



Measurement of nonlinear coefficients of crystals at terahertz frequencies via High-Field THz at the FELIX FEL

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14. ABSTRACT We investigated the possibility of determining the nonlinear properties of Si, GaAs and GaSex-1Sx and other nonlinear optical crystals in the FIR and THz regimes using the classic Maker Fringe and Z-scan techniques. The Z-scan measurements were conducted successfully, in both the FIR and THz regimes, and the measurement results in the FIR were found to be in reasonable agreement with the available literature [7, 8, 9]. We can conclude that multi-photon absorption in the FIR is no more an impediment to pumping, GaSe based, nonlinear frequency conversion devices that in the near-IR.					
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Measurement of $\chi^{(3)}$ Optical Nonlinearities

in the Far Infra-Red (FIR) and THz regimes by the Z-scan method

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Introduction

We report on attempts to determine the key nonlinear optical property $\chi^{(3)}$ in the Far Infra-Red (FIR) and THz regimes for a range of semiconductor crystals by the classic Z-scan method, pumped using the Felix free electron laser.

Measurements of these properties, which are crucial for designing of efficient nonlinear optical frequency conversion sources, have to date been largely confined to around 1.064 μm . However, at longer wavelengths, where, from the Manly-Rowe relations, the efficiency of generation of THz radiation is increased, the value of n_2 has not typically been measured. Little direct data exist for the Mid Infra-Red (Mid-IR) or FIR, with the exception of measurements in and around 10 μm by means of CO₂ lasers [1]. Nothing at all exists for the THz regime. Generally the values are inferred from the system performance in the Mid-IR range, the values of nonlinear coefficients in FIR & THz regime are presumed to follow on from those. Furthermore even in well-established nonlinear materials such as Potassiumtitanylphosphate KTi₂PO₄ (KTP) and LiNbO₃ the value of n_2 varies considerably [2], likely a result of variation in the production process, as Armstrong noted "not all KTP crystals are created equal" [3].

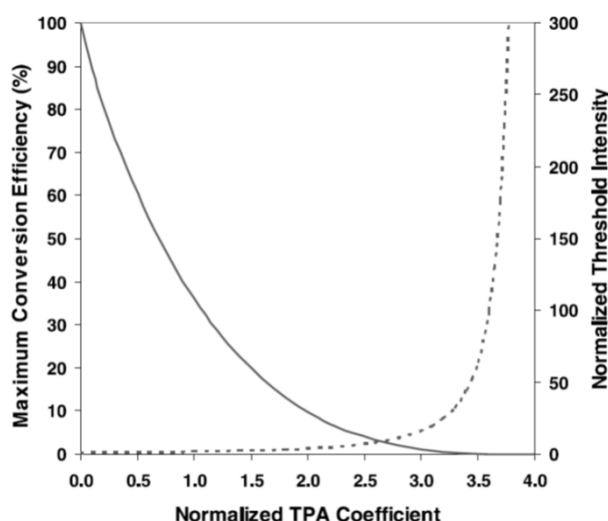


Fig. 1 The effect of two-photon absorption on the frequency conversion efficiency and oscillation threshold of a backward OPO [4]

$\chi^{(3)}$ nonlinearity, is responsible for the Kerr Effect, two-photon absorption and self-focusing, which can lead to laser induced damage of materials. As such, plays an important role in the design, operation and efficiencies of nonlinear frequency conversion sources. Zotova & Ding showed that two-photon absorption increases the threshold oscillation for OPOs and at high values prevents oscillation entirely [4], the increased threshold $I_{\text{Thres};B}$ being described by the relation:

$$I_{Thres,B} = \left(\frac{4}{4-\beta'} \right)^2 I_{Thres} \quad (1)$$

where β' is the two-photon absorption for the given pump intensity and length of the nonlinear medium and I_{Thres} is the oscillation threshold in the absence of two-photon absorption.

