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Pulse Propagation Through Different Materials User-Friendly Simulation Software

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Lund Reports on Atomic Physics, LRAP-310
Lund, August 2000

Abstract

High-intensity ultrashort laser pulses are affected by many different effects passing through materials. There is often a need to estimate these effects, especially the impact on the pulse duration and the spectrum. This estimation is not a trivial problem and needs to be simulated numerically. In this report a user-friendly simulation program that simulates the propagation of pulses through several different materials is presented.

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1 Introduction

The rapid development within the field of lasers has resulted in shorter and shorter laser pulses with an increasing peak power. Such ultrashort and high power pulses will be affected in different ways when passing through materials. In many experimental set-ups the pulses pass through at least one, some times several, optical component(s) such as windows, lenses etc. It is often of crucial importance to know the pulse duration at the site of the interaction, but not possible to measure it there. In these cases there is a need for calculating/estimating the pulse duration knowing the input pulse parameters and the design of the set-up. Normally, a rough estimate is done taking only dispersion into account. For an ultrashort, high power laser pulse this estimate is not sufficient. At least the combined effect of group velocity dispersion (GVD) and self phase modulation (SPM) has to be accounted for. For extremely short or very high power pulses, higher order effects must be taken into account as well.

A simulation program that takes most of the effects into account has been developed. The simulations in the program are based on the theory derived in ref [1] using the numerical method called split step Fourier method explained in the same reference. A short summary of the theory of pulse propagation and the numerical method is given in appendices A and B.

2 Short about propagation in different materials

The effects on the laser pulses propagating through a material are mainly due to two different phenomena:

- a short pulse consists of many frequencies that are affected differently by the material
- the high intensity will change the refractive index.

The refractive index in a material depends on the frequency. A short pulse consists of many different frequencies and will therefore be affected by different refractive index. The speed with which the pulse propagates in the material is called the group velocity and given by:

$$v_g = \left(\frac{d\omega}{d\beta} \right)_{\beta=\beta_o} \quad (1)$$

Where β is the wave number and β_o is the central wave number. The refractive index (and the wave number since $\beta = \frac{2\pi n(\lambda)}{\lambda}$) does not change linearly with frequency and different frequencies will therefore have different group velocities. This is known as group velocity dispersion, GVD and will stretch the pulse as it propagates through the material. In ordinary materials the low frequency components travel faster than the high frequency components resulting in a linear change of frequency across the pulse. A frequency change across the pulse is known as a chirp. For very short pulses with very broad spectra even higher order contributions become significant and can no longer be neglected.

The refractive index of the material also depends on the intensity of the pulses according to:

$$n(t) = n_o + n_2 I(t) \quad (2)$$

where n_2 is known as the non-linear refractive index. In most cases $n_2 > 0$ so that the refractive index increases with increasing intensity. Since the intensity of a laser pulse changes across the pulse, different parts will experience different refractive index. This effect is known as self phase modulation (SPM) and will affect the phase of the pulses according to

$$\phi = \omega_o t - \beta z = \omega_o t - \frac{\omega_o n_o z}{c} - \frac{\omega_o n_2 I(t) z}{c}. \quad (3)$$

Since the instantaneous frequency is given by the time derivative of the phase this too will depend on the intensity

$$\omega = \frac{\partial \phi}{\partial t} = \omega_o - \frac{\omega_o n_2}{c} \frac{\partial I}{\partial t} z. \quad (4)$$

Due to the time-dependent profile of the pulse new frequencies will be generated depending on the sign of the derivative. The leading edge of the pulse, where $\frac{\partial I}{\partial t} > 0$, will be shifted to lower frequencies while the trailing edge will be shifted to higher frequencies. The central part remains unaffected since $\frac{\partial I}{\partial t} \approx 0$ here. The total effect is a broadening of the spectrum with a change in frequencies across the pulses from lower to higher frequencies. $\frac{\partial I}{\partial t}$ does not change linearly across the pulse resulting in a non-linear chirp.

In any material the two above mentioned effects will act simultaneously and the combined effect will be greater than the sum of the separate contributions. SPM will generate a broader spectrum and enhance the effect of GVD. In a normal material SPM will generate new low frequency components at the leading edge that will travel faster than the rest of the pulse due to GVD (and vice versa for the trailing edge) and the temporal stretching of the pulse will therefore increase.

