Necklace beam generation in nonlinear colloidal engineered media

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Modulational instability is a phenomenon that reveals itself as the exponential growth of weak perturbations in the presence of an intense pump beam propagating in a nonlinear medium. It plays a key role in such nonlinear optical processes as supercontinuum generation, light filamentation, rogue waves, and ring (or necklace) beam formation. To date, a majority of studies of these phenomena have focused on light–matter interactions in self-focusing Kerr media existing in nature. However, a large and tunable nonlinear response of a colloidal suspension can be tailored at will by judiciously engineering the optical polarizability. Here, we analytically and numerically show the possibility of necklace beam generation originating from spatial modulational instability of vortex beams in engineered soft-matter nonlinear media with different types of exponential nonlinearity.

Metamaterials offer a strong potential to enable a plethora of novel nonlinear light–matter interactions and even new nonlinear media. Linear and nonlinear properties of metamaterials can be suitably tailored by properly adjusting the dimensions, periodicity, and other parameters of the meta-atoms [1]. Metamaterials were predicted to drastically change fundamental nonlinear processes, including second-harmonic generation, soliton propagation, four-wave mixing, modulation instability (MI), and optical bistability, to name a few [2].

To date, the majority of studies in the field of metamaterials have focused on solid-state nanotranstructures. However, engineering of optical properties in soft matter offers new degrees of freedom for designing optical polarizabilities. While the first ideas of using interplay of optical forces [3] and nonlinear self-action effects [4] were proposed in the 70s and 80s, recently it was proposed that colloidal suspensions can offer a promising platform for engineering polarizabilities and large, and tunable nonlinearities [5–13]. Recently, it was demonstrated that the nonlinear response of colloidal suspensions can be tailored at will by judiciously engineering their optical polarizability [6]. Such systems may provide new opportunities for fundamental studies and applications of such nonlinear effects as self-focusing and related beam-breakup effects.

First described by Askar’yan [14], self-focusing and filamentation in various nonlinear media have been studied by many researchers [15–18]. In particular, transverse spatial periodic breakup of an optical beam due to self-focusing has been theoretically predicted and experimentally observed. It was shown to be related to supercontinuum generation, filamentation, rogue waves, and necklace beam generation [2,19–25].

The necklace beams, first studied by Soljacic et al. [26–28], are ring-shaped beams with intensities that are azimuthally periodically modulated [29–34]. In these studies, it was theoretically predicted and experimentally demonstrated that necklace beams with and without the orbital angular momentum (OAM) launched in self-focusing Kerr medium maintain their shape, in spite of the inherent instability in such a medium, and that their diameters increase during propagation at a predictable rate. In another series of studies, self focusing of optical vortices in Kerr media was investigated, and the azimuthal MI for ring-shaped vortices was shown to result in the beam breakup into a ring of filaments [35–37]. Importantly, a majority of these studies focused on light-matter interactions in self-focusing Kerr medium. Recently, the optical nonlinearity of colloidal suspensions, despite the initial belief that it was of the Kerr type, was found to be exponential [6]. Moreover, this exponential nonlinearity can be saturable or supercritical with intensity, depending on the sign of the particle polarizability in the suspension.

Therefore, in this Letter, we analytically and numerically study spatial MI in soft-matter nonlinear media with these two distinct types of nonlinearity using beams with an OAM. Our results may be of interest for imaging and spectroscopic applications using light propagating in highly scattering biological and chemical colloidal media.

To understand the reason behind these different types of nonlinearity, we note that colloidal suspensions can be created by two kinds of particles; with positive polarizabilities (PP) corresponding to a particle refractive index greater than the background index ($n_p > n_b$) or with negative polarizability (NP), with the corresponding refractive index of a particle lower than that of the background material ($n_p < n_b$) [6]. PP
dielectric particles are attracted toward the high-intensity regions of the beam, whereas NP particles are repelled. At the same time, in the PP case, the nonlinear scattering losses increase in the high-intensity region of the beam. In contrast, in the NP case, the nonlinear losses decrease with the increase of intensity since the particle concentration decreases.

