Nonlinear Optics (NLO)

V 2.0 (Feb. 2017)

Most of the experiments performed during this course are perfectly described by the principles of *linear optics*. This assumes that interacting optical beams (i.e. beams crossing paths) will not affect each other. As the light intensity is increased, however, this assumption no longer holds; we are forced to consider the possibility of *nonlinear* interactions. Nonlinear optical effects manifest in a a variety of phenomena including frequency (wave) mixing, harmonic generation, self-focusing, and multi-photon absorption. The NLO experiments in this lab are designed to introduce students to some of these interactions using simple setups. Before diving into the experimental procedures, a student must have a basic understanding of the foundational principles of nonlinear optics. See, for example : http://phys.strath.ac.uk/12-370/ .

An important concept is the time response of a light-matter interaction. Although any physical interaction cannot be instantaneous, we can reasonably assume the response to be instantaneous if it occurs within (i.e. less than) the period of an optical cycle. In that case, we can express the total polarization of the material when irradiated by an optical field *E(t)* to contain a nonlinear response represented by nonlinear susceptibility $\chi^{(2)}$, $\chi^{(3)}$, .., in addition to the usual linear susceptibility $\chi^{(1)}$:

$$P = \varepsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \right] \quad (1)$$

Here $\chi^{(n)}$ represents the n-th order nonlinear susceptibility and ε_0 is the permittivity of free space. Recall that the linear response $\chi^{(1)}$ contains the refractive index of the material: $n_0 = \sqrt{1 + \Re\{\chi^{(1)}\}}$ and the (linear) absorption coefficient: $\alpha_0 = \left(\frac{4\pi}{\lambda_0}\right)\sqrt{1 + \Im\{\chi^{(1)}\}}$ where λ_0 is the wavelength of light in vacuum.

In the most general case, assuming $E(t) = \sum_j A_j cos\left(\omega_j t - k_j . r\right); j = 1,2, \ldots$, the nonlinear polarization terms in Eq. (1) can result in many combinations of frequencies. For example, the second order response $\chi^{(2)}$ can produce new optical fields with frequencies $2\omega_j$ (second harmonic), $\omega = 0$ (a DC term corresponding to optical rectification), and $\omega_j \pm \omega_m$ (m#j, sum and difference frequencies). The third-order nonlinearity $\chi^{(3)}$ will result in $3\omega_j$ (third harmonic), and a variety of sum and difference frequency generation terms that are often described as four-wave mixing. Only materials with a broken centro-symmetry have a nonzero $\chi^{(2)}$. All even-order coefficients vanish in symmetric systems. In solids, only crystals that lack inversion symmetry have nonzero

$\chi^{(2)}$ tensor elements. Every optical material must have a nonzero $\chi^{(3)}$ and higher odd-order response. In practice, it is rare and difficult to experimentally investigate nonlinearities that are higher order than 3.

(a) **Second Harmonic Generation (SHG)**

The NLO experiments in this lab will use a single monochromatic beam of light (laser) with frequency ω_1. The 2nd order nonlinear polarization will give rise to $2\omega_1$ and a DC term. You are required to investigate the second harmonic generation (SHG) process in a nonlinear crystal (not the DC term) which describes optical rectification. The SHG process is a useful for generating high power coherent beams at the visible and ultraviolet wavelengths starting from readily available infrared laser sources. The conversion efficiency and other characteristic of SHG are governed by the Maxwell equations, once the nonlinear polarization is identified:

$$\nabla^2 E - \frac{1}{c_0^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2} \quad (2)$$

Details are left for advanced courses such as PHYC 568. Results of this analysis are highly pertinent to your experiments.

Energy conservation requires that each second harmonic photon is generated at the expense of two photons from the fundamental field; $\omega+\omega\rightarrow 2\omega$. The fundamental photons are annihilated. Momentum conservation demands that the wave-vectors obey the following vector addition: $\vec{k}(\omega) + \vec{k}(\omega) = \vec{k}(2\omega)$. When all the fields are co-propagating, we can drop the vector notation and use the amplitude only: $|k(\omega)|=n(\omega)\omega/c$. Any deviation from the perfect matching condition is described by

$$\Delta k = 2k(\omega_1) - k(2\omega_1) = \left(\frac{2\omega_1}{c}\right)\left(n(\omega_1) - n(2\omega_1)\right) \quad (3)$$

Wave-vector conservation ($\Delta k=0$) is not possible in general due to normal dispersion. Fortunately, many nonlinear crystals are anisotropic: the refractive index depends on the direction of polarization (birefringence). A uniaxial birefringent crystals is characterized by $n_o(\omega)$ (ordinary) and $n_e(\omega)$ (extra-ordinary) refractive indices that correspond to polarization normal to and parallel to the axis of symmetry of the crystal (c-axis), respectively. Further fine tuning is provided by the angle ($\theta=\theta_m$) between the direction of

propagation and the c-axis that leads to $n_o(\omega)$ and $n_e(\omega,\theta)$. From crystal optics analysis, one finds that for a uniaxial crystal:

$$\frac{cos^2\theta}{n_o^2} + \frac{sin^2\theta}{n_e^2} = \frac{1}{n_e^2(\theta)} \quad (4)$$

