Exact description of self-focusing in highly nonlinear geometrical optics

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Abstract

We demonstrate that laser beam collapse in highly nonlinear media can be described, for a large number of experimental conditions, by the geometrical optics approximation within high accuracy. Taking into account this fact we succeed in constructing analytical solutions of the eikonal equation, which are exact on the beam axis and provide: i) a first-principles determination of the self-focusing position, thus replacing the widely used empirical Marburger formula, ii) a benchmark solution for numerical simulations, and iii) a tool for the experimental determination of the high-order nonlinear susceptibility. Successful comparison with several experiments is presented.

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Nonlinear light self-focusing is a self-induced modification of the optical properties of a material which leads to beam collapse at a certain point $z_{sf}$ in the media. This effect, first observed in the 1960s, plays nowadays a key role in all scientific and technological applications related to the propagation of intense light beams [1], like material processing [2], environmental sciences [3], femtochemistry in solutions [4], macromolecule chromatography [5], medicine [6], etc.

Usually, $z_{sf}$ is estimated using the empirical Marburger formula [1, 7, 8], which has been constructed via fitting the results of extensive numerical simulations obtained for the case when the refractive index $n$ is a linear function of the electric field intensity $n = n(I) = n_0 + n_2 I$, $(n_2 > 0)$ [9, 10, 11, 12]. In most modern experiments, however, high beam intensities are used for which the linear approximation breaks down, and further contributions to $n(I)$ must be considered [7, 8, 13]. For these cases no general mathematical condition for the behavior of $z_{sf}$ and the filament intensity has been derived so far. Most theoretical results are based on numerical studies, or on variational calculations assuming a fixed beam profile inside the medium (see e.g. [7] and Refs. therein). An analytical theory, able to accurately describe beam collapse in highly nonlinear optics, is still missing. Moreover, it is widely believed that the exact treatment of beam propagation in a highly nonlinear medium can only be done numerically [1].

In this letter, we construct for the first time analytical solutions for the eikonal equations with highly nonlinear forms of the refractive index avoiding any a priori assumptions on the form of the beam during propagation. The results obtained are exact on the beam axis within the geometrical optics approximation, which we demonstrate to be accurate for many of the situations taking place in modern experiments. Our approach permits not only to obtain exact expressions for $z_{sf}$ for different nonlinear functions $n(I)$, but also to find a general mathematical framework which corrects traditionally used formulas for the filament intensity calculation [7]. Since the accuracy of the semi-classical approximation can be easily estimated, we can determine and control the error in our calculations, which is not possible in the case of the Marburger formula.

Based on these results we are also able to propose experiments to precisely determine the high order nonlinear susceptibility of different materials. Moreover, our results yield a natural explanation of the experimentally observed [14] deviation of the scaling law $z_{sf} \sim I^{-1/2}$ for high beam intensities.

We consider the propagation of a linearly polarized laser beam of initially Gaussian shape. Starting from the nonlinear wave equation and assuming that the light beam is almost monochromatic and that the envelope varies slowly in space and time, one obtains a generalized nonlinear

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FIG. 1: (Color online.) Accuracy $L \equiv L_{nl}/L_{diff}$ of the geometrical optics approximation for different media as a function of the laser pulse power $P_{in}$. The green, black and blue curves refer to water, air and fused silica, respectively. The pulse wavelength is assumed to be 800nm. $P_{in}$ is given in units of TW for air, and of GW for water and fused silica.

Schrödinger equation (NLSE) of the form \[1\]:

\[i\partial_z \mathcal{E} + \frac{1}{2k_0}\partial_{xx} \mathcal{E} + k_0 n(|\mathcal{E}|^2) \mathcal{E} = 0.\]

where $\mathcal{E}$ is the electric field, $z$ the propagation length and $k_0$ the wave vector. The second term describes wave diffraction on the transverse plane. $n(|\mathcal{E}|^2)$ is the nonlinear refractive index. The magnitude of the contributions of the diffraction and the nonlinear effects to the beam propagation can be estimated through the comparison of the characteristic distances $L_{diff}$ and $L_{nl}$, at which the beam suffers considerable changes \[7\]. Then, $L \equiv L_{nl}/L_{diff}$ is a measure of the accuracy of the geometrical optics approximation: if $L \ll 1$, diffraction can be neglected.