$\chi^{(3)}$ effects are most easily observed and measured through nonlinear refractive index n_2 via the Kerr effect. The Kerr effect is the induced change of refractive index of a material due to the effect of an intense beam of light propagating through it. The intensity dependent refractive index is defined as:

$$n = n_0 + \bar{n}_2 |E(\omega)|^2 \quad (2)$$

where n_0 is the linear refractive index, $\bar{n}_2$ is the time averaged nonlinear refractive index and $E(\omega)$ is the incident electric field. The imaginary component of nonlinear refractive index, i.e. nonlinear or multi-photon absorption, has been widely studied at $1.064 \mu\text{m}$ [5, 6, 7, 8, 9] in order to determine the performance of ns and ps Nd:YAG pumped OPOs and other nonlinear schemes [10]. Little work is to be found relating to the real component of nonlinear refractive index n_2 and in some case, the authors designed the experimental set up to negate its observation [5] or failed to report the results [6].

Measurement of the nonlinear coefficients in the THz regime has, without doubt, been hampered by the lack of suitable sources. The pump source for such measurements has several requirements. The laser should be of high power to enable the measurement of nonlinear phenomena. It is desirable that the source laser is short pulsed to achieve high peak power while reducing the impact of heating on the crystal and, hence, the resulting thermal lensing that may distort the measurements. Tunability and narrow linewidth are required to select a number of pump wavelengths while avoiding absorption features that would negatively effect the measurement. Currently, only free electron lasers are the source that can readily meet those requirements in the THz range, see Fig 2.

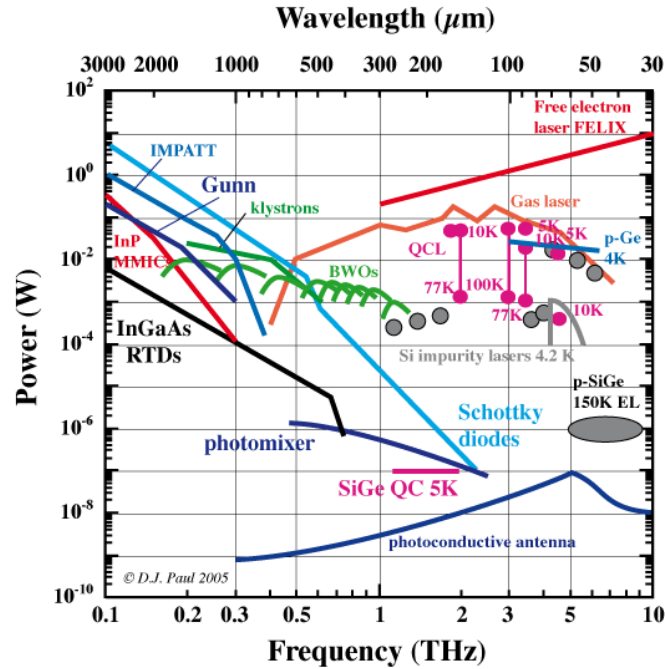


Fig. 2 Power and tunability of the Felix free electron laser at Radboud University, Nijmegen, the Netherlands, compared against other current THz sources [5].

FELIX is a free electron laser at Radboud University, Nijmegen, the Netherlands; run as a European user facility it provides continuously tunable sub-picosecond pulse of radiation from 3 to 1500 μm , and peak power of up to 100 MW, to user in the fields of Physics, Chemistry and Medicine. FELIX is one of the few available light sources that is suitable for conducting nonlinear optical experiments in the THz regime. While station staff are on run the light source, but users must design, build and conduct their own experiments.

To access beam time on FELIX, we submitted proposals in a competitive process to “*determine the nonlinear coefficients of a range of semiconductor and nonlinear crystals.*” Following review of our proposal by the FELIX’s external Programme Advisory Committee, we were awarded two sets of beam time, each 4 x 8 hour shifts, on the FELIX and FLARE instruments. Following the analysis of the data from these initial experiments we were able to redesign our experimental procedures to control sources of noise and improve the results. A further proposal was then submitted including these improvements and following review we were again awarded of two sets of beam time again each 4 x 8 hours. An outline of the work conducted is presented below.

$\chi^{(3)}$ Nonlinearity

As we have seen above in Eqn (2) the refractive index of a material n can be altered by an intense beam of light propagating through it. The measured value of the nonlinear refractive index can be used to derive the value of $\chi^{(3)}$. Measurements of n_2 can be performed in a number of ways such as by optical phase conjugation and optical Kerr gating. However, by far the simplest and most straightforward to analyse method is the Z-scan technique developed by Sheik-Bahae *et al* in their classic 1989 paper and whose method we will follow and summarise below [11, 12]. Z-scan allows the magnitude and sign of $\chi^{(3)}$ to be measured, with a relatively straightforward set up and not overly involved interpretations [12].