The Type-I phase-matching condition suggests that we choose the angle $\theta=\theta_m$ such that $n_o(\omega)=n_e(2\omega,\theta_m)$ or $n_o(2\omega)=n_e(\omega,\theta_m)$ depending on the type of the birefringence. Crystals with $n_e>n_o$ are called positive uniaxial, and those with $n_e<n_o$ are known as negative uniaxial. There is also Type-II phase matching that has $n_o(\omega)+n_e(\omega,\theta_m)= 2n_e(2\omega,\theta_m)$ or $n_o(\omega)+n_e(\omega,\theta_m)= 2n_o(2\omega)$, again depending on the type of birefringence. In this case, the polarization of fundamental beam is rotated by 45^o (or crystal is re-oriented) such that there are equal amount of ordinary and extra-ordinary components in the fundamental beam. For more details, see this link. Given a nonlinear crystal and its empirical Sellmeier equations for its wavelength-dependent principal refractive indices, you can calculate the phase matching angle. You may also download the SNLO software and find phase matching conditions for a variety of crystals and all types of wave-mixing processes.

Perfect phase matching with a sufficiently long nonlinear crystal at high enough intensities can, in principle, result in 100% optical power conversion from fundamental to the SHG wavelength. For low SHG conversion (known as *low depletion limit*), there is a simple solution to Eq. (1) that gives the intensity of the second harmonic at 2ω in terms of the fundamental intensity $I(\omega)$ for a crystal of length l:

$$I_{2\omega} = \frac{2\omega^2 d_{eff}^2 l^2}{n^3 c^3 \epsilon_0}\left(\frac{sin\left(\frac{\Delta kl}{2}\right)}{\frac{\Delta kl}{2}}\right)^2 I_\omega^2 , \quad (5)$$

where d_{eff} ($d=\chi^{(2)}/2$) reflects the *tensor product* of $\chi^{(2)}$ and E-field at the fundamental frequency. Consult the SNLO software for d_{eff} for a given nonlinear crystal with Type-I or Type-II phase matching.

Question: *The above equation was derived assuming plane waves. Ignoring diffraction, what is the power conversion efficiency assuming $I_\omega=I_0exp(-r^2/w^2)$? If the incident laser is also pulsed such that $I_\omega=I_0exp(-r^2/w^2)\times exp(-t^2/t_p^2)$, what is the energy conversion efficiency per pulse?*

(c) **Optical Kerr Effect (OKE)**

Third-order nonlinearities are the next higher order, resulting in $\chi^{(3)}$ effects. As with the second-order $\chi^{(2)}$ interactions, a variety of frequency mixing can result from 3rd order nonlinearities. When the incident beam is a single monochromatic field $E=A_0\cos(\omega_1 t-k_1 z)$, the 3rd order polarization $\chi^{(3)}E^3$ results in two terms only: a $3\omega_1$ term (third-harmonic generation) and a term at the fundamental frequency ω_1. The total polarization at ω_1 is the sum of the linear and nonlinear terms:

$$P(\omega_1) = \epsilon_0\left(\chi^{(1)} + 3\chi^{(3)}A_0^2\right)E \ . \ (6)$$

The term in the bracket simply implies that the $\chi^{(1)}$ is now modified by an intensity dependent term ($I \propto A_0^2$). Using arguments similar to those following Eq. (1), the refractive index n will vary with the light intensity:

$$n = n_0 + n_2 I, \ (7)$$

Question: What is the relation between n_2 and Re{$\chi^{(3)}$}?

Intensity-dependent refraction is also known as the optical Kerr effect (OKE). This has many applications in photonics including controlling light with light (optical switching), optical solitons in fibers, mode-locking, and ultrashort pulse generation in solid-state lasers.

Mode-locking relies on the spatial consequence of OKE. For example, a Gaussian laser beam where $I=I_0 exp(-2r^2/w^2)$ produces a gradient index profile n(r) when inserted into Eq. (7). Elementary optics shows that a gradient index profile will lead to lensing. This self-lensing (also known as Kerr-lensing) can cause self-focusing ($n_2>0$) or self-defocusing ($n_2<0$). These so-called "self-action" phenomena can also be used as tools for measuring the n_2 coefficient in materials. A sensitive and popular technique known as Z-scan uses the self-lensing effect to measure the nonlinear refraction in a simple setup. A nonlinear sample is scanned along the z-axis near the focus of a lens and its far-field transmission through an aperture is measured. See this online demo. You will be asked to perform Z-scan experiments to measure various nonlinearities including two-photon absorption as described below.

(d) **Two-Photon Absorption (TPA or 2PA)**

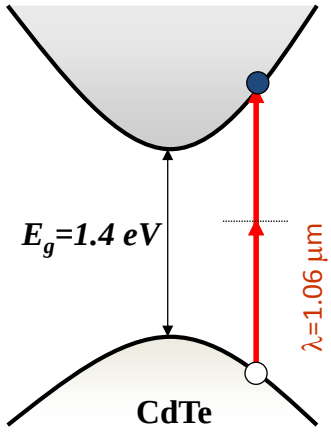
In general $\chi^{(3)}$ is a complex quantity (like $\chi^{(1)}$) with its imaginary component associated with "absorption"; it becomes nonzero only at or near an electronic resonance of the material. In the nonlinear response, this is associated with a multi-photon resonance: the combined energy of two or more photons are needed to make a resonant transition. In the case of $\chi^{(3)}$, when $2\omega=\omega_0$ (ω_0 is the material's resonance), we encounter two-photon absorption where material is excited by simultaneously absorbing two photons of the incident beam. Similar to equation (7), we can write an intensity-dependent absorption coefficient:

$$\alpha = \alpha_0 + \alpha_2 I. \quad (8)$$

Here α_2 describes the two-photon absorption coefficient.

