drives MI within a finite band of momenta [see Figs. 1(c), 1(d)]. This condition provides a challenge for most nonlinear optics experiments where nonlocality typically arises from transport processes [32–38] that naturally yield a sign-definite nonlinear response. Overcoming this obstacle, we consider a combination of a focusing and defocusing nonlinearity [see Fig. 1(b)] and describe a physical realization of the proposed response function in atomic vapor. We derive simple conditions for the emergence of stable ordered states and show that signatures of such “crystals” are observable in the propagation of light through the medium.

Specifically, we study the evolution of a wave function $\psi(\mathbf{r}, z)$, representing the slowly varying envelope of the electric field component of a light beam. Its propagation is governed by the nonlinear Schrödinger equation

$$i\partial_z\psi(\mathbf{r}, z) = -\Delta_{\perp}\psi(\mathbf{r}, z) + U(\mathbf{r})\psi(\mathbf{r}, z) - \frac{i}{\ell}\psi(\mathbf{r}, z) - \int R(|\mathbf{r} - \mathbf{r}'|)|\psi(\mathbf{r}', z)|^2 d^2r' \psi(\mathbf{r}, z), \quad (1)$$

with \mathbf{r} and z denoting generalized transverse and longitudinal (propagation) coordinates, respectively. The amplitude $\psi(\mathbf{r}, z)$ and all other parameters in Eq. (1) represent dimensionless quantities as obtained from proper length and time scaling of the specific realization given in the Supplemental Material [39]. The parameter ℓ is the linear

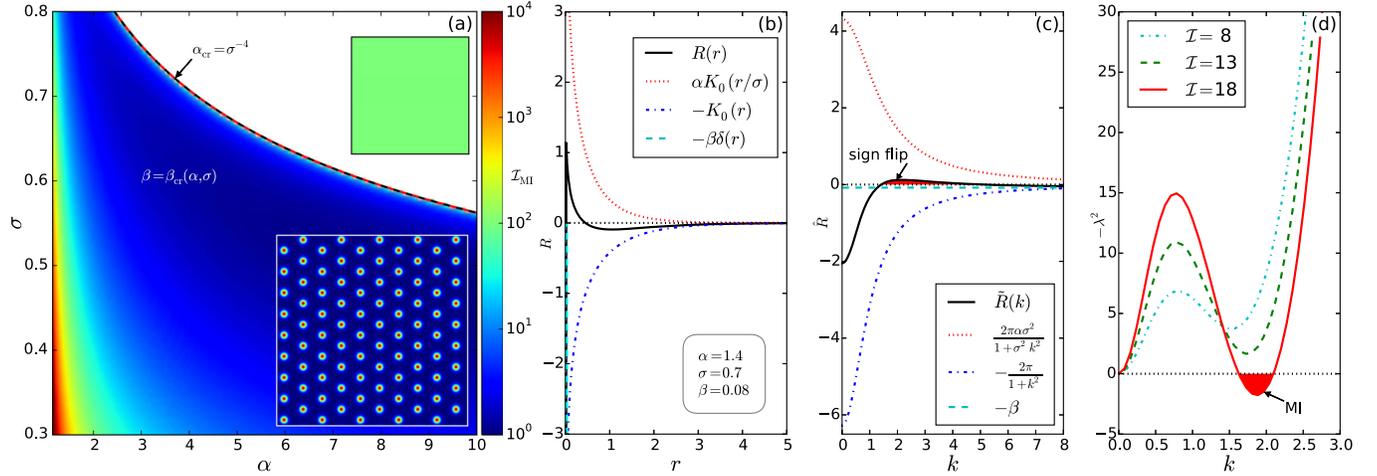


FIG. 1. (a) Where MI occurs in a finite momentum range and ordered intensity patterns (lower inset) are possible. The color coding for $\alpha < \alpha_{\text{cr}}$ shows the minimum intensity for MI, while plane wave solutions remain stable for $\alpha > \alpha_{\text{cr}}$. (b) Position [Eq. (2)] and (c) momentum space [Eq. (4)] form of the nonlinear response function (solid line) arising from a combination of a nonlocal focusing (dotted line), nonlocal defocusing (dashed-dotted line), and local defocusing nonlinearity (dashed line). Panel (d) shows the corresponding dispersion relation, Eq. (3), of periodic perturbations with momentum k .

