Characterization of optical third-order non-linearities by prism coupling and pulse shape analysis on a ps timescale

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Abstract

Materials with an intensity dependent index of refraction and absorption coefficient—third-order optical non-linear (ONL) effects—offer the possibility of all-optical signal processing. Prism coupling is a well-known tool to investigate the intensity dependent refractive index, however, such experiments are often obscured by thermal effects. To avoid these we have studied the influence of the ONL effects on the shape of 70 ps pulses in non-linear prism coupling. The full width at half maximum (FWHM) of the in and out coupled pulses is compared simultaneously with the FWHM of the pulses of a reference beam. By measuring at various angles of incidence around that for optimal coupling it is possible to measure the sign and value of the change in both the refractive index and absorption coefficient. As a function of the coupling angle, the first leads to an asymmetric line shape for the ratio of the two FWHMs mentioned above, whereas the second leads to a symmetric one. From a comparison of simulations with experimental data, the values of the non-linear constants can be derived.

Introduction

There is a variety of methods to characterize the third-order optical non-linear coefficient, which describe the intensity dependent refractive index and absorption coefficient: the intensity dependent complex refractive index. We mention four wave mixing, pump and probe techniques and interferometric methods. The induced changes in the optical constants may have several physical origins depending on the material properties. Mostly various effects compete which can often be separated on their different response in time. Prism coupling is a well-known tool to investigate the non-linearity but is very sensitive to thermal effects. Not only does the thermo-optical effect influence the process, but also the coupling structure itself changes due to expansion of the layers. To be sure that only the fast electronic effect is measured we use 70 ps pulses in a prism coupling setup and look at the influence on the pulse shape in the time domain. Thermal effects can be ruled out as the time constants of the thermal effects are much larger than the pulse width [1].
Theory

With the aid of simulations one is able to relate the measured change of the full width at half maximum (FWHM) of the pulses to values of the real and imaginary part of $\chi^{(3)}$. We have to distinguish between a change in the refractive index and in the absorption coefficient. The first influences the phase matching in the coupling process, which means that it has to be described with the theory of the non-linear prism coupler. The influence of an intensity induced absorption change depends on the propagation distance in the non-linear layer structure, which is essentially the distance between in and output coupler.

**Influence of non-linear refraction**

The theory of (non-linear) prism coupling of light into a planar structure has been described in refs. 2 and 3. Assuming that neither the modal field shape nor the coupling constants depend on the intensity of the modal field, the coupling process can be described by

$$\frac{dA_m}{dz} = -k_0(K_m + \beta_i)A_m + t_mK_mk_0E_0 \exp\{i(\Delta\beta z - \varphi_r)\}$$  \hspace{1cm} (1)

The amplitude of the modal field, $A_m$, is related to the electric modal field, $E_m$, by

$$E_m(x, z) = A_m(z)\xi_m(x) \exp(-i\beta_mz)$$ \hspace{1cm} (2)

The field shape $\xi$ is defined such that $|A_m|^2$ represents the power of the mode per unit length in the $y$ direction. The modal absorption is given by $\beta_i$, $K_mk_0$ and $t_mK_mk_0$ are the output and input coupling coefficients. The mismatch of the propagation constants of the incoming light parallel to the $z$ axis, $\beta_0$, and the mode $m$, $\beta_m$, is given by $\Delta\beta: \Delta\beta = \beta_0 - \beta_m$. The change in the real part of the refractive index is described with the non-linear phase shift $\varphi_r$, which is the integral over $z$ of the change in the real part of the propagation constant $\text{Re}\{\Delta\beta_{NL}\}$. This change is, for TE modes, related to $\chi^{(3)}$ and the electric field [3, 4] in the following way

$$\Delta\beta_{NL} = \frac{\omega}{4P} \int 3\varepsilon_0\chi^{(3)}(-\omega; \omega, \omega - \omega |E_y(\omega)|^4 \, dx$$ \hspace{1cm} (3)

where $P$ is the power of the mode. The coupling efficiency can now be calculated by numerical integration of eqn. (1). To obtain a large effect on the non-linear phase shift the interaction length and coupling efficiency need to be large. This means that the coupling should be weak with, for a good coupling efficiency, a large waist of the incoming beam. The non-linearity is assumed to be instantaneous and diffusion effects are not taken into account.

In the case of a negative non-linear coefficient an increase of power will decrease the effective index of the waveguide. If the incoming beam
is tuned to an effective refractive index just a little below that of the waveguide, the coupling efficiency will increase with power. This results in a decrease of the FWHM of the pulses as the effect is the largest in the peak of the pulse. Tuning at an effective index a little bit above the waveguide index leads to an increase of the FWHM.