NOTE: Many authors or texts may use β instead of α_2

Question: What is the relation between α_2 and Im{$\chi^{(3)}$}?



Two-photon absorption in semiconductors: Example: CdTe under irradiation by λ=1064 nm laser pulses. Show that 2PA is also allowed for λ=1.55 μm.

In semiconductors, this is often observable when the incident photon energy $\hbar\omega$ exceeds half the band-gap energy E_g (or $2\hbar\omega>E_g$) *and* the irradiance is high enough. The propagation of a beam experiencing two-photon absorption is then governed by a nonlinear Beer's law:

$$\frac{dI}{dz} = -\alpha_0 I - \alpha_2 I^2 \quad (9)$$

At the same time, one can track the population of the excited state. In the case of semiconductors, this is the time-evolution of the density of electrons in the conduction band (ΔN_e) or holes in the valance band ($\Delta N_h=\Delta N_e\equiv\Delta N_{eh}$):

$$\frac{d\Delta N_{eh}}{dt} = \frac{\alpha_0 I}{\hbar\omega} + \frac{\alpha_2 I^2}{2\hbar\omega} - \frac{\Delta N_{eh}}{\tau} \quad (10)$$

where τ is the effective electron-hole recombination lifetime. In cases where TPA is studied in the transparency regime of a semiconductor ($2\hbar\omega>E_g>\hbar\omega$), it is quite reasonable to ignore linear absorption (i.e. assume α_0~0).

(e) **Non-instantaneous Nonlinearities:**

Nonlinear refraction and absorption as described by Equations (7) and (8) imply the response is instantaneous which, in this case, means the it should be faster than the laser pulsewidth. This often requires relatively high peak powers and tightly focused pulsed lasers. There are however other mechanism that are relatively slow and therefore cumulative. These types of nonlinearities are in principle a cascading process and more easily observed. For example, the refractive index can also vary with temperature due to various factors. This is known as the thermo-optic effect and it is quantified by the thermo-optic coefficient $\frac{dn}{dT}$:

$$n = n_0 + \frac{dn}{dT}\Delta T \text{ , (11)}$$

where ΔT is the temperature change. An intensity-dependent effect then results when we consider laser induced heating due to absorption in the sample:

$$C_v \frac{d\Delta T}{dt} = \alpha_0 I + \kappa \nabla^2 T \text{ (12)}$$

Where C_v (J/cm^3/K) is the heat capacity, and κ is the coefficient of thermal conductivity. It is also assumed that all the absorbed power is converted to heat. For cases where only a fraction (φ<1) is converted to heat (i.e. the rest is fluorescence), then we must use $\varphi\alpha_0$. If the laser pulsewidth (t_p) is shorter than the thermal diffusion time ($\tau_d \sim w^2 C_v/\kappa$, we ignore the diffusion term and estimate $\Delta T(t) \approx \frac{1}{C_v}\int_{-\infty}^{t} \alpha_0 I(t')dt'$. Time averaging over the duration of a Gaussian pulse further simplifies this to $< \Delta T > \approx \frac{\sqrt{\pi}}{2C_v}\alpha_0 I_0 t_p$. With the use of (11), one can now write:

$$\langle n \rangle \approx n_0 + \left[\frac{dn}{dT}\frac{\sqrt{\pi}}{2C_v}\alpha_0 t_p\right] I_0 \text{ (13)}$$

Notice that this mimics the optical Kerr effect and result in self-action observable in, for example, Z-scan experiments. Thermal lensing can be used in determine small amounts of absorption in materials.

In the case of CW laser radiation or when long pulses (with duration >>τ_d) are used, the steady-state approximation to Eq. (12) gives: $\Delta T \approx \frac{1}{\kappa}\alpha_0 I_0 w^2 = \frac{2}{\pi\kappa}\alpha_0 P_0$, where P_0 is the laser power. This, in turn leads to refractive index changes of $n \approx n_0 + \left[\frac{dn}{dT}\frac{2}{\pi\kappa}\alpha_0\right] P_0$ and subsequently a thermal phase shift $\theta = \frac{\alpha_0 PL}{\lambda\kappa}\frac{dn}{dT}$. We will see shortly how this can be measure in a thermal Z-scan measurement.

Another cascading effect may result from the change of refractive index due to changes in population density in various states. For semiconductors, this is written as:

$$n = n_0 + \frac{dn}{d\Delta N_e}\Delta N_{eh} \equiv n_0 + \sigma_{FCR}\Delta N_{eh} \quad (14)$$

where σ_{FCR} is the free carrier refraction coefficient. From simple models that include the band-filling (saturation) effects, it has been shown to follow an approximate scaling law: $\sigma_{FCR}(cm^{-3}) \approx \frac{5\times10^{-21}}{n_0 E_g^3} H(\frac{\hbar\omega}{E_g})$ where E_g (eV) is the bandgap energy and $H(x)=[x^2(x^2-1)]^{-1}$.

