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Analysis of beam deflection measurements in the presence of linear absorption

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Abstract: We develop a series of analytical approximations allowing for rapid extraction of the nonlinear parameters from beam deflection measurements. We then apply these approximations to the analysis of cadmium silicon phosphide and compare the results against previously published parameter extraction methods and find good agreement for typical experimental conditions.

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OCIS codes: (190.4400) Nonlinear optics, materials; (190.3270) Kerr effect.

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Introduction

Knowledge of the temporal response of the Nonlinear Refraction (NLR) and Nonlinear Absorption (NLA) of materials is key for understanding of the physical mechanisms underlying the Nonlinear Optical (NLO) properties [1]. Various experimental techniques have been developed for measuring this response [2]. Commonly used methods such as pump–probe [3, 4] provide the temporal response of the NLA, the refraction via the induced birefringence as in the Optical Kerr Effect (OKE) experiment [5] or with use of a local oscillator as with four wave mixing [6]. The Beam Deflection (BD) method was developed as a high sensitivity, easy to implement time and polarization resolved technique for simultaneous measurement of NLR and NLA [7].

Previously, techniques for analyzing BD data for instantaneous and non-instantaneous nonlinearities in the presence of Group Velocity Mismatch (GVM) have been determined [8, 9]. This method, however is limited in that it only treats materials in the undepleted excitation approximation, where the absorption is sufficiently small so that irradiance throughout the sample is constant. This is a significant limitation, especially in probing 2-D, plasmonic and metamaterials which typically have high linear and nonlinear absorption [10, 11]. These materials have come to be of interest to the NLO community for a variety of application such as photonic-electronic interconnects, all optical switching and computing and hybrid silicon photonics [12].

Additionally, depending on the spatial and temporal resolution required, this method can be very time consuming and makes the extraction of NLO parameters tedious, especially for materials with multiple mechanisms acting together. In this work we develop an analytic approach that accounts for change in the excitation throughout the sample due to linear, absorption. To demonstrate the validity of these approximations, we will compare them against previous extraction methods to show good agreement for typical experimental conditions. We then apply our approximations to measurements of Cadmium Silicon Phosphide (CSP), a material with large index dispersion and linear absorption.

1. Analysis of beam deflection data

The BD method operates by using a strong Gaussian excitation beam to generate an index change $\Delta n$ within the material as seen in Fig. 1. If the spot size of the probe is much smaller than the excitation, the probe, displaced from the peak by half the beam waist of the excitation, experiences a prism-like index gradient. This transient prism deflects the beam by some angle $\Delta \theta$. This deflection angle is in turn measured by the quadrant cell diode as a
change in the differential energy signal of the probe $\Delta E_p[x_o] = E_{p,L} - E_{p,R}$, where $E_{p,(L,R)}$ are the probe signal measured from the left and right sides of the quadrant diode and $x_o$ is the delay between the excitation and probe. This signal is normalized by the total energy of the probe $E_p[x_o] = E_{p,L} + E_{p,R}$ so that the signal $\Delta E_p / E_p[x_o]$ is proportional to $\Delta n$ [13]. Similarly, the NLA can be extracted using total energy signal $E_p[x_o]$ which can be normalized to calculate the transmission.

Following the method outlined in Reichert et al [9], we start with the propagation equation in the absence of Group Velocity Dispersion (GVD) in normalized coordinates.

$$\frac{\partial a_e[r,z,t]}{\partial z} + \rho \frac{\partial a_e[r,z,t]}{\partial \tau} + (-\sigma_e + iG[r,z,t])a_e[r,z,t] = 0$$

(1)

where $a_e[r,z,t]$ is the dimensionless field of the probe, $z = z / L$ is the normalized propagation distance, $L$ is the sample length, $\tau = t / \tau_e$ is the normalized temporal coordinate, $\tau_e$ is the excitation pulse duration, $\sigma_p = a_pL / 2$ is the normalized linear absorption of the probe, $\rho = \Delta n_p / (c\tau_e)$ is the GVM parameter, $\Delta n_p$ is the difference in the group indices of the excitation and probe, and $c$ is the speed of light in vacuum and $a_p$ is the linear absorption of the probe.