The main contribution to $n(|\mathcal{E}|^2)$ is usually given by the Kerr cubic term $n_2 |\mathcal{E}|^2$. Therefore it is natural to define a nonlinear length $L_{nl} = 1/(k_0 n_2 I_0)$, where $I_0$ is the intensity of the beam at the entry plane of the nonlinear medium. The diffraction length is defined as $L_{diff} = n_0 k_0 w_0^2/2$, where $w_0$ is the initial beam radius \[7\]. In Fig.1 we plot $L$ as a function of the initial beam power $P_{in}$ ($P_{in} = I_0 w_0^2 \pi/2$) for different media. In many recent experiments $L \sim 0.05$ or smaller (see \[7\] and Refs. therein), thus making the geometrical optics approximation valid.

We represent the electric field $\mathcal{E}$ in Eq. (1) in the eikonal form: $\mathcal{E} = \sqrt{I} \exp(i k_0 S)$. Neglecting
the diffraction term we obtain
\[ \partial_z S = -\frac{1}{2} (\partial_z S)^2 + n(I), \quad \partial_z I = -\partial_z (I \partial_z \varphi). \] (2)

As a next step we rewrite these equations in a dimensionless form by considering \( x \) and \( z \) in units of \( w_0 \) and the beam intensity \( I \) in units of \( I_0 \). We also introduce the dimensionless variable \( v \equiv \partial_x S \) and differentiate the first of Eqs. (2) with respect to \( x \). The refractive index term yields \( \partial_x n(I) \equiv a \varphi \partial_x I \), where \( \varphi = \varphi(I) \equiv \partial_I n(I) \) and \( a \equiv n_2 I_0 \) is a dimensionless parameter. The order of magnitude of \( a \) in a large number of modern experiments lies below \( 10^{-5} \) \([7]\). It is therefore reasonable to consider \( a \) as a small parameter.

The boundary value problem to be solved can be summarized as
\[ \partial_z v + v \partial_x v - a \varphi \partial_x I = 0, \quad \partial_z I + v \partial_x I + I \partial_x v = 0, \]
\[ v(0, x) = 0, \quad I(0, x) = \exp(-x^2), \] (3)
which describes the propagation of an initially collimated Gaussian beam in a nonlinear medium. Solutions of Eqs. (3) and their derivatives can exhibit singularities for particular values of \( z \). Analyzing these points, we obtain the nonlinear self-focusing position \( z_{sf} \) of the laser beam \([15]\).

Following Ref. \([12]\), one can notice that the system (3) is linear with respect to the first order derivatives. Therefore, it is convenient to use a hodograph transformation \([16]\) in order to transform it into a linear system of the form
\[ \partial_w \tau - \frac{I}{\varphi} \partial_I \chi = 0, \quad \partial_w \chi + a \partial_I \tau = 0, \] (4)
where \( \tau = I z, \chi = x - vz, w = v/a \). The boundary conditions are transformed as: for \( w = 0, \chi = \sqrt{\ln(1/I)} \) and \( \tau = 0 \).

Taking into account the smallness of \( a \) we solve Eqs. (4) by proceeding in two steps \([17, 18]\). First, accounting for the boundary conditions, from the first of Eqs. (4), we find \( \tau \) as a function of \( \chi, I \) and \( w \):
\[ \tau = -w/(2\chi \varphi). \] (5)
Then, substituting Eq. (5) into the second of Eqs. (4), we obtain a closed partial differential equation for the variable \( \chi \)
\[ \partial_w \chi + \frac{aw}{2\chi^2 \varphi} \partial_I \chi + \frac{aw}{2\chi^2 \varphi^2} \partial_I \varphi = 0. \] (6)
Integration of Eq. (6) results in two invariants

\[ \chi \varphi = \Psi_1, \quad \int \varphi^{-1} dI - a \tau^2 = \Psi_2. \]  

(7)

Then, the final steps for the construction of the desired solutions of Eqs. (3) can be summarized as follows. With the help of Eqs. (7) we express \( I \) and \( \chi \) as functions of the integration invariants: \( I = I(\Psi_1, \Psi_2) \) and \( \chi = \chi(\Psi_1, \Psi_2) \). Then, we require that according to the boundary conditions for \( \tau = 0 \) the equation \( I(\Psi_1, \Psi_2) = \exp[-\chi(\Psi_1, \Psi_2)^2] \) must be fulfilled. Finally, substituting \( \Psi_1 \) and \( \Psi_2 \) into the relation above and returning to the original variables, we get the solution of Eqs. (3).

The scheme presented above allows us to find analytical solutions of the optics equations for different types of nonlinearities \( \varphi(I) \). For high field intensity, the refractive index of most materials contains nonlinear contributions additional to the Kerr-term \( n_2 I \). Usually, they are modeled as a power function of the intensity in the general form \( \beta I^K \). Physically, this term can be attributed to the fifth order nonlinear susceptibility \( n_4 I^2 \) [19, 20] or to the material ionization \( \sigma_K I^K \), where \( K \) is the number of photons absorbed, and \( \sigma_K \) being the multiphoton ionization (MPI) cross section [17].

