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Stationary Self-Trapping of Optical Beams in Dense Media with Lorentz Local-Field Corrections*

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We describe a method for finding expressions for nonlinear electrical susceptibilities of dense media which are "exact" to all orders of the field strength, in the context of the Lorentz local-field theory. The method requires as a starting point the corresponding susceptibility to all orders for a dilute gas. We apply this method to determine the nonlinear susceptibilities responsible for self-trapping of optical beams in CS_2 and C_6H_6. A graphical method for finding approximate filament sizes from the susceptibility curve is described, and the self-trapping equation is solved numerically. Our results for the susceptibility and for the filament sizes differ, in the low-power region, from previous work. We find *smaller* filaments containing *less* power.

I. INTRODUCTION

SEVERAL recent theories of the intense filaments of light observed when a high-power laser beam focuses itself in a nonlinear medium have employed expressions for the refractive index which include all powers of the optical intensity.[1,2] Such expressions are derived from the equations of classical equilibrium statistical mechanics in the manner of the Debye theory of polar gases,[3] which leads to a susceptibility proportional to the Langevin function $\mathcal{L}(x) = \coth x - x^{-1}$, where x is a dimensionless field strength. Since the experiments are performed in liquids, one may expect the appropriate susceptibilities to differ markedly from those derived for dilute gases because of local-field effects. However, these effects have been taken into account only roughly in theories of self-trapping.

In this paper, we show that "exact" expressions for the nonlinear susceptibilities of liquids may be obtained easily in the context of the Lorentz local-field theory once the corresponding expressions are known for dilute gases. Section II is a brief review of the theory for gases, and Sec. III includes a discussion of the Lorentz local-field theory and the transcendental equations for the susceptibility to which it leads. The solution of the equation for prolate nonpolar molecules shows deviations from a similar approximate formula due to Gustafson *et al.*[2] in the important low-field region. In Sec. IV we present an approximate theory of stationary self-trapping, which shows qualitatively how the properties of the trapped beams depend upon the features of the nonlinear susceptibility curve. We find that for all powers there is only one stable trapped beam. Section V contains the results of numerical solutions of the self-trapping equation with the corrected saturable susceptibilities. Significant deviations from the results of Refs. 1 and 2 appear, as expected, only in the low-power regime. The corrected susceptibility leads to *smaller* filaments containing *less* power than given by previous work. A short discussion of the relevance of this work to actual experimental situations is given in Sec. VI.

II. NONLINEAR SUSCEPTIBILITIES FOR DILUTE GASES

For simplicity, we consider axially symmetric molecules. If these are polar, then the electric dipole vector $\boldsymbol{\mu}$ must point along the symmetry axis. In thermal equilibrium at temperature $T = (k_B\beta)^{-1}$, one such molecule possesses a mean dipole moment $\langle m \rangle$ along

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[1] J. D. Reichert and W. G. Wagner, IEEE J. Quant. Electron. **QE-4**, 221 (1968).

[2] T. K. Gustafson, P. L. Kelley, R. Y. Chiao, and R. G. Brewer, Appl. Phys. Letters **12**, 165 (1968).

[3] P. Debye, *Polar Molecules* (Dover Publications, Inc., New York, 1929).

an applied electric field E, where

$$\langle m\rangle = Z^{-1}\int_{-1}^{1}\left[\mu\cos\theta+(\alpha_\perp\sin^2\theta+\alpha_{\shortparallel}\cos^2\theta)E\right] \times e^{-\beta W} d\cos\theta. \quad (1)$$

Here $\alpha_{\shortparallel}$ and $\alpha_\perp$ are the principal axes of the molecular polarizability tensor along and perpendicular to the symmetry axis. Z is the partition function and W is that part of the molecular energy depending upon θ, the angle between $\boldsymbol{\mu}$ and $\mathbf{E}$:

$$W=-\mu E\cos\theta-\tfrac{1}{2}(\alpha_\perp\sin^2\theta+\alpha_{\shortparallel}\cos^2\theta)E^2. \quad (2)$$

If E is a high-frequency field, each term must be replaced by its average over many cycles. For optical fields the term in μ does not contribute, because the torque arising from this term changes direction before the molecule can respond. If E is an optical field, we assume it is linearly polarized.