In the Z-scan technique, a Gaussian laser beam is brought to a focus while passing through the nonlinear sample of interest as shown in Figure (A). The light then passes through an aperture placed in the far field at z_{ap} , where $z_{\text{ap}} \gg z_R$, and $z_R = \frac{\pi \omega_0^2}{\lambda}$ is the Rayleigh range, sometimes called the diffraction length. ω_0 is the $\frac{1}{e^2}$ beam radius at the focal point. Typically, the aperture limits the transmission of the beam S to 10% - 50% [12]. The sample to be measured is moved along the beam axis, through the focus and the transmitted radiation recorded as a function of z , the distance to the focal plane. As the sample moves along the z -axis, the intensity of the incident radiation increases in a known fashion, and, at high intensities, a change in refractive index is induced. The change in refractive index alters the beam propagation and more or less radiation is transmitted through the aperture.

In order to improve the accuracy of the results, it is important to remove the measurement background. In these measurements, the background was removed by recording the transmission of the circular sample holder as a function of distance from the focal plane (z) and dividing the measurement data by the result. The laser power was monitored on a separate channel to compensate for pulse to pulse variation.

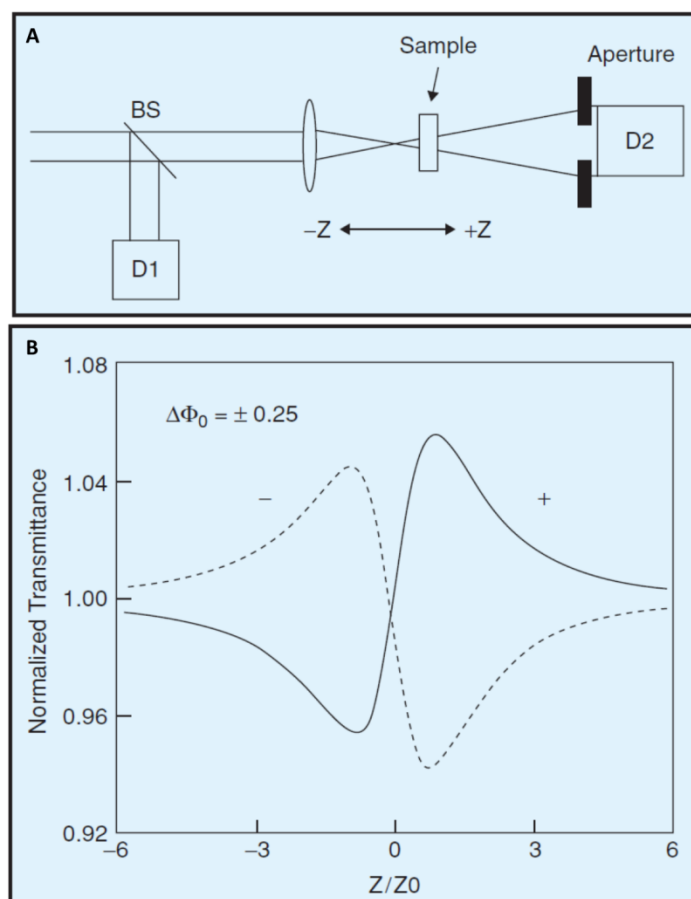


Fig. 3 (A) Sheik-Bahae's experimental set-up for Z-scan measurements, (B) Calculated Z-scan transmission for both positive and negative 3rd order nonlinearity [11]

The presence of strong nonlinear absorption prevents the accurate determination of n_2 . Two-photon absorption dominates the nonlinear absorption as typically $\alpha_{2PA} \gg \alpha_{3PA}, \alpha_{4PA} \dots$. The effect of the nonlinear absorption, β can be mitigated by performing an open aperture scan [11]. The sample is placed in the beam and transmission recorded as it is scanned along the z-axis. The aperture is opened or removed so that the beam transmission is uninhibited and all radiation is collected. Having performed open aperture scans to remove the distortion of nonlinear absorption on the n_2 measurement, we may also find the third order nonlinear absorption coefficient β . The radiation induced absorption leads to a change in the transmitted radiation incident on the detector $\Delta T(z)$.