absorption length and the external potential $U(\mathbf{r})$ may represent an additional optical waveguide. We consider a response function of the cubic nonlinearity

$$R(r) = \alpha K_0\left(\frac{r}{\sigma}\right) - K_0(r) - \beta\delta(r), \quad (2)$$

that is composed of three terms. The first and the second term describe a focusing and defocusing nonlocal nonlinearity, respectively, while the third corresponds to a local defocusing nonlinearity as given by the Dirac delta function, $\delta(r)$. The parameter $\beta > 0$ represents its strength and K_0 denotes the modified Bessel function of the second kind. Scaling with respect to the defocusing nonlinearity leaves two parameters, describing the strength ($\alpha > 0$) and spatial range ($0 < \sigma < 1$) of the focusing nonlinear response relative to that of the defocusing term [39]. While our general findings do not depend qualitatively on the shape of the nonlocal kernel, the function $K_0(r)$ plays an important role in diverse optical settings. For example, it describes light propagation in nematic liquid crystals with orientational nonlinear response [40], and was used to model the nonlinearity of thermal media [41,42]. Although most of these situations only yield a single sign-definite response, a combination of both appears possible [43,44].

Here, we suggest that the complete response function, Eq. (2), can be realized in alkali metal vapor. One can obtain a cubic Kerr nonlinearity whose nonlocal character emerges from diffusive atomic motion. In fact, the formation of nonlocal solitons due to a response function $\sim K_0(r)$ in such systems has already been demonstrated experimentally [35]. As we show in Ref. [39], the simultaneous coupling of light to near-resonant transitions involving two incoherently coupled hyperfine levels can give rise to competing nonlinearities as given in Eq. (2). Choosing the frequency detuning of the propagating light just in

between the corresponding hyperfine splitting yields a blue and red detuned transition and, thereby, two nonlocal nonlinearities of opposite sign. Moreover, the devised approach naturally provides a third, local nonlinearity, which plays a critical role for the emergence and stability of regular patterns, as we discuss below.

To this end, it appears appropriate to first consider $U(\mathbf{r}) = \ell^{-1} = 0$. The aforementioned MI refers to linear instability of plane wave solutions $\psi_{\text{pw}}(\mathbf{r}, z) = A_0 \exp(i\mu z)$ with respect to periodic modulations $a(\mathbf{r}, z) = a_1 \exp(i\mathbf{k}\mathbf{r} + \lambda z) + a_2^* \exp(-i\mathbf{k}\mathbf{r} + \lambda^* z)$ [45], where $\mu = A_0^2 \int R(|\mathbf{r}|) d^2r$ is the propagation constant. Linearization in terms of the perturbation amplitudes $a_{1,2}$ then yields the growth rate, λ ,

$$\lambda^2 = -k^2(k^2 - 2\mathcal{I}\tilde{R}(k)), \quad (3)$$

of a given mode with wave vector \mathbf{k} , and $\mathcal{I} = |A_0|^2$ is the plane wave intensity. The Fourier transform, $\tilde{R}(\mathbf{k})$, of the response function Eq. (2) reads

$$\tilde{R}(k) = \frac{2\pi\alpha\sigma^2}{1 + \sigma^2 k^2} - \frac{2\pi}{1 + k^2} - \beta. \quad (4)$$

Wherever $\tilde{R}(k) > 0$, one can find MI, i.e., a real and positive growth rate λ , for a sufficiently large intensity, \mathcal{I} , of the initial plane wave solution. In particular, if $\tilde{R}(0) < 0$ and $\tilde{R}(k)$ changes sign at a finite value of $k = k_0 > 0$, MI only occurs in a *finite* band of wavelengths $< 2\pi/k_0$ [46]. Figure 1(d) shows a typical spectrum and illustrates the onset of MI as \mathcal{I} is increased above the critical intensity \mathcal{I}_{MI} . The resulting wave number filtering is important as it yields an additional length scale emerging from initial white-noise perturbations which are typically present in experiments. On the contrary, more common long-wavelength MI requires overall focusing nonlinearities [$\tilde{R}(0) > 0$] and includes arbitrarily small wave numbers

in the instability interval. This results in an semi-infinite band of unstable wavelengths, associated with random filamentation and, ultimately, the formation of bright solitons or collapse [47,48].

For our choice of response function, $-\lambda^2(k)$ exhibits a local maximum followed by a minimum [Fig. 1(d)], which bears analogies to the known maxon-roton structure of excitation spectra of superfluid helium [49,50] and studied for Bose-Einstein condensates with finite-range interactions [51,52]. The roton minimum and the associated instability in quantum fluids may appear as a precursor to a solid phase [53,54], but can also usher in a transition to a modulated fluid described by a single-particle amplitude ψ [52,55].

