Spatial Pattern Formation in Fused Silica Under UV Irradiation

Problem Presenter

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Preface

At the 30th Annual Workshop on Mathematical Problems in Industry (MPI), Leslie Button of Corning presented a problem concerning the formation of spatial patterns or microchannels in fused silica fibers exposed to ultraviolet radiation.

This manuscript is really a collection of reports from teams in the group working on several aspects of the problem. Here is a brief summary of each:

1. Edwards *et al.* outline the general problem, scale the relevant variables, and present a linear stability analysis for transverse perturbations from the plane wave in various cases.
2. McCollom and Witelski generalize the wave equation to the case of time-varying index of refraction.
3. Gambino presents some numerical simulations of the problem.
4. Zyskin performs some perturbation analysis of the Gaussian beam solution.

In addition to the authors of these reports, the following people participated in the group discussions:

- Yuxin Chen, Northwestern University
- John Cummings, University of Tennessee
- Roy Goodman, New Jersey Institute of Technology
- Michael Mazzoleni, Duke University
- Colin Please, Oxford University
- Marisabel Rodriguez, Arizona State University
- Tural Sadigov, Indiana University
- Donald Schwendeman, Rensselær Polytechnic Institute
- Cheng Yuan, University of Buffalo
- Jielin Zhu, University of British Columbia

Special recognition is due to John Cummings, James Gambino, Brittany McCollom, Jimmy Moore, and Tural Sadigov for making the group's oral presentations throughout the week. We also wish to acknowledge Tom Witelski for writing the group's executive summary.

Governing Equations; Linear Stability Analysis

David A. Edwards, University of Delaware
Richard O. Moore, New Jersey Institute of Technology
Thomas P. Witelski, Duke University

Section 1: Introduction

This problem was presented on June 23, 2014 at the 30th annual Mathematical Problems in Industry workshop held at the New Jersey Institute of Technology. Les Button, the industry representative from Corning Corporation, presented the following problem in which the transmission of ultraviolet (UV) radiation through fused silica lenses gradually degrades and ultimately damages these optical components. Corning is a global supplier of optical and ceramic materials across various industries and is particularly interested in this damage mechanism as it affects a number of its customers. A greater understanding of laser/material interactions of UV photons within silica lenses could mitigate or eliminate the damage mechanism.

UV damage is especially problematic in the fabrication of microchips and integrated circuits. Here, a process known as *photolithography* is used to etch wafers coated with photosensitive chemicals. A series of chemical treatments then either engraves the exposure pattern into the wafer, or enables deposition of a new material in the desired pattern. Repeating this process several times (tens to hundreds of cycles) allows for the creation of highly complex integrated circuits. Fused silica lenses are used to steer and focus laser light used in this process.

As transistor densities have increased, so has the need for finer etching resolution. This has pushed the industry to use smaller-wavelength light, which comes with an increase in photon energy. At UV wavelengths (100–300 nm), photon energies begin to be high enough to interact with silica molecules in the lens. Excimer lasers, which operate solely within the UV range, are used extensively in the semiconductor industry and numerous papers report measurable and permanent changes in lens characteristics across the illuminated area. These effects (local changes in density, physical shrinkage of lens material) develop after millions of pulses [1], which (given normal duty of 1000 pulses/sec), correspond to time scales as short as a few hours.

Due to photolithography's increasingly stringent resolution requirements, any degradation in beam quality is highly undesirable. Local changes in the lens density ρ even on the order of parts-per-million (such as those imparted by UV-silica interactions) are significant enough to measurably affect optical characteristics. In particular, they generate interference through refractive gradients and nanometer changes in path length from physical shrinkage of the lens (see Fig. 1.1).

The proposed mechanism of these changes is through two-photon absorption. On their own, UV photons lack the necessary energy to interact meaningfully with silica molecules. However, if two photons collide with an atom at once, the simultaneous energy transfer is enough to change the orientation of the silica molecules into a tighter packing, a locally more dense arrangement. This is referred to as "densification" or "compaction" in the literature [2, 3]. Here, incoming light collides with silica molecules and progressively changes the density of the lens in various places throughout the illuminated region. These local changes in density create material stresses within the lens where the compressive forces of the densified regions generate tension with the unaffected material around it (see right

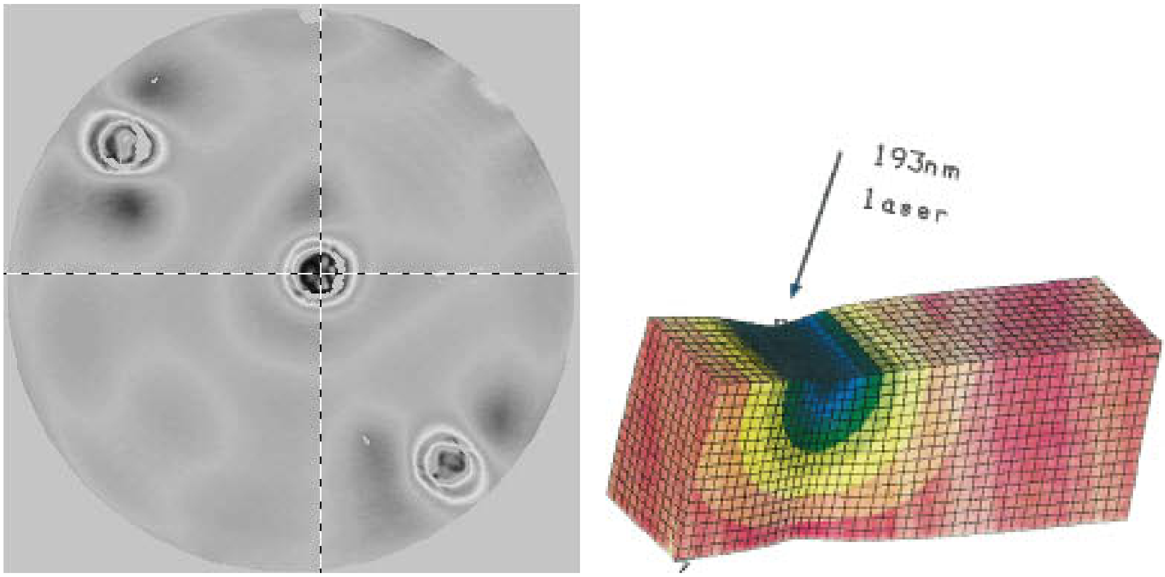

Figure 1.1. Lens damage due to UV irradiation. Left: Interferogram showing stress birefringence [1]. Right: Contours of isostrain on a finite element grid [4].

of Fig. 1.1). Densification is an accumulative process which continues as the lens is used; thus lens stress continues to grow over time.

Experimental observations show that a few months to a year after a lens has begun densifying, small cylindrical voids (called microchannels) begin to form at the exit face of the lens, and grow towards the front [4] (see Fig. 1.2).

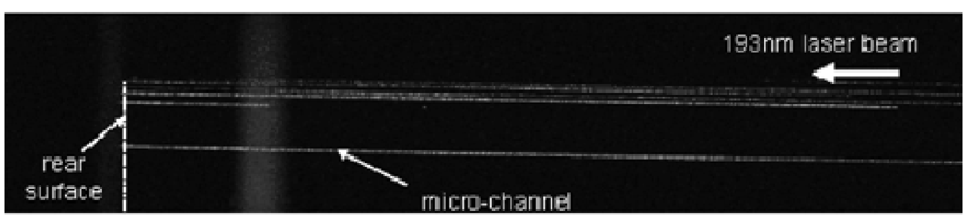

Figure 1.2. Formation of microchannels at the exit face [4].

At this time, the mechanism for microchannel formation is unknown, along with any causal relationship to the earlier densification process. The report details our efforts to model the development and evolution of these dynamics and aid in the understanding of these complex phenomena.

Section 2: Governing Equations

Fused silica glass is used in the optical train of lithography equipment for microelectronics. Pulses of electromagnetic radiation are sent through the glass. In order to increase the image resolution, pulses with high-energy photons are used, which correspond to shorter ultraviolet wavelengths ($\lambda \approx 193$ nm). In practice, sources are pulsed for just a short time ($T_{\mathrm{p}} \approx 2$ ns—here the "p" subscript denotes "pulse"), with a somewhat longer time between pulses ($T_{\mathrm{r}} \approx 20$–$200$ ns—here the "r" subscript denotes "rest").

Very long-term exposure to this ultraviolet energy (on the order of hours–years), even at moderate intensities, causes small but measurable changes to the glass. In particular, the glass permanently compacts (densifies), increasing its refractive index n. Experiments suggest that two-photon processes are important in the compaction process; hence we assume that the relative compaction scales with the total "two-photon dose" $\tilde{D}$: [3, 4, 5]:

$$\frac{\delta\rho}{\rho} = \kappa \tilde{D}^b, \qquad \tilde{D} = \frac{I_{\mathrm{e}}^2 N}{T_{\mathrm{p}}} \tag{2.1}$$

where ρ is the local density, I_{e} is the (constant) *energy* intensity of the pulse (hence the subscript "e"), N is the number of pulses, b is an exponent experimentally determined to be in the range $0.5 \leq b \leq 0.7$ [4], and κ is a constant of proportionality.

Under certain conditions, an even more dramatic effect called "microchanneling" can occur. In microchanneling, the irradiated glass contracts, leaving small voids or microchannels in the fused silica. These channels begin at the end away from the incident wave. At very long exposures, tiny cylindrical channels (on the order of microns) develop. They are parallel to the beam, much smaller in diameter than the beam, and typically start at the exit side of the sample and grow towards the beam [4].

We hypothesize that there is a feedback mechanism connecting the connection and microchanneling phenomena. In particular, suppose that a steady transverse non-uniformity in the beam creates transverse and axial intensity gradients within the medium. Over a long time scale, these gradients can cause compaction, which changes the refractive index. But these in turn will cause more nonuniformity in the beam. Could this feedback loop cause self-focusing, intensity enhancement, and ultimately damage?