3 The program and how to use it

To be able to run the program Matlab (ver 5.3 or higher) has to be installed on the computer. The folder containing the program should be copied and added to the Matlab-path. To add a program to the Matlab-path use the **File** pop-up menu in the command window and choose **set path**. Then choose **path**, **add to path** and select the copied folder. To start the program type *PropagatePulse* in the command window and figure (1) will be displayed.

First add the input parameters to the program:

- Material. Choose a material in the list through which you would like to simulate the propagation?
- Pressure. If the chosen material is a gas, what is the pressure? If the material is not a gas this parameter will not be used.

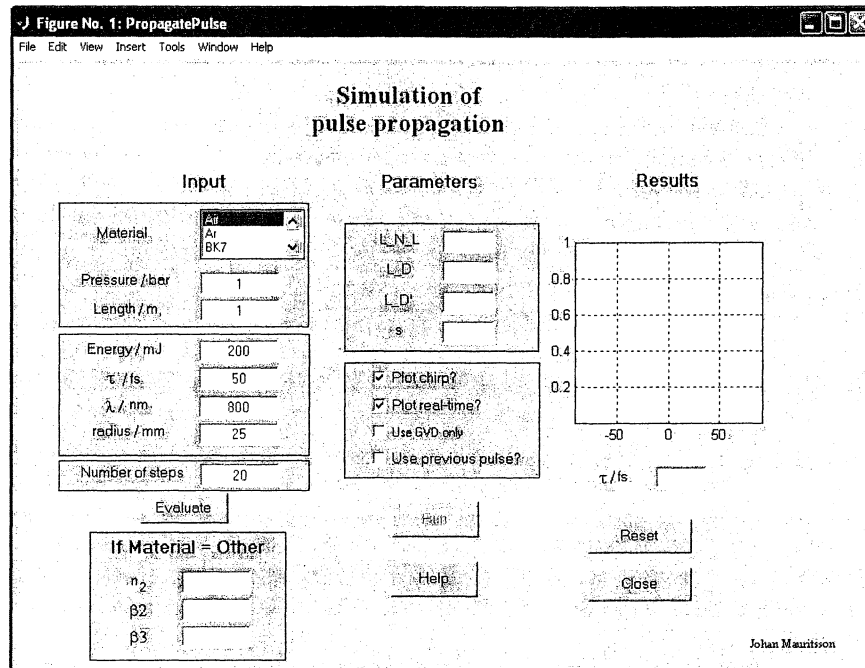


Figure 1: The program window as it looks when the program is first started.

- Length. The distance the pulses will travel through the given material?
- Energy. The energy per pulse?
- τ / fs. The time duration of the pulses (full width half maximum).
- λ / nm. The central wavelength of the pulses.
- Radius. The radius of the beam ($\text{Intensity}/e^2$).
- Number of steps. How many steps the calculations should take. Many steps makes it more accurate but it will take longer time, normally 20 steps is a good number to use.

If you would like to use a parameter not listed you can choose 'other material', but then you have to know the values of n_2 , β_2 and β_3 . After the parameters are inserted press the **evaluate** button and the program will calculate and display the parameters used in the simulation. Now you can choose if you would like the program to display the results in real-time (takes a little longer) or if you just want the final result. You can also tell the program to calculate the chirp, the result is not calibrated and will therefore only give the shape of the chirp.

After this you press the **run** button and the simulation starts. When the simulation is completed the pulse duration will be displayed in the box labelled ' τ / fs'. The following data will be displayed in this manner:

- Figure PropagatePulse. The pulse shapes of the input and output pulses are displayed.
- Figure Spec Freq. The input and output spectra as a function of frequency (relative intensity).
- Figure Spec Lambda. The input and output spectra as a function of wavelength (relative intensity).
- Figure Spec Lambda Norm. The input and output spectra as a function of wavelength, both normalized.
- Figure Phase. The temporal phase of the pulse.
- Figure Chirp. If you have chosen to calculate the chirp it will be displayed in this figure.

