Optical spatial solitons in soft-matter: mode coupling theory approach

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We predict that spatial self-trapping of light can occur in soft matter encompassing a wide class of new materials such as colloids, foams, gels, fractal aggregates etc. We develop a general nonlocal theory that allows to relate the properties of the trapped state of Maxwell equations to the measurable static structure factor of the specific material. We give numerical evidence for stable trapping in fractal aggregates and suggest also the possibility of soliton spectroscopy of soft-matter.

Self-trapping of light beams, predicted forty years ago, is still a subject of great interest. Observation of optical spatial solitons (OSS) at low (down to mW) power levels, demonstrated in photoreflective or liquid crystals, makes OSS attractive candidates in several applications of emerging photonics technology, and have driven successful efforts to understand them. Yet, the description of light trapping seems still incomplete, and several applications of emerging photonics technology, or liquid crystals, makes OSS attractive candidates in bio-matter where OSS can find new applications (e.g., laser surgery, optical manipulation of nano-particles). Roughly speaking, softness is generally due to a characteristic mesoscopic (i.e., larger than atomic) length scale of the constituents, and an energy scale comparable to room temperature thermal energy. As a consequence, SM properties can be easily tailored via external field of different (mechanical, electrical, magnetic, thermal, ...) origin. SM includes colloidal suspensions, emulsions, and foams (all involving different constituents in a host fluid), and typical examples are polymers in a liquid, glues, liposomes, blood, and all sort of bio-matter.

Historically, the use of aerosols and water suspension of dielectric spheres as nonlinear media dates back to the early 80’s. However, in these materials electrostriction has been described in the framework of simple models that treat the diluted constituents as a gas of non-interacting particles, in terms of local index change of the Kerr type (∆n = n2I where I is the local intensity). As a consequence existence of stable OSS in two transverse dimensions is ruled out by the occurrence of the well-known catastrophic self-focusing instability. Phenomenologically, stabilization can be expected from the index saturation arising from the maximum packing fraction of the dispersed particles. However, before reaching such a regime, the physics of these materials, and in general of other SM systems, is well-known to be affected by particle-particle interactions. In particular, this occurs whenever the particle-particle correlation function g(r) is structured on a length scale comparable to the laser beam waist. Under these conditions a nonlocal model for self-focusing in SM must be considered.

In this letter, we propose a novel general approach to stationary self-focusing, linking for the first time the electrostrictive nonlocal response of SM to its static structure factor S(q) (roughly speaking, the Fourier transform of the particle-particle correlation function), usually measured by means of scattering experiments. This allows us (i) to predict stable propagation of two-dimensional OSS in a new wide class of condensed matter, and (ii) to assess the importance that ultra-focused laser light can have to investigate the properties of SM. Both issues are of paramount importance in order to go towards a more general description of solitons in complex media (fractal aggregates, structured and supercooled liquid, etc.) and their application in bio-photonics as well as to develop a new spectroscopic tool for the investigation of SM properties.

Assuming a linearly polarized beam and exploiting isotropy of system, we start from the unidirectional scalar wave equation written for a monochromatic beam with complex amplitude E(x, y, z) propagating along z

\[ i \frac{\partial \xi}{\partial z} + \sqrt{k^2 + \nabla_z^2} \xi + \frac{\omega}{2\varepsilon_0} \rho_a(E) = 0. \]  

where \( k^2 = \frac{\omega^2(\varepsilon)/c^2 \propto \omega^2 n_0^2/c^2, n_0 \) is the SM bulk refractive index, and \( \nabla_z^2 = \partial_z^2 + \partial_y^2 \). We further assume that the nonlinear polarization is responsible for a refractive index change \( \Delta n \), i.e., \( \rho_a(E) = \Delta \chi E = 2n_0 \Delta n \xi \), which is dominated by an electrostrictive contribution (fast electronic nonlinearities as well as index change due to thermal heating by optical absorption are usually negligible, and we also neglect scattering). This legitimates our scalar (polarization-independent) approach, regardless of beam spectral content [at variance with Kerr effect of electronic origin where the non-paraxial regime at very high intensities requires to account for vectorial effects (see e.g., [13]).]