To answer this question, we begin by writing down the standard wave equation for the energy field $\tilde{E}$ in the case of an index of refraction that can vary with time:

$$\frac{1}{c^2}\frac{\partial^2(n^2\tilde{E})}{\partial\tilde{t}^2} = \frac{\partial^2\tilde{E}}{\partial x^2} + \frac{\partial^2\tilde{E}}{\partial y^2} + \frac{\partial^2\tilde{E}}{\partial\tilde{z}^2}, \tag{2.2}$$

where c is the speed of light. (For more details, see the chapter by McCollom and Witelski.) We assume that the glass occupies the half-space $\tilde{z} > 0$, and that incident upon it is a simple plane wave:

$$\tilde{E}(x,y,\tilde{z},\tilde{t}) = R_0 A(x,y,z,t) e^{i(\omega\tilde{t} - n_0 k\tilde{z})}; \qquad R_0 \in \mathcal{R}, \quad z = \epsilon_z n_0 k\tilde{z}, \quad t = \epsilon_t \omega\tilde{t}. \tag{2.3}$$

where ω is the frequency of the light wave, n_0 is the index of refraction in vacuum, k is the wavenumber of the light wave, and ϵ is a small parameter. Here R_0 is a scaling factor to be determined later. Note from the choice of variables that A is a slowly-varying amplitude (envelope) function. The changes to the refractive index are caused by the variance in the amplitude; hence we have that n depends on the slow time scale t, not the fast time scale $\tilde{t}$.

Substituting (2.3) into (2.2), we have

$$\frac{1}{c^2}\frac{\partial}{\partial \tilde{t}}\left\{\left[\epsilon_t\omega\left(n^2\frac{\partial A}{\partial t}+A\frac{\partial(n^2)}{\partial t}\right)+i\omega A n^2\right]e^{i(\omega\tilde{t}-n_0k\tilde{z})}\right\}=\frac{\partial^2 A}{\partial x^2}e^{i(\omega\tilde{t}-n_0k\tilde{z})} + \frac{\partial^2 A}{\partial y^2}e^{i(\omega\tilde{t}-n_0k\tilde{z})}+\frac{\partial}{\partial\tilde{z}}\left[\left(\epsilon_z n_0 k\frac{\partial A}{\partial z}-in_0kA\right)e^{i(\omega\tilde{t}-n_0k\tilde{z})}\right],$$

which can be rearranged to obtain

$$\frac{\omega^2}{c^2}\left\{\epsilon_t^2\left[n^2\frac{\partial^2 A}{\partial t^2}+2\frac{\partial A}{\partial t}\frac{\partial(n^2)}{\partial t}+A\frac{\partial^2(n^2)}{\partial t^2}\right]+2i\epsilon_t\left[n^2\frac{\partial A}{\partial t}+A\frac{\partial(n^2)}{\partial t}\right]-An^2\right\} = \frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}+n_0^2k^2\left(\epsilon_z^2\frac{\partial^2 A}{\partial z^2}-2i\epsilon_z\frac{\partial A}{\partial z}-A\right). \quad (2.4)$$

Note here that k as defined relates to the variation of the incoming plane wave in vacuum, so we have $k = \omega/c$. Also, note that while the x- and y-length scales have not been chosen, the choice of time and z-scales means that we may neglect the second derivative terms in (2.4), yielding

$$k^2n^2A\left(2i\epsilon_t\frac{\partial A}{\partial t}-A\right)+2ik^2\epsilon_tA\frac{\partial(n^2)}{\partial t}=\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}-n_0^2k^2\left(2i\epsilon_z\frac{\partial A}{\partial z}+A\right). \quad (2.5)$$

We expect that changes in n due to the light wave will be small; therefore we write

$$n = n_0 + \delta n, \quad (2.6)$$

where we consider δn to be small. Hence the last term on the left-hand side of (2.5) may also be neglected, and we have (showing various steps of the simplification by clarity)

$$\begin{aligned}
k^2n^2A\left(2i\epsilon_t\frac{\partial A}{\partial t}-A\right) &= \frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}-n_0^2k^2\left(2i\epsilon_z\frac{\partial A}{\partial z}+A\right)\\
2ik^2\left(n_0^2\epsilon_z\frac{\partial A}{\partial z}+n^2\epsilon_t\frac{\partial A}{\partial t}\right) &= \frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}+k^2(n^2-n_0^2)A\\
2ik^2\left[n_0^2\epsilon_z\frac{\partial A}{\partial z}+(n_0+\delta n)^2\epsilon_t\frac{\partial A}{\partial t}\right] &= \frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}+k^2[(n_0+\delta n)^2-n_0^2]A\\
2ik^2n_0^2\left(\epsilon_z\frac{\partial A}{\partial z}+\epsilon_t\frac{\partial A}{\partial t}\right) &= \frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}+k^2[2n_0\delta n+(\delta n)^2]A. \quad (2.7)
\end{aligned}$$

We now pose the problem (2.7) in the region $z \geq 0$ with boundary condition

$$A(x, y, 0, t) = 1. \tag{2.8}$$

As an energy density for a single pulse, I_e is the product of the regular intensity for that pulse I_p and the width of the pulse. Therefore, substituting this result into (2.1), we have

$$I_\mathrm{e} = I_\mathrm{p} T_\mathrm{p} \qquad \Longrightarrow \qquad \tilde{D} = I_\mathrm{p}^2 N T_\mathrm{p}. \tag{2.9}$$

We wish to consider the case of variable intensity pulses. In this case we would make the following generalization:

$$I_\mathrm{p}^2 N T_\mathrm{p} = \sum_{j=1}^{N} \tilde{I}^2(\tilde{t}_j)(\Delta\tilde{t})_j. \tag{2.10}$$

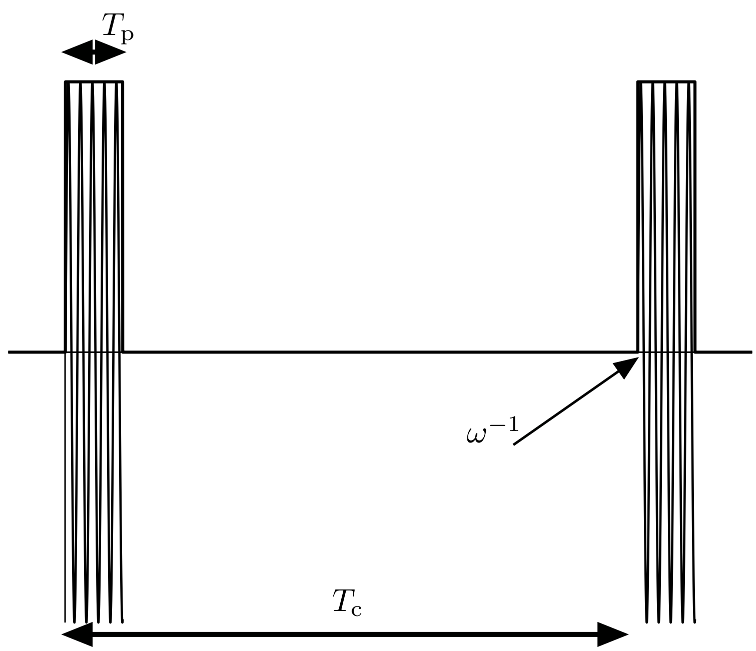

Figure 2.1. Schematic of various time scales. T_n as defined in (2.21) is several orders of magnitude larger than T_c.

Now to make the intensity variable, we just take the limit as $N \to \infty$ and $(\Delta\tilde{t})_j \to 0$, in which case (2.10) is just a Riemann sum. Hence we have

$$\tilde{D} = \int_0^{\tilde{t}} \tilde{I}^2(\tilde{t}')\, d\tilde{t}'. \tag{2.11}$$

However, we must introduce a normalization. Assume that in the finite formulation, we have only one pulse of intensity I_p with duration T_p and time between pulses T_c (see Fig. 2.1). We want to introduce a scaling

$$I = \frac{\tilde{I}}{I_0} \tag{2.12}$$

such that the photon dose is equivalent at $\tilde{t} = T_\mathrm{c}$ when the average of I^2 is 1. In other words,

$$I_0^2 \int_0^{T_\mathrm{c}} \tilde{I}^2 \, d\tilde{t}' = I_0^2 T_\mathrm{c} = I_\mathrm{p}^2 T_\mathrm{p}$$
$$I_0^2 = \frac{I_\mathrm{p}^2 T_\mathrm{p}}{T_\mathrm{c}} \tag{2.13}$$

The relationship between intensity and the electric field is given by [6]:

$$\tilde{I} = \frac{cn\varepsilon_0}{2}|\tilde{E}|^2, \tag{2.14a}$$

which motivates the following relationship between I_0 and R_0:

$$I_0 = \frac{cn\varepsilon_0}{2}R_0^2 \qquad \Longrightarrow \qquad \tilde{I}(\tilde{t}) = I_0^2|A(\tilde{t})|^4, \tag{2.14b}$$

where we have used (2.3). Substituting (2.14b) into (2.11), we have

$$\tilde{D} = I_0^2 \int_0^{\tilde{t}} |A(\tilde{t}')|^4 \, d\tilde{t}' = \frac{I_\mathrm{p}^2 T_\mathrm{p}}{T_\mathrm{c}} \int_0^{\tilde{t}} |A(\tilde{t}')|^4 \, d\tilde{t}', \tag{2.15}$$

where we have used (2.13). In the literature, only I_e is given; hence using (2.9) in (2.15), we obtain the following:

$$\tilde{D} = \frac{I_\mathrm{e}^2}{T_\mathrm{p} T_\mathrm{c}} \int_0^{\tilde{t}} |A(\tilde{t}')|^4 \, d\tilde{t}'. \tag{2.16}$$

We keep the time scale arbitrary for now by letting

$$t_n = \frac{\tilde{t}}{T_n}. \tag{2.17}$$

Substituting (2.17) into (2.16), we obtain

$$\tilde{D} = \frac{I_\mathrm{e}^2 T_n}{T_\mathrm{p} T_\mathrm{c}} D, \qquad D = \int_0^{t_n} |A(t')|^4 \, dt'. \tag{2.18}$$

Next we consider the constitutive equation for δn. From [7], we have that

$$\frac{\delta n}{n_0} = \alpha \frac{\delta \rho}{\rho} = \alpha\kappa\tilde{D}^b = \alpha\kappa \left(\frac{I_\mathrm{e}^2 T_n}{T_\mathrm{p} T_\mathrm{c}}\right)^b D^b, \tag{2.19}$$

where α is a constant and we have used (2.18). We want the δn term in (2.7) to balance with the terms on the left-hand side, hence we want

$$\delta n = n_0 \epsilon_z D^b, \tag{2.20}$$

which implies that

$$\epsilon_z = \alpha\kappa \left(\frac{I_\mathrm{e}^2 T_n}{T_\mathrm{p} T_\mathrm{c}} \right)^b \qquad \Longrightarrow \qquad T_n = \frac{T_\mathrm{p} T_\mathrm{c}}{I_\mathrm{e}^2} \left(\frac{\epsilon_z}{\alpha\kappa} \right)^{1/b}. \tag{2.21}$$

In other words, T_n should be the time scale on which the δn terms become important and refractive index changes occur.