For more details, you may consult the literature in the following links: Said et. al. & Sheik-Bahae/van Stryland.

A corresponding nonlinearity can be written for the absorption:

$$\alpha = \alpha_0 + \sigma_{FCA}\Delta N_{eh} \quad .(15)$$

where σ_{FCA} is known as the free-carrier absorption (FCA) cross-section. In intrinsic (undoped) semiconductors, FCA is often dominated by transition between heavy-hole and light-hole valence bands, and scales with λ^p (p~2-3).

Experiments:

For all the experiments described below, you will have two pulsed lasers at your disposal:

Laser 1. A modelocked diode laser/fiber amplifier system by Raydiance. See Appendix-A for more details.

- Pulse energy: 1 μJ to 5 μJ
- Wavelength: 1.55 μm
- Pulse rate: 1Hz to 500 kHz
- Pulse width: ~800 fs (1ps)
- Fully software controlled
- Peak Power (calculate it)
- Spot Size (?)

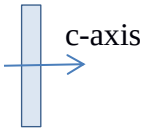
Laser 2. A diode-pumped Q-switched Nd:YAG laser with the following characteristics by the manufacturer (CNI): Wavelength: λ_0=1064.59 nm

- Average Power: 2.35W
- Pulse Duration: 2.4 ns (*measure it as it varies!)*
- Repetition Rate: 10kHz
- Peak Power*: You calculate it!*
- Pulse Energy: *You calculate it!*
- Beam Spot <2mm

Changchun New Industries Optoelectronics Tech. Co.
Contact: +86-431-85603799 *http://www.cnilaser.com, sales@cnilaser.com*

Note: Use Laser-1 for your experiments unless (a) it is not working or (b) you have time and are interested to repeat the experiments at 1.06 μm and nanosecond pulses.

i) **SHG Experiments:**



NLO Crystal: β-Barium Borate (β-BaB_2O_4)- known as BBO

Thickness l=3mm. The crystal is cut so the c-axis is normal the surfaces as shown.

Data:

Refractive index n (λ in μm) http://www.unitedcrystals.com/BBOProp.html

$$n_o(\lambda) := \sqrt{2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2}, \quad n_e(\lambda) := \sqrt{2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2}$$

Nonlinear coefficient:

$$d_{eff}^{(I)} = d_{31}sin(\theta) + \left(d_{11}cos(\phi) - d_{22}sin(3\phi)\right)cos(\theta)$$

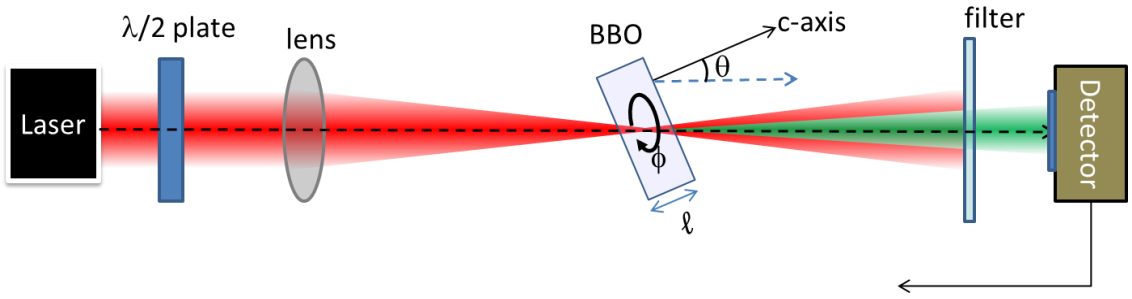
$$d_{eff}^{(II)} = \left(d_{11}sin(3\phi) + d_{22}cos(3\phi)\right)cos^2(\theta)$$

where d_{11}=0, d_{22}=-2.2, d_{31}=0.08 (pm/V) for BBO crystal (Group 3m)

Laser damage threshold (λ=1064 nm): 5 GW/cm^2 (10 ns); 10 GW/cm^2 (1.3 ns)

Reference: Eckardt et al, IEEE J. QE-26, 992 (1990),
also see https://www.coherent.com/downloads/BBO_DS.pdf

Experimental Set-up:



Procedure:

a) Calculate type-I PM angle (θ_m) (Note: Beware of the Snell's law)

b) Phase matching angle θ is adjusted by rotating around an axis normal to the optical table as depicted in Fig. above that shows the top view. Determine the correct polarization (S or P) of the incident fundamental and the generated second harmonic fields. Choose appropriate polarizer and waveplate to achieve the correct polarization.

c) From above, calculate the (external) acceptance angle Δθ (i.e. the angular spread that the crystal is phase-matched). What limitation does this impose on the spot size of the beam? (Hint: think diffraction and beam divergence)

d) Measure I(2ω) vs θ around θ_m

e) Describe the shape of the SH beam. If not circular, explain how is this related to Part (b) above?