While a majority of previous studies investigated nonlinear propagation of Gaussian beams in colloidal suspensions [6–13], here, we analyze the azimuthal MI for optical vortices with different topological charges propagating in the nanocolloidal system. We show that different types of the exponential optical nonlinearity in the PP and NP cases lead to different MI gain for the same perturbation and to the formation of different spatial distributions of the necklace beams.

Let us consider an optical beam propagating in the nanocolloidal system. The nonlinear Schrödinger equation governing the evolution of the slowly varying electric field envelope $\phi$ can be written as [10,11]

$$i \frac{\partial \phi}{\partial z} + \frac{1}{2k_0n_0} \nabla^2_x \phi + k_0(n_p - n_b)V_p \rho_0 e^{i\sigma_0 |\phi|^2} \phi + i\frac{2}{\sigma_0} \sigma_0 e^{i\sigma_0 |\phi|^2} \phi = 0. \tag{1}$$

The particle polarizability is denoted by $\alpha$, and $k_0T$ is the thermal energy. $V_p$ is the volume of a particle, $\rho_0$ is the unperturbed particle concentration, $\sigma$ is the scattering cross section [11], and $k_0 = 2\pi/\lambda_0$ is the wavenumber. After normalization, Eq. (1) reads:

$$i \frac{\partial U}{\partial \xi} + \frac{\nabla^2_x U}{1} + (a + i\delta) \exp[a|U|^2]U = 0, \tag{2}$$

where $U$ is the normalized field amplitude, $\delta$ is normalized loss coefficient, and $\xi$ is the normalized propagation distance. For PP particles, the normalized nonlinear parameter $a = 1$. Consequently, the nonlinearity in the PP case is super-critical. For NP particles $a = -1$ and the nonlinearity is saturable. The MI is analyzed by applying a method involving a linear stability analysis [2] to Eq. (2). The azimuthal phase dependency of the vortex beams can be easily expressed in the cylindrical coordinate frame. Therefore, it is convenient to express the transverse Laplacian in Eq. (2) in this frame:

$$i \frac{\partial U}{\partial \xi} + \left( \frac{1}{r} \frac{\partial U}{\partial r} + \frac{\partial^2 U}{\partial \varphi^2} + \frac{1}{r^2} \frac{\partial^2 U}{\partial \theta^2} \right) + a \exp[a|U|^2] U = 0. \tag{3}$$

In Eq. (3), we neglect the loss term to facilitate the analytical studies of the MI. The normalized propagation distance is $\xi = Z/(2k_0n_0L^2)$, the normalized radial coordinate $r = \sqrt{X^2 + Y^2}/L$, and $L = (2k_0^2n_0n_p - n_b|V_p\rho_0|)^{-1/2}$.

Before we present the results of the studies of the OAM beam propagation, it should be mentioned that propagation of Gaussian beams with zero OAM was analyzed in soft-matter systems with the supercritical/saturable nonlinearities [6–13]. These studies predicted that the 2D beams can form solitons in both PP and NP particle-based suspensions. Experimental study showed a collapse of 3D Gaussian beams in the PP case and a stable soliton propagation with self-induced transparency in the NP case. Here, we study the evolution of the OAM beams in nanocolloidal suspensions with supercritical/saturable nonlinearities. A careful comparison of the terms in the Taylor expansion of the nonlinear term in Eq. (3) shows that the higher-order terms are non-negligible and that the nonlinear response of the medium considered in this study cannot be approximated by the pure Kerr nonlinear term.