Since the electrostriction \( \Delta n = \rho(\partial n/\partial \rho)_{p=0} \) is proportional to the particle number density change \( \rho \) (from equilibrium value \( p_0 \)), Eq. (1) must be coupled to an evolution equation for \( \rho \). A widely accepted and largely applicable theory for SM is the so-called Mode-Coupling...
Theory (MCT) \[14\] [15] [16] which relies on the so-called Zwanzig-Mori formalism (given some observable, like \(\rho\), it allows to write closed equations for it and for its correlations functions [17]). By exploiting MCT (see also [18]), we find that the density perturbation \(\tilde{\rho}(q,t)\) obeys the dynamical equation:

\[
\ddot{\tilde{\rho}}(q,t) + q^2 \frac{k_B T}{\eta_0} \tilde{\rho} + q^2 \int_0^1 m(t - t') \dot{\tilde{\rho}}(q,t') dt' = \frac{f(q,t) + \frac{1}{2} \gamma_q \eta_q^2 I(q)}{f(q,t)},
\]

(2)

where \(\tilde{\rho}\) denotes 3D spatial Fourier transform, \(\eta = (\mu_0 / c_0 n_0^2)^{1/2}\) is the impedance, \(k_B\) is the Boltzman constant, \(T\) is the temperature, \(m(t)\) is the memory function of the system (for a simple liquid \(m(t)\) is a Dirac delta function times the viscosity), and \(S(q)\) is the static structure factor. In Eq. \(2\), \(f\) is a Langevin term describing random forces (see e.g. [18]).

Eq. \(2\) has been always considered without the deterministic forcing term weighted by the electrostrictive coefficient \(\gamma_q = \rho \partial e / \partial \rho\) [21], and provides one of the most successful approaches to structural phase transitions of soft-matter. Here we extend it by accounting for the presence of an external optical field [coupled through Eq. \(1\)], which induces an electrostrictive force with potential proportional (in configurational space) to \(\Delta I\). This model can be thought of as a generalization of the acoustic wave equation [12], which has been previously employed to determine the electrostrictive correction to electronic nonlinearity of silica glass [19]. The latter case considered here, MCT accounts for the elastic deformation of a medium made of interacting particles which results in a non-homogeneous response weighted by \(S(q)\).

Additionally, though here we deal with electrostriction, MCT is a powerful approach that can be generalized to account for other types of nonlinearity, e.g. reorientational mechanisms, by looking at different observables. As such, it provides a general framework for studying nonlinear optics in SM, going beyond the idealized Kerr (local) limit [4, 5] (which is, nevertheless, correctly retrieved for non-interacting particles, as shown below). In the following, we address specifically the properties of spatial solitons.

Assuming that the random fluctuations are negligible with respect to the driving electrostrictive term, the stationary state solution (\(\partial_t = 0\) of Eq. \(2\)) yields

\[
\tilde{\rho}(q) = \frac{\gamma_q \eta S(q)}{2 k_B T} \frac{\partial}{\partial q} I(q),
\]

(3)

which shows that \(S(q)\) plays the role of a transfer function from the optical intensity to the density. Incidentally, Eq. \(4\) can be also obtained by starting from a different model of SM employing so-called generalized hydrodynamics equations [22] such as those typically adopted to modelling inelastic light scattering spectra (ISTS) [21]. Obviously, in real space, Eq. \(4\) corresponds to a differential equation for \(\rho = \rho(x,y,z)\). While this equation, as well as Eq. \(1\), involves three dimensions, OSS imply by definition a \(z\)-independent intensity. Therefore, in this case, \(\rho\) does not depend on \(z\) and Eq. \(4\), coupled to Eq. \(1\), will be interpreted henceforth as a 2D transverse equation where \(q = (q_x, q_y)\).