f) Calculate Type-II PM angle

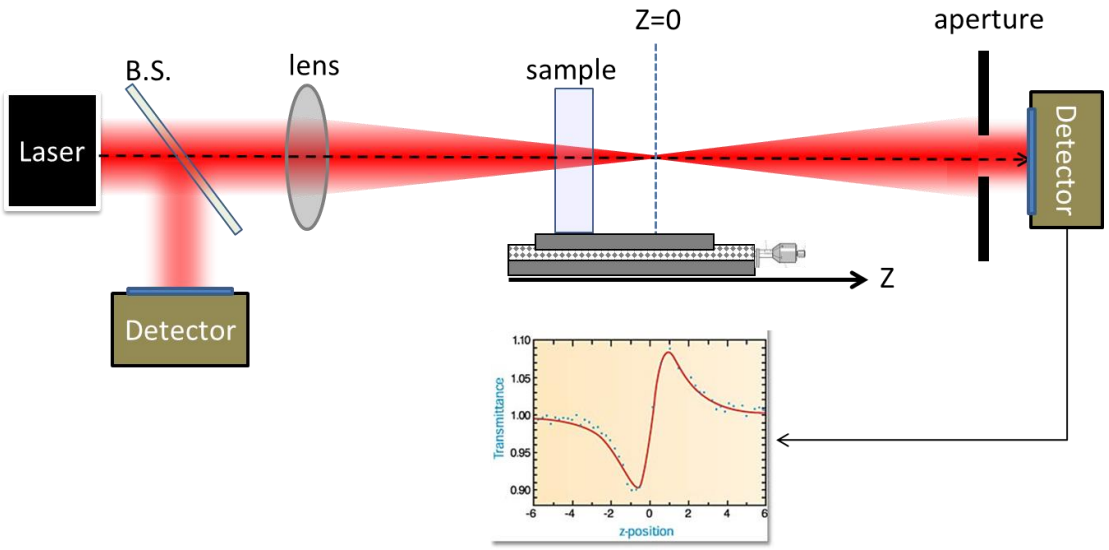
g) Measure and plot I(2ω) vs θ *(The current BBO crystal lateral dimensions may limit you in performing this experiment)*

h) Measure and plot I(2ω) vs azimuthal angle ϕ (around the c-axis)

i) For optimum value of ϕ, determine the conversion efficiency P(2ω)/P(ω) versus θ and compare it with Eq. (5). Note that in addition to Δk(θ), the effective length of the crystal also varies with ℓ as it is given by l/cos(θ') where θ' is the internal angle (use the Snell's law).

j) Measure and compare the *beam radius* and the *pulse width* for ω and 2ω beams. Explain your results.

ii) **Z-scan Experiment:**



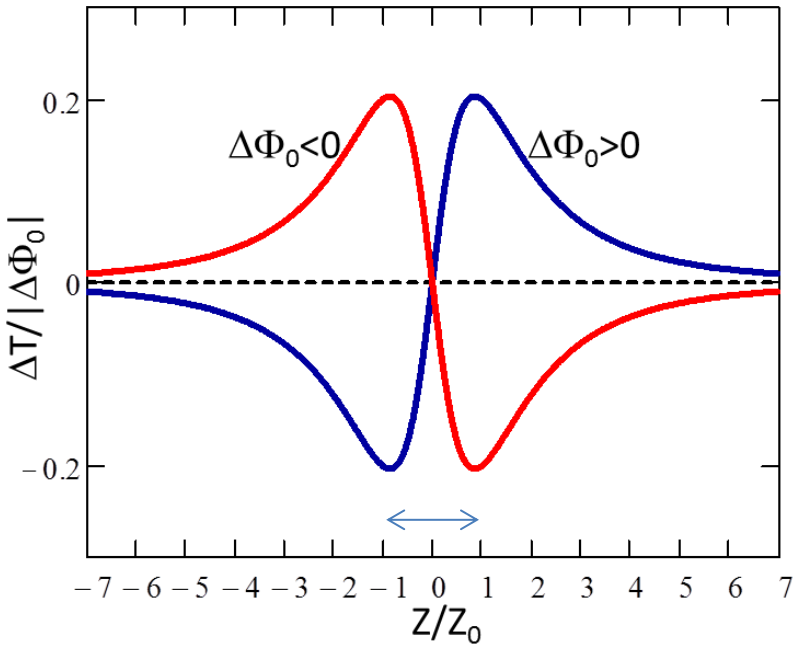
In the following experiments you will observe the effects of nonlinear absorption (NLA) and nonlinear refraction (NLR) using the Z-scan technique. You will be then able to infer the nonlinear optical coefficients and potentially other material parameters from your data. The standard Z–scan set-up is shown in figure above where we measure the far-field transmission of the laser beam through an aperture while scanning the sample along the propagation axis (z).

Nonlinear refraction causes beam distortion (self-action) but not absorption loss. If the aperture is removed or fully opened, nonlinear refraction should not cause any change of transmitted power as the sample is translated along z. In the presence of nonlinear absorption such as TPA, however, an open aperture Z-scan will reveal position-dependent nonlinear transmission as described by the nonlinear Beer's law in Eq. (9).