Experimental Setup

The experimental set up is depicted schematically below in Fig. 4. After the FELIX output port, a beam splitter was installed to pick off a portion off the beam to monitor laser performance. The light was focused on to a pyro-electric detector and the laser power recorded in parallel with measurements. A HeNe laser propagates collinearly with the FELIX beam to ease the processes of alignment.

A clean Gaussian beam is desired for simple analysis of the results. To achieve a good beam profile the available FELIX beam was focused to a tight spot using an Off-Axis Parabolic mirror (M3). The beam profile is then spatially filtered using a pinhole, slightly smaller than the first minimum of the diffraction limit point spread function, transmitting approx. 90% of the incident energy. The pinhole was mounted on a xyz translation stage to allow precise alignment at the focal point. The beam was

then re-collimated by another OAP mirror (M4). In the measurement position between OAP mirrors M6 and M7, the beam is brought to a focus.

The samples were mounted on top a xyz translation stage to enable placement of the samples in the focus of the beam and translation along the axis of propagation between mirrors M6 and M7. The transmitted radiation was re-collimated by M7 and focused on a pyro-electric detector, by OAP mirror (M8).

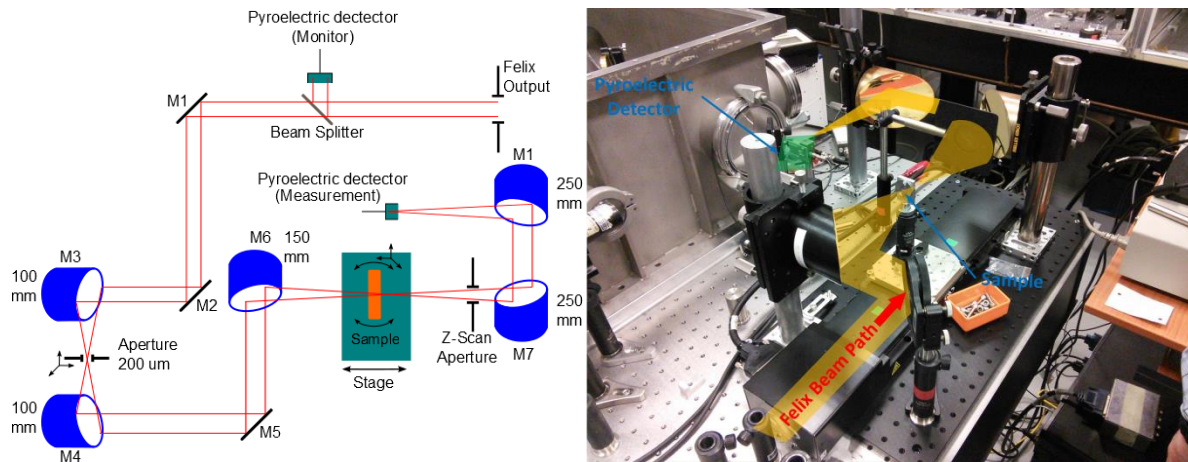


Fig. 4 Schematic & Photo of the experimental setup for Z-scan measurements.

The FELIX instrument was run in 20 MHz mode. The macropulse energy E_{macro} is dependent on the operating wavelength of the laser, measured at the sample position $E_{\text{macro}} = 0.7 \text{ mJ}$ for $\lambda = 20 \text{ }\mu\text{m}$ and $E_{\text{macro}} = 0.15 \text{ mJ}$ for $\lambda = 87 \text{ }\mu\text{m}$.

Measurements

A range of semiconductor crystals including Si, GaAs and GaSe were measured by the Z-Scan method at $20 \text{ }\mu\text{m}$ and $87 \text{ }\mu\text{m}$ using the FELIX instrument. It is necessary to have sufficiently large crystals so that the beam may pass through the crystal without been clipped, preferably for a distance $z > 5Z_R$ [12]. The crystals should also be thin, with the crystal thickness $l \leq Z_R$ though it is possible to use thick crystals with a more complex analysis. Example results are presented for $\text{GaSe}_{x-1}\text{S}_x$ $x = 0.11$, at $\lambda = 20 \text{ }\mu\text{m}$.

