

Exact solutions of the eikonal equations describing self-focusing in highly nonlinear geometrical optics

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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 mathematical condition for obtaining the filament intensity, (iii) a benchmark solution for numerical simulations, and (iv) a tool for the experimental determination of the high-order nonlinear susceptibility. Successful comparison with experiment 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], such as 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 $n=n(I)=n_0+n_2I$ ($n_2>0$) [9]. Under the geometrical optics approximation, and for the same form of refractive index, exact analytical expressions for z_{sf} have been obtained in Refs. [10–13]. 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 [7,8,14]. 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 references 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 paper, we construct 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 $n(I)$ in (1+1) and (1+2) dimensions, but also to find a general mathematical framework which corrects traditionally used formulas for the filament intensity [7] and approximate expression for z_{sf} [15]. Since the accuracy of the semiclassical 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.

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 Schrödinger equation (NLSE) of the form [1]

$$i\partial_z\mathcal{E} + \frac{1}{2k_0}\nabla_{\perp}^2\mathcal{E} + k_0n(|\mathcal{E}|^2)\mathcal{E} = 0, \quad (1)$$

where \mathcal{E} is the electric field, z is the propagation length, and k_0 is 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, $L \equiv L_{\text{nl}}/L_{\text{diff}}$ is a measure of the error 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 $L_{\text{nl}}=1/(k_0n_2I_0)$, where I_0 is the intensity of the beam at the entry plane of the nonlinear medium. The diffraction length is defined as $L_{\text{diff}}=n_0k_0w_0^2/2$, 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} ($P_{\text{in}}=I_0w_0^2\pi/2$) for different media. In many recent experiments $L \sim 0.05$ or smaller (see [7], and references therein), thus making the geometrical optics approximation valid [16,17].

Thus we consider Eq. (1) under the semiclassical approximation. We represent the electric field \mathcal{E} in Eq. (1) in the eikonal form $\mathcal{E}=\sqrt{I}\exp(ik_0S)$, and introduce a dimensionless variable $\mathbf{v} \equiv \nabla_{\perp}S$. The operator ∇_{\perp} acts in the plane perpendicular to the z axis; the vector \mathbf{v} has a single nonzero component v in this plane. Omitting the high-order derivative

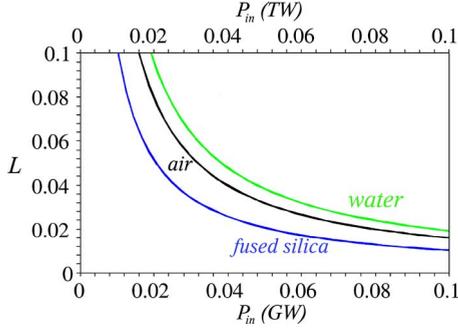


FIG. 1. (Color online) Error $L \equiv L_{nl}/L_{diff}$ of the geometrical optics approximation for different media as a function of the laser pulse power P_{in} . The green, black, and blue curves refer to water, air, and fused silica, respectively. The pulse wavelength is assumed to be 800 nm. P_{in}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

term related to diffraction, we get the boundary value problem

$$\begin{aligned} \partial_z I + v \partial_x I + I \partial_x v + (v-1)Iv/x &= 0, \\ \partial_z v + v \partial_x v - \varphi(I) \partial_x I &= 0, \\ I(0, x) = I_0 \exp(-x^2/w_{in}^2), \quad v(0, x) &= 0, \end{aligned} \quad (2)$$

where φ is related to the refractive index by $\varphi(I) \equiv \partial n(I)$. $\nu=1$ and $\nu=2$ correspond to the (1+1)- and (1+2)-dimensional case, respectively. Equations (2) describe the propagation of an initially collimated Gaussian beam with waist $w_{in} = w_0/\sqrt{2}$ [8] in an arbitrary nonlinear medium. Solutions of Eqs. (2) 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 [18].

For convenience we introduce dimensionless variables $\tilde{I} \equiv I/I_0$, $\tilde{x} \equiv x/w_{in}$, $\tilde{z} \equiv z/w_{in}$, a function $\tilde{\varphi}(\tilde{I}) \equiv \varphi(I)/n_2$, and a parameter $a \equiv n_2 I_0$. The order of magnitude of a in a large number of modern experiments lies below 10^{-5} [7]. It is therefore reasonable to view a as a small parameter. First, we consider (1+1) dimensions ($\nu=1$). Following Ref. [10], one can notice that in (1+1) dimensions the system of Eqs. (2) is linear with respect to the first-order derivatives. Therefore, it is convenient to use a hodograph transformation [19] in order to transform it into a linear system of the form

$$\partial_w \tau - \tilde{I} \tilde{\varphi}^{-1} \partial_{\tilde{I}} \chi = 0, \quad \partial_w \chi + a \partial_{\tilde{I}} \tau = 0, \quad (3)$$

where $\tau = \tilde{I} \tilde{z}$, $\chi = \tilde{x} - v \tilde{z}$, $w = v/a$. The boundary conditions are transformed as $w=0$, $\chi = [\ln(1/\tilde{I})]^{1/2}$, and $\tau=0$.