The partition function may be evaluated in terms of known functions for this model[1,2,4]:

$$\begin{aligned}Z&=\int_{-1}^{1}e^{-\beta W}d\cos\theta\\&=(E_s/E)\exp\left[\tfrac{1}{2}\beta E^2\alpha_\perp\pm(E/E_s)^2\right]\{e^{E/E_d}G_\pm[(E/E_s)\\&\quad\pm(E_s/2E_d)]e^{-E/E_d}G_\pm[(E/E_s)\mp(E_s/2E_d)]\}. \quad (3)\end{aligned}$$

Here the "saturation fields" E_s and E_d are given by

$$E_s\equiv(\tfrac{1}{2}\beta|\alpha_{\shortparallel}-\alpha_\perp|)^{-1/2},$$
$$E_d\equiv(\beta\mu)^{-1},$$

and the subscripts $+$, $-$ refer to prolate ($\alpha_{\shortparallel}>\alpha_\perp$) and oblate ($\alpha_{\shortparallel}<\alpha_\perp$) molecules, respectively. For prolate (cigar-shaped) molecules we have

$$G_+(x)\equiv\int_0^x e^{y^2-x^2}dy$$

(which is tabulated as Dawson's integral in Ref. 5) and for oblate (disk-shaped) molecules,

$$\begin{aligned}G_-(x)&\equiv\int_0^x e^{x^2-y^2}dy\\&=(\tfrac{1}{2}\sqrt{\pi})e^{x^2}\operatorname{erf}(x).\end{aligned}$$

A closed expression for the mean dipole moment may be obtained from (3) via the relation

$$\langle m\rangle=\frac{1}{\beta}\frac{\partial\ln Z}{\partial E},$$

but we shall give the final result only for nonpolar molecules ($\mu=0$). The nonlinear local-field theory appropriate for polar liquids is prohibitively difficult, and this case will not be discussed further.

We take $CS_2(\alpha_{\shortparallel}/\alpha_\perp=2.73)$ as typical of prolate and $C_6H_6(\alpha_{\shortparallel}/\alpha_\perp=0.52)$ of oblate spheroidal molecules. For either case we may write

$$\langle m(E)\rangle=\alpha E+\tfrac{1}{2}|\alpha_{\shortparallel}-\alpha_\perp| \times\{\mp\tfrac{2}{3}-\mathcal{E}_0^{-2}+[\mathcal{E}_0G_\pm(\mathcal{E}_0)]^{-1}\}E, \quad (4)$$

where $\alpha=\frac{1}{3}(2\alpha_\perp+\alpha_{\shortparallel})$ and $\mathcal{E}_0=E/E_s$. The upper signs refer to prolate, the lower signs to oblate molecules.

III. LOCAL-FIELD CORRECTIONS

A good discussion of the Lorentz local-field theory may be found in Kittel's *Introduction to Solid State Physics*.[6] Here we simply state the features of this theory, which allow a painless solution of the nonlinear local-field problem of interest to us. First, the local field $\mathbf{E}_l$ (that experienced by a single molecule) is taken to be parallel to the macroscopic field $\mathbf{E}$ and unaffected by the orientation of the molecule in question. Thus the extra energy introduced into the problem by considering mutual interactions among molecules is independent of θ and may be neglected in evaluating the mean moment $\langle m\rangle$. Second, the expression for the Lorentz local field depends only upon the component of the dielectric tensor ϵ along the direction of the applied field. Thus, the induced anisotropy of the medium does not affect the local-field problem. Finally, the linearity of the formal relation between the local and the "applied" field simplifies the problem considerably:

$$E_l=\tfrac{1}{3}(\epsilon+2)E\equiv L(\epsilon)E. \quad (5)$$

None of these features is shared by more sophisticated theories, e.g., those based upon the work of Onsager for polar molecules.[7,8]

In Gaussian units (commonly used in tabulating α_i and μ), the relation between ϵ and $\langle m\rangle$ is (introducing the number density ρ)

$$\epsilon=1+4\pi\rho\langle m\rangle E^{-1}.$$

Replacing each field E in the expression $\langle m(E)\rangle$ for dilute gases (4), by the local field E_l of (5), we find the following transcendental equation for ϵ:

$$\epsilon(E)=1+4\pi\rho\langle m(LE)\rangle E^{-1}.$$

This may be transformed, using (5), to

$$E=s-\tfrac{4}{3}\pi\rho\langle m(s)\rangle, \quad (6)$$

which is an explicit expression for E as a function of

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[4] W. G. Wagner, H. A. Haus, and J. H. Marburger, Phys. Rev. **175**, 256 (1968).