A strong broad induced increase in the transmission curves is observed for all crystals at both $\lambda = 20$ and $87 \text{ }\mu\text{m}$. When normalised to the transmission profile of the sample holder this broad increase is found to have nominally a Gaussian profile. We assumed it to be the result of thermal lensing in the crystals [11] and remove it in data processing, using a Gaussian fit as shown in Fig. 5.

With the background and the effect of thermal lensing removed we are left with the multiphoton absorption curves, as presented in Figure 6, which is fitted with a Lorentzian function [14]. By measuring the depth of the absorption peak, we may calculate the third order nonlinear absorption coefficient β .

To obtain the nonlinear refractive index, we divide the normalised crystal transmission for closed aperture by the normalised crystal transmission for the open aperture; the resulting curves for nonlinear refractive index are presented below in Figure 6.

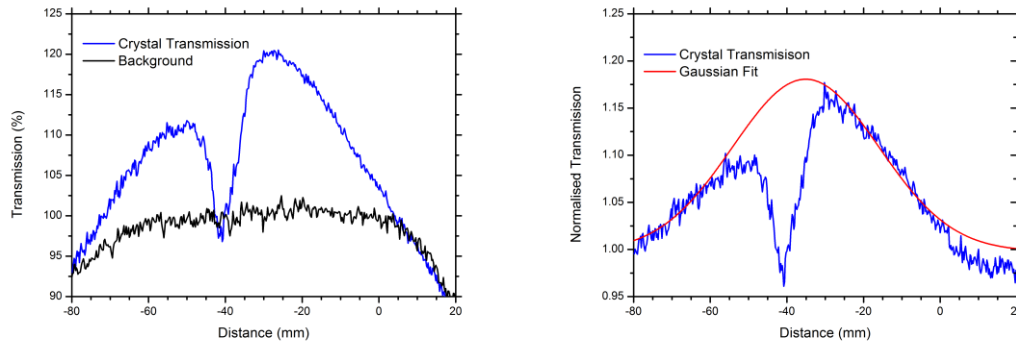


Fig. 5 The effect of thermal lensing on crystal transmission, and normalised transmission with background removed, in the presence of thermal lensing.

By inspection we can see that the sign of the change in refractive index is positive, the transmission first decreases before the focus and increases after in all samples, in keeping with Sheik-Bahae's results depicted in Figure 6 [13, 14]. This is analogous to placing a thin positive lens in the beam path; in advance of the beam focus, the additional focusing of thin lens increases divergence in the far field, while after the focus the thin lens collimates the beam, increasing transmission [13]. By measuring the peak-to-valley change in transmission, ΔT_{pv} , the magnitude of the nonlinear refractive index, n_2 can be calculated, and hence obtain of $\chi^{(3)}$

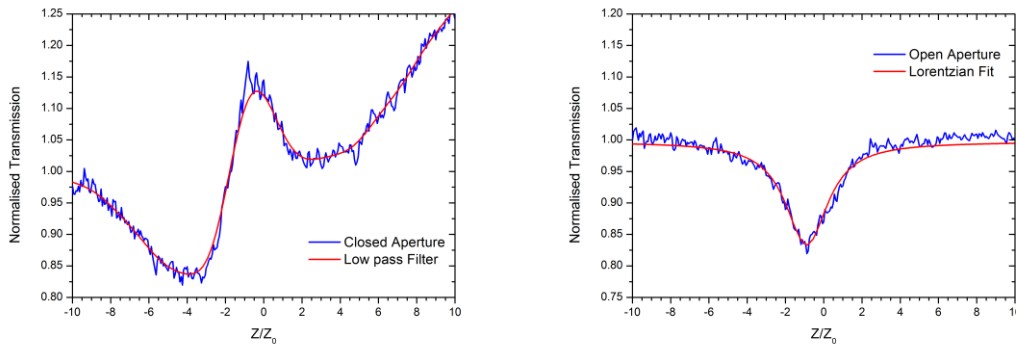


Fig. 6 Normalised transmission through the crystal samples for both closed and open aperture Z scan measurements, showing the effect of nonlinear refractive index (n_2) and nonlinear absorption (β) on the normalised transmission of $\text{GaSe}_{x-1}\text{S}_x$, $x = 0.11$, at $\lambda = 20 \mu\text{m}$.