We solve Eqs. (3) by proceeding in two steps [20,21]. First, setting $a=0$ and assuming that $\partial_w \chi=0$, we find τ as a function of χ , \tilde{I} , and w from the first of Eqs. (3),

$$\tau = -w/(2\chi\tilde{\varphi}). \quad (4)$$

Then, substituting Eq. (4) into the second of Eqs. (3), we obtain a closed partial differential equation for the variable χ ,

$$\partial_w \chi + \frac{aw}{2\chi^2 \tilde{\varphi}} \partial_{\tilde{I}} \chi + \frac{aw}{2\chi \tilde{\varphi}^2} \partial_{\tilde{I}} \tilde{\varphi} = 0. \quad (5)$$

Integration of Eq. (5) results in two invariants

$$\chi \tilde{\varphi} = \Psi_1, \quad \int \tilde{\varphi}^{-1} d\tilde{I} - a\tau^2 = \Psi_2. \quad (6)$$

With the help of Eqs. (6) we express \tilde{I} and χ as functions of the integration invariants: $\tilde{I} = \tilde{I}(\Psi_1, \Psi_2)$ and $\chi = \chi(\Psi_1, \Psi_2)$. Then, we require that, according to the boundary conditions, for $\tau=0$ the equation $\tilde{I}(\Psi_1, \Psi_2) = \exp[-\chi(\Psi_1, \Psi_2)^2]$ must be fulfilled.

The scheme presented above allows us to find analytical solutions of the optics equations for different types of nonlinearities $\tilde{\varphi}(\tilde{I})$. 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 βI^K . Physically, this term can be attributed to the fifth-order nonlinear susceptibility $n_4 I^2$ [8,24] or to the material ionization $\sigma_K I^K$, where K is the number of photons absorbed, and σ_K is the multiphoton ionization (MPI) cross section [7].

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 $\tilde{\varphi} = 1 - \beta \tilde{I}$, where $\beta \equiv 2n_4 I_0/n_2$. Substituting $\tilde{\varphi}$ into Eqs. (6), we obtain $\Psi_2 = -1/\beta \ln(1 - \beta \tilde{I}) - a\tau^2$ and $\Psi_1 = \chi(1 - \beta \tilde{I})$. Thus, the solution to Eqs. (2) in two dimensions reads as

$$(1 - e^{-\beta \Psi_2}) = \beta e^{\Psi_1} e^{\beta \Psi_2}. \quad (7)$$

We return to the original variables in Eqs. (4) and (7), differentiate them with respect to \tilde{x} and \tilde{z} , and solve the obtained system of four algebraic equations with respect to $\partial_{\tilde{x}} \tilde{I}$, $\partial_{\tilde{z}} v$, etc. Substituting the obtained expressions into Eqs. (2) for $\nu=1$, one can verify that the obtained approximate solutions [Eqs. (4) and (7)] are exact on the beam axis ($x=0$, $v|_{x=0}=0$). As a rule, due to the symmetry, the behavior on the beam axis reflect the major properties of the beam such as self-focusing, defocusing, filamentation, etc. [22].

Based on the results of the renormalization-group analysis of Refs. [10,13] (for $\tilde{\varphi}=1$), we make the Ansatz that, when going from (1+1) to (1+2) dimensions, the variables scale as $z \rightarrow z/\sqrt{2}$, $v \rightarrow v\sqrt{2}$; thus the solution reads as

$$1 - (1 - \beta \tilde{I}) e^{a \tilde{I}^2 \tilde{z}^2 \beta/2} = \beta \exp\left(\frac{-\tilde{x}^2 e^{-a \beta \tilde{I}^2 \tilde{z}^2}}{[1 - a \tilde{I} \tilde{z}^2 (1 - \beta \tilde{I})]^2}\right),$$

$$v = -a \tilde{I} \tilde{x} (1 - \beta \tilde{I}) / [2 - 2a \tilde{I} \tilde{z}^2 (1 - \beta \tilde{I})]. \quad (8)$$

Again, after direct substitution of Eqs. (8) into Eqs. (2) for $\nu=2$, we can verify that this solution is exact at the beam axis.