If you after the simulation would like the output pulse to be input in a new simulation (for instance if the pulses pass two different windows) check the '**use previous pulse**' check box, change the input parameters needed to match the new material, press **evaluate** and press **run**. This can be repeated as many times as you like. The program flow is depicted in figure 2.

If the energy of the pulses is low enough ($L_{NL} \gg L$) the effect of SPM is insignificant and dispersion is the only pulse broadening effect that has to be taken into account. In this case there is an analytic solution to the problem that makes the calculation much faster. If you want to know the pulse duration for pulses only affected by GVD (no higher order dispersion), check the check-box labelled 'use GVD only' before clicking the evaluate button.

3.1 Limitations

To be able to simulate the pulse propagation some approximations are needed. This will limit the range of situations covered by the program. The design is such that it will optimize the performance if the pulses are from a Ti:Sapphire laser centered around 800 nm with pulse duration of 30 - 150 fs.

- In the program the pulses are approximated with the envelope of the electric field. This approximation is valid as long as the pulse duration is much longer than the oscillation period of the electric field. For visible and near infrared light this corresponds to pulses longer than ~ 10 fs. The program cannot handle pulses shorter than this correctly.
- The wavelength range covered by the program is approximately 400-1000 nm. The lower limit depends on the absorption edge for the different materials.
- The program is one-dimensional and does not take effects such as diffraction and self-focusing into account. Different parts of a real pulse are affected differently passing through a material so that the pulse duration changes across the pulse. In the program the intensity used is the average intensity of the pulse and no spatial variations are included.

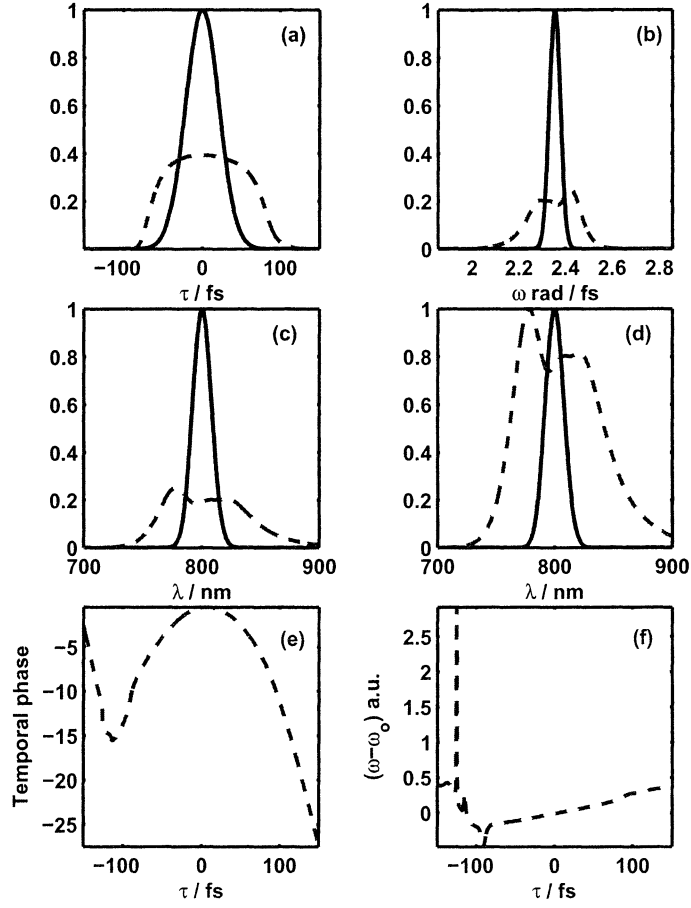


Figure 3: a) Temporal structure of the input (blue, solid line) and output (green, dashed line) pulses. b) Spectrum before and after propagation as function of angular frequency. c) Same as in b) but as a function of wavelength. d) Same as in c) but with both spectra normalized. e) The temporal phase. f) The chirp of the pulse.