Substituting (2.20) into (2.7), we obtain the full evolution equation

$$2ik^2 n_0^2 \left(\epsilon_z \frac{\partial A}{\partial z} + \epsilon_t \frac{\partial A}{\partial t} \right) = \frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} + k^2 n_0^2 [2\epsilon_z D^b + \epsilon_z^2 D^{2b}] A,$$

which can be simplified in the case that $\epsilon_z \ll 1$ to

$$2ik^2 n_0^2 \left(\epsilon_z \frac{\partial A}{\partial z} + \epsilon_t \frac{\partial A}{\partial t} \right) = \frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} + 2k^2 n_0^2 \epsilon_z D^b A. \tag{2.22}$$

Lastly, for future purposes it is convenient to introduce the following lemma:

Lemma. Let $|A_0| = 1$. Then

$$|A_0(1 + \epsilon A_1)|^{2\gamma} \sim 1 + \gamma\epsilon(A_1 + \overline{A_1}), \qquad 0 < \epsilon \ll 1. \tag{2.23}$$

Proof.

$$\begin{aligned} |A_0(1 + \epsilon A_1)|^{2\gamma} &= |A_0|^{2\gamma} (1 + \epsilon A_1)^\gamma \overline{(1 + \epsilon A_1)}^\gamma \sim (1 + \epsilon\gamma A_1)(1 + \epsilon\gamma \overline{A_1}) \\ &\sim 1 + \epsilon\gamma(A_1 + \overline{A_1}), \end{aligned}$$

as required. ✔

Section 3: Linearization of Quasisteady Case, No Explicit Time

As a first approximation, we assume that $\epsilon_t \ll \epsilon_z$, so the first-order time derivative in (2.22) may be neglected:

$$2in_0^2k^2\epsilon_z\frac{\partial A}{\partial z} = \frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} + 2k^2n_0^2\epsilon_z D^b A. \tag{3.1}$$

We now wish to perform a linear stability analysis to see if transverse perturbations from the plane wave will grow, thus perhaps causing the type of behavior for which we are searching. At leading order, the solution will not depend on x or y, since there are no transverse perturbations. Hence we assume a solution of the form

$$A(x, y, z, t) = A_0(z, t)[1 + \epsilon A_1(x, y, z, t)]. \tag{3.2}$$

Here we assume that though $\epsilon \ll 1$, it is large enough that we need not consider the other terms we have neglected in §2. Substituting (3.2) into (3.1) yields, to leading orders,

$$\begin{aligned} 2in_0^2k^2\epsilon_z\frac{\partial}{\partial z}\left\{A_0\left[1+\epsilon A_1\right]\right\} = \epsilon A_0\left(\frac{\partial^2 A_1}{\partial x^2} + \frac{\partial^2 A_1}{\partial y^2}\right) \\ + 2k^2n_0^2\epsilon_z\{(D^b)_0A_0 + \epsilon[(D^b)_0A_1 + (D^b)_1A_0]\}, \end{aligned} \tag{3.3a}$$

where here we have introduced the notation

$$[D(A_0(1+\epsilon A_1))]^b = (D^b)_0 + \epsilon(D^b)_1. \tag{3.3b}$$

Expanding out the terms at each order, we obtain at $O(1)$:

$$2in_0^2k^2\epsilon_z\frac{\partial A_0}{\partial z} = 2k^2n_0^2\epsilon_z(D^b)_0A_0 \tag{3.4a}$$

$$i\frac{\partial A_0}{\partial z} = (D^b)_0A_0, \qquad A_0(0, t) = 1. \tag{3.4b}$$

At $O(\epsilon)$, we obtain the following:

$$\begin{aligned} 2in_0^2k^2\epsilon_z\epsilon\left(\frac{\partial A_0}{\partial z}A_1 + A_0\frac{\partial A_1}{\partial z}\right) &= \epsilon A_0\left(\frac{\partial^2 A_1}{\partial x^2} + \frac{\partial^2 A_1}{\partial y^2}\right) + 2k^2n_0^2\epsilon_z\epsilon[(D^b)_0A_1 + (D^b)_1A_0] \\ 2in_0^2k^2\epsilon_z\epsilon A_0\frac{\partial A_1}{\partial z} &= \epsilon A_0\left(\frac{\partial^2 A_1}{\partial x^2} + \frac{\partial^2 A_1}{\partial y^2}\right) + 2k^2n_0^2\epsilon_z\epsilon(D^b)_1A_0 \\ 2in_0^2k^2\epsilon_z\frac{\partial A_1}{\partial z} &= \frac{\partial^2 A_1}{\partial x^2} + \frac{\partial^2 A_1}{\partial y^2} + 2k^2n_0^2\epsilon_z(D^b)_1. \end{aligned} \tag{3.5}$$

where in going from the first line to the second we have used (3.4a).

Now we consider the *quasisteady* case. In this case, we assume that A varies slowly with respect to the t_n time scale. This doesn't make a lot of sense, as we would expect A to vary *on* the pulse time scale t_n, but it serves as a reasonably simple first approximation. In that case, we may treat $|A|$ in (2.18) as a constant, so we have

$$D(A) = |A|^4 t_n. \tag{3.6}$$

With this result, we see from (3.4b) that A_0 is always independent of t, so we let

$$A_0(z,t) = r_0(z)e^{i\theta_0(z)}, \qquad r_0(0) = 1, \quad \theta_0(0) = 0 \tag{3.7}$$

in (3.4b) to obtain

$$\begin{aligned} i\left(\frac{dr_0}{dz}e^{i\theta_0} + i\frac{d\theta_0}{dz}r_0e^{i\theta_0}\right) &= (D^b)_0 r_0 e^{i\theta_0} \\ i\frac{dr_0}{dz} - \frac{d\theta_0}{dz}r_0 &= (D^b)_0 r_0. \end{aligned} \tag{3.8}$$

We see from (3.6) that the right-hand side of (3.8) is real, so $dr_0/dz = 0$ and

$$r_0(z) = 1 \qquad \Longrightarrow \qquad D(A_0) = t_n, \tag{3.9a}$$

where we have used (3.6). Then substituting (3.9a) into the real part of (3.8), we have

$$\begin{aligned} -\frac{d\theta_0}{dz} &= t_n^b, \qquad \theta_0(0) = 0 \\ \theta_0(z) &= -t_n^b z. \end{aligned} \tag{3.9b}$$

Note that since the problem (at this stage) has no transverse variation, we expect no focusing and just a phase shift which increases in z.

Since $|A_0| = 1$, we may use the lemma in §2 with $\gamma = 2b$ to find that

$$\begin{aligned} D^b &= t_n^b |A_0(1+\epsilon A_1)|^{4b} \sim t_n^b[1 + 2\epsilon b(A_1 + \overline{A_1})]t_n^b \\ (D^b)_1 &= 2bt_n^b(A_1 + \overline{A_1}). \end{aligned} \tag{3.10}$$

To perform the linear stability analysis, we perturb the plane wave by a transverse complex exponential:

$$A_1(x,y,z,t) = A_+(z,t)\Phi(x,y), \quad \Phi(x,y) = e^{i(k_x x + k_y y)}. \tag{3.11}$$

Substituting (3.10) and (3.11) into (3.5), we obtain

$$2in_0^2k^2\epsilon_z\frac{\partial A_+}{\partial z}\Phi = (-k_x^2 - k_y^2)A_+\Phi + 2k^2n_0^2\epsilon_z[2bt_n^b(A_+\Phi + \overline{A_+\Phi})]. \tag{3.12}$$

Equation (3.12) yields a natural physical spatial scale to determine ϵ_z. In particular, if we choose

$$\epsilon_z = \frac{k_x^2 + k_y^2}{2n_0^2 k^2}, \tag{3.13}$$

then many of the coefficients in (3.12) will simplify. However, there is a problem in that we also wish to cancel the Φ terms, which we cannot do with the conjugate in the final term. In particular, the fact that D [and hence $(D^b)_1$] is real [as shown in (3.10)] messes up the cancellation. Therefore, we replace our *ansatz* in (3.11) with

$$A_1(x, y, z, t) = A_+(z, t)\Phi(x, y) + A_-(z, t)\overline{\Phi(x, y)}. \tag{3.14}$$

(We did previous work with just assuming a single cosine, rather than a complex exponential. Though it obtained the results given below, such a method is a special case of the more generic analysis performed here.)

Substituting (3.13) into (3.5) and using the replacement in (3.14), we obtain the following:

$$i(k_x^2+k_y^2)\left(\frac{\partial A_+}{\partial z}\Phi + \frac{\partial A_-}{\partial z}\bar{\Phi}\right) = (-k_x^2-k_y^2)A_+\Phi+2bt_n^b(k_x^2+k_y^2)(A_+\Phi+\overline{A_+\Phi}+A_-\bar{\Phi}+\overline{A_-\bar{\Phi}}). \tag{3.15}$$

Since the forcing term depends on t_n, not t, we replace ∂ with d in the subsequent analysis. Collecting coefficients of the positive and negative exponentials, we have from the positive exponential that

$$i\frac{dA_+}{dz} = -A_+ + 2bt_n^b\left(A_+ + \overline{A_-}\right), \tag{3.16a}$$

while from the negative exponential we have

$$\begin{aligned} i\frac{dA_-}{dz} &= -A_- + t_n^b\left(2bA_- + 2b\overline{A_+}\right) \\ -i\frac{d\overline{A_-}}{dz} &= -\overline{A_-} + 2bt_n^b\left(\overline{A_-} + A_+\right), \end{aligned} \tag{3.16b}$$

where in the last line we have taken the complex conjugate so we have a system in $\{A_+, \overline{A_-}\}$.