In order to further analyze the present system, we consider the ground state of Eq. (1), i.e., the minimizer of the Hamiltonian density,

$$H = \frac{1}{V} \int |\nabla_{\perp} \psi_{\text{st}}(\mathbf{r})|^2 d^2 r - \frac{1}{2V} \iint R(\mathbf{r} - \mathbf{r}') |\psi_{\text{st}}(\mathbf{r})|^2 |\psi_{\text{st}}(\mathbf{r}')|^2 d^2 r' d^2 r, \quad (5)$$

in the limit of a large integration area $V \rightarrow \infty$. Since we are looking for a stationary solution, $\psi_{\text{st}} = A_{\text{st}}(\mathbf{r})e^{i\mu z}$, the Hamiltonian is only affected by the transverse profile $A_{\text{st}}(\mathbf{r})$. The analysis of Eq. (5) reveals a rich ground state behavior, including plane waves, hexagonal intensities patterns as well as bright soliton solutions. Figure 2(a) illustrates the emergence of these different phases from the plane wave solution as a function of the plane wave intensity \mathcal{I} and the strength β of the local defocusing nonlinearity. For $0 < \sigma < 1$ and $\alpha > 1$, the nonlocal part of

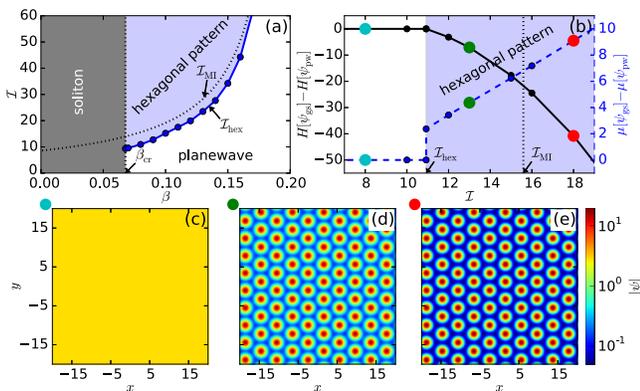


FIG. 2. (a) Phase diagram for $\alpha = 1.4$, $\sigma = 0.7$ illustrating the emergence of three different phases from the plane wave solution as a function of \mathcal{I} and β . (b) Difference in Hamiltonian density H (solid line) and propagation constant μ (dashed line) between plane wave solutions ψ_{pw} and numerically computed ground states ψ_g versus plane wave intensity \mathcal{I} ($\beta = 0.08$). Pattern formation at the threshold intensity \mathcal{I}_{hex} is accompanied by a jump in the propagation constant, and occurs well below the critical intensity for MI. Exemplary ground states for different plane wave intensities \mathcal{I} obtained from imaginary propagation (see text) are shown in (c)–(e).

the kernel Eq. (2) diverges to positive values as $r \rightarrow 0$, which inevitably leads to the existence of a bright soliton as ground state under the sole action of the nonlocal nonlinearity. Fortunately, the additional local nonlinearity $\sim \beta$ tends to diminish this short-distance focusing behavior and ultimately allows us to suppress the soliton solution upon exceeding a critical local defocusing β_{cr} . We can estimate this critical value from below through a variational analysis of the minimizer of Eq. (5), assuming a Gaussian form of $A_{\text{st}}(\mathbf{r})$ (see Ref. [39] for further details). This calculation yields a good estimate of the exact β_{cr} obtained from numerical simulations, e.g., $\beta_{\text{cr}}^{(\text{var})} \approx 0.0654$ and $\beta_{\text{cr}}^{(\text{num})} \approx 0.0678$ in Fig. 2(a).

Having obtained β_{cr} as a function of α and σ we can calculate the critical intensity \mathcal{I}_{MI} necessary to induce finite- k MI at the minimum value of $\beta = \beta_{\text{cr}}$. The result, shown in Fig. 1(a), indeed yields an extended range of parameters where a modulated ground state is possible without contracting to a single bright soliton. We find that the transition line which separates MI from the region where an initial plane wave will remain stable for every value of \mathcal{I} follows a simple relation which can be derived from the following argument. Noting that the nonlocal response asymptotically decreases as $2\pi(\alpha - 1)/k^2 > 0$ it needs to exhibit a local minimum at $k = 0$ in order to allow for a finite- k sign change through the addition of the local defocusing nonlinearity. Formally, this requirement corresponds to $\partial_k^2 \tilde{R}(k)|_{k=0} > 0$ and, thus, yields $\alpha_{\text{cr}} = \sigma^{-4}$. Alternatively, we can determine the transition line by excluding the possibility of long-wavelength MI which implies $\tilde{R}(0) \leq 0$. Since both criteria are equivalent, their combination yields the critical $\beta_{\text{cr}} = 2\pi(\sigma^{-2} - 1)$ along the transition line. This expression matches our numerical results and coincides with the variational analysis described above (see Ref. [39]).