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Appendices

A The propagation equation

To study the behavior of light propagating in any material one have to study Maxwell's equations

$$\nabla \times E = -\frac{\partial B}{\partial t}, \quad (5)$$

$$\nabla \times H = J_f + \frac{\partial D}{\partial t}, \quad (6)$$

$$\nabla \cdot D = \rho_f, \quad (7)$$

$$\nabla \cdot B = 0. \quad (8)$$

The flux densities D and B is due to the medias response to E and H and can be rewritten as:

$$D = \varepsilon_o E + P, \quad (9)$$

$$B = \mu_o H + M, \quad (10)$$

for a non-magnetic media $M = 0$. Using equations (6), (9) and (10) equation (5) can be rewritten in the following way by taking the curl of equation (5):

$$\begin{aligned} \nabla \times \nabla \times E &= \nabla \times \left(-\frac{\partial B}{\partial t} \right) \\ &= -\frac{\partial}{\partial t} (\nabla \times B) \\ \{M = 0\} &= -\mu_o \frac{\partial}{\partial t} (\nabla \times H) = \\ \{J_f = 0\} &= -\mu_o \frac{\partial}{\partial t} \left(\frac{\partial D}{\partial t} \right) = -\mu_o \frac{\partial^2}{\partial t^2} (\varepsilon E + P) \\ \left\{ \mu_o \varepsilon_o = \frac{1}{c^2} \right\} &= -\frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} - \mu_o \frac{\partial^2 P}{\partial t^2} \end{aligned} \quad (11)$$

and using the fact that

$$\nabla \times \nabla \times E = \nabla (\nabla \cdot E) - \nabla^2 E. \quad (12)$$

In a media without charge density, $\rho_f = 0$, (12) can be written as:

$$\nabla \times \nabla \times E = -\nabla^2 E, \quad (13)$$

resulting in

$$\nabla^2 E = \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} + \mu_o \frac{\partial^2 P}{\partial t^2}. \quad (14)$$

This equation is known as the wave equation and to know the behavior of the light it has to be solved. This is, however, not an easy task. Fortunately some approximations are possible to make that simplifies matter. One such approximation is to only include the nonlinear effects governed by $\chi^{(3)}$, then the polarization can be written as:

$$P(r, t) = P_L(r, t) + P_{NL}(r, t), \quad (15)$$

where P_L is the linear part and P_{NL} is the non-linear part. The wave equation can now be rewritten as:

$$\nabla^2 E - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \mu_o \frac{\partial^2 P_L}{\partial t^2} + \mu_o \frac{\partial^2 P_{NL}}{\partial t^2}. \quad (16)$$

The next approximation is to study the slowly varying amplitude of the envelope instead of the electric field. This approximation holds as long as the pulses are significantly longer than an optical cycle, i.e. pulses longer than around 10 fs if the wavelength is in the visible region [1]. If the slowly varying envelope is separated from the rapidly changing parts, the electric field and the polarization components can be written as:

$$E(r, t) = \frac{1}{2} \hat{x} [\bar{E}(r, t) \exp(-i\omega_o t) + c.c.], \quad (17)$$

$$P_L(r, t) = \frac{1}{2} \hat{x} [\bar{P}_L(r, t) \exp(-i\omega_o t) + c.c.], \quad (18)$$

$$P_{NL}(r, t) = \frac{1}{2} \hat{x} [\bar{P}_{NL}(r, t) \exp(-i\omega_o t) + c.c.], \quad (19)$$

where c.c. stands for complex conjugate. The non-linear part of the polarization is often small compared to the linear parts so that it can be considered as a perturbation. According to [1] it is possible to show that the refractive index is intensity dependent and can be written as:

$$\bar{n}(\omega) = n(\omega) + n_2 |\bar{E}|^2, \quad (20)$$

where the non-linear part, n_2 , can be shown to be [2]

$$n_2 = \frac{3}{8n} \chi_{xxxx}^{(3)}. \quad (21)$$

Another parameter that has to be part of the equation is the wave-number, $\beta = \frac{2\pi}{\lambda}$. This parameter depends on the frequency and to simplify matter it is expanded in a Taylor series centered around the central frequency ω_o

$$\beta(\omega) = \beta_o + (\omega - \omega_o) \beta_1 + \frac{1}{2} (\omega - \omega_o)^2 \beta_2 + \frac{1}{6} (\omega - \omega_o)^3 \beta_3 + \dots \quad (22)$$

where

$$\beta_n = \left(\frac{d^n \beta}{d\omega^n} \right)_{\omega=\omega_o}. \quad (23)$$