Equation (3.16) can be written as a matrix-vector system. But we are interested in the eigenvalues, rather than the eigenvectors. Therefore, it is enough to posit that

$$A_+ = c_+e^{i\mu z}, \qquad \overline{A_-} = c_-e^{i\mu z}, \tag{3.17}$$

and find the eigenvalues μ. Substituting (3.17) into (3.16), we have

$$\begin{aligned} -\mu c_+e^{i\mu z} &= -c_+e^{i\mu z} + 2bt_n^b\left(c_+e^{i\mu z} + c_-e^{i\mu z}\right) \\ 0 &= c_+(\mu + 2bt_n^b - 1) + 2bt_n^b c_-, \end{aligned} \tag{3.18a}$$

$$\begin{aligned} \mu c_-e^{i\mu z} &= -c_-e^{i\mu z} + 2bt_n^b\left(c_-e^{i\mu z} + c_+e^{i\mu z}\right) \\ 0 &= 2bt_n^b c_+ + c_-(-\mu + 2bt_n^b - 1), \end{aligned} \tag{3.18b}$$

The system (3.18) has a nontrivial solution only when

$$\begin{aligned}(\mu + 2bt_n^b - 1)(-\mu + 2bt_n^b - 1) - (2bt_n^b)^2 &= 0 \\ (2bt_n^b - 1)^2 - \mu^2 - (2bt_n^b)^2 &= 0 \\ -4bt_n^b + 1 &= \mu^2 \qquad (3.19a) \\ \mu &= \pm\sqrt{1 - 4bt_n^b}. \qquad (3.19b)\end{aligned}$$

Therefore, only two values of μ are allowed. At t_n^b, they are equal to 1, but after some time, the eigenvalues become imaginary, which causes exponential growth in z. That time is

$$4bt_n^b = 1 \qquad \Longrightarrow \qquad t_n = (4b)^{-1/b}.$$

Using the parameters in the Appendix, we have that the transition occurs at $t_n = 1/4$, or around 3 minutes.

We now briefly discuss what happens if $\epsilon = \epsilon_z$. In that case, the ϵ_z^2 terms in (2.4) would appear in our equation for the next term in the expansion. The problem is that these expressions are constant in x and y. Hence they would force at only a single mode—$\mu = 0$, which doesn't occur in the analysis.

Section 4: Linearization of Unsteady Case, No Explicit Time

Next we consider the more realistic case where $t_n = t$. However, we still assume that $\epsilon_t \ll \epsilon_z$, so we neglect the $\partial/\partial t$ term in (2.22). In that case, we may follow the analysis in §3, but we keep the full form of D in (2.18). Nevertheless, since D^b is still real, equations (3.9) hold with t_n replaced by t.

Moreover, we may again use the lemma in §2, though this time with $\gamma = 2$, to yield

$$
\begin{aligned}
D^b &= \left(\int_0^t |A_0(1+\epsilon A_1)|^4\,dt'\right)^b \sim \left[\int_0^t 1 + 2\epsilon(A_1 + \overline{A_1})\,dt'\right]^b = \left[t + 2\epsilon\int_0^t A_1 + \overline{A_1}\,dt'\right]^b \\
&\sim t^b\left(1 + \frac{2\epsilon}{t}\int_0^t A_1 + \overline{A_1}\,dt'\right)^b \sim t^b\left(1 + \frac{2b\epsilon}{t}\int_0^t A_1 + \overline{A_1}\,dt'\right) \\
(D^b)_1 &= 2bt^{b-1}\int_0^t A_1 + \overline{A_1}\,dt'. \qquad (4.1)
\end{aligned}
$$

Note that in the last expression of the second line above we have used a binomial expansion, and this expansion [and hence (4.1)] will hold only if t is not $O(\epsilon)$.

Again $(D^b)_1$ is real, so we must use both A_+ and A_- in our analysis, as in §3. Substituting (4.1) and (3.14) into (3.5), we have

$$
i\frac{\partial}{\partial z}\left(A_+\Phi + A_-\bar{\Phi}\right) = -\left(A_+\Phi + A_-\bar{\Phi}\right) + 2bt^{b-1}\int_0^t A_+\Phi + A_-\bar{\Phi} + \overline{A_+\Phi + A_-\bar{\Phi}}\,dt', \quad (4.2)
$$

where we have used (3.13). Equation (4.2) is analogous to (3.15). Hence we again separate positive and negative exponential coefficients:

$$
\begin{aligned}
i\frac{\partial A_+}{\partial z} &= -A_+ + 2bt^{b-1}\int_0^t A_+ + \overline{A_-}\,dt', \qquad (4.3a)\\
i\frac{\partial A_-}{\partial z} &= -A_- + 2bt^{b-1}\int_0^t A_- + \overline{A_+}\,dt' \\
-i\frac{\partial \overline{A_-}}{\partial z} &= -\overline{A_-} + 2bt^{b-1}\int_0^t \overline{A_-} + A_+\,dt'. \qquad (4.3b)
\end{aligned}
$$

Note that we have again taken the complex conjugate to make this a linear system in $\{A_+, \overline{A_-}\}$. In the case where A_- and A_+ are constant, then (4.3) would reduce to (3.16) with t replacing t_n.

The form of the normal modes is more complicated; we try functions of the form

$$
A_+ = c_+ e^{i\mu z} F(t), \qquad \overline{A_-} = c_- e^{i\mu z} F(t), \qquad (4.4)
$$

where $F(t)$ is to be determined. Substituting (4.4) into (4.3a) yields the following:

$$\begin{aligned}
-\mu c_+ e^{i\mu z} F(t) &= -c_+ e^{i\mu z} F(t) + 2bt^{b-1} \int_0^t F(t') e^{i\mu z}(c_+ + c_-)\, dt' \\
0 &= (\mu - 1)c_+ F(t) + 2bt^{b-1}(c_+ + c_-) \int_0^t F(t')\, dt'. \qquad (4.5)
\end{aligned}$$

In order for this to reduce to an algebraic equation, we must have that the $F(t)$ terms cancel; one way for this to happen is for

$$F(t) = \frac{bt^{b-1}}{\phi} \int_0^t F(t') \qquad (4.6a)$$

$$\begin{aligned}
\frac{d(Ft^{1-b})}{dt} &= \frac{b}{\phi} F \\
t^{1-b}\frac{dF}{dt} + (1-b)t^{-b}F - \frac{b}{\phi}F &= 0 \\
\frac{dF}{dt} + \left[(1-b)t^{-1} - \frac{bt^{b-1}}{\phi}\right] F &= 0
\end{aligned}$$

$$F = \exp\left(-\left[(b-1)\log t - \frac{t^b}{\phi}\right]\right) = t^{b-1} e^{t^b/\phi}. \qquad (4.6b)$$

Note that F diverges as $t \to 0$, which is consistent with our discussion before regarding how our expansion breaks down.

Substituting (4.6a) into (4.5), we obtain

$$\begin{aligned}
0 &= \frac{(\mu - 1)c_+}{\phi} + 2(c_+ + c_-) \\
0 &= (\mu - 1 + 2\phi)\, c_+ + 2c_-\phi. \qquad (4.7a)
\end{aligned}$$

Note that (4.7a) is just (3.18a) with bt_n^b replaced by ϕ. Hence by direct extension (4.3b) becomes

$$0 = 2c_+\phi + (-\mu - 1 + 2\phi)\, c_-. \qquad (4.7b)$$

The system (4.7) has a nontrivial solution only when

$$\begin{aligned}
-4\phi + 1 &= \mu^2 \\
\phi &= \frac{1 - \mu^2}{4}. \qquad (4.8)
\end{aligned}$$

The system (4.7) has solutions for all positive μ, so we always have oscillations in z. For $\mu < 1$ (which corresponds to high frequencies), we see that $\phi > 0$, which corresponds to exponential growth in time. Note also that the form of the solution is quite odd, since as $\mu \to 1^-$, ϕ^{-1} (which is the coefficient of t^b) blows up. However, recall that the purpose of the blowup in the linearized model is to show when the nonlinearity must be taken into account.

One question to investigate would be the spectral representation of a train of square pulses. This should have components in every mode, so it should grow in time.

We now briefly discuss what happens if $\epsilon = \epsilon_z$. In that case, the ϵ_z^2 terms in (2.4) would appear in our equation for the next term in the expansion. The problem is that these expressions are constant in x and y. Hence they would force at only a single mode—$\mu = 0$, which doesn't occur in the analysis.

Section 5: Explicit Time Dependence

We now return to the consideration of the quasisteady case, but now we take $\epsilon_t = \epsilon_z/c_\epsilon$, where $c_\epsilon = O(1)$. Therefore, (2.22) is replaced by

$$2in_0^2k^2\epsilon_z\left(\frac{\partial A}{\partial z}+\frac{1}{c_\epsilon}\frac{\partial A}{\partial t}\right)=\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}+2k^2n_0^2\epsilon_z D^b A. \tag{5.1}$$

But by defining

$$\tau = t - \frac{z}{c_\epsilon} \tag{5.2}$$

and writing $A(x,y,z,t)$ as $A(x,y,z,\tau)$, we have

$$\frac{\partial}{\partial t}\to\frac{\partial}{\partial \tau}, \qquad \frac{\partial}{\partial z}\to\frac{\partial}{\partial z}-\frac{1}{c_\epsilon}\frac{\partial}{\partial \tau}$$
$$2in_0^2k^2\epsilon_z\frac{\partial A}{\partial z}=\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}+2k^2n_0^2\epsilon_z D^b A,$$

which is just (3.1). Therefore, all our results from §3 hold with t replaced by τ, since in this case D is independent of t.

We now return to the consideration of the unsteady case where D does depend on t. We again perform a linear stability analysis. Therefore, substituting (3.2) into (5.1), we obtain, to leading order,

$$i\left(\frac{\partial A_0}{\partial z}+\frac{1}{c_\epsilon}\frac{\partial A_0}{\partial t}\right)=(D^b)_0A_0, \tag{5.3}$$

analogous to (3.4b). However, now we have a PDE, so we need both boundary and initial conditions:

$$A_0(0,t)=1, \qquad A_0(z,0)=0. \tag{5.4}$$

We may posit a solution of the form (3.7), but in this case both r_0 and θ_0 must be functions of time. Therefore, we have

$$A_0(z,t)=r_0(z,t)e^{i\theta(z,t)}; \qquad r_0(0,t)=1, \quad \theta_0(0,t)=0; \quad r_0(z,0)=0, \tag{5.5}$$

where we have used (5.4). Note that $\theta_0(z,0)$ is undetermined. Substituting (5.5) into (5.3), we obtain

$$i\left[\left(\frac{\partial r_0}{\partial z}+\frac{1}{c_\epsilon}\frac{\partial r_0}{\partial t}\right)e^{i\theta_0}+i\left(\frac{\partial \theta_0}{\partial z}+\frac{1}{c_\epsilon}\frac{\partial \theta_0}{\partial t}\right)r_0e^{i\theta_0}\right]=(D^b)_0r_0e^{i\theta_0}$$
$$i\left(\frac{\partial r_0}{\partial z}+\frac{1}{c_\epsilon}\frac{\partial r_0}{\partial t}\right)-\left(\frac{\partial \theta_0}{\partial z}+\frac{1}{c_\epsilon}\frac{\partial \theta_0}{\partial t}\right)r_0=(D^b)_0r_0, \tag{5.6}$$

analogous to (3.8).