To determine the ground state ψ_{gs} we solve Eq. (1) for an imaginary propagation coordinate ($z \rightarrow -iz$) with periodic boundary conditions and $U(\mathbf{r}) = \ell^{-1} = 0$, starting from a plane wave, $\psi(\mathbf{r}, 0) = \mathcal{I}^{1/2} + \varepsilon(\mathbf{r})$, perturbed by small amplitude white noise, $\varepsilon(\mathbf{r})$. Above the threshold intensity $\mathcal{I}_{\text{hex}} < \mathcal{I}$ we find that the ground state ψ_{gs} acquires hexagonal intensity pattern as shown in Figs. 2(d) and 2(e). This threshold value \mathcal{I}_{hex} is significantly smaller than the critical intensity for MI. While the plane wave solution remains stable for $\mathcal{I}_{\text{hex}} < \mathcal{I} < \mathcal{I}_{\text{MI}}$, it, consequently, ceases to be the lowest-energy state in this intensity region. We can detect the ground state transition by monitoring Hamiltonian density $H[\psi_{\text{gs}}]$ and propagation constant $\mu[\psi_{\text{gs}}]$ relative to those of the plane wave solution ψ_{pw} . The found behavior, shown in Fig. 2(b) is consistent with a first order phase transition as expected for two-dimensional systems [56,57]. As a result, intensity modulations in ψ_{gs} set in abruptly upon crossing \mathcal{I}_{hex} rather than growing continuously.

While MI, hence, represents a sufficient, but not necessary criterion for structured ground states, the phase transition occurs as a precursor of the instability and does not take place in systems which do not feature

finite-wavelength MI. We also note that the intensity patterns can neither be interpreted in terms of conventional bright solitons, nor do they represent dark solitons since the found state does not feature any phase structure which is typical for the latter. These observations underline again the importance of the competition between the nonlocal nonlinearities to observe the described phenomena.

Let us now study signatures of these stationary properties in the propagation of light, that would potentially be observable in experiments. We begin with the real space propagation of Eq. (1) in a hollow-core optical waveguide, which we model by a simple harmonic potential $U(\mathbf{r}) = (r/4)^2$. As the initial condition, we choose a Thomas-Fermi profile $\psi(\mathbf{r}, 0) = \mathcal{I}^{1/2} \sqrt{1 - \frac{r^2}{w^2}} + \varepsilon(\mathbf{r})$, whose width $w = 4\sqrt{-\mathcal{I}\tilde{R}(0)}$ is determined by the confining potential, the intensity \mathcal{I} , and $\tilde{R}(0) = \int R(r)d^2r < 0$. Figure 3 shows intensity profiles obtained for different input intensities \mathcal{I} below and above \mathcal{I}_{MI} . While the former case preserved the rotational symmetry and yields a nearly stationary intensity profile [Fig. 3(a)], the higher intensity results in the formation of regularly spaced filaments [Fig. 3(b)]. Dynamically, pattern formation is preceded by MI leading to rapid formation of filaments. Because of the nonlocal nonlinearity and its overall defocusing character the formed filaments experience effective repulsive interactions and eventually settle into a hexagonal lattice structure. Note, that we have set $\ell^{-1} = 0$ in order to study the dissipationless propagation dynamics. Nevertheless, ordering is still possible since the associated Hamiltonian density is dissipated into phase gradients [39] that predominantly emerge in the low-intensity regions between the filaments [Fig. 3(c)].

In Fig. 4, we show the propagation dynamics for an input beam $\psi(\mathbf{r}, z = 0) = \mathcal{I}^{1/2} \exp[-\frac{r^2}{w^2}] + \varepsilon(\mathbf{r})$, with $\mathcal{I} = 40$ and $w = 500$, for $U = 0$ and $\ell = 5.3$. Again one finds fast

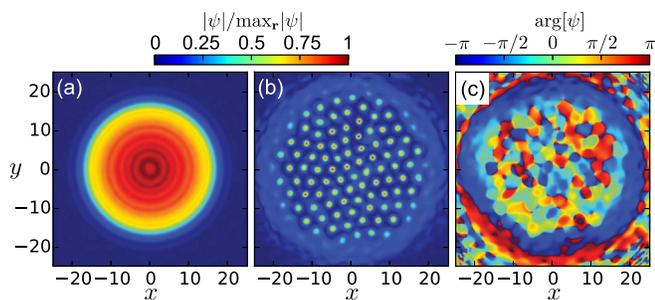


FIG. 3. Guided light propagation for $\alpha = 1.4$, $\sigma = 0.7$, $\beta = 0.08$, $U(\mathbf{r}) = (r/4)^2$, $\ell^{-1} = 0$ and two different intensities of (a) $\mathcal{I} = 10 < \mathcal{I}_{\text{MI}}$ and (b),(c) $\mathcal{I} = 20 > \mathcal{I}_{\text{MI}}$ after a propagation length of $z = 10$. Panel (c) indicates the inhomogeneous phase evolution accompanying the emergence of hexagonal intensity patterns shown in (b). (a) Below \mathcal{I}_{MI} the intensity profile develops a weak ring structure due to the initial noise. See Ref. [39] for further details.