**Influence of non-linear absorption**

The influence of an intensity dependent absorption coefficient increases with the propagation distance of the mode. The attenuation can be written as

\[
\frac{d |A_m|^2}{dz} = -2(\beta_i |A_m|^2 + n''_2 |A_m|^4)
\]

We use this equation only in the limit where the absorption change is small compared to the linear absorption coefficient. The non-linear coefficient \( n''_2 \) is related to \( \text{Im}\{\chi^{(3)}\} \) with eqn. (3) by \( n''_2 = \text{Im}\{\Delta\beta_{NL}\}/P \). Solving eqn. (5) we obtain

\[
|A_m(z)|^2 = \frac{\beta_i |A_m(0)|^2}{(\beta_i + n''_2 |A_m(0)|^2) \exp(2\beta_i z) - n''_2 |A_m(0)|^2}
\]

As the pulse corresponds to an intensity distribution, the change in pulse width can be related directly to the value of the imaginary part of \( \chi^{(3)} \) under the condition that the linear attenuation of the mode is known.

**Experimental**

In the experiment, pulses that are coupled in and out of the non-linear layer structure are simultaneously compared with undisturbed pulses from the same source. To do this part of the laser beam is split off while most is coupled into the non-linear layer structure. The excited mode is coupled out and both beams are combined and detected as shown in Fig. 1. We used a fast detector, 10 GHz bandwidth and a 50 GHz sampling scope. Because the path lengths of the beams differ the pulses can be detected separated in time. The measurements are performed with a mode-locked Nd:YLF-laser at 1313 nm having 70 ps pulses with a repetition rate of 76 MHz and peak power of 600 W. The layer structure consists of an optically non-linear substrate with a linear PECVD-deposited SiO\(_x\)N\(_y\) guiding layer. For the non-linear material we used semiconductor doped glass, Schott RG 850.

To characterize the linear structure, first the refractive index of the RG 850 substrate is determined by measuring the critical angle for refraction from the prism to the glass substrate. The SiON layer is characterized with the prism coupling method. It is important that the simulated shape of the coupling curve is well fitted to the measured one.
Fig. 1. Experimental setup. In the circle the refractive index and the thickness in μm of the layered structure are given, respectively.

Using a one parameter fit of the coupling strength with the gap thickness, a good agreement between experiment and simulations can be obtained. The structure is shown in the circle in Fig. 1. The laser beam is focused in the y direction by a cylindrical lens to 45 μm. The coupling efficiency of the TE₀ mode is determined to be 16% and the attenuation is 4 dB/cm. The distance between the prism is 14 mm. The peak power of the incoming pulses is 160 W and their FWHM about 70 ps. The experimental results are given in Fig. 2. With the described models we obtain

\[
\chi^{(3)} = -4 \times 10^{-18} - i2 \times 10^{-20} \text{ (m/V)}^2
\]

This represents a negative change of the refractive index and a decrease of the absorption coefficient.

Fig. 2. (a) Measured and (b) simulated relative pulse width change as a function of the effective index of the incoming beam. The experimental result is decomposed into a symmetrical and asymmetrical part.
Discussion

We found a decrease of the refractive index and absorption coefficient with intensity, which is what may be expected for the electronic non-linearity of a semiconductor at wavelengths above the gap wavelength [5]. The non-linearity of semiconductor doped glasses for wavelengths around the gap is extensively investigated and found to be in the order of $10^{-18} \text{(m/V)}^2$. Cotter and Manning [6] found, like ourselves, a large non-linearity at 1064 nm of $|\chi^{(3)}| = 10^{-20} \text{(m/V)}^2$, with $\text{Re}\{\chi^{(3)}\}/\text{Im}\{\chi^{(3)}\} = 9$ and a relaxation much faster than 75 ps. For the real part of the non-linearity we find an even larger coefficient. However it is difficult to reproduce the results on the same sample, probably due to darkening of the sample [7]. The non-linearity of a darkened sample is reduced by about a factor of 20. On behalf of this technique we conclude that the experiment is simple because only one beam needs to be coupled into the guiding wave structure, however a reference beam is needed for reliable measurements. There will only be an effect on the pulse shape if the non-linearity is fast, so slow thermal effects are cancelled out. The time dependence of the non-linearity cannot be measured, only an upper limit can be given. For ONL organic polymers, which are promising for device applications [8], the time response of the electronic non-linearity is much faster than the pulse width of the mode-locked pulses. Both a change in refractive index and absorption coefficient can be measured this way.

Acknowledgements

We thank A. Hollink for technical support. This investigation was supported by the Dutch Foundation for Fundamental Material Research (FOM) and the Innovative Research Program of the Dutch Ministry of Economic Affairs (IOP).

References