The on-axial beam intensity distribution in (1+2) dimensions is given by

$$a\tilde{I}^2\tilde{z}^2 = \frac{2}{\beta} \ln\left(\frac{1-\beta}{1-\beta\tilde{I}}\right). \quad (9)$$

Analyzing the function $\tilde{z}(\tilde{I})$ in Eq. (9) one finds points where $\partial\tilde{z}(\tilde{I})=0$ corresponding to self-focusing. There is only one such point when $\beta=\beta_c\sim 0.175$. For $\beta>0.175$, Eq. (9) has no special points; the on-axial intensity monotonically increases approaching a saturation value \tilde{I}_{sat} . By studying the asymptotic behavior of $\tilde{I}=\tilde{I}(\tilde{z})$, we obtain $\tilde{I}_{\text{sat}}=1/\beta$. Note that this value fulfills the condition $1-\beta\tilde{I}_{\text{sat}}=\tilde{\varphi}(\tilde{I}_{\text{sat}})=0$. For $\beta<0.175$ there is an interval $[\tilde{z}_1, \tilde{z}_2]$ on the beam axis where the solution $\tilde{I}(\tilde{z})$ is not unique. The first point \tilde{z}_1 corresponds to the development of a short-range modulational instability in the beam. At this point, several filaments can appear, which further merge into a single filament with \tilde{I}_{sat} at the point \tilde{z}_2 .

For materials described by $n(I)=n_2I-n_6I^3$, we have $K=3$, $\tilde{\varphi}=1-\beta\tilde{I}^2$, $\beta=3n_6I_0^2/n_2$, and the on-axial intensity distribution is given by

$$2 \operatorname{arctanh}(\sqrt{\beta\tilde{I}}) - a\tilde{I}^2\tilde{z}^2\sqrt{\beta} = 2 \operatorname{arctanh}(\sqrt{\beta}). \quad (10)$$

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

Notice that the values of I_{sat} for $K=2$ and 3 obtained here are different from previous theoretical estimates [7,8,15], 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,

$$\partial n(I)|_{I_{\text{sat}}} = 0. \quad (11)$$

This condition to obtain the filament intensity is general, independent of the medium or material, and represents one of the central predictions of this paper, which should serve as a basis for future calculations. Note that such a general mathematical condition cannot be obtained from the 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 references therein). The nonlinear refractive index of air is taken in the following widely used form;

$$n = n_2\mathcal{R}(t) - n_4I^2 - \frac{\rho(I)}{2\rho_c}, \quad (12)$$

where the first term describes the Kerr response involving a delayed (Raman) contribution $\mathcal{R}(t)=(1-\theta)I + \theta(1+\omega_R^2\tau_d^2)\omega_R^{-1}\tau_d^{-2}\int_{-\infty}^t e^{-(t-t')/\tau_d} \sin[\omega_R(t-t')]I(t')dt'$, with $\theta\approx 0.5$, $\omega_R\approx 1.6\times 10^{13}$ s⁻¹ and $\tau_d\approx 77$ fs [23]. The magnitude

TABLE I. Comparison of the predictions for z_{sf} and the filament fluence in air calculated using Eq. (9) and Eq. (11) with the results of experiments and numerical simulations.

Pulse duration	z_{sf} (m)		Fluence (J/cm ²) 50 fs
	50 fs	450 fs	
Experiment [26]	3	5.5	0.6
Numerical [23]	3	6.5	0.6–1.4
This work	3	6.2	0.66

of n_4 for air is unknown, its most accepted estimates lie around $\sim 10^{-32}$ cm⁴/W² [7,24]. In the last term of Eq. (12), $\rho(I)$ refers to the density of free electrons and $\rho_c=1.7\times 10^{21}$ cm⁻³ denotes the critical density above which the plasma becomes opaque. A rough estimate yields $\rho(I)\sim \sigma_K I^K \rho_{\text{at}} t_p$, where ρ_{at} is the atom density $\rho_{\text{at}}=2\times 10^{19}$ cm⁻³ and t_p the pulse duration. $K=8$ for the MPI with a pulse of 800 nm, and $\sigma_8=3.7\times 10^{-96}$ (cm¹⁶/W⁸)/s [7,27].