After some manipulations the wave equation can be rewritten as:

$$\frac{\partial A}{\partial z} + \beta_1 \frac{\partial A}{\partial t} + \frac{i}{2} \beta_2 \frac{\partial^2 A}{\partial t^2} + \frac{\alpha}{2} A = i\gamma |A|^2 A, \quad (24)$$

where A is the slowly varying envelope of the pulse and γ is known as the nonlinearity coefficient and is defined as:

$$\gamma = \frac{n_2 \omega_o}{c A_{eff}} \quad (25)$$

and A_{eff} is given by $\pi \omega^2$ if a capillary is studied but by $\frac{\pi \omega^2}{2}$ in most other cases¹.

¹The program uses $A_{eff} = \frac{\pi \omega^2}{2}$.

Since the program should be able to handle very short pulses some of the approximations made are not strictly valid and a few more terms are needed in the wave equation. When these terms are added, the frame of reference is also changed to a frame moving along with the pulse

$$T = t - \frac{z}{v_g} = t - \beta_1 z. \quad (26)$$

This finally gives a wave-equation valid for pulses down to ~ 10 fs, [1]

$$\frac{\partial A}{\partial z} + \frac{\alpha}{2} A + \frac{i}{2} \beta_2 \frac{\partial^2 A}{\partial T^2} - \frac{1}{6} \beta_3 \frac{\partial^3 A}{\partial T^3} = i\gamma \left[|A|^2 A + \frac{2i}{\omega_o} \frac{\partial}{\partial T} (|A|^2 A) - T_R A \frac{\partial |A|^2}{\partial T} \right]. \quad (27)$$

This equation is also known as the generalized nonlinear Schrödinger equation.

For inert gases the response time T_R is very fast (< 5 fs) and can therefore be neglected if the time duration of the laser pulses are much longer than this². The time scale and the slowly varying envelope can be normalized in the following way:

$$\tau = \frac{T}{T_o}, \quad (28)$$

$$A(z, \tau) = \sqrt{P_o} \exp\left(-\frac{\alpha z}{2}\right) U(z, \tau). \quad (29)$$

Using this and rearranging the terms in equation (27) results in:

$$i \frac{\partial U}{\partial z} = \frac{\text{sgn}(\beta_2)}{2L_D} \frac{\partial^2 U}{\partial \tau^2} + i \frac{\text{sgn}(\beta_3)}{6L'_D} \frac{\partial^3 U}{\partial \tau^3} - \frac{e^{-\alpha z}}{L_{NL}} \left[|U|^2 U + is \frac{\partial}{\partial \tau} (|U|^2 U) \right], \quad (30)$$

where:

$$L_D = \frac{T_o^2}{\text{sgn}(\beta_2)}, \quad (31)$$

$$L'_D = \frac{T_o^3}{\text{sgn}(\beta_3)}, \quad (32)$$

$$L_{NL} = \frac{1}{\gamma P_o}, \quad (33)$$

$$s = \frac{2}{\omega_o T_o}. \quad (34)$$

Equation (30) is the equation used in the program to simulate the pulse propagation.

B Numerical model

To study the behavior of the propagating pulses equation (30) has to be simulated since it is not analytically solvable. The method used is known as the Split-step Fourier method [1].

²Even though the program is to be used for other materials than inert gases this term is excluded, since it in most cases has a minor effect on the pulses. It is however possible to add it to the program if that should be needed.