$G[r,z,t] = \int_{-\infty}^{\infty} R[\tau - \tau'] |a_e[r,z,\tau']|^2 d\tau'$ is the convolution of the nonlinear response function $R[\tau] = (\eta + i\Gamma)\tau$ and the dimensionless excitation irradiance $|a_e[r,z,\tau]|^2$. The response function $r[\tau]$ is normalized such that $\int_{-\infty}^{\infty} r[\tau] d\tau = 1$. The dimensionless nonlinear refraction and absorption parameters are $\eta = (4\pi / \lambda_0)n_2I_0,eL$ and $\Gamma = \alpha s_2I_0,eL$ where $\lambda_0$ is the probe wavelength, $n_2$ is the nonlinear index of refraction, $I_0,e$ is the peak irradiance of the excitation and $\alpha_s$ is the nonlinear absorption coefficient. For bandwidth limited Gaussian spatiotemporal profiles we have

$$a_e[r,z,\tau] = e^{-(x-x_0)^2/2} e^{-2\sigma_e}$$

(2)

$$a_p[r,0,\tau] = Ae \left( \frac{y^2 + z^2}{w^2} \right) e^{-2\tau^2}$$

(3)

where $a_e[r,z,\tau]$ is the field of the excitation, shifted so the probe is centered over the maximum gradient of the spatial envelope ($X_0 = \frac{1}{2}$), $a_p[r,0,\tau]$ is the field of the probe at the front of the sample, and $X = x / w_x$, $Y = y / w_e$ are the normalized spatial coordinates, $\sigma_e = a_eL / 2$ is the normalized linear absorption parameter of the excitation, $\sigma_s$ is the linear absorption of the excitation, $T = \tau_p / \tau_e$ is the ratio of the pulse durations, $W = w_{0,p} / w_e$ is the ratio of the spot sizes of the beams, and $w_{0,p}$ and $w_e$ are the 1 / $e^2$ spot sizes of the probe and excitation. We can solve for the field at the back of the sample $a_p[r,1,\tau]$ in the linear ($L << z_{0,p}$) and nonlinear ($L << z_{0,p} / \Delta \phi_0$) thin sample approximations where $z_{0,p}$ is the Rayleigh range of the probe and $\Delta \phi_0$ is the peak induced phase shift. Typically, $\Delta \phi_0 < < 1$ and $L$ can be selected to meet these conditions giving us
where $H[τ]$ is the integral for the pulse overlap in the sample accounting for GVM and depletion of the excitation due to linear absorption. The real part $H[τ]$ corresponds to the change in the absorption $Δα_p$ and $H''[τ]$ corresponds to the nonlinear phase accumulation $Δφ_p$.

As in previous work the signal can be calculated by Fresnel propagating $a_p[r,1,τ]$ and spatially integrating over both sides of the detector and temporally over the pulse duration. In this analysis we use the convenience that the Fresnel propagation to the detector will yield another Gaussian expanded in size, deflected by an angle $Δθ$ due to $H''[τ]$ and attenuated due to $H'[τ]$.

The complex transmission $t[r,τ]$ applied to the probe by the material response induced by the excitation is $t[r,τ] = Q[r,τ] \exp[iφ[r,τ]]$. If $W$ is sufficiently small ($W << 1$) and the deflection is small ($Δθ << 1$) we can expand the magnitude and phase transmissions as

$$
\phi[r,τ] = \tan\left[ e^{-2(x-x_0)^2} X X S H \right] = e^{-2(x-x_0)^2} X X H \approx e^{-2x_0^2} X X H \tag{7}
$$

As shown in Fig. 2, the effect of this attenuation is to make it appear as if the probe beam has been translated slightly, without distorting its shape significantly. Thus while $φ[r,τ]$ acts as the prism that deflects the probe an angle $Δθ$, $Q[r,τ]$ attenuates and slightly translates the probe yielding a small deflection signal due to NLA.

The average deflection angle of the probe is $Δθ[r,τ] = 4e^{-2x_0^2} X_H \approx (k_{0,p} w_o) \sqrt{1 + 2W^2} \left[ e^{-2x_0^2} X_H \right] R / \left[ k_{0,p} w_o \right]$, where $R = e^{-2x_0^2} X_H \approx (1 + 2W^2)^{1/2}$. With $Q[r,τ]$ and $Δθ[τ]$ we can use Gaussian beam propagation instead of Fresnel propagation to calculate the probe field at the detector a normalized distance $D = d / z_{0,ρ}$ away from the sample

$$
a_p[r,τ] = A \cdot \exp\left[ -\left( \frac{X^2 + Y^2}{W^2} \right) - \left( \frac{(ρ - τ - τ_0)^2}{2T^2} \right) - \sigma_p - e^{-2(x-x_0)^2 -2r^2} H'[τ] \right] \tag{4}
$$

$$
H'[τ] = \int_{0}^{∞} \left[ iG[0, Z, τ + ρ(-1 + Z')] \right] dZ = H'[τ] + iH^*[τ] \tag{5}
$$