With the definition of D in (2.18), the right-hand side of (5.6) is still real, so the imaginary part of (5.6) is

$$\frac{\partial r_0}{\partial z} + \frac{1}{c_\epsilon}\frac{\partial r_0}{\partial t} = 0,$$
$$r_0(z,t) = r_0(z - c_\epsilon t) = \begin{cases} 1, & z < c_\epsilon t, \\ 0, & z > c_\epsilon t, \end{cases}$$

where we have used the initial and boundary conditions in (5.5). Continuing to simplify, we obtain

$$r_0(z,t) = H(c_\epsilon t - z), \tag{5.7a}$$
$$r_0(z,\tau) = H(\tau). \tag{5.7b}$$

Substituting (5.7a) and the definition of D into the real part of (5.6), we have

$$-\left(\frac{\partial \theta_0}{\partial z} + \frac{1}{c_\epsilon}\frac{\partial \theta_0}{\partial t}\right) = \left(\int_0^t |A_0|^4\,dt'\right)^b = \left(\int_0^t |r_0(z - c_\epsilon t')|^4\,dt'\right)^b$$
$$\left(\frac{\partial \theta_0}{\partial z} + \frac{1}{c_\epsilon}\frac{\partial \theta_0}{\partial t}\right) = -H(c_\epsilon t - z)\left(\int_{z/c_\epsilon}^t r_0^4(z - c_\epsilon t')\,dt'\right)^b = -H(c_\epsilon t - z)\left(t - \frac{z}{c_\epsilon}\right)^b. \tag{5.8}$$

Then writing θ_0 as a function of τ and using (5.2), we obtain the following:

$$\frac{\partial \theta_0}{\partial z} = -\tau^b H(\tau); \qquad \theta(0,\tau) = 0$$
$$\theta_0(z,\tau) = -\tau^b H(\tau) z, \tag{5.9}$$

where we have used (5.5). Note that (5.9) is (3.9b) with t replaced by τ, multiplied by $H(\tau)$.

At next order, we have that the integral term will be of the form

$$\int_0^t |r_0(z - c_\epsilon t')|^4(\cdots)\,dt' = H(c_\epsilon t - z)\int_{z/c_\epsilon}^t |r_0(z - c_\epsilon t')|^4(\cdots)\,dt'$$
$$= H(\tau)\int_0^\tau |r_0(-\tau)|^4(\cdots)\,d\tau' = H(\tau)\int_0^\tau (\cdots)\,d\tau',$$

where the $(\cdots)$ term contains terms only in the unknown A_1. The transverse derivatives just yield a constant times A_1, and when going to the τ variable, the left-hand side of the operator just becomes $\partial/\partial z$. Hence we believe (but didn't have time to check) that our analysis in §4 holds with t replaced by τ.

Section 6: Conclusions and Further Research

When using fused silica lenses for photolithography and other uses, it is critical to maintain the optical integrity of the lens for as long as possible. The desire for finer beam control has led to the use of smaller wavelengths in the UV range. Unfortunately, these wavelengths correspond to higher intensities. These higher intensities increase the two-photon dosage imparted by the beam. The increased dosage, in turn, yields to compaction or densification of the lens, and eventually the formation of microchannels.

In this work, we have derived the governing equations using a theory for the dose-compaction relationship given in [3, 4, 5]. We examined the case of a slightly perturbed plane wave moving through the fused silica. The leading-order solution satisfies both the linear and nonlinear terms. Then transverse perturbation leads to a linear stability analysis.

We examined both the quasisteady case, where the dosage is assumed to be occurring on a different time scale from the slowly varying amplitude, and the unsteady case, where both processes are assumed to occur on the same time scale. We began by considering the case where the explicit time dependence is suppressed.

In the quasisteady case, the stability analysis leads to an estimate for the time at which the modes become unsteady and begin to grow exponentially in time. This time compares favorably with experimental and simulated results. In the unsteady case, the form of the time-dependent eigenfunctions is more complicated, and do not hold for small time. In this analysis, the growth rate of the mode depends on its wavelength, with high-frequency waves having the fastest growth rate.

We then considered the case where time is explicitly included. In the quasisteady case, we were able to establish that our previous results held with t replaces by the shifted time variable τ. We have strong evidence that the same result holds in the unsteady case, but we were unable to verify this in the time provided.

In addition to the work in this report, the other avenues were also explored:

Yuxin Chen worked on coding up the solution as in the Wright paper. She used both x- and y-directions with periodic boundary conditions. She doesn't have the integral term in her calculations; just the t^b term.

With the numerics, we need to verify exactly how δn is being calculated. From (2.20), we would have that

$$\left(\frac{\delta n}{n_0 \alpha \epsilon_z}\right)^{1/b} = \int_0^{t_n} |A|^4 \, dt'_n$$

$$\frac{d}{dt_n}\left(\frac{\delta n}{n_0 \alpha \epsilon_z}\right)^{1/b} = |A|^4.$$

This seems to be different from what was previously described in the presentations, namely that the equation used was

$$\frac{d(\delta n)}{dt} = C|A|^{4b}.$$

Les did not seem particularly concerned about this, and suspects that it won't affect the structure of the solution that much.

Roy Goodman attempted to find a similarity solution to the problem, but he discovered the operator does not admit one.

Recall that in the plane-wave solution of the paraxial equation (with constant refractive index), the amplitude does not vary with z. Hence we were able to use the plane-wave solution as A_0 and still satisfy the paraxial equation with the nonlinear term added to represent the time-varying refractive index.

Another solution of the paraxial equation (with constant refractive index) is the Gaussian beam, which has the form

$$A(r,0) = e^{-(r/r_c)^2} \tag{6.1}$$

in cylindrical coordinates, where r_c is some characteristic width of the Gaussian (called the waist). In contrast to the plane-wave solution, this beam focuses, which causes its amplitude to vary with z. Our original plan was to use this beam as A_0. However, since its amplitude varies with z, this meant that the nonlinear terms which appear at leading order cannot easily be satisfied.

We next thought about retaining the plane wave as A_0, but using the Gaussian as our perturbation to replace (3.11). Though all the details haven't been worked out yet, it seems that since the Gaussian satisfies the paraxial equation with constant refractive index, it may be difficult to track the form of the solution including the nonlinearity. (Zyskin has some work on this topic in his chapter.)

Richard O. Moore found a paper [8] that considers acoustics within an optical fiber. In particular, there is the following equation for $\Delta\tilde{\rho}$, which is proportional to δn:

$$\frac{\partial^2 \Delta\tilde{\rho}}{\partial t^2} - \Gamma' \nabla^2 \frac{\partial \Delta\tilde{\rho}}{\partial t} - \nu^2 \nabla^2 \Delta\tilde{\rho} = -\frac{\gamma_e}{4\pi} \nabla_\perp^2 |\bar{\mathbf{E}}|^2.$$

Moore thought that perhaps when calculating our electric field, we might be forcing this equation on a harmonic, which would then cause $\Delta\tilde{\rho}$ to grow as long as the damping coefficient Γ' was small.

Nomenclature

Units are listed in terms of mass (M), pulses (N), length (L), and time (T). If a symbol appears both with and without tildes, the symbol with tildes has units, while the one without is dimensionless. Equation numbers where a variable is first defined is listed, if appropriate.

A: slowly-varying amplitude of transmitted wave (2.3).
b: exponent in compaction law (2.1).
c: speed (variously defined), units L/T.
$\tilde{D}$: two-photon dose, units M^2/T^5 (2.1).
$\tilde{E}$: energy field, units M/T^2 (2.2).
$F(t)$: function used to derive amplitude perturbations in unsteady case (4.4).
f: frequency, units T^{-1} (A.6a).
$\tilde{I}$: intensity per pulse, units variously defined.
j: integer used to index pulses (2.10).
k: wavenumber of light wave, units L^{-1} (2.3).
N: number of pulses, units N (2.1).
n: refractive index (2.2).
R_0: characteristic scale for slowly-varying amplitude, units M/T^2 (2.3).
r: radius of complex function (3.7) or radial coordinate (6.1).
T: period of portion of pulse, units T/N (2.1).
t: time, units T (2.2).
x: transverse distance, units L (2.2).
y: transverse distance, units L (2.2).
$\tilde{z}$: propagation distance, units L (2.2).
$\mathcal{Z}$: the integers.
α: proportionality constant in δn law (2.19).
γ: arbitrary constant (2.23).
ϵ: small parameter, variously defined.
ε_0: vacuum permittivity (2.14a).
θ: argument of complex function.
κ: constant of proportionality in $\delta\rho$ law (2.1).
λ: wavelength, units L.
μ: spatial eigenvalue (3.17).
ρ: density of fused silica, units M/L^3 (2.1).
τ: shifted t variable (5.2).
$\Phi(x, y)$: Fourier mode in linear analysis (3.11).
ϕ: constant in exponent of $F(t)$ (4.6b).
ω: frequency of light wave, units T^{-1} (2.3).

Other Notation

c: as a subscript on T, refers to the cycle (2.13).
e: as a subscript on I, refers to energy density (2.1).
$n \in \mathcal{Z}$: as a subscript, used to indicate a perturbation series in ϵ (2.23).
n: as a subscript, used to indicate a time scale that balances the δn term (2.17).
p: as a subscript, used to indicate properties of the pulse (2.1).
r: as a subscript, used to indicate properties of the rest period between pulses.
ϵ: as a subscript on c, used to indicate a ratio (5.1).
0: as a subscript on n, used to indicate vacuum; otherwise, characteristic scale (2.3).
$-$: as a subscript, used to indicate negative exponential (3.14).
$+$: as a subscript, used to indicate positive exponential (3.11).
$\bar{\ }$: used to indicate complex conjugate (2.23).

Appendix: Parameter Values

Now we want to gather the parameter values from the literature so that we can actually calculate some values. The parameters will come from [2, 7, 9]. First we begin by calculating ϵ_z. We want to write (3.13) in terms of wavelengths, so we have

$$\epsilon_z = \frac{\lambda^2}{2n_0^2(\lambda_x^2 + \lambda_y^2)}. \tag{A.1}$$

From Wright [7], we have that

$$\lambda = 193 \text{ nm}, \qquad n_0 = 1.5. \tag{A.2}$$

Les told us that typically the transverse wavelength were tens of microns, so we choose

$$\lambda_x = \lambda_y = 50\,\mu\text{m}. \tag{A.3}$$

Substituting (A.2) and (A.3) into (A.1), we obtain the following:

$$\epsilon_z = \frac{(1.93 \times 10^{-1}\,\mu\text{m})^2}{2(1.5)^2[2(5 \times 10\,\mu\text{m})^2]} = \frac{3.73}{225} \times 10^{-4} = 1.66 \times 10^{-6}, \tag{A.4}$$

which is small, as theorized.