filamentation, as indicated by the peak amplitude dynamics shown in Fig. 4(c). Subsequently, the filaments start to form short-range ordered structures. However, this self-organized state cannot be sustained against intensity loss due to absorption and beam spreading. It ultimately disintegrates once the average intensity, $\tilde{\mathcal{I}}_0(z) = V_0^{-1} \int_{V_0} |\psi(\mathbf{r}, z)|^2 d^2r$, in the central area, V_0 , approaches \mathcal{I}_{hex} .

We finally want to relate these findings to the proposed experimental realization in atomic media. As further detailed in the Supplemental Material [39], the parameters used in Figs. 3 and 4 can be obtained for a sodium vapor at a density of $9 \times 10^{13} \text{ cm}^{-3}$ where incoherent hyperfine pumping with a rate of $2\pi \times 0.9 \text{ MHz}$ transfers population from the $|F = 1\rangle$ to the $|F = 2\rangle$ state, and vice versa with a rate of $2\pi \times 3.9 \text{ MHz}$. Coupling the light field detuned by $2\pi \times 14 \text{ MHz}$ from the D_1 transition then yields $\alpha = 1.4$, $\sigma = 0.7$, and a dimensionless absorption length of $\ell = 5.3$. The dimensionless intensity $\mathcal{I} = 40$ then gives the reasonable value of 300 W/cm^2 . For a diffusion constant of $30 \text{ cm}^2/\text{s}$ the dimensionless unit length corresponds to $10 \mu\text{m}$, making the predicted patterns observable with conventional imaging techniques. Generally, the number of tunable parameters entails considerable flexibility, allowing us to find viable experimental conditions for other combinations of α and σ as well.

In summary, we have investigated the emergence of crystalline intensity patterns due to a competition of non-local optical nonlinearities with different signs and ranges. The phenomenon was traced back to a first-order phase transition between ground states of the underlying propagation equation. Yet, we showed that it should be observable in the unidirectional propagation of light, facilitated by Hamiltonian density dissipation into phase gradients. We

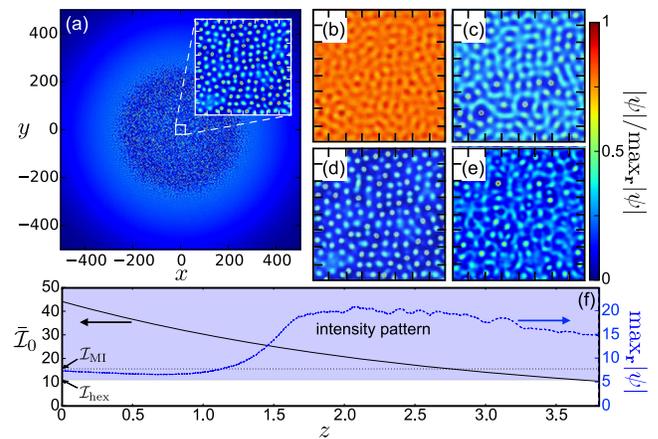


FIG. 4. Free propagation for $\alpha = 1.4$, $\sigma = 0.7$, $\beta = 0.08$, $U = 0$, $\ell = 5.3$. During propagation regular intensity patterns form in the beam center, as shown in the inset of (a). Panels (b)–(e) show the propagation dynamics in this central region of area V_0 for different propagation lengths $z = 1$ (b), 1.7 (c), 2.3 (d), and 3.7 (e). (f) Evolution of the peak amplitude, $\max_r |\psi|$, and average intensity, $\tilde{\mathcal{I}}_0$, in the central area V_0 . See Ref. [39] for further details.

have devised a physical implementation in dilute atomic vapor that realizes the proposed model. However, the presented analysis also applies to other media in which light propagation is adequately described by Eq. (1). We hence expect this work to be relevant to such systems where competing nonlocal nonlinearities may arise from different transport mechanisms, including particle or heat diffusion or reorientation of induced dipoles.

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