Conclusion

We investigated the possibility of determining the nonlinear properties of Si, GaAs and $\text{GaSe}_{x-1}\text{S}_x$ and other nonlinear optical crystals in the FIR and THz regimes using the classic Maker Fringe and Z-scan techniques. The Z-scan measurements were conducted successfully, in both the FIR and THz regimes, and the measurement results in the FIR were found to be in reasonable agreement with the available literature [7, 8, 9]. We can conclude that multi-photon absorption in the FIR is no more an impediment to pumping, GaSe based, nonlinear frequency conversion devices that in the near-IR. The results of this work will be published in a peer reviewed, English language, journal.

References

- [1] K. Kato and N. Umemura, "Sellmeier equations for GaS and GaSe and their applications to the nonlinear optics in $\text{GaS}_x\text{Se}_{(1-x)}$," *Optics Letters*, vol.36, pp. 746-7, Mar 2011.
- [2] K. Allakhverdiev, F. Ismailov, L. Kador, and M. Braun, "Second-Harmonic Generation in GaS Crystals," *Solid State Communications*, vol.104, no.1, pp. 1-3, 1997.
- [3] G. B. Abdullaev, K. R. Allakhverdiev, R. K. Nani, E. Y. Salaev, and M. M. Tagyev, "Neutron diffraction, infrared, and Raman scattering investigations of the layered $\text{GaS}_x\text{Se}_{1-x}$ system," *physica status solidi (a)*, vol.53, no.2, pp. 549-555, 1979.
- [4] I. Shoji, T. Kondo, and R. Ito, "Tutorial review Second-order nonlinear susceptibilities of various dielectric and semiconductor materials," *Optical and Quantum Electronics*, vol.34, pp.797-833, 2002.
- [5] D. J. Armstrong, W. J. Alford, T. D. Raymond, and A. V. Smith, "Absolute measurement of the effective nonlinearities of KTP and BBO crystals by optical parametric amplification," *Applied Optics*, vol.35, no.12, pp.2032-40, 1996.
- [6] I. B. Zotova and Y. J. Ding, "Optical parametric oscillators in the presence of strong two-photon absorption for extended applications of nonlinear optical materials," *Optical Communications*, vol.198, November, pp.453-458, 2001.
- [7] K. Vodopyanov, S. Mirov, V. Voevodin, and P. Schunemann, "Two-photon absorption in GaSe and CdGeAs_2 ," *Optics Communications*, vol. 155, October, pp. 47–50, 1998.
- [8] M. Yuksek, A. Elmali, M. Karabulut, and G. M. Mamedov, "Switching from negative to positive nonlinear absorption in p-type 0.5 at% Sn doped GaSe semiconductor crystal," *Optical Materials*, vol.31, no.11, pp.1663-1666, 2009.
- [9] M. Yuksek, A. Elmali, M. Karabulut, and G. M. Mamedov, "Nonlinear absorption in undoped and Ge doped layered GaSe semiconductor crystals," *Applied Physics B: Lasers and Optics*, vol.98, no.1, pp.77-81, 2010.
- [10] V. Petrov, G. Marchev, P. G. Schunemann, A. Tyazhev, K. T. Zawilski, and T. M. Pollak, "Subnanosecond, 1 kHz, temperature-tuned, noncritical mid-infrared optical parametric oscillator based on CdSiP_2 crystal pumped at 1064 nm," *Optics Letters*, vol.35, no.8, pp.1230-1232, 2010.
- [11] M. Sheik-Bahae, A. A. Said, and E. W. Van Stryland, "High-sensitivity, single-beam n_2 measurements," *Optics Letters*, vol.14, no.17, p.955, 1989.
- [12] W. N. Herman and L. M. Hayden, "Maker fringes revisited: second-harmonic generation from birefringent or absorbing materials," *Journal of the Optical Society of America B*, vol.12, no.3, p.416, 1995.
- [13] L. Kador, M. Braun, K. Allakhverdiev, and E. Salaev, "Second-harmonic generation in GaSe crystals investigated with the Maker fringe technique," *Optics Communications* vol. 143, November, pp. 62-66, 1997.
- [14] E. W. Van Stryland and M. Sheik-Bahae, *Z-scan measurements of optical nonlinearities*, Ch.3, pp.655-692. Marcel Dekker, 1998.