The idea behind the Split-step Fourier method is to treat the operators that accounts for the dispersion and absorption and the operators for the nonlinearities separately. Equation(27) can now be written as

$$\frac{\partial A}{\partial z} = (\hat{D} + \hat{N}) A, \quad (35)$$

where:

$$\begin{aligned} \hat{D} &= -\frac{i}{2}\beta_2 \frac{\partial^2}{\partial T^2} + \frac{1}{6}\beta_3 \frac{\partial^3}{\partial T^3} - \frac{\alpha}{2}, \\ \hat{N} &= i\gamma \left[|A|^2 + \frac{2i}{\omega_o A} \frac{\partial}{\partial T} (|A|^2 A) - T_R \frac{\partial |A|^2}{\partial T} \right]. \end{aligned} \quad (36)$$

If the two operators are treated as if they were independent equation (35) can be solved in two steps. The first step is to solve the equation if the nonlinearities acts alone, $\hat{D} = 0$ and the second step is to solve the equation when dispersion acts alone, $\hat{N} = 0$. The solution is then given by:

$$A(z+h, T) \simeq \exp(h\hat{D}) \exp(h\hat{N}) A(z, T). \quad (37)$$

The exponential operator $\exp(h\hat{D})$ is calculated in the Fourier domain:

$$\exp(h\hat{D}) B(z, T) = \left\{ F^{-1} \exp[h\hat{D}(i\omega)] F \right\} B(z, T), \quad (38)$$

$\hat{D}(i\omega)$ is obtained by replacing all the $\frac{\partial}{\partial T}$ by $i\omega$ in (36)

$$\hat{D}(i\omega) = \frac{i}{2}\beta_2 \omega^2 - \frac{i}{6}\beta_3 \omega^3 - \frac{\alpha}{2}. \quad (39)$$

It can be shown that this method is accurate to the second order in the step size h . To improve the method it has to be taken into account that \hat{N} varies across h , until now we have just added the nonlinearities at the boundaries of each section, one way to do this is to rewrite equation (37) as:

$$A(z+h, T) \simeq \exp\left(\frac{h}{2}\hat{D}\right) \exp\left(\int_z^{z+h} \hat{N}(z') dz'\right) \exp\left(\frac{h}{2}\hat{D}\right) A(z, T). \quad (40)$$

This is called the symmetrized split-step Fourier method. The integral accounts for the z -dependence of the nonlinearities. It can be shown that this method is accurate to the third order of the step size h . This expression is however more complicated to calculate, even if we use a very rough estimate of the integral (the trapezoidal rule)

$$\int_z^{z+h} \hat{N}(z') dz' \simeq \frac{h}{2} [\hat{N}(z) + \hat{N}(z+h)], \quad (41)$$

we end up with an expression that we have to iterate. However, since the accuracy is higher we can use bigger steps, h , and gain time anyhow. The method can be visualized in the following way: think of the media, in which the pulse propagates, as divided in equispaced segments (separated by h). The pulse propagates the distance $h/2$ only effected by dispersion, then it's multiplied with a term that represents the nonlinear effect over the entire segment and after that it propagates the remaining $h/2$ only effected by dispersion.

In the program the symmetrized split-step Fourier method is used to simulate the propagation.

C The refractive index

For most solid materials the Sellmeir equation has been used to calculate the dispersion. The equation used is:

$$n^2 = 1 + \frac{B_1\lambda^2}{\lambda^2 - C_1} + \frac{B_2\lambda^2}{\lambda^2 - C_2} + \frac{B_3\lambda^2}{\lambda^2 - C_3} \quad (42)$$

B_n and C_n are constants for the different materials.

For the gases the following formula has been used instead:

$$n^2 = 1 + k_1 \left(1 + \frac{k_2}{\lambda^2} + \frac{k_3}{\lambda^4} + \frac{k_4}{\lambda^6} + \frac{k_5}{\lambda^8} + \frac{k_6}{\lambda^{10}} + \dots \right) \quad (43)$$

k_n are constants for the different materials.

The refractive index is also intensity dependent, that is, very high intensities will change the refractive index according to

$$n = n_o + n_2 I, \quad (44)$$

where n_o is the refractive index for low intensities, I is the intensity and n_2 is called the nonlinear refractive index. There is a relation between the nonlinear refractive index and the nonlinear susceptibility, $\chi^{(3)}$, [2] given by

$$n_2 = \frac{12\pi^2}{n_o^2 c} \chi^{(3)}. \quad (45)$$

The intensity dependance of the refractive index is what causes effects like SPM and self focusing to occur.