Next we compute the wave train time scale. From Wright [7], we have that

$$T_\text{p} = 20\,\frac{\text{ns}}{\text{pulse}}, \qquad I_\text{e} = 50\,\frac{\text{mJ}}{\text{cm}^2 \cdot \text{pulse}}, \qquad b = 0.5, \tag{A.5a}$$

while from Piao [2], we have the following:

$$T_\text{p} = 30\,\frac{\text{ns}}{\text{pulse}}, \qquad I_\text{e} = 10\text{–}20\,\frac{\text{mJ}}{\text{cm}^2 \cdot \text{pulse}}, \qquad b = \frac{2}{3}. \tag{A.5b}$$

The value of $b = 2/3$ is also given by Primak [3, 5]. In both these manuscripts, T_p is denoted τ, I_e is denoted I, and b is denoted c. The value of T_c is hard to discern from Wright, since it's a numerical simulation. Les gave a value of the frequency:

$$f_\text{c} = \frac{1000 \text{ pulses}}{\text{s}} \qquad \Longrightarrow \qquad T_\text{c} = 10^{-3}\,\frac{\text{s}}{\text{pulse}}, \tag{A.6a}$$

while in Piao, they have

$$f_\text{c} = 330 \text{ Hz} \qquad \Longrightarrow \qquad T_\text{c} = 3.03 \times 10^{-3}\,\frac{\text{s}}{\text{pulse}}. \tag{A.6b}$$

κ is slightly more complicated, as Wright [7] just gives a value of

$$\kappa = 0.6 \times 10^{-6},$$

which can't be right because of the units in (2.1). However, digging into the paper, we find

1. This is the true value of κ, as expressed in the text. In the definition of κ, it says that it is expressed in ppm (parts per million), but the values in Piao [2] (see below) indicate that the 10^{-6} component of κ has already been baked in.
2. For κ to have this value, N has to be expressed in millions of pulses (Mpulses), $I_{\mathrm{e},0}$ has to be expressed in mJ/cm^2/pulse, and T_{p} has to be expressed in ns/pulse.

Therefore, from (2.1) we have that

$$\kappa = 0.6 \times 10^{-6} \left[\frac{(\mathrm{mJ/cm^2/pulse})^2 \cdot \mathrm{Mpulses}}{\mathrm{ns/pulse}} \right]^{-b} = 0.6 \times 10^{-6} \left(\frac{\mathrm{cm^4 \cdot ns}}{10^6\ \mathrm{mJ^2}} \right)^{b}. \tag{A.7}$$

Wright [7] also has a value of

$$\alpha = 0.3. \tag{A.8}$$

Piao [2] defines $A' = \alpha\kappa$, whose value is given in the range

$$0.275 \times 10^{-6} \le A' \le 0.435 \times 10^{-6}. \tag{A.9}$$

Note that the values given in Piao's paper, which are just the decimals, are given in parts per million (ppm), so we have inserted the 10^{-6} terms. But this verifies the proper size of κ above. The same convention is used regarding the units.

We begin by using all the values in Wright and the value of T_{c} from Les. Then we have

$$\begin{aligned}
T_n &= \frac{T_{\mathrm{p}} T_{\mathrm{c}}}{I_{\mathrm{e},0}^2} \left(\frac{\epsilon_z}{\alpha\kappa} \right)^{1/b} \\
&= \left(20\,\frac{\mathrm{ns}}{\mathrm{pulse}} \right) \left(10^{-3}\,\frac{\mathrm{s}}{\mathrm{pulse}} \right) \left(50\,\frac{\mathrm{mJ}}{\mathrm{cm^2 \cdot pulse}} \right)^{-2} \left(\frac{1.66 \times 10^{-6}}{0.3} \right)^{2} \\
&\qquad \div (0.6 \times 10^{-6})^2 \left(\frac{\mathrm{cm^4 \cdot ns}}{10^6\ \mathrm{mJ^2}} \right) \\
&= \frac{(2 \times 10^{-2})(5.53 \times 10^{-6})^2}{(50)^2 (3.6 \times 10^{-13})} \left(\frac{\mathrm{ns \cdot s \cdot cm^4}}{\mathrm{mJ^2}} \right) \left(\frac{10^6\ \mathrm{mJ^2}}{\mathrm{cm^4 \cdot ns}} \right) \\
&= \frac{2(5.53)^2}{(25)(3.6)} \times 10^3\ \mathrm{s} = 6.80 \times 10^2\ \mathrm{s}.
\end{aligned}$$

Note that T_n corresponds to about 6.8×10^5 pulses. In Wright [7], they observe their first "hot spots" around 6×10^6 pulses, so at least this is in the right ball park.

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Deriving an improved paraxial wave equation

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Maxwell's equations in the absence of free charges and currents are given by

$$\nabla \cdot \mathbf{D} = 0, \qquad \nabla \cdot \mathbf{B} = 0, \tag{1a}$$

$$\nabla \times \mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{B}}{\partial t}, \qquad \nabla \times \mathbf{H} = \frac{1}{c}\frac{\partial \mathbf{D}}{\partial t}, \tag{1b}$$

$$\mathbf{D} = \varepsilon \mathbf{E}, \qquad \mathbf{B} = \mu \mathbf{H}, \tag{1c}$$

where $\mathbf{E}$ and $\mathbf{B}$ are the electric and magnetic fields, $\mathbf{D}$ and $\mathbf{H}$ are the displacement and magnetizing fields, ε is the permittivity, and μ the permeability of the medium.

For the problem at hand, assume $\mu = 1$ and that the permittivity varies with space and time. The permittivity is related to the index of refraction by $n^2 = \varepsilon$. Using this relation, and combining (1b) with (1c) gives

$$\nabla \times \mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{B}}{\partial t} \tag{2}$$

$$\nabla \times \mathbf{B} = \frac{1}{c}\frac{\partial (n^2\mathbf{E})}{\partial t}. \tag{3}$$

Taking the curl of (2) and using the definition $\nabla \cdot \mathbf{B}$, the following is obtained:

$$\nabla \times \nabla \times \mathbf{E} = -\frac{1}{c^2}\frac{\partial^2 (n^2\mathbf{E})}{\partial t^2}. \tag{4}$$

Recall the identity

$$\nabla \times \nabla \times \mathbf{E} = \nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}. \tag{5}$$

Taking the divergence of the constitutive equation $\mathbf{D} = n^2\mathbf{E}$, and using the fact that $\nabla \cdot \mathbf{D} = 0$, we find that

$$\nabla \cdot \mathbf{E} = -\frac{1}{n^2}\mathbf{E} \cdot \nabla n^2; \tag{6}$$

thus the final PDE for $\mathbf{E}$ is given by

$$\nabla\left(\frac{1}{n^2}\mathbf{E}\cdot\nabla n^2\right)+\nabla^2\mathbf{E}=\frac{1}{c^2}\frac{\partial^2(n^2\mathbf{E})}{\partial t^2}. \tag{7}$$

Note that when n is a constant, the first term on the right hand side of (7) is zero, and we recover the classic form of the wave equation. Equation (7) may also be written as

$$\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\left(n^2\mathbf{E}\right)=\nabla\left(\mathbf{E}\cdot\nabla[\ln(n^2)]\right)+\nabla^2\mathbf{E}. \tag{8}$$

Given this equation as a starting point, we may re-derive the paraxial wave approximation, to initially retain all contributions from the variation of the amplitude with respect to space and time.

As an intermediate step towards this goal, we retained the contributions from the time-dependence of the index of refraction, but neglected the influence of its spatial variations, expecting those to be very small. This yields a paraxial wave equation of the form

$$i2n_0k\left(\frac{\partial A}{\partial z}+\frac{1}{c_*}\frac{\partial}{\partial t}\left(n^2A\right)\right)=\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}+2k^2n_0\delta nA, \tag{9}$$

where the dependence of the index of refraction on the amplitude is taken to be

$$\delta n\equiv n(x,y,z,t)-n_0=\alpha\left(\int_0^t |A(x,y,z,\tau)|^4\,d\tau\right)^b. \tag{10}$$

Expanding out the time-derivative term in (9) yields

$$\frac{\partial A}{\partial z}+\frac{n^2}{n_0c^*}\frac{\partial A}{\partial t}+\frac{2nA}{n_0c^*}\frac{\partial n}{\partial t}=-\frac{i}{2n_0k}\left(\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}\right)-ik\alpha\left(\int_0^t |A|^4\,d\tau\right)^b,$$

which can be made more explicit with respect to its dependence on A by using the derivative of n from (10),

$$\frac{n^2}{n_0c^*}\frac{\partial A}{\partial t}+\frac{\partial A}{\partial z}+\frac{2\alpha bn}{n_0c^*}\left(\int_0^t |A|^4\,d\tau\right)^{b-1}|A|^4A=-\frac{i}{2n_0k}\left(\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}\right)-ik\alpha\left(\int_0^t |A|^4\,d\tau\right)^bA. \tag{11}$$

If we consider nearly uniform plane waves, we can seek solutions describing the complex amplitude in phase form as

$$A(x,y,z,t)=R(z,t)e^{i[\phi(z,t)+k_xx+k_yy]},$$

with R,ϕ being real functions and (k_x,k_y) being real transverse wavenumbers. Separating real and imaginary parts of the resulting equation yields transport equations for R,ϕ:

$$\frac{n^2}{n_0c^*}\frac{\partial R}{\partial t}+\frac{\partial R}{\partial z}+\frac{2\alpha bn}{n_0c^*}\left(\int_0^t R^4\,d\tau\right)^{b-1}R^5=0, \tag{12a}$$

$$\frac{n^2}{n_0 c^*}\frac{\partial \phi}{\partial t} + \frac{\partial \phi}{\partial z} = -\frac{k_x^2 + k_y^2}{2n_0 k} - k\alpha\left(\int_0^t R^4\, d\tau\right)^b, \tag{12b}$$

with

$$n = n_0 + \alpha\left(\int_0^t R^4\, d\tau\right)^b.$$

Note that in this form, the equation for $R(z,t)$ can be solved first, then that solution can be plugged into the equation for the phase.