$$
a_p[r,τ,τ] = 2^d A^d \frac{\exp[-2x_0^2]}{Dn!} \frac{g[t]}{Dn} \left( e^{-2x_0^2} H'[τ] \right)^d \left[ 1 + 8e^{-2x_0^2} X_0 (-X + ΔS[τ]) H'[τ] \right] \tag{8}
$$

where $ΔS[τ] = Δθ[τ] d / w_o$ is the normalized lateral beam displacement on the detector due to the deflection and $d$ is the sample – detector distance. Integrating and then re-summing the probe irradiance $|a_p[r,τ,τ]|^2$ we calculate the difference in the power between the left and right sides $ΔP[τ, τ_d]$ and the total power $P[τ, τ_d]$.
\[
\Delta P[\tau, \tau_d] = 2A^2 e^{-2\chi_0 \rho^2/\tau^2} \left( H[\tau] + H'[\tau] \right) / D \\
\]

To calculate \( \Delta E_p[\tau_d] \) and \( E_p[\tau_d] \) we integrate \( \Delta P[\tau, \tau_d] \) and \( P[\tau, \tau_d] \) over \( \tau \), which we then divide to yield the normalized signal \( \Delta E_p / E_p[\tau_d] \). The transmission \( Q[\tau_d] \) is calculated by normalizing \( E_p[\tau_d] \) by its value at some large negative delay. Because this calculation does not involve performing Fresnel propagation at every time \( \tau \) and delay \( \tau_d \), this method an order of magnitude faster, allowing for automated parameter fitting.

For an instantaneous response \( (r[t] = \delta[t]) \) with negligible GVM \( (\rho = 0) \) the overlap integrals reduce to

\[
\Delta E_p / E_p[\tau_d] = \frac{1}{2} \frac{2W}{\sqrt{\pi}} \left( \frac{2W}{\Gamma + R\eta} \right) F \\
\]

\[
F = -\frac{1 + e^{-2\sigma_e}}{2\sigma_e} \\
\]

In the case of no linear absorption \( (\sigma_e = 0, F \to 1) \), excitation much larger than the probe \( (W << 1, R \to 1) \), and negligible NLA \( (\Gamma \to 0) \), Eq. (11) reduces to the previously derived expression for the signal in transparent material [14]. Note that by setting \( X_0 = 0 \), Eq. (10) can be used to calculate the expression for the transmission in excite–probe experiments [8]

\[
Q[\tau_d] = \frac{1}{\sqrt{\pi T}} \int e^{-2\chi_0 \rho^2/\tau^2} \left( H[\tau] + H'[\tau] \right) d\tau \\
\]

Taking the ratio of the deflection signal due to the refraction \( (\Delta E_p / E_p[\Gamma = 0]) \) and absorption \( (\Delta E_p / E_p[\eta = 0]) \) we define the contamination factor \( C = (\Delta E_p / E_p[\eta = 0]) / (\Delta E_p / E_p[\Gamma = 0]) \sim 1 / D \). As we seen in Fig. 2 the effect of a small translation is reduced by the expansion of the probe as it propagates to the detector, while the translation due to the deflection scales with distance. For a typical configuration \( (D > 15) \) we can effectively set \( H[\tau] = 0 \) in Eq. (9). This makes the BD method particularly well suited for measuring materials with large NLA since the refraction and absorption signals are essentially independent of each other. This approach has been applied to BD measurements of thin film refractory metal nitrides, which possess both high linear and nonlinear absorption [15].
2. Comparisons to previous models

In Fig. 3 we compare the method from Reichert et al and our analysis for various values of $\rho$, along with our approximations compared to the Fresnel propagation method. The agreement between the Fresnel propagation and the analytical expression is very good, with a difference at the peak signal of less than 2.0%.

As seen in Fig. 4, the effect of GVM is to extend the temporal range of the signal, as one would expect as it is possible for the excitation pulse to walk entirely through the probe pulse over a wide range of delays [8]. The effect of the excitation depletion is to reduce the signal at negative delays due to the excitation catching up to the probe at the back of the sample after it has been significantly attenuated.
Fig. 4. a) Transmission and b) refraction signals for various values of linear absorption \( \sigma_e \) with \( \rho = 10 \). The simulation parameters are \( W = 0.175, T = 1.09, \eta = 0.118, I = 0, D = 16.78 \) with \( \sigma_e \) variable. Increasing \( \sigma_e \) reduces the signal at negative delay due to the excitation catching up to the probe at the back of sample, after it has been attenuated by propagation through the sample.