In a single-beam Z-scan experiment, the sample is scanned in the vicinity of a focused laser beam. The scan range is typically from z = $-6Z_0$ to + $6Z_0$ where $Z_0=\pi w_0^2/\lambda$ is the Rayleigh range with w_0 being the focal (minimum) spot size. In the presence of optical Kerr effect ($n=n_0+n_2I$), the normalized transmittance T of a thin sample ($L<Z_0$) through a partially closed far-field aperture is approximated as:

$$T(x) \cong 1 + \Delta\Phi_0 \frac{4x}{(x^2+1)(x^2+9)} \quad ,(16)$$

where $\Delta\Phi_0 = \frac{2\pi}{\lambda} n_2 I_0 L$ is the on-axis nonlinear phase shift at the focus, and $x = Z/Z_0$. The above expression is valid for $\Delta\Phi_0 < 1$, preferably ≤0.5.



The resulting behavior is shown in the above figure above for both positive and negative nonlinearities. Both the magnitude and sign of n_2 can be obtained. The signature characteristic of a Z-scan is the presence of a peak and valley. Their separation in z is obtained from Eq. (16):

$$\Delta Z_{p-v} \approx 1.7 Z_0 \quad (17)$$

The amplitude difference between peak and valley can also be obtained from Eq. (16):

$$\Delta T_{p-v} \approx 0.406 \times \Delta\Phi_0$$

The above analysis works when the aperture is nearly closed. In general, the transmission set by the aperture (S) can be anywhere in the range 0 < S < 1 and the simple formulas are no longer valid. The following empirical expression best fits the exact theoretical calculation for the Z-scan transmittance in the general case:

$$\Delta T_{p-v} \approx 0.406 \times \Delta\Phi_0 \, (1 - S)^{0.27} \quad (18)$$

When nonlinear absorption (e.g. TPA) is appreciable and accompanies nonlinear refraction, the Z-scan transmittance curves are altered by the nonlinear loss (smaller T) in the region of the focus. To quantify this, one first performs an open aperture (S=1) Z-scan. The transmitted signal is insensitive to beam distortion caused by nonlinear refraction, i.e. the Z-scan curve is only caused by nonlinear loss. The lowest order change in the open-aperture Z-scan transmittance of a Gaussian beam is then:

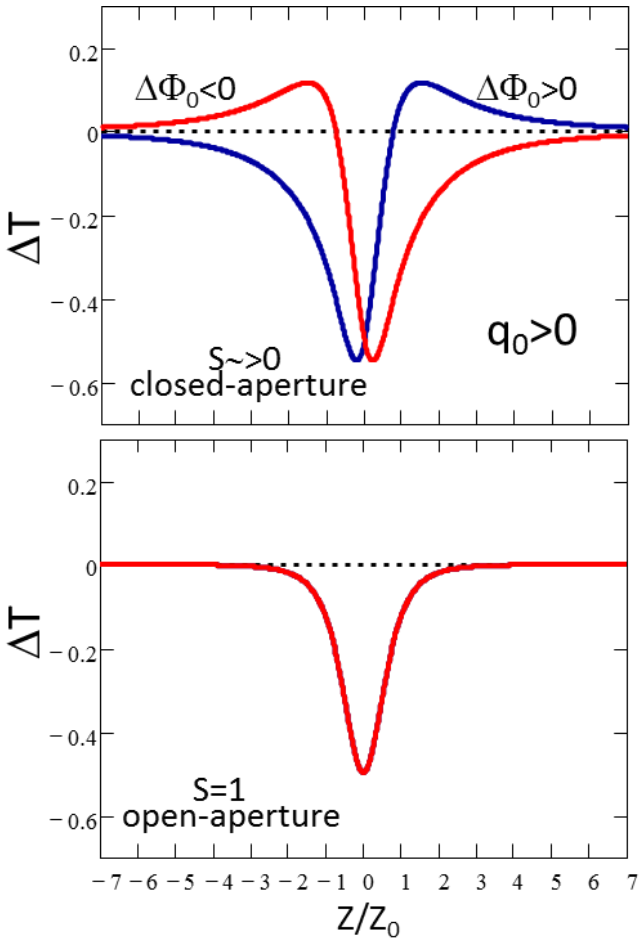
$$T(x, S=1) \cong 1 - \frac{q_0}{2(x^2+1)^2} \quad ,(19)$$

where $q_0=\beta I_0 L$. The above expression is valid for $\Delta T<15\%$. Temporal averaging (assuming Gaussian pulses), leads to $< \Delta\Phi_0 >= \Delta\Phi_0/\sqrt{2}$ and $< q_0 >= q_0/\sqrt{2}$.

It is therefore a good approximation to take a closed-aperture Z-scan transmittance as:

$$T(x, S) \cong 1 + \Delta\Phi_0 \frac{4x(1-S)^{0.27}}{(x^2+1)(x^2+9)} - q_0 \frac{1}{2(x^2+1)^2}$$

Note that by performing an open aperture (S=1) scan and then subtracting it from the closed-aperture we can retrieve (separate) the nonlinear refraction component from which Δn can be inferred. Example calculations of open-aperture and closed aperture Z-scan traces (ΔT) are given below for $q_0>0$ (eg. two-photon absorption) for positive and negative n_2.



Thermo-Optic Z-scan:

In the presence of loss from linear and/or nonlinear absorption, optical energy is deposited into the material, which in turn leads to heating. We neglect the possibility of cooling on the timescale of the experiment here, although this is not always valid. The ensuing thermal lensing becomes the dominant nonlinear refraction for CW or relatively long laser pulses and/or when the absorption coefficient is large.