Returning to the general equation (11), another approach to analyzing this equation is to use perturbation methods to examine the influence of the variation of the index of refraction in the limit of weak densification effects, namely $\alpha \to 0$, where $n \sim n_0$ to leading order as $\alpha \to 0$. Consider expanding the amplitude function as

$$A = A_0 + \alpha A_1 + O(\alpha^2);$$

then at $O(1)$, equation (11) reduces to

$$\frac{\partial A_0}{\partial t} + \frac{c_*}{n_0}\frac{\partial A_0}{\partial z} + \frac{ic_*}{2n_0^2 k}\left(\frac{\partial^2 A_0}{\partial x^2} + \frac{\partial^2 A_0}{\partial y^2}\right) = 0, \tag{13a}$$

and at $O(\alpha)$ the correction to the solution due to compaction effects satisfies

$$\frac{\partial A_1}{\partial t} + \frac{c_*}{n_0}\frac{\partial A_1}{\partial z} + \frac{ic_*}{2n_0^2 k}\left(\frac{\partial^2 A_1}{\partial x^2} + \frac{\partial^2 A_1}{\partial y^2}\right) = -\left(\int_0^t |A_0|^4\, d\tau\right)^b\left(\frac{2}{n_0}\frac{\partial A_0}{\partial t} + \frac{2b}{n_0}\left(\int_0^t |A_0|^4\, d\tau\right)^{-1}|A_0|^4 A_0 + \frac{ic_*}{n_0}\right). \tag{13b}$$

At leading order, separation of variables yields exact solutions as modulated traveling waves

$$A_0(x,y,z,t) = F(z - (c_*/n_0)t)\exp\left(i\left[k_x x + k_y y - \frac{(k_x^2 + k_y^2)c_*}{2n_0^2 k}t\right]\right), \tag{14}$$

for any traveling wave profile F (wave packet shape). Since (13a) is a Schrodinger equation, the Gaussian beam should also be an exact solution for A_0. To make some comments on going to next order in the calculations, we assume that the solution is uniform in the transverse directions, i.e. $A = A(z,t)$ or $k_x = k_y = 0$, then $A_0 = F(z - (c_*/n_0)t)$ and (13b) reduces to

$$\frac{\partial A_1}{\partial t} + \frac{c_*}{n_0}\frac{\partial A_1}{\partial z} = -\left(-\frac{n_0}{c_*}\int_z^{z-\frac{c_*}{n_0}t}|F(\zeta)|^4\, d\zeta\right)^b \times \left(\frac{2c_*}{n_0^2}F'(z - \tfrac{c_*}{n_0}t) + \frac{2b}{n_0}\left(-\frac{n_0}{c_*}\int_z^{z-\frac{c_*}{n_0}t}|F(\zeta)|^4\, d\zeta\right)^{-1}|F|^4 F + \frac{ic_*}{n_0}\right). \tag{15}$$

Solutions of this equation for A_1 can be expected to grow with time since the equation includes a (complicated) inhomogeneous forcing term that moves with the underlying wave speed. This means that the naive perturbation expansion, $A \sim A_0 + \alpha A_1$ will break down after A_1 grows enough so that $O(\alpha A_1) = O(A_0)$. An improved perturbation expansion solution would make use of multiple scales, in terms of a slow time $T = \alpha t$ and corresponding slow traveling wave spatial variable $R = \alpha\rho$ where $\rho \equiv z - (c_*/n_0)t$, to write the solution as $A(z,t) \sim A_0(\rho, R, T)$ and obtain a solvability equation for A_0 from the Fredholm alternative applied to equation (15).

Further work on analysis of a complete paraxial wave equation that also retains the terms for spatial dependence n is of interest.

Numerical Simulation

James Gambino
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We simulated a coupled system of a spacial PDE for the field amplitude, A, and an evolution equation for δn, the perturbation of the index.

$$A_z = \frac{i}{2kn_0}\nabla_T^2 A + ik_0\delta n A \tag{1}$$

$$\delta n_t = \gamma |A|^{4b} \tag{2}$$

After nondimensionalization of the equations we obtain

$$A_z = \mu\nabla_T^2 A + \nu\delta n A \tag{3}$$

$$\delta n_t = \Gamma |A|^{4b}. \tag{4}$$

Where

$$\mu = \frac{iL_z}{2kn_0L_x^2} \qquad \nu = ik_0L_z \qquad \Gamma = \gamma t_{\text{ref}} \tag{5}$$

with L_x, L_z the reference lengths in transverse and propagation directions respectively. The values of these dimensionless parameters were calculated from physical parameters found in Wright [1] and are shown in Table 1.

μ	8.4×10^{-3}	ν	1.22×10^{6}
Γ	3.7×10^{-7}		

Table 1: Parameters calculated from Wright [1].

The numerical solution was found using a two stage process similar to what was used in [1]. First, we fix the value of δn. We choose an initial condition for A at $z = 0$ and the integrate treating z as the time-like direction. (3) is solved using the fourth order Runge Kutta method and finite differencing for the transverse derivatives. This process results in a solution for A over the entire spacial grid at a given time step.

Next, a time step is taken by integrating (4) using a forward Euler step. This process is done pointwise at every grid point using the value of A calculated in the first stage. The entire process is then repeated until a set number of time steps have been taken.

Numerical Results

The numerical simulation was conducted with both one and two transverse dimensions. In the 2D case we examined the affects of a perturbed plane wave initial condition and in the 3D case we examined a smoothed hat initial condition.

2D results

In both of the following 2D cases 1600 grid points were used on the z-axis and 200 grid points were used on the x-axis. The first case we look at has the initial condition

$$A(x,0) = \frac{\sqrt{2}}{2}\left(1 + \frac{e^{i2\pi x}}{200}\right)$$

Figure 1 shows the intensity of the wave at time 0, corresponding to $\delta n(x,z) = 0$. We can see there is only a slight diffraction in the wave. Figure 2 shows the intensity of the wave

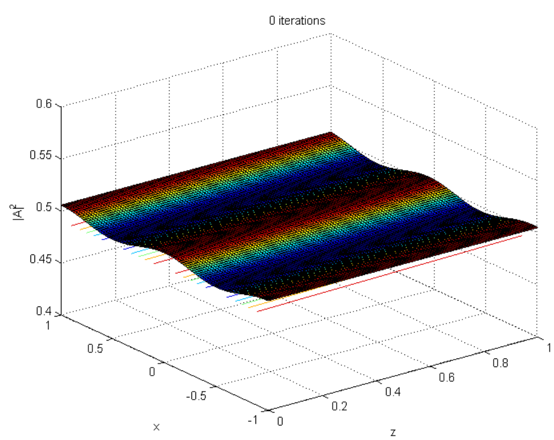

Figure 1: Intensity of perturbed plane wave at time 0

at time 20. The small intensity variation in the initial condition becomes prominent in the back wall of the lens at $z = 1$. Time 80, shown in Figure 3 shows that this process results in increasingly large intensity variation at the exit wall of the lens.

The second case we look at has the initial condition

$$A(x,0) = \frac{\sqrt{2}}{2}\left(1 + \frac{e^{i4\pi x}}{200}\right)$$

Figure 4 shows the intensity of the wave at time 0. As before, we can see there is only a slight diffraction in the wave. Figures 5 and 6 show the intensity at times 20 and 80 respectively. At time 20 we again see a small increase in intensity at the exit wall, while at time 80 the intensity dramatically increases at the exit wall of the lens.

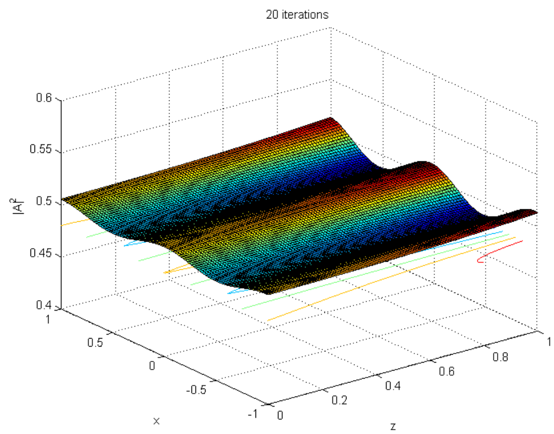

Figure 2: Intensity of perturbed plane wave at time 20

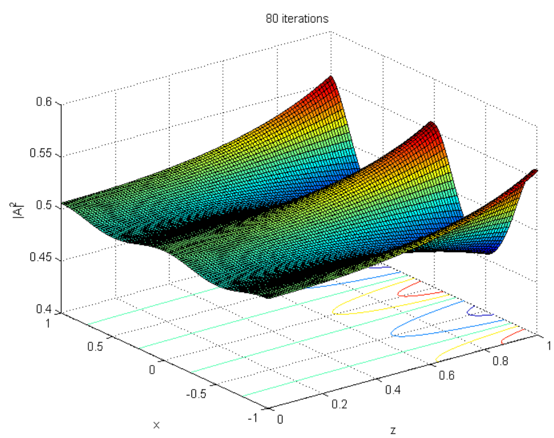

Figure 3: Intensity of perturbed plane wave at time 80

3D results

In the 3D case, due to time and computational constraints only 800 grid points were used in the z-axis and 100 grid points were used on the x and y axes. The initial condition used in this case was

$$A(x, y, 0) = \frac{\sqrt{2}}{4}(\tanh(20(.5 - r) + 1)$$

At time 0, shown in Figure 7, we can see that some diffraction does occur, as expected. We see in Figure 8, at time 20, we see higher intensity rings begin to form along the edge of the hat initial hat function.

Due to computational and time constraints, in order to examine further times we used a coarser grid of only 400 grid points in the z-axis and 50 on the x and y axes. Figures 9 and 10 show how the coarse grid causes numerical artifacts to appear along the edge of

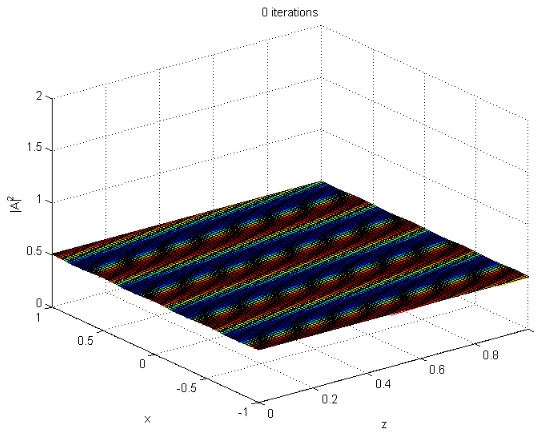

Figure 4: Intensity of perturbed plane wave at time 0

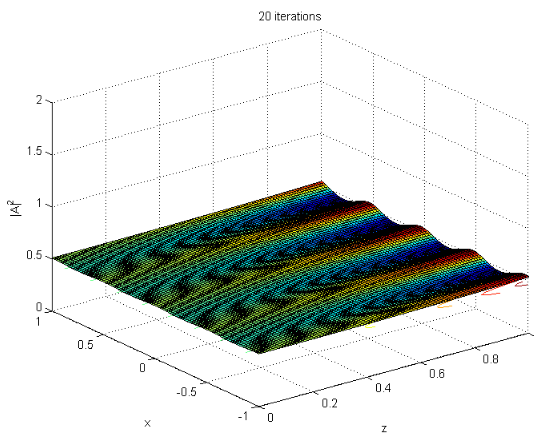

Figure 5: Intensity of perturbed plane wave at time 20

the aperture. At time 80, there are significant number of numerical irregularities appearing along the edge of the aperture.