[5] *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (U. S. Government Printing Office, Washington, D. C., 1964).

[6] C. Kittel, *Introduction to Solid State Physics* (Wiley-Interscience, Inc., New York, 1966), 3rd ed.

[7] C. J. F. Bottcher, *Theory of Electric Polarization* (Elsevier Publishing Company, Inc., New York, 1952).

[8] J. Frenkel, *Kinetic Theory of Liquids* (Dover Publications, Inc., New York, 1955).

the parameter $s=LE$. For a given value of s, ϵ is

$$\epsilon=3sE^{-1}-2$$
$$=\frac{3s+8\pi\rho\langle m(s)\rangle}{3s-4\pi\rho\langle m(s)\rangle}. \qquad (7)$$

Using (6), (7), and the function $\langle m(s)\rangle$ from (4), one may determine $\epsilon(E)$ exactly. In particular, we have for the nonlinear part, $\delta\epsilon\equiv\epsilon-\epsilon_0$, of the dielectric constant

$$\delta\epsilon=\frac{2\pi\rho\Delta L_0{}^2\Xi_\pm(t)}{1-(4\pi\rho\Delta L_0/6)\Xi_\pm(t)}, \qquad (8)$$

$$L_0\mathcal{E}_0=t[1-(4\pi\rho\Delta L_0/6)\Xi_\pm(t)]. \qquad (9)$$

Here, $t=L\mathcal{E}_0$ and

$$L_0=\tfrac{1}{3}(\epsilon_0+2)=[1-(\tfrac{4}{3}\pi\rho\alpha)]^{-1},$$
$$\Xi_\pm(t)=\mp\tfrac{2}{3}-t^{-2}+[tG_\pm(t)]^{-1}, \qquad (10)$$
$$\Delta=|\alpha_{\parallel}-\alpha_{\perp}|,$$
$$t=L\mathcal{E}_0.$$

In Fig. 1, we have plotted the dimensionless susceptibilities

$$X_\pm\equiv\delta\epsilon/2\pi\rho\Delta L_0{}^2 \qquad (11a)$$

versus $\mathcal{E}^2\equiv(L_0\mathcal{E}_0)^2$. The initial slope of all curves shown is 8/45, which in our units is the value of the nonsaturable index employed in earlier self-trapping computations.[9,10] Also shown is $X_+{}^G$, the approximate form employed by Gustafson *et al.*[2] (who did not discuss oblate molecules):

$$X_+{}^G=\frac{\Xi_+(\mathcal{E})}{1-(4\pi\rho\Delta L_0/6)\Xi_+(\mathcal{E})}. \qquad (11b)$$

This may be obtained from (1) by ignoring the field dependence of the local-field factors in the Boltzmann probability $Z^{-1}e^{\beta W}$. On the same graph we show the approximate forms of Reichert and Wagner[1]

$$X_\pm{}^{RW}=\Xi_\pm(\mathcal{E}), \qquad (11c)$$

in which Δ in the expression for dilute gases is replaced everywhere by $L_0{}^2\Delta$. The physical implications of the differences among these susceptibilities are discussed in Sec. IV.

IV. PROPERTIES OF SELF-TRAPPED OPTICAL BEAMS

A rough idea of the relation of the properties of self-trapped optical beams to the susceptibilities shown in Fig. 1 may be obtained from the "paraxial-ray" theory of self-focusing as developed by Wagner *et al.*[4] In this theory, the transverse optical intensity distribution near the axis is assumed to remain Gaussian with distance dependent variance $a(z)$. This quantity obeys an equation which is formally equivalent to Newton's second law:

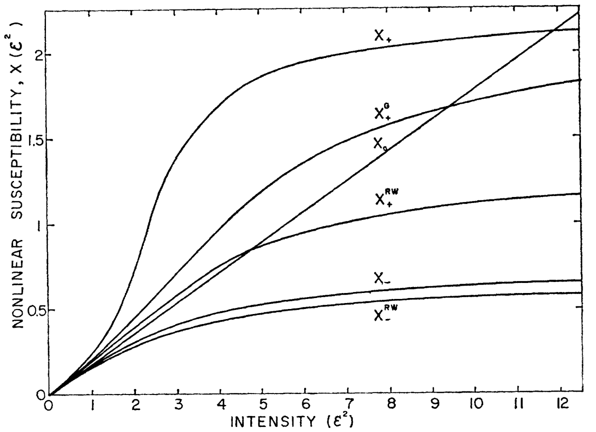

FIG. 1. Dimensionless susceptibilities defined in Eq. (11) versus dimensionless intensity. Subscript + refers to CS_2, − to C_6H_6. $H_\pm{}^{RW}$ are the susceptibilities employed in Ref. 1, and $X_+{}^G$ used in Ref. 2. $X_\pm$ are "exact" in the Lorentz local-field theory.

$$\frac{d^2a}{dz^2}=-\frac{\partial U}{\partial a}, \qquad (12)$$

where z is the axial distance and U is related to $\delta\epsilon(E)$ via

$$U=(2k^2a^2)^{-1}-\delta\epsilon(q/a)/2\epsilon_0$$

with

$$q^2=4P/c\epsilon_0{}^{1/2}.$$

P is the total power in the beam, k the wave number in the medium, and c the speed of light.

The values $a=a_c$ for which dU/da vanishes are the radii of beams trapped in stationary "modes." Because of the constant shape and paraxial-ray assumptions, modes with several off-axis intensity maxima[10] are not given by this theory.

From a graph of $\delta\epsilon$ versus E^2, one may find the stationary diameters by a simple construction. Rewriting the potential in terms of the dimensionless susceptibilities $X(\mathcal{E}^2)$ plotted in Fig. 1, we find by straightforward differentiation that the slope of $U(a)$ vanishes whenever

$$\frac{\partial X}{\partial\mathcal{E}^2}=\frac{8}{45}\frac{P_1}{P}. \qquad (13)$$

Here $P_1=45c\epsilon_0{}^{3/2}/32\pi k^2\rho\beta\Delta^2L_0{}^4$ is a "critical power" for self-focusing which is less than P_{CGT}, the value given by the theory of Chiao *et al.*,[9] by the factor 0.273. (We are using cgs units and mean fields.) Numerical solu-

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[9] R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Letters **13**, 479 (1964).

[10] H. A. Haus, Appl. Phys. Letters **8**, 128 (1966); Z. K. Yankauskas, Izv. VUZ Radio Fizika **9**, 412 (1966) [English transl.: Soviet Radiophysics **9**, 261 (1966)]; Zh. Eksperim. i Teor. Fiz., v Redaktsiya Pisma **5**, 335 (1967) [English transl.: Soviet Phys.—JETP Letters **5**, 275 (1967)].

tion of the self-focusing equation derived by Kelley[11] shows that for an equiphase Gaussian input beam with $P_1 \leq P < P_{\mathrm{CGT}}$, the on-axis intensity rises initially but decreases again before the beam comes to a focus, while for $P > P_{\mathrm{CGT}}$ the beam always focuses itself.[12]

To find the stationary diameter (in this case the variance) for a given power, it is only necessary to find the value of $\mathcal{E}_c$ for which the slope of $X(\mathcal{E}_c^2)$ in Fig. 1 is $8P_1/45P$. The variance is then given (in cm) by

$$a_c = (c\epsilon_0^{1/2}/2L_0^2\beta\Delta P)^{1/2}\mathcal{E}_c. \tag{14}$$

Applied to Fig. 1 this construction shows that for prolate molecules there are two equilibrium diameters for powers not too much less than the critical power, a possibility first recognized by Chiao *et al.*[13] But the larger radius is unstable because, as a short computation shows, the curvature of $U(a)$ at an extremum is opposite that of $X(\mathcal{E}^2)$. This is illustrated in Fig. 2 where potential curves are plotted for powers greater than and less than P_1. Notice that for trapping at powers less than P_1 the initial diameter must already be quite small, or rapidly decreasing (corresponding to a converging beam).

For oblate molecules such as benzene and, approximately, nitrobenzene, there are no stable solutions for beam powers $P < P_1$. Moreover, the nonlinear susceptibility for such liquids begins saturating even at small field strengths and therefore decreases to a given slope more rapidly than the corresponding function for prolate molecules. According to our construction this means that, other parameters being equal, the radii of trapped beams in liquids of oblate molecules will be larger than those in liquids of prolate type.