In the following we use our approach to determine under which conditions self-focussing takes place or can be prevented by the high order nonlinearity, and arrive at a new general mathematical equation to obtain the filament intensity, which improves previous theories. Let us first consider a system having a nonlinear part of the refractive index of the form \( n(I) = n_2 I - n_4 I^2 \) (i.e., \( K = 2 \)). In this case \( \varphi = 1 - \beta I \), where \( \beta \equiv 2n_4 I_0/n_2 \). Substituting \( \varphi \) into Eqs. (7), we get \( \Psi_2 = -1/\beta \ln(1 - \beta I) - a \tau^2 \) and \( \Psi_1 = \chi(1 - \beta I) \). Thus, the solution to Eqs. (3) reads

\[
1 - (1 - \beta I)e^{aI^2z^2\beta} = \beta \exp\left(\frac{-x^2e^{-2aI^2z^2}}{[1 - 2aIz^2(1 - \beta I)]^2}\right),
\]

\[
v = -2aIzx(1 - \beta I)/[1 - 2aIz^2(1 - \beta I)].
\]

After differentiating these expressions with respect to \( x \) and \( z \), solving the obtained system of four algebraic equations with respect to \( \partial_x I, \partial_t v \) etc., and substituting the resulting expressions into Eqs. (3), one can verify that the obtained solutions are exact on the beam axis \( (x = 0, v|x=0 = 0) \) [21]. The on-axial beam intensity distribution is given by

\[
aI^2z^2\beta = \ln \left[ \frac{1 - \beta}{1 - \beta I} \right],
\]

(8)

Analyzing the Eq. (8) we find a critical value \( \beta_c \sim 0.175 \). For \( \beta_c > 0.175 \), Eq. (8) has no special points, the on-axial intensity monotonically increases and reaches a saturation value \( I_{sat} \).
By analyzing the asymptotic behaviour of \( I = I(z) \) we obtain \( I_{\text{sat}} = 1/\beta \). Note that this value fulfills the condition \( 1 - \beta I_{\text{sat}} = \varphi(I_{\text{sat}}) = 0 \). For \( \beta_c < 0.175 \) there is an interval \([z_1, z_2]\) on the beam axis where the solution \( I(z) \) is not unique. The first point \( z_1 \) can correspond to development of an instability in the beam, while the second point \( z_2 \) corresponds to \( z_{sf} \), where the intensity sharply increases, and the beam is compressed into a single filament with \( I_{\text{sat}} \).

For materials described by \( n(I) = n_2 I - n_6 I^3 \), we have \( K = 3 \), \( \varphi = 1 - \beta I^2 \), \( \beta = 3n_6 I_0^2/n_2 \) and the on-axial intensity distribution is given by

\[
\text{arctanh}(\sqrt{\beta}I) - aI^2 z^2 \sqrt{\beta} = \text{arctanh} \sqrt{\beta}. \tag{9}
\]

From the analysis of the asymptotic behavior \((z \to \infty)\) we obtain \( \beta_c \sim 0.05 \), \( I_{\text{sat}} = 1/\sqrt{\beta} \). By inspection, we realize again that, as for \( K = 2 \), the intensity of the beam saturates when \( \varphi = 0 \).

Notice that the values of \( I_{\text{sat}} \) for \( K = 2 \) and 3 obtained here are different from previous theoretical estimates \([7, 19, 22]\), which were obtained assuming that the intensity in the filament saturates when the nonlinear terms in \( n(I) \) compensate each other \([7]\). From the present results we see, however, that this is not the case. Upon propagation, the beam tends to reach the on-axial value of the intensity which maximizes the index of refraction at the beam axis. In other words, not the nonlinear refractive index, but its variation should be zero:

\[
\partial_I n(I)|_{I_{\text{sat}}} = 0. \tag{10}
\]

This new condition is general, independent of the medium or material and represents one of the predictions of this letter, which should serve as a basis for future calculations. Note that such a general mathematical condition would have been impossible to obtain on the basis of numerical simulations.