CW Thermal Lensing: In the case of **CW** laser radiation, steady-state thermal lensing driven by transverse thermal heat conduction has been shown to give a Z-scan transmission trace that follows:

$$T(x) = 1 + \theta \,\text{atan}\left[\frac{2x}{3+x^2}\right], \quad (20)$$

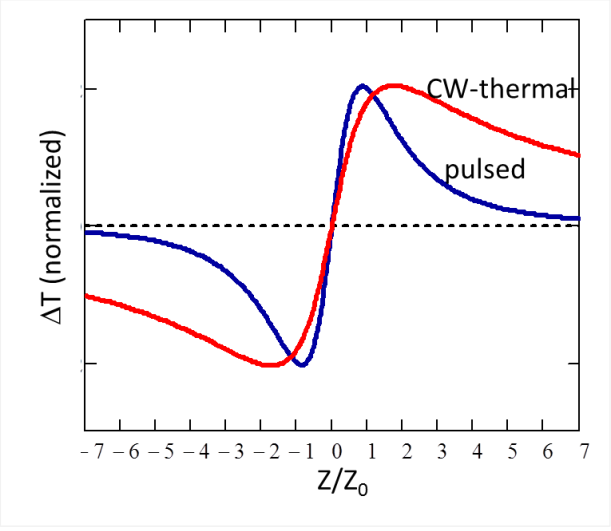
where x = Z/Z_0, and $\theta = \frac{\alpha_0 PL}{\lambda\kappa}\left[\frac{dn}{dT} + (n-1)\frac{1}{L}\frac{dL}{dT}\right]$ is the thermal induced on-axis phase shift. Note that here we also included the longitudinal thermal expansion of the material $\frac{1}{L}\frac{dL}{dT}$ that will introduce additional thermal lensing. See references below:

https://www.osapublishing.org/josab/abstract.cfm?uri=josab-19-6-1342

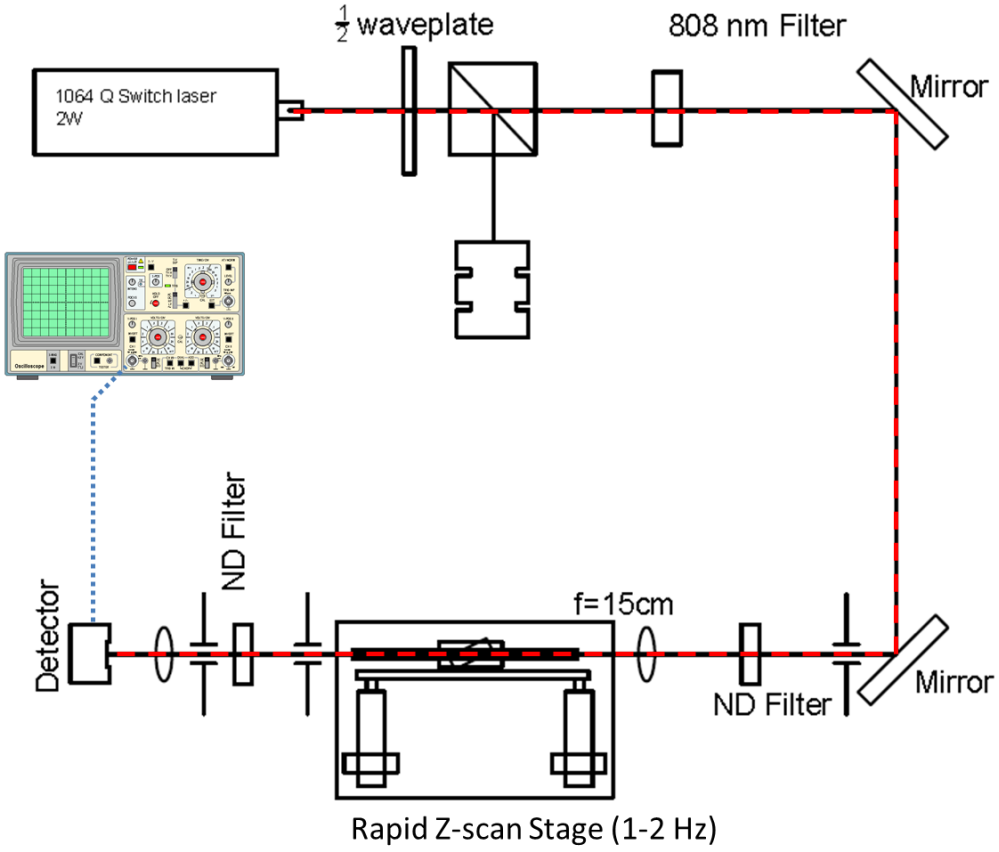
http://www.sciencedirect.com/science/article/pii/S0022309306007277

Pulsed Thermal Lensing: As described earlier, when short laser pulses are used, thermal conduction may be ignored and, as deduced from Eq. (13), we obtain $< n_2^{eff} > \approx \frac{dn}{dT}\frac{\sqrt{\pi}}{2C_v}\alpha_0 t_p$. This is the case that you most likely encounter in the lab experiments (*you should verify this*). Under this assumption, the "normal" Z-scan analysis, as given by Eq. 16-18, can be used to analyze the data by simply using $\langle \Delta\Phi_0 \rangle = \frac{2\pi}{\lambda}\frac{dn}{dT}\frac{\sqrt{\pi}}{2C_v}\alpha_0 t_p I_0 L$.