Equations \(1\) \(3\) allow us to develop a general non-local model for trapping in SM. First we consider the paraxial (Fresnel) regime, which corresponds to expanding the transverse operator in the \(q\)-Fourier transform of Eq. \(1\) as \(\sqrt{k^2 - q^2} \approx k[1 - q^2/(2k^2)]\). Equation \(1\) becomes, in terms of the slowly varying envelope \(E(x,y,z) = \mathcal{E}(x,y,z) \exp(-i k z)\),

\[
2 i k \frac{\partial E}{\partial z} + \nabla_x^2 E + \frac{2 k^2}{n_0} \left( \frac{\partial n}{\partial \rho} \right) \rho E = 0.
\]

(4)

In Eq. \(1\) the nonlinearity arises from the term \(\rho E\). Indeed, back-transforming Eq. \(4\) to real space and assuming azimuthal symmetry, we obtain, after some algebra, a self-consistent nonlinear non-local wave equation

\[
i 2 k \frac{\partial E}{\partial z} + \nabla_x^2 E + \chi E \int_0^{2 \pi} G(r, \theta') E(r', z) r' dr' = 0,
\]

(5)

where \(r \equiv \sqrt{x^2 + y^2}\), and we have defined the kernel

\[
G(r, \theta') \equiv \int_0^{2 \pi} \frac{S(Q)}{S_0} J_0(Qr) J_0(Qr') QdQ,
\]

(6)

and the coefficient \(\chi \equiv k^2 \frac{\partial n}{\partial \rho} \gamma_\eta S_0 / 2 k_B T n_0\) (note that \(S(q)\) is scaled to \(S_0 = S(0)\) representing the ratio between the compressibility of the material and that of the ideal gas [22]). We seek for bound states of Eqs. \(4\) \(5\) in the form \(E(z, r) = (\chi w_0)^{-1/2} u(\sigma) \exp(i \beta \zeta)\), where \(\beta\) is the nonlinear correction to the wavevector \(k\), which is determined self-consistently in the numerical simulations, and \(\sigma = r / w_0\), \(\zeta = z / z_0 = z / 2 k w_0^2\) are dimensionless radial and longitudinal variables, in units of beam width \(w_0\) and diffraction (Rayleigh) length \(z_0\), respectively. The OSS (bound state) profile \(u(\sigma)\) obeys the non-local eigenvalue equation (we set \(w_0^2 \nabla^2 = \nabla^2 = d^2 / d\sigma^2 + \sigma^{-1} d / d\sigma\))

\[
\nabla^2 u - \beta u + u \int_0^{2 \pi} g(\sigma, \sigma') u^2(\sigma') \sigma' d\sigma' = 0,
\]

(7)

where the kernel \(g(\sigma, \sigma')\) can be obtained (at least numerically), once \(S(Q)\) is known, from the integral

\[
g(\sigma, \sigma') = \int_0^{2 \pi} S(\theta / w_0) J_0(\sigma \theta) J_0(\sigma \theta) \theta d\theta.
\]

(8)

From Eqs. \(7\) \(8\) the ideal local (Kerr) limit is recovered for \(S(q) = S_0 = \text{constant}\), which yields \(g(\sigma, \sigma') = (\sigma -...
\[ n_2 = \frac{4\pi^2 \eta k_B}{c n_0^3} \left( \frac{\varepsilon_s - \varepsilon_h}{\varepsilon_s + 2\varepsilon_h} \right)^2 \rho_0 S_0 \frac{1}{k_B T}. \]  \tag{9}

This limit, however, is well known to lead to unstable (so-called Townes after Ref. [1]) OSS. Conversely, in the general case \( S(q) \neq \text{constant} \), we expect solutions of Eqs. (7-8) to be stabilized by non-locality \[23, 24, 25, 26, 27\]. We are also naturally brought to consider deviation from paraxiality due to strong focusing, and argue for the existence of OSS in this case. In fact, narrow OSS in SM may be important both in specific applications (e.g. laser surgery) and in order to establish OSS as a mean for probing the static structure factor \( S(q) \) of SM when the latter extends to high spatial frequencies. Deviations from paraxiality can be accounted for by considering the next order in the expansion of the transverse operator in Eq. (4). By adopting the normalization employed for the paraxial case, we cast the new bound state equation in the form

\[ \nabla_\sigma^2 u - \varepsilon \nabla^4 u - \beta u + u \int_0^\infty g(\sigma, \sigma') u^2(\sigma') \sigma' d\sigma' = 0, \]  \tag{10}

where the degree of non-paraxiality is measured by a single dimensionless parameter \( \varepsilon = (\lambda/4\pi n w_0)^2 \) fixed by the ratio between wavelength \( \lambda \) and beam width scale \( w_0 \).