3. Measurements of cadmium silicon phosphide

To test the validity of our analysis, we fit measurements of CSP using both the above expressions. CSP is a II-IV-V\(_2\) chalcopyrite semiconductor with a high 2nd order nonlinear coefficient (\( d_{36} \)) of 84.5 pm/V in the mid-infrared spectrum [16] with sufficient birefringence for 2 \( \mu \)m to mid-IR wavelength conversion [17]. Grown by the horizontal gradient freeze technique, CSP shows significantly improved transparency (i.e., lower optical absorption), unfortunately combined with a somewhat lower thermal conductivity, as compared to the more established Zinc Germanium Phosphide (ZGP) [18]. These properties suggest that CSP will display a higher thermal-lensing threshold and thus enable higher power mid-IR laser output than possible with ZGP [19, 20]. Previous works have demonstrated CSP based high power femtosecond Optical Parametric Amplifiers (OPA) pumped at 1064 nm [21].

BD measurements are made using a Ti:Sapphire amplified system (KM Labs Wyvern 1000-10) producing 4.2 mJ, 35 fs (FWHM) pulses at 790 nm operating at a 1 kHz repetition rate. The strong excitation pulse is obtained by splitting off \( \sim 5 \) \( \mu \)J of the fundamental with a beam splitter. An optical parametric generator/amplifier (Light Conversion TOPAS-Prime) is pumped by the fundamental to generate the 650 nm, 55 fs probe pulses, which is then spatially filtered to produce a Gaussian irradiance profile. The probe is focused to a spot size, \( w_p \sim 3 - 5 \) times smaller than \( w_e \), both which were determined by knife-edge scans. The probe is displaced from the peak of the excitation by \( \Delta x = \frac{1}{2} w_e \) to the maximized irradiance gradient where the probe experiences an induced refractive index gradient, causing it to be deflected by a small angle \( \Delta \theta \). The deflection signal is measured using a quad-segmented Si photodiode (OSI QD50-0-SD) which simultaneously measures \( \Delta E_p[\tau_d] \) and \( E_p[\tau_d] \), each of which is detected via lock-in detection (Stanford Systems SR-830). A mechanical optical chopper (Thorlabs MC-2000) synchronized with the excitation repetition rate is used to modulate the excitation at 286 Hz.
Fig. 5. Fit of CSP data using analytic approximation for a) transmission and b) deflection. Note that the reduction in the signal between $\tau_d = 5$ and $\tau_d = 25$ is due to the depletion of the excitation. The difference in the slope of the rise and fall of the signal is due to GVD, which is not accounted for. Inset) absorption coefficient $\alpha_e$ vs. peak excitation irradiance $I_{0,e}$. Plots have been shifted vertically by 2.5% for clarity.

Figure 5 shows measurements of CSP at $\lambda_p = 650$ nm and $\lambda_e = 800$ nm. Using the expressions Eq. (9) – 10 we fit values for the nondegenerate parameters $n_2 = 115 \times 10^{-19} \text{ m}^2/\text{W}$ and $\alpha_2 = 8.8 \times 10^{-11} \text{ m/W}$, with $\rho = 33$ and values of $\alpha_e$ as shown in the inset of Fig. 5 a). The increase in the linear absorption of the excitation as a function of irradiance follows the form of an effective 2PA of the excitation modeled as $\alpha_e[I] = \alpha_0 + \alpha_2 e I$. Fitting the data we find an linear absorption coefficient $\alpha_0 = 1.22 \text{ cm}^{-1}$ and an effective 2PA coefficient of $\alpha_2 e = 0.23 \times 10^{-11} \text{ m/W}$. The extracted nonlinear parameters are on the same order ($\alpha_2 = 2.4 \times 10^{-11} \text{ m/W}$) as previously reported degenerate measurements at 1 µm [22]. The deviation of the fit from the data at negative delay is due to the GVD, which broadens the pulses as they propagate through the material, thus reducing the irradiance and the induced nonlinear effect. This effect is not modeled in order to derive an analytic solution for Eq. (1). It may be possible to determine the GVD either through ellipsometry measurements or by BD measurements of differing thicknesses of material.

4. Conclusions

We have extended the analysis of BD data to include depletion of the excitation due to linear absorption. Additionally, we have applied a series of approximations in order to arrive at analytic expressions that are much quicker to evaluate than the standard Fresnel propagation based methods. We have shown that the approximations have very good agreement with the Fresnel propagation method, with a difference of less than 2%. Lastly we have applied these expressions to the extraction of the nonlinear properties of CSP.