C.1 Material parameters

C.1.1 Refractive index for different materials

Material constants for material calculated using

$$n^2 = 1 + \frac{B_1\lambda^2}{\lambda^2 - C_1} + \frac{B_2\lambda^2}{\lambda^2 - C_2} + \frac{B_3\lambda^2}{\lambda^2 - C_3}, \quad [\lambda] = \mu m$$

Material	B_1	B_2	B_3	C_1	C_2	C_3	Ref
Air	$5.7378 \cdot 10^{-4}$	—	—	0.0059526	—	—	
BK7	1.03961212	0.231792344	1.01046945	0.00600069867	0.0200179144	103.560653	[3]
Fused Silica	0.6961663	0.4079426	0.8974794	0.00467914826	0.0135120631	97.9340025	[3]
LiF	0.61388327	0.30609975	0.00274702	0.00775673	0.00113901	2.88235102	[4]
MgF ₂ eo	0.01173533	0.90388658	0.00502180	0.016499869	0.005374722	3.823524155	[3]
MgF ₂ oo	0.01937139	0.86427500	0.00432548	0.011587826	0.005202665	3.729621083	[3]
Sapphire eo	1.50397590	0.550691410	6.59273790	0.00548041129	0.0147994281	402.895140	[3]
Sapphire oo	1.43134930	0.650547130	5.34140210	0.00527992610	0.0142382647	325.017834	[3]
SF11	1.73848403	0.311168974	1.17490871	0.0136068604	0.0615960463	121.922711	[5]

$$n^2 = A + \frac{B}{\lambda^2 - C} + \frac{D\lambda^2}{\lambda^2 - E}, \quad [\lambda] = \mu m$$

Material	A	B	C	D	E	Ref
KDP oo	2.2576	0.0101	0.0142	1.7623	57.8984	[6]
KDP eo	2.1295	0.0097	0.0014	0.7580	127.0535	[6]
DKDP oo	2.2409	0.0097	0.0156	2.2470	126.9205	[6]
DKDP eo	2.1260	0.0086	0.0120	0.7844	123.4032	[6]

$$n^2 = 1 + k_1 \left(1 + \frac{k_2}{\lambda^2} + \frac{k_3}{\lambda^4} + \frac{k_4}{\lambda^6} + \frac{k_5}{\lambda^8} + \frac{k_6}{\lambda^{10}} + \dots \right), \quad [\lambda] =$$

Material	k ₁	k ₂	k ₃	k ₄	k ₅	k ₆	Ref
He	6.927·10 ⁻⁵	2.24·10 ⁵	5.94·10 ¹⁰	1.72·10 ¹⁶	—	—	[7]
Ne	1.335·10 ⁻⁴	2.24·10 ⁵	8.09·10 ¹⁰	3.56·10 ¹⁶	—	—	[7]
Ar	5.547·10 ⁻⁴	5.15·10 ⁵	4.19·10 ¹¹	4.09·10 ¹⁷	4.32·10 ²³	—	[7]
Kr	8.377·10 ⁻⁴	6.70·10 ⁵	8.84·10 ¹¹	1.49·10 ¹⁸	2.74·10 ²⁴	5.10·10 ³⁰	[7]
Xe	1.366·10 ⁻³	9.02·10 ⁵	1.81·10 ¹²	4.89·10 ¹⁸	1.45·10 ²⁵	4.34·10 ³¹	[7]

C.1.2 The nonlinear refractive index

Material	n_2 ($10^{-20} m^2/W$)	Reference
MgF ₂	1.15 ± 0.20	[8]
Suprasil	2.82 ± 0.30	[8]
SiO ₂	2.88	[9]
Sapphire	3.2	[10]
Herasil	3.21	[8]
BBO	3.4	[10]
BK7	3.75	[8]
Ruby	4.15	[8]
Water	5.7	[8]
Methanol	6.7	[8]
CCl ₄	19	[8]
KTP	23.4	[10]
Benzene	24	[8]
CS ₂	150	[8]

Gas	n_2 ($10^{-23} m^2/W$)	Reference
Ar	0.980	[11]
Ne	0.1	[10]
H ₂	1.11	[11]
Kr	2.8	[10]
N ₂	4.52	[11]
Air	5.57	[11]
Xe	8.1	[10]
O ₂	9.68	[11]