A comparison of Z-scan trances for CW-thermal and pulsed (instantaneous) nonlinearities is depicted in Fig. below:



Experiments: Rapid Z-scan Set Up:



The above set up assumes the 1064 nm laser. It is essentially the same when using the 1.55 μm laser.

A. Pulsed Thermo-Optic Z-scan:

Samples: ND-filters (absorbing) with various absorption losses. Material: BK7

-Knowing (estimating) the beam radius at the focus, find the thermal diffusion time $\tau_d \approx w^2 C_v/\kappa$ for BK7. Assure that $t_p << \tau_d$.

-The rapid-scan stage operates at .2-1 Hz. Compare the thermal diffusion time with the scan period (peak-valley scan time) and discuss the potential complications if these time-constants become comparable.

-Record Z-scan traces. Measure ΔZ_{p-v} and obtain Z_0. Compare with expected value from laser data.

B. Instantaneous Z-scan

Samples: ZnSe (polycrystalline), 3mm, CdTe (crystalline), 2mm

- Perform open and closed aperture Z-scans on both samples.
- Plot the transmittance changes (T_{min} for S=1, and ΔT_{p-v} for S=0.1) as a function of incident power (for 5 different powers).
- Deduce n_2, β, and any possibly other nonlinear coefficients defined earlier.

Useful references:

M. Sheik-Bahae and E.W. Van Stryland, Optical Nonlinearities in the Transparency Region of Bulk Semiconductors, in *Nonlinear Optics of Semiconductors*, E. Garmire and A. Kost, Eds., Volume 58 of Semiconductor and Semimetals, pp. 257-318, Academic Press (1998)

A. A. Said, M. Sheik-Bahae , D.J. Hagan, T.H. Wei, J. Wang, J. Young, E.W. Van Stryland, "Determination of Bound and Free-Carrier Nonlinearities in ZnSe, GaAs, CdTe, and ZnTe," J. Opt. Soc. Am. B. 9 , 409 (1992)

Z-Scan, E. W. Van Stryland and M. Sheik-Bahae, , in *Characterization Techniques and Tabulations for Organic Nonlinear Materials,* M. G. Kuzyk and C. W. Dirk, Eds., page 655-692, Marcel Dekker, Inc., 1998

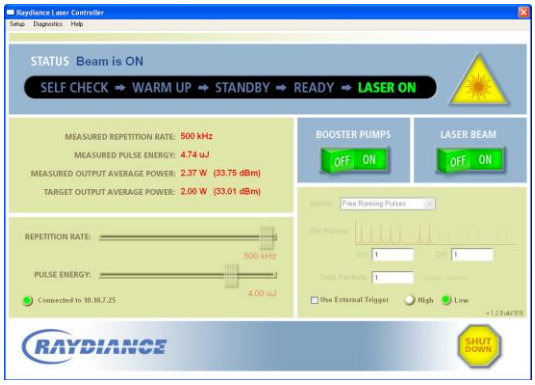
Appendix A: Operation of Raydiance Fiber Laser

The experiment uses a commercial mode-locked diode-laser/fiber amplifier made by Raydiance, now part of Coherent Inc. Key parameters are $\lambda = 1.55$ μm and pulse duration ~ 800 fs. The energy and repetition rate of the laser are adjustable and the beam is elliptically polarized.

DANGER: Although there is not enough average laser power to burn clothing, skin, or equipment, the peak power in a short pulse can permanently damage the retina. The laser used in this experiment is not eye-safe, which requires that PROTECTIVE EYEWEAR MUST BE WORN whenever the laser is on. Only approved, specially marked laser safety glasses available in the lab are acceptable. An added complication is that the beam is infrared and thus not visible. It is the responsibility of the experimenters to secure the lab and control access to it.

Turn on the ThermoCube chiller; switch can be found on the left side of the enclosure. Push the Start/Stop button. You should see – TEMP in the display. The chiller should quickly reach the 17C setpoint. If *TEMP is displayed, no cooling is occurring and the laser will overheat.

The Raydiance laser is comprised of two blue boxes. The smaller box is the oscillator and the large lower box is the amplifier. Turn on the amplifier box first; its switch is located on the upper-right back panel. Wait 30 seconds, then turn on the oscillator switch located in the same position on its rear panel.



Observe the front panel display and wait for SYSTEM OFF to appear. This will take several minutes. Next launch the Raydiance Laser System application found on the desktop. You should see WARM UP flashing and "Warming up" on the panel display. Warm-up will take several minutes, after which the system will emit a loud beep. When warm-up is complete, reach around the periscope and open the shutter by sliding the knob on the front panel to the right. Be careful not to disturb the periscope optics.

Set the desired pulse energy and repetition rate. Activate the Booster Pumps by pressing the switch on the program user interface. Wait ~ 1 minute for the system to activate and then turn the laser beam on. NOTE: The laser and booster pumps must be switched off and re-started to change the pulse energy. The shutdown procedure is the reverse of the startup.