Since the nitrobenzene molecule is not axially symmetric, we must justify its inclusion in the class of oblate spheroidal molecules for the purposes of this discussion. The dilute-gas theory for molecules of arbitrary symmetry[14] yields for $\delta\epsilon$ (through E^4)

$$\delta\epsilon = 2\pi\rho\beta\alpha^2\Delta' E^2 + \pi\rho\beta^2\alpha^3\Gamma E^4, \tag{15}$$

where

$$\Delta' \equiv (2/45\alpha^2)[(\alpha_1-\alpha_2)^2+\cdots],$$

$$\Gamma \equiv (8/945\alpha^3)[2(\alpha_1^3+\cdots)-3(\alpha_1^2\alpha_2+\cdots)+12\alpha_1\alpha_2\alpha_3].$$

The dots indicate cyclic permutations of the first term and $3\alpha = \alpha_1+\alpha_2+\alpha_3$, α_i being the ith principal value of the polarizability tensor. The sign of Γ distinguishes between "thin" ($\Gamma > 0$) and "fat" ($\Gamma < 0$) moleucles. For the molecules CS_2, C_6H_6, and $C_6H_5NO_2$, Γ is 1.33, −0.193, and −0.0593, respectively.[15] The ratio of the coefficient of E^4 in (15) for nitrobenzene to that for benzene is 0.525 at 300°K. Thus, the nonlinear refractive index for nitrobenzene does not saturate as rapidly as that for benzene, but neither does it show the "negative saturation" effect characteristic of prolate molecules such as CS_2. Unfortunately, it is difficult to obtain an expression to all orders for the susceptibility of nonaxially symmetric molecules and, therefore, we do not give reliable estimates of filament sizes for nitrobenzene in the context of our theory.

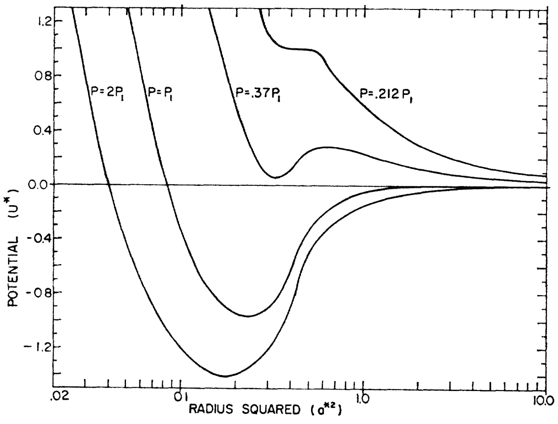

FIG. 2. Dimensionless potentials which determine the radius of a self-focusing beam in the paraxial-ray theory. All are derived from X_+ shown in Fig. 1. Notice that the larger of the two equilibrium radii for $P = 0.37P_1$ is unstable.

V. NUMERICAL SOLUTIONS

The graphical method described above cannot be relied upon to predict the finer features of the trapped intensity distribution. As shown by Reichert and Wagner (Figs. 1 and 2 of Ref. 1), the stationary intensity distributions are not at all Gaussian and possess shapes sensitively dependent upon the trapped power in violation of the assumptions which lead to (12). For this reason, it is necessary to solve the stationary self-trapping equation numerically to determine the precise influence of the susceptibility upon the trapped intensity distribution.

The equation we wish to solve is derived and discussed in some detail in Refs. 1, 2, and 9. It is simply the scalar wave equation for a cylindrically symmetric mean electric field of amplitude $E(r)$, where the total peak field is $2E(r)\cos(k_z z - \omega t)$. In our dimensionless variables, this equation is

$$\frac{\partial^2\mathcal{E}}{\partial R^2}+\frac{1}{R}\frac{\partial\mathcal{E}}{\partial R}-[\nu - X_\pm(\mathcal{E})]=0, \tag{16}$$

where

$$R^2 \equiv 2\pi L_0^2\rho\Delta\epsilon_0^{-1}k^2r^2,$$

$$\nu = \frac{\epsilon_0}{2\pi L_0^2\rho\Delta}\frac{k_z^2-k^2}{k^2},$$

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[11] P. L. Kelley, Phys. Rev. Letters **15**, 1005 (1965).

[12] E. Dawes and J. Marburger, Phys. Rev. **179**, 862 (1969).