We start with performing a standard linear stability analysis of the OAM beam propagating in a nanocolloidal system. We apply azimuthal perturbations to the steady-state solution $U_0(r, \vartheta)$ of Eq. (3). Here, perturbations are applied only to the azimuthal field distribution taken at the radial distance for which the intensity is constant $U_0(\vartheta) = U_0(r = r_m, \vartheta)$, where $r_m$ defines a mean radius of the steady-state solution and it is calculated using Eq. (21) in Ref. [20]. The perturbed field distribution is given by

$$U_p(\xi, \vartheta) = ([U_0] + a_1 e^{-i(M0 + \mu)\xi} + a_2 e^{-i(M0 + \mu^* \xi)} e^{i\xi + i\mu \vartheta}), \tag{4}$$

where $[U_0]$ is the amplitude of the steady-state solution, $a_1, a_2$ are the amplitudes of the small perturbations $(a_1, a_2 \ll |U_0|)$, and $m$ and $M$ are the azimuthal indices of the steady-state solution [topological vortex charge $(m = 0, 1, 2, \ldots)$ and the perturbation, respectively, $\lambda$ is the propagation constant of the steady-state solution, and $\mu$ is the propagation constant correction for the perturbation.

Substituting Eq. (4) into Eq. (3) and linearizing the resulting equation in $a_1$ and $a_2$, we obtain coupled equations for perturbation amplitudes that can be written as an eigenvalue problem in the following matrix form:

$$\begin{bmatrix} A + \mu & B \\ -B & C + \mu \end{bmatrix} \begin{bmatrix} a_1 \\ a_2 \end{bmatrix} = 0, \tag{5}$$

where

$$A = -\lambda - \frac{(m + M)^2}{r_m^2} + f(|U_0|^2) + f'(|U_0|^2)|U_0|^2;$$

$$B = f'(|U_0|^2)|U_0|^2; \quad \text{and}$$

$$C = \lambda + \frac{(m - M)^2}{r_m^2} - f(|U_0|^2) - f'(|U_0|^2)|U_0|^2.$$ 

Here, $f(|U|^2) = a \exp[a|U|^2]$ and a prime denotes the derivative with respect to $|U|^2$. Equation (5) has nontrivial solutions when the matrix determinant is equal to zero. This condition yields two expressions: (i) for the propagation constant $\lambda = -\frac{d^2}{d\xi^2} + f(|U_0|^2)$, and (ii) for the propagation constant correction $\mu$ associated with the MI. The MI gain is given by the imaginary part of $\mu$:

$$\text{Im}(\mu) = M/r_m \times \text{Im} \sqrt{(M/r_m)^2 - 2|U_0|^2 \exp[a|U_0|^2]).} \tag{6}$$

The difference between the gain amplitude $\text{Im}(\mu)$ in the PP and NP case is the consequence of a different value of the parameter $a$ depending on the sign of polarizability.

Equation (6) is used in the following to predict the MI gain for vortices propagating in the nanocolloidal medium. We study the propagation of light with the wavelength $\lambda_0 = 532$ nm in two systems: (i) the PP suspension composed polystyrene particles ($n_p = 1.56$) randomly dispersed in water ($n_b = 1.33$) with the volume filling fraction $(f = V_p/\rho_0) = 1.4 \times 10^{-3}$; (ii) the NP suspension made of low-refractive-index particles (air bubbles with $n_p = 1$) dispersed in water with the volume filling fraction $f = 10^{-3}$. In both cases, the radius of the particles is assumed to be 50 nm. For these two sets of the parameters, Fig. 1 shows the gain curves $\text{Im}(\mu)$ as a function of the
perturbation azimuthal index $M$ for different values of the vortex charge $m$.

We observe that, for a fixed vortex charge $m$, the MI gain is higher in the PP particle-based system than in the NP particle-based system due to the different type of nonlinearity. As a result, for a fixed initial power, the MI onsets for a shorter propagation distance for PP than for the NP case. Additionally, we observe that the number of peaks in the necklace beam, associated with the perturbation azimuthal index $M$ for which $\text{Im}(\mu)$ is maximal, is larger in the PP case than in the NP case, for a fixed initial vortex charge.