References

[1] E. M. Wright, M. Mansuripur, V. Liberman, and K. Bates, "Spatial pattern of microchannel formation in fused silica irradiated by nanosecond ultraviolet pulses," *Appl. Opt.*, vol. 38, pp. 5785–5788, 1999.

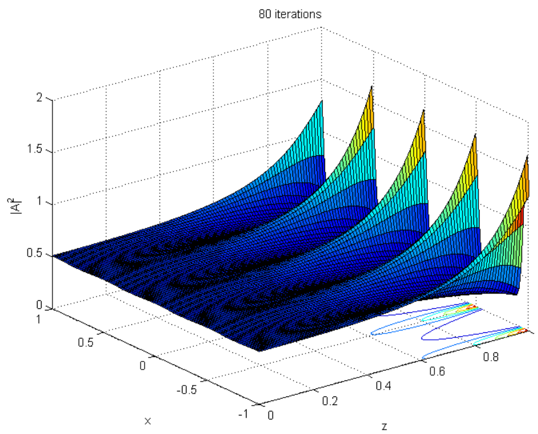

Figure 6: Intensity of perturbed plane wave at time 80

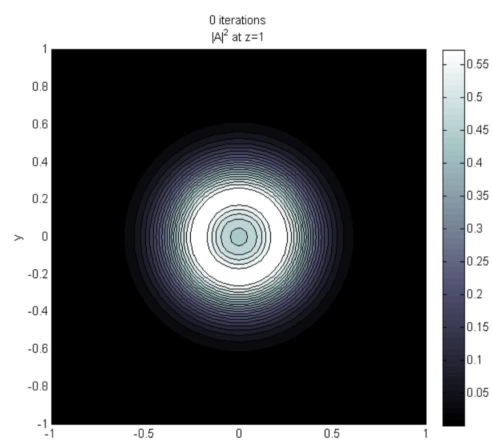

Figure 7: Intensity at z=1 at time 0 for 3D case.

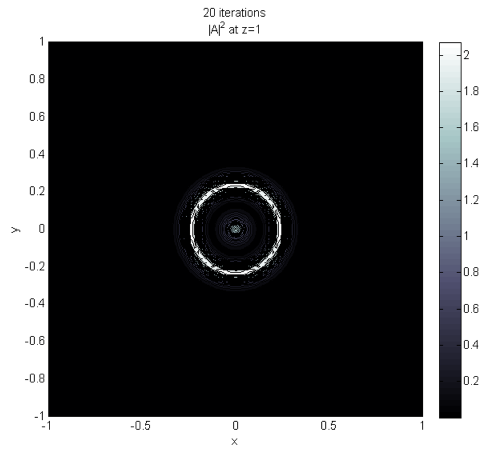

Figure 8: Intensity at z=1 at time 20 for 3D case.

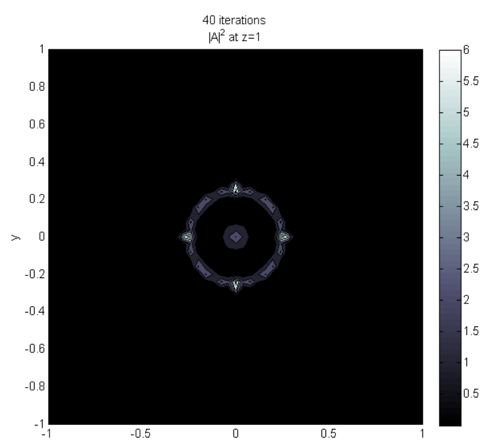

Figure 9: Intensity at z=1 at time 40 for 3D case.

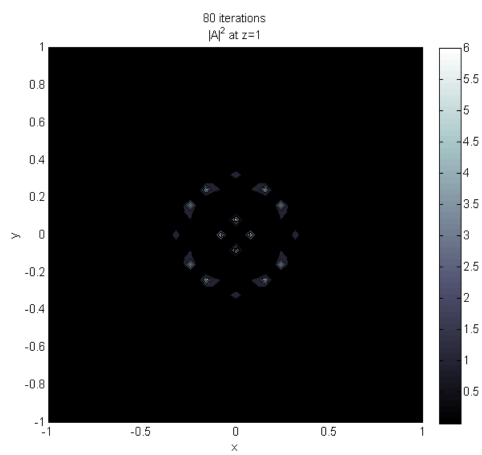

Figure 10: Intensity at z=1 at time 80 for 3D case.

Perturbative Gaussian Solution

Maxim Zyskin
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We consider the equation

$$\begin{aligned}
&\frac{1}{\imath\gamma}\frac{\partial A}{\partial z} = \Delta_2 A + \beta A \int_0^t |A(\tau)|^4\, d\tau,\\
&\Delta_2 \equiv \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}, \quad A = A(x,y,z,t),\\
&A(x,y,0,t) = A_0(x,y).
\end{aligned} \tag{1}$$

The solution of this system may be represented by

$$A(\ldots,z,t) = e^{\imath\gamma z\Delta_2}A_0 + \int_0^z e^{\imath\gamma(z-w)\Delta_2}\left[\beta A \int_0^t |A(\tau)|^4\, d\tau\right](\ldots,w,t)\,dw, \tag{2}$$

where $e^{\imath\gamma z\Delta_2}$ may be understood, for example, via the Fourier transform.

We will be interested in the case of

$$A_0(x,y) = \exp\left(-\alpha(x^2+y^2)\right). \tag{3}$$

We note that

$$e^{\imath\gamma z\Delta_2}\exp\left(-\phi(z)(x^2+y^2)\right) = \frac{1}{1+4\imath z\phi(z)\gamma}\exp\left(-\frac{\phi(z)}{1+4\imath z\phi(z)\gamma}(x^2+y^2)\right). \tag{4}$$

(This may be obtained via Fourier transform, or noting that the right hand side solves an appropriate equation and initial condition at $z=0$.)

Using (2), (4), a perturbative (Picard's iteration) solution may be obtained for the Gaussian case (3), in principle, to any order.

We illustrate this to the first order in perturbation theory.

We have that

$$a_0(x,y,z,t) = e^{\imath\gamma z\Delta_2}A_0 = \frac{1}{1+4\imath z\alpha\gamma}\exp\left(-\frac{\alpha(x^2+y^2)}{1+4\imath z\alpha\gamma}\right) = \frac{1}{1+4\imath\zeta}\exp\left(-\frac{\rho}{1+4\imath\zeta}\right), \tag{5}$$

where

$$\zeta \equiv \alpha\gamma z, \quad \rho \equiv \alpha(x^2+y^2). \tag{6}$$

(Note that $a_0(x, y, z, t)$ does not depend on t.) In the next order, we use a_0 in the nonlinear part of (2). Upon substituting $w = sz$, $0 < s < 1$, after a little computation and using (4) we obtain in the first order of perturbation

$$a_1(x, y, z, t) = (\beta t z) \int_0^1 \frac{(i - 4s\zeta)^2(-1 + 4is\zeta)^3}{1 - 20i\zeta + 20is\zeta - 16s\zeta^2 + 32s^2\zeta^2} \exp\left[\frac{i\rho(5i + 4s\zeta)}{1 - 20i\zeta + 20is\zeta - 16s\zeta^2 + 32s^2\zeta^2}\right] ds. \tag{7}$$

Analysis of the integral may be performed by various methods (for example, expanding exponent in a series and using partial fractions, or steepest descent method. We will consider this elsewhere).

It is clear that nonlinearity leads to focusing; there is also the usual dispersion of the beam.

As a quick illustration, we computed $I = |a_0(x, y, z, t) + a_1(x, y, z, t)|^2$ for the case when $\tau \equiv \beta t/\alpha\gamma = 0.5$. We have that I depends on ζ, ρ, and τ; results are shown in the figure below.

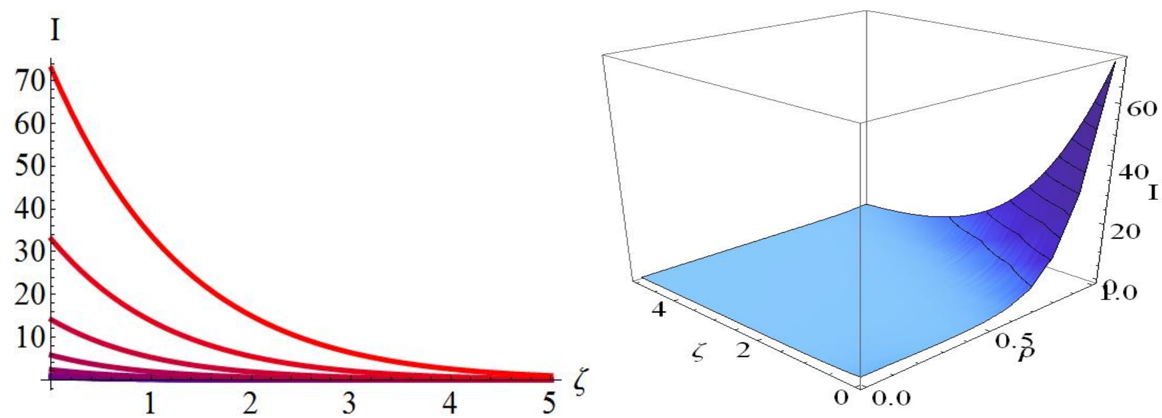

Figure 1: (a) I as a function of ρ, for consecutive ζ from 0 to 1 (blue to red color) (b) 3D plot of $I(\rho, \zeta)$. Here $\tau \equiv \beta t/\alpha\gamma = 0.5$.

It appears that we see the focusing effect, even in the first perturbation order.