We now apply our theoretical scheme to study the concrete problem of femtosecond laser pulse propagation in air, which is relevant due to a large number of applications and whose description is still a subject of discussion (see, e. g. Refs. \([7, 8]\) and Refs. therein). The nonlinear refractive index of air is taken in the following widely used form

\[
n = n_2[I + f(I)] + n_4[I^2 + f(I^2)] - \frac{\rho(I)}{2\rho_c}, \tag{11}
\]

where the first term describes the Kerr response involving a delayed (Raman) contribution \( f(I) = \tau_K^{-1} \int_{-\infty}^{t} e^{-(t-t')/\tau_K} I(t')dt' \). \( n_2 \) and \( \tau_K \) are known to be equal to \( 3.2 \times 10^{-19} \text{ cm/W}^2 \) and 70 fs \([23]\), respectively. \( n_4 \equiv \chi^{(5)}/2n_0 \), where \( \chi^{(5)} \) is the fifth order nonlinear susceptibility its exact value is a
subject of some controversy \cite{8,20}. The most accepted estimates lie around $\sim 10^{-32}$ cm$^4$/W$^2$ \cite{7}. In the last term of Eq. (11), $\rho(I)$ refers to the density of free electrons and $\rho_c$ denotes the critical density above which the plasma becomes opaque. A rough estimate yields $\rho(I) \sim \sigma K^2 \rho_{at} t_p$, where $\rho_{at}$ is the atom density $\rho_{at} = 2 \times 10^{19}$ cm$^{-3}$ and $t_p$ the pulse duration. $K = 8$ for the MPI with a pulse of 800 nm, and $\sigma_8 = 3.7 \times 10^{-96}$ cm$^{16}$/W$^8$/s \cite{7}.

A numerical solution of the NLSE for the pulse with $P_{in} \sim 0.08$ TW and $w_0 = 3$ mm with $n(I)$ given by Eq. (11) with $n_4 = 0$ gives $z_{sf,I} = 128.18$ cm \cite{24}. Note that if $n(I) = n_2 I$, the self-focusing distance is given by the Kovalev formula \cite{11,12}: $z_{sf} = w_0/(2\sqrt{n_2 I_0})$, which is exact under the geometrical optics approximation and for initially Gaussian beam shape. For the experimental conditions of Ref. \cite{24} this yields $z_{sf,I} = 127.66$ cm. This confirms our initial statement that for many experiments the diffraction (and in this case also the plasma defocusing) can be neglected.

Recently reported experimental results on air clearly indicate that $z_{sf}$ scales as $1/\sqrt{P_{in}}$ for a relatively low initial pulse power $P_{in}$. However, for powers above 500 GW ($I_0 \sim 5 \times 10^{12}$ W/cm$^2$) a qualitative change is observed and $z_{sf}$ depends on the power as $\sim 1/P_{in}$ \cite{14}. This behavior was attributed by authors to noise effects in the beam. On the other hand one can notice that the value of 500 GW in the experiment of Ref. \cite{14} corresponds to the case where the terms $n_2 I_0 = 1.7 \times 10^{-6}$ and $n_4 I_0^2 \sim 3.1 \times 10^{-7}$ in the refractive index (11) become comparable. Therefore, we believe that the change in the power dependence is mainly driven by the contribution of the highly nonlinear terms (fifth order susceptibility). Moreover, from the analysis of the experiment of Ref. \cite{14} within our theoretical scheme we can draw important conclusions regarding the form of the nonlinear refractive index.

Using the value of $I_0$ given in Ref. \cite{14}, and assuming $n_4 \sim \pm 10^{-32}$ cm$^4$/W$^2$ \cite{7}, we get $\beta \simeq \pm 0.35$. Comparing this value with $\beta_c \sim 0.175$, we see that, if $n_4 < 0$, then no self-focusing would be observed. Since self-focusing is indeed observed, we conclude that $n_4 > 0$. The behavior of $z_{sf}$ as a function of the initial beam intensity obtained from our theory is shown in Fig. 2 and is qualitatively compared to the measured $z_{sf}$. Note that one could use our results to find an accurate value of $n_4$ by performing a similar experiment with a controlled initial Gaussian beam profile (i.e., without noise) and fitting the measured curve $z_{sf}(I_0)$ to the solution of Eq. (8).