The theoretical predictions based on the analytical expression for the MI gain [Eq. (6)] are verified by a direct numerical solution of Eq. (1). This equation is solved using a 3D split-step Fourier algorithm [38,39] to study the vortex dynamics in nanocolloidal systems. The input field in the numerical algorithm corresponds to the steady-state solution of Eq. (1) $U_m(r, \theta)$ for the $m$th order optical vortex:

$$
\psi(r, \theta, z = 0) = A_m(r/\alpha_0)^m \exp^{i z^2/(2 r_m^2) + i m \phi},
$$

where the width $\alpha_0$ for the $m$th order stable vortex is related to the averaged radius $r_m$ as $\alpha_0 = r_m/(m + 1)$ and the amplitude $A_m$ can be deduced from the total power of the stable solution

$$
P_m = \frac{4 \pi k_B T 2^{2m+1} m! (m + 1)!}{|\alpha|} L^2 \varepsilon_0 c n \mu,
$$

where $c$ denotes the speed of light in vacuum and $\varepsilon_0$ is the vacuum permittivity. To accelerate the growth of the MI, 10% of random noise is added to the input field.

Figures 2 and 3 show the dynamics of light propagation of vortex beams with various topological charges $m$ for PP and NP particle-based systems, respectively. The first column presents the input steady-state solution with the noise, the second column shows the vortex at half of the distance corresponding to the MI onset, and the third column presents the generated necklace beam immediately after the onset of the MI breakup. Detailed numerical studies show that there is good qualitative agreement between the dynamics of the beam evolution in our case and the predictions of Ref. [26]. Indeed, the radius increases slowly at the beginning of the propagation of the necklace beams, and then, the increase is almost linear. However, quantitatively, the results differ owing to the different type of nonlinearity and the presence of losses.

There is a good agreement between the analytical predictions shown in Fig. 1 and the numerical simulations. Indeed, for a given charge $m$, the number of maxima observed in the necklace beam generated by the numerical simulations corresponds to the azimuthal frequency $M$ with the highest growth rate predicted analytically. Moreover, in excellent agreement with analytical predictions, numerical simulations show that the necklace-beam formation occurs at shorter distances in the PP case than in the NP case. In both PP and NP cases, the distance at which the MI appears decreases with the increase of the vortex topological charge. The distance required to observe the MI for the PP case is more than 3 times shorter than that for the NP case. However, the analytical predictions suggest that this distance should be only two times shorter. Another discrepancy between the analytical predictions and the numerical calculations is the power required to observe the steady-state solution in the case of NP particles. These discrepancies can be explained based on the assumptions made in the analytical calculations.

The values of the power corresponding to the steady-state solutions in the NP system calculated using Eq. (8) are $P_1 = 3.39 \, \text{W}$, $P_2 = 6.78 \, \text{W}$, $P_3 = 10.86 \, \text{W}$, and $P_6 = 26 \, \text{W}$. These values were obtained neglecting losses and taking only three first terms of the Taylor expansion of the nonlinear function $f(|U|^2)$. In the system with loss and a full exponential nonlinearity, the steady-state solutions computed in the approximate analytical way are not fully stable. The width of these solutions oscillates during the propagation, and they
experience loss. Therefore, to observe the MI in the numerical simulations, we need to modify the stable solutions found analytically. We found out that increasing the analytically predicted input power for each topological charge by the same factor of 2.4 while keeping all the other parameters unchanged suffices to observe the beam breakup. The input power levels used in the numerical simulations were $P_1 = 8.25$, $P_2 = 16.5$, $P_3 = 26.4$, and $P_6 = 63$ W. These powers correspond to the continuous-wave laser light peak intensity of the order of $1 \text{ MW/cm}^2$, which is comparable to that used in recent experimental studies of similar nonlinear colloidal suspensions [6].

In summary, we investigated the phenomenon of spatial MI of OAM beams in colloidal nonlinear media consisting of PP or NP particles and predict the formation of the necklace beams. We show that different types of exponential nonlinearity (saturable in the NP case and supercritical in the PP case) lead to different MI gains for the same perturbation. As a result, for the fixed input-beam power, the MI onsets at a shorter propagation distance for PP as compared to the NP case. Also, the number of peaks in the necklace beam, associated with the perturbation azimuthal index $M$, is larger for the PP case than for the NP case, for a fixed initial vortex charge. These results may be of great importance to future studies of nonlinear propagation of structured light beams and filamentation in liquids. They can also be useful in applications of light propagation in highly scattering biological and chemical colloidal media.

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