[13] R. Chiao, J. Dodson, D. Irwin, and J. Gustafson, Bull. Am. Phys. Soc. **12**, 686 (1967). Also see Refs. 1 and 2.

[14] J. Marburger (to be published).

[15] We use values for α_i given by Landolt and Bornstein, in *Zahlenwerte und Funktionen*, 6th ed., Vol. 1, part 3, p. 510.

and, as defined in Sec. III,

$$\mathcal{E}^2 = \tfrac{1}{2}\beta L_0^2 \Delta E^2.$$

The parameter ν of this nonlinear equation plays the role of $\Gamma^2 = k_z^2 - k^2$ in the work of Ref. 2, and fixes the power within the beam. For a more detailed discussion, see Ref. 1.

We found numerical solutions of (16) with a single maximum at $r = 0$ for various values of ν. Other solutions with off-axis maxima exist but were not investigated. The results which are of most interest are summarized in Fig. 3. Here the diameter of the trapped field profile at its half-intensity point is plotted in microns as a function of the total trapped power in kilowatts.

The most striking feature of this figure is the disparity between the minimum diameters in benzene and carbon disulfide, the latter possessing filaments roughly a third the size of the former. In most respects, the predictions of all three theories with saturation differ little, the only significant new feature being the much lower power at which stable trapped filaments may form in CS_2.

Curve B in Fig. 3 is plotted from the numerical solutions reported by Gustafson *et al.*,[2] normalized to agree with the other curves for CS_2 in the low-power regime. The actual numerical values of our curves differ slightly from those obtained in Ref. 2 because we used slightly different values of the physical constants employed in converting to dimensional units.

Comparison of Eqs. (8) and (11) shows that the saturated values of the susceptibilities X_+ and X_+^G are the same. It is therefore of interest that in the range of powers shown in Fig. 3 the trapped beam diameters for the two susceptibilities differ by a nearly constant amount. This difference must be attributed to the more rapid saturation of X_+ in this power regime.

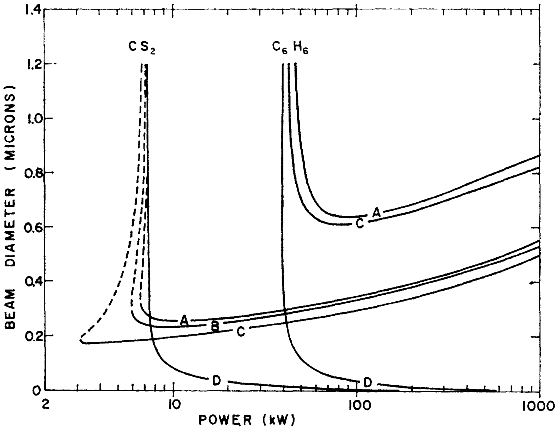

FIG. 3. Diameters of trapped beams versus power for carbon disulfide and benzene. All are from numerical solutions of the stationary self-trapping equation: A from Ref. 1 using $X_\pm^{RW}$ of Fig. 1, B from Ref. 2 using X_+^G (B is normalized to agree with our choice of physical constants), C from this work using $X_\pm$, and D (also normalized) from the theory of Ref. 9 and Fig. 1 of Ref. 1 using X_0.

VI. DISCUSSION

Our aim has been to present a theory of the saturation of the nonlinear refractive index which is complete in the context of the Lorentz local-field theory, and to study the influence of local-field corrections on the properties of self-trapped light filaments. We do not pretend that the theory presented here is capable of predicting the sizes of small-scale trapped light filaments actually observed experimentally (for example in Ref. 16). Moreover, setting aside for a moment the difficult problem of constructing a realistic theory of self-trapping, we doubt that the Lorentz local-field theory leads to the correct form of the susceptibility even for static electric fields. It is well known that other more sophisticated theories[7,8,17] lead to deviations from the results of the Lorentz theory.

Therefore, the primary value of this work is the demonstration in a well-investigated context that the nonlinear susceptibility is exceptionally sensitive to local-field corrections and that the accuracy of future quantitative theories of self-trapping must remain questionable until the local-field problem is resolved.

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[16] R. G. Brewer, J. R. Lifsitz, E. Garmire, R. Y. Chiao, and C. H. Townes, Phys. Rev. **166**, 326 (1968).

[17] R. W. Hellwarth, Phys. Rev. **152**, 156 (1966).