In order to discuss various OSS supported by different types of SM, we make specific examples. We start considering hard spheres in a host liquid (solvent). In the limit of diluted, non-interacting spheres, \( S(q) \) is constant and this yields, once again, unstable OSS. In a more refined approximation, \( S(q) \) can be described by a parabolic law in the framework of the Percus-Yevick model \[22\]

\[ S(q) = S_0 + K q^2. \]  \tag{11}

After Eq. (4), the corresponding expression for \( \rho \) (as stated, we assume \( \rho \) to follow adiabatically \( I \) along \( z \)) is

\[ \rho(r, z) = \frac{\gamma e \eta}{2k_B T} \left[ S_0 I(r, z) - K \nabla^2 \left( I(r, z) \right) \right] \]  \tag{12}

which, once inserted in Eq. (4), gives a model for weakly non-local solitons that has interdisciplinary interest (plasma physics, matter waves, transport in DNA, see Ref. [28] and references therein). Stable soliton solutions of this model have been reported \[22\] and, in this context, represent 1+2D OSS in SM, when its static structure factor can be well approximated by Eq. (11). To this end, consider that, using Percus-Yevick model, the parabolic approximation of \( S(q) \) breaks down around \( q r_s \approx 5 \).

Since \( q \) can be reasonably estimated to be \( q \sim w_0^{-1} \), the weakly non-local model starts to lose its validity when the spheres have size comparable with beam width \( w_0 \) (in this regime, a microscopic description of the molecular dynamics is needed). Vice versa, when Eqs. (11) hold valid, the nonlocality that, generally speaking, provides the stabilizing mechanism of OSS against catastrophic self-focusing \[22\] stems from the particle-particle correlation function \( g(r) \), which is proportional to the Fourier transform of \( S(q) - 1 \) \[22\], as anticipated. Importantly, since \( S(q) \) is not uniform, stable OSS not only exist, but provide information on the material [for a given optical power, the width of OSS is determined by the constant \( K \) in Eq. (11)].

Further models for \( S(q) \) can be discussed. A very intriguing case is that in which the suspended particles of colloidal SM develop self-similar aggregates with fractal dimension \( D \), described by the function \[22\]

\[ S(q) = 1 + \frac{D}{(q r_s)^2 \sin[(D - 1)\tan^{-1}(q\xi)]}, \]  \tag{13}

where \( \Gamma \) is the Gamma function, \( r_s \) is the sphere radius, and \( \xi \) gives the spatial extension of the aggregate. Incidentally, when \( D = 2 \), Eq. (13) yields \( S(q) = 1 + 2(\xi/r_s)^2 (1 + q^2 \xi^2)^{-1} \), which entails the sum of a Kerr contribution and a non-local one with Lorentzian lineshape. In the limit \( q \xi \ll 1 \), Eq. (13) yields

\[ S(q) = \Gamma(D + 1) \frac{\xi}{r_s} \left[ 1 - \frac{D(D + 1)}{6 q^2 \xi^2} \right], \]  \tag{14}

and the corresponding model for \( \rho \) reads as:

\[ \left[ 1 - \frac{D(D + 1)}{6 \xi^2 \nabla^2} \right] \rho = \frac{\eta c \Gamma(D + 1)(\xi/r_s) I}{2k_B T}. \]  \tag{15}

From Eq. (15), it is readily seen that \( \rho \) spatially decays (when \( I = 0 \)) as the modified Bessel function \( K_0 \) of argument \( \xi \sqrt{D(D + 1)}/6r \) which depends on the fractal dimension. Recalling that \( \sigma = r/w_0 \) and that the degree of nonlocality is the ratio between the spatial decay rate of the optically induced index perturbation and that of the self-trapped beams, our result implies that the degree of optical non-locality scales basically as the fractal dimension of the material. Noteworthy, Eqs. (6) \[15\] define another well known model for non-local OSS, which applies in the case of nematic liquid crystals \[29\].