Based on the results of this work it is clear that one is able to determine experimentally the nonlinear optical constants by measuring $I_{sat}$. As we discussed above, the last term in Eq. (11) does not change $z_{sf}$ significantly. However, its value is crucial for $I_{sat}$. In Ref. \cite{25} $z_{sf}$ and $I_{sat}$...
FIG. 2: (Color online.) Dependence of the self-focusing position on the beam intensity. The blue curve refers to the dependence $1/\sqrt{I}$ (obtained for low intensities), whereas the black curve shows the deviation for high intensities due to influence of the fifth-order nonlinearity $n_4 = 10^{-32} \text{cm}^4/W^2$. Inset: Experimental points for $z_{sf}(P_{in})$ from Ref. [14].

for air were measured. In the experimental setup a collimated beam with FWHM diameter of $d \sim 4 \text{ mm} (w_0 = d/\sqrt{2\ln 2})$, a pulse duration (FWHM) of 450 fs and a pulse energy $E_{in} \sim 20 \text{ mJ}$ was applied. We took these values and used them to calculate the on-axial intensity distribution from our analytic equations. The comparison between our theory and experimental results of Ref. [25] is shown in Fig. 3. The temporal pulse compression due to MPI is estimated along the similar lines with Ref. [23]. For the refractive index given by Eq. (11), we calculate $I_{sat}$ on the basis of Eq. (10). The value of $n_4$ chosen in a way to provide a satisfactory fit of experimental date for both $z_{sf}$ and $I_{sat}$ is equal to $8 \times 10^{-32} \text{ cm}^4/W^2$. It is necessary to point out that for a more accurate determination additional experimental studies are required.

In Ref. [25] an additional experiment was reported using a shorter pulse of duration (FWHM) 50 fs and energy 19 mJ. For the same model input-parameters, we obtain: $I_{sat} = 1.26 \text{ W/cm}^2$ and the fluence $F = 0.67 \text{ J/cm}^2$ which are in a good agreement with the experimental results $I_{sat} = 1.3 \text{ W/cm}^2$ and $F \sim 0.6 \text{ J/cm}^2$, correspondingly.

In Table I we demonstrate the influence of high order nonlinearities on the self-focusing position. The Marburger Formula (MF) reads $z_{sf} = 0.367 z_0 \left( (\sqrt{P_{in}/P_{cr}} - 0.852)^2 - 0.0219 \right)^{-1/2}$; here $z_0 = \pi w_0^2/\lambda \sim 45\text{m}$, $P_{cr} = \lambda^2/2\pi n_2 \simeq 3.3\text{GW}$. The “effective critical power” mentioned in the table means a replacement $P_{cr} \rightarrow P_{cr}'$, where the relation $P_{cr}' = 2P_{cr}$ was used [24]. From the Table one can see that other analytical approaches which do not account for high order nonlin-
FIG. 3: Filamentation in air. On-axial fluence versus the propagation distance. Circles are the experimental results from Ref. [25]. The black curve represents our analytical solution.

TABLE I: Comparison of the predictions for $z_{sf}$ in air calculated using different theoretical approaches and compared with the experimental data of Ref. [25].

| Experiment                                      | $z_{sf}$  |
|------------------------------------------------|-----------|
| Marburger formula (MF) [9]                     | 6.1 m     |
| MF with an effective critical power [7]        | 9.9 m     |
| Kovalev formula ($n_4 = 0$) [11]                | 9 m       |
| Eq. (8) for $n_4 = 8 \times 10^{-32} \text{cm}^4/\text{W}^2$ | 8.3 m     |

earities cannot provide a satisfactory prediction of $z_{sf}$ in modern high-intensity experiments.

Finally, and for the sake of completeness, we present results obtained by applying our theory to other types of nonlinearities in order to predict the behavior of self-focusing for other cases of current physical interest [13]. If the nonlinear refractive index has the form $n(I) = aI/(1 + \beta I)$, the on-axial intensity distribution is given by the expression

$$[(1 + \beta I)^3 - a3\beta I^2 z^2]^{1/3} - 1 = \beta.$$  

For $n(I) = 1 - e^{-\beta I/a}$, the on-axial intensity is given by

$$e^{\beta I} - a^2 z^2 \beta^2 I^2 = e^\beta,$$

and for a polynomial form $n(I) = I^k$ ($k \neq 2$) by

$$I^2[I^{-k} + k(k - 2)az^2] = 1.$$

No intensity saturation has been observed.
The singularities in solutions of equations above correspond to the $z_{\text{sf}}$ for the given $n(I)$. It should be noted that the approach suggested in this letter can be generalized to arbitrary $n(I)$. In general, the solutions can be found semi-analytically by interpolation of the integrals in Eqs. (7).

Summarizing, exact solutions for the self-focusing length $z_{\text{sf}}$ and the filament intensity $I_{\text{sat}}$ for several different forms of the nonlinear refractive index in the framework of geometrical optics were obtained. Depending on the experimental conditions, these solutions can be very accurate, describe the essential physics of the problem and explain different independent measurements. The analytical expressions obtained for the dependence $I = I(z)$ constitute a clear improvement with respect to the empirical Marburger formula.

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