A numerical solution of the NLSE for the focused beam with $f=2$ m, $P_{\text{in}}\sim 0.08$ TW, and $w_0=3$ mm using $n(I)$ given by Eq. (12) with $n_2=3.2\times 10^{-19}$ cm/W², $n_4=0$ and $\theta=0$ gives $z_{\text{sf},f}=128.2$ cm [25]. Note that if $n(I)=n_2I$, the self-focusing distance is given by the Kovalev formula [11,13] $z_{\text{sf}}=w_{\text{in}}/\sqrt{2n_2I_0}$, which is exact under the geometrical optics approximation and for initially Gaussian beam shape. For the experimental conditions of Ref. [25] it yields $z_{\text{sf},f}=127.6$ cm. This confirms our initial statement that for many experiments diffraction (and in this case also the plasma defocusing) can be neglected.

Now we compare our results with the recent experiment and numerical results of Refs. [23,26], where a collimated beam with full width at half-maximum (FWHM) diameter of $d\sim 4-5$ mm ($w_0=d/\sqrt{2\ln 2}$), with a pulse energy $E_{\text{in}}\sim 20$ mJ and different pulse durations (FWHM) was used. In Ref. [23], n_2 was taken to be $n_2=2.5\times 10^{-19}$ cm²/W for the pulse duration 50 fs, and $n_2=6\times 10^{-19}$ cm²/W for 450 fs, $n_4=2.5\times 10^{-33}$ cm⁴/W were fixed and independent on the pulse duration. Substituting this set of parameters into Eq. (9) and estimating the delayed response as an integral over the pulse duration, we obtain z_{sf} and the on-axial fluence $F[F=\int I(t)dt]$ which are presented in Table I.

From Table I, one can see that in spite of used simplifications, our results are in good agreement with experimental and numerical results.

It is important to point out that the values of all parameters in Eq. (12) are the subject of controversy. For example, the magnitude of n_2 is taken as $n_2=3.2\times 10^{-19}$ cm/W² in Refs. [7,27] and as $n_2=4\times 10^{-19}$ cm/W² in Ref. [8]. The value $\sigma_8=2.9\times 10^{-99}$ (cm¹⁶/W⁸)/s from Ref. [8] is three orders of magnitude larger than the one used in Ref. [7]. With such an uncertainty in these parameters and fluctuations of the experimental data (compare the results of Refs. [23,26] by the same group), a single experiment is probably not sufficient in order to adjust all the parameters of the refractive index equation (12).

Therefore, and based on Eq. (9), we suggest the following scaling measurement. In Fig. 2 we show our results for the

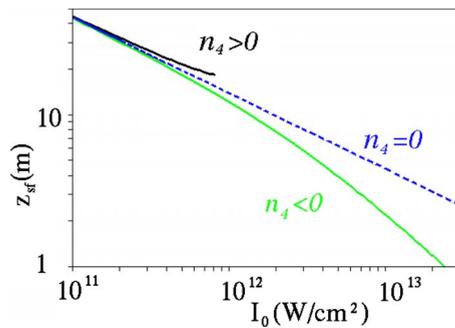


FIG. 2. (Color online) Dependence of the self-focusing position on the beam intensity. The blue dashed curve refers to the dependence $1/\sqrt{I_0}$ (obtained for low intensities), whereas the black and green curves show the deviation for high intensities due to influence of the fifth-order nonlinearity $n_4 = \pm 10^{-32} \text{ cm}^4/\text{W}^2$. The solid black curve ends at a point for which the sharp self-focusing is no longer observed, since the derivative I_z becomes positive.

dependence of z_{sf} on the initial pulse intensity. We obtain considerable qualitative (and quantitative) differences depending on the sign and magnitude of n_4 . One can use this result to design an experiment with varying beam power to find accurate values of n_2 and n_4 by fitting the measured experimental curve $z_{\text{sf}}(I_0)$ with z_{sf} obtained from Eq. (9). Afterwards, Eq. (11) derived from first principles can be used for determining the plasma response by fitting the value of I_{sat} .

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 [14]. If the nonlinear refractive index has the form $n(I) = n_2 I / (1 + \beta I)$, the on-axial intensity distribution in (1+1) dimensions is given by the expression

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

For $n(I) = n_2(1 - e^{-\beta I})$, the on-axial intensity is given by

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

and for a polynomial form $n(I) = n_{2K} I^K$ ($K \neq 2$) by

$$\tilde{I}^2 [I^{-K} + aK(K-2)z^2] = 1.$$

No intensity saturation has been observed. The singularities in the solutions of the above equations correspond to the z_{sf} for the given $n(I)$. Note that our approach can be generalized to arbitrary forms of $n(I)$. In general, the solutions can be found semianalytically by interpolation of the integrals in Eqs. (6).

Summarizing, exact solutions for the self-focusing length z_{sf} and the filament intensity I_{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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