In order to show that OSS exist also in the general case [Eq. (10)], with features directly linked to the fractal dimension \( D \), we resort to numerical integration of Eqs. (4) \[16\] (or Eqs. (13) in the non-paraxial regime) using finite difference discretization in \( \sigma \) and Newton-Rapson iterations. To fix the ideas, we show results for the characteristic values of the following length scale ratios (between aggregate dimension \( \xi \), particle radius \( r_s \), beam width \( w_0 \)) \( \xi = 100 \) and \( \kappa = \xi/w_0 = 0.1 \). In Fig. 1(a) we show existence curves, i.e. the soliton normalized peak intensity against the normalized soliton width \( \max[u^2] \) vs. std of \( u^2 \) parametrized by \( \beta \), obtained for three values of \( D \). As shown the features of OSS change...
with fractal dimension $D$. This is even more clear from Fig. 1 (b), where we display the OSS normalized power (i.e., the norm $Q = 2\pi \int u^2 r dr$) and width as a function of the fractal dimension $D$ (here we fix $\beta = 2$). In Fig. 2 we show the effect of non-paraxiality. The features of OSS starts to exhibit significant deviations when the soliton width decreases significantly, and non-paraxial effects are no longer negligible.

It is interesting to observe that for a fractal medium the equation for $p$ in the configurational space includes fractional derivatives. Soft matter, not only provide a un-precedented framework to study nonlocality with a taylorable structure factor, but also opens the may to new mathematical models, as we will discuss in future publications.

To prove that stable self-trapping can be achieved for input conditions that do not exactly match the OSS profile, we have run beam propagation simulations for the paraxial and non-paraxial models. In Fig. 3 we show the spatial evolution of an input Gaussian $\text{TEM}_{00}$ laser beam $u(\sigma, \zeta = 0) = A \exp(-\sigma^2)$, whose parameters do not exactly match the existence condition. The resulting longitudinal beam oscillations, which depend on beam power (for a fixed width) and degree of non-paraxiality, have been reported previously for other non-local solitons [3]. They are connected with the fact that non-local solitons are absolutely stable [2], and can be related to excitation of "internal modes" of the soliton [2, 31], or, in the framework of the highly non-local approximation [i.e., when $I(q)$ varies on a q-scale much broader than that of $S(q)$] to the existence of exact breathing solutions [30, 32, 33]. Notably, in the latter regime, $I(q) \simeq I(0) \equiv P$ in Eq. [30], with $P$ the optical beam power. In this case, Eq. [30] takes the form of the linear Schrödinger equation for a quantum particle in a 2D potential well with shape dictated by particle autocorrelation $g(r)$ and power $P$, and OSS reduce to the bound states, which can be found by standard techniques. Such results let us envisage a broad physical setting for the observation of deeply oscillating nonlocal solitons and show that their existence is not restricted to the paraxial regime.

In summary we have shown that optical beams can be self-trapped in soft matter both in the paraxial and tightly focusing regimes, thus opening new perspectives for their applications in biological materials and as a mean to probe properties of condensed matter. For example, fixing the input beam waist and adjusting the incoming power to find the appearance of a soliton allows one to directly measure the fractal dimension of the aggregates. Assessing the role of time dynamics and thermal contributions as well as the validity of the present approach in other condensed matter systems (e.g., supercooled liquids, where optical trapping is unexplored to date) will be natural extensions of this work.

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[34] Specific expressions for $\gamma_e$ follow from the dependence of the dielectric constant on particle density $\epsilon(\rho)$, e.g. for a suspension of dielectric spheres of radius $r_s$, $\gamma_e = \rho_0 4\pi r_s^2 (\epsilon_h - \epsilon_s)/(\epsilon_h + 2\epsilon_s)$, where subscripts $h$ and $s$ refer to the host medium and the spheres, respectively.
[35] The 2D approach is justified also for input beams that do not match exactly the OSS profile, since changes in $z$ occur usually on a length scale much longer than the transverse dependence of $\rho$ that yields the trapped state.