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Kerr induced limit of effective interaction length of photoinduced SHG

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A new theoretical model for the length dependence of second-harmonic generation in fibres is presented. The model assumes the interaction of non-monochromatic (pulse) radiations in fibres. The origin of second-order susceptibility is described by means of the general phenomenological formula. It is shown that the effective interaction length is restricted mainly due to self-phase and cross-phase modulation effects. The theoretical predictions for the effective interaction length are found to be in a very good agreement with the experimental results.

The discovery of self-organized second-harmonic generation (SHG) is doped-glass fibres [1], [2] has attracted much attention because in theory all such second-order nonlinear optical processes in media possessing centrosymmetric structure are disallowed. The importance of SHG in doped glasses increases due to the possibility to develop cheap miniature nonlinear elements which could replace the conventional nonlinear crystals and could also be utilized in optoelectronic devices.

The fibres are usually prepared for self-organized SHG by launching both the fundamental and SH rediations. Their interaction causes "writing" of second-order susceptibility ($\chi^{(2)}$) grating into fibre. The SHG can be observed during subsequent "reading" process when only fundamental radiation is launched. The effective self-organized SHG can be observed for limited fibre lengths only, being of the order of 10 cm.

On principle, the following factors can limit the effective fibre length.

i) *Walk-off effect* occurs at the interaction of optical pulses due to the dispersion of group velocities [3]. For most experiments the separation of fundamental and SH pulses occurs at the distance $l \gtrsim 1$ m and thus the walk-off effect seems not to be the dominant factor that limits effective fibre length.

ii) *Kerr nonlinearities* cause the phase modulation of interacting waves [3]. In the picture of monochromatic wave interaction, if considering the $\chi^{(2)}$ writing and reading processes with the same pumping intensity, the same Kerr nonlinearities occur in both processes. They contribute to the total phase mismatch and they cause a small variation of $\chi^{(2)}$ grating period, but the total efficiency of self-organized SHG remains unchanged in this case.

In experiments, non-monochromatic pulse-form radiations are used for conditioning fibres or bulk glass samples to SHG. Owing to the non-uniform spectral distribution of light intensity, there is a frequency dependent Kerr-induced phase mismatch, which causes the different wavelengths to be converted with various efficiencies and phase. The phase mismatch is now intensity dependent, which leads to a gradual fuzzing of the $\chi^{(2)}$ grating and limits the effective interaction length [4], [5].

iii) *Phase fluctuations* between the pump and SH radiation during the writing process can drastically affect the efficiency of self-organized SHG [6] and shorten the effective fibre length.

iv) *Frequency fluctuations* in the pump laser beam cause a drift of the central frequency emission during the writing process, which would induce various $\chi^{(2)}$ gratings with different periods [7]. This effect leads to the fuzzing of the resulting $\chi^{(2)}$ grating and, consequently, to the shortening of the effective fibre length.

v) *Competition of secondary and tertiary $\chi^{(2)}$ gratings* being formed due to the $\pi/2$ phase shift of the SH writing radiation and SH radiation generated is believed to cause a multiple grating interference effect that reduces the total efficiency of self-organized SHG [8].

vi) *Saturation of second-harmonic generation* limits the maximum conversion efficiency [9], [10] and can be used to explain the length dependence of SHG [9].

The influence of none of the limiting factors mentioned above on effective fibre length has been studied more deeply yet.

The aim of this paper is to introduce a new phenomenological model of $\chi^{(2)}$ grating formation and $\chi^{(2)}$ grating reading by optical pulses that involves the Kerr nonlinearity effect and the walk-off effect.

The interaction of strong fundamental and weak SH optical pulses propagating in a quadratic dispersive medium, including Kerr nonlinearities, is described in the approximation of nondepleted pump by means of two partial differential equations

$$\frac{\partial E_\omega}{\partial z}+\frac{1}{v_{g\omega}}\frac{\partial E_\omega}{\partial t} = -i\left(\frac{\mu_0}{\varepsilon_0}\right)^{1/2}\frac{\omega}{2n_\omega}\chi_{\mathrm{eff}}^{(3)\mathrm{self}}E_\omega E_\omega^* E_\omega, \tag{1}$$

$$\frac{\partial E_{2\omega}}{\partial z}+\frac{1}{v_{g2\omega}}\frac{\partial E_{2\omega}}{\partial t} = -i\left(\frac{\mu_0}{\varepsilon_0}\right)^{1/2}\frac{\omega}{n_{2\omega}}[\chi_{\mathrm{eff}}^{(2)}(t,z)E_\omega^2\exp(i\Delta kz)+\chi_{\mathrm{eff}}^{(3)\mathrm{cross}}E_\omega E_\omega^* E_{2\omega}] \tag{2}$$

where $E_\omega = E_\omega(t,z)$ and $E_{2\omega} = E_{2\omega}(t,z)$ are slowly varying complex field amplitudes, ε_0 is the electric permittivity and μ_0 – magnetic permeability, n_ω and $n_{2\omega}$ are the refractive indices of fundamental and SH waves, ω is the frequency of fundamental field, $\chi_{\mathrm{eff}}^{(2)}(t,z)$ is the effective quadratic susceptibility, $\chi_{\mathrm{eff}}^{(3)\mathrm{self}}$ and $\chi_{\mathrm{eff}}^{(3)\mathrm{cross}}$ are the effective third-order susceptibilities describing the self-phase and cross-phase modulation, respectively, and $\Delta k = k_{2\omega}-2k_\omega$ represents the wavevector mismatch (k_ω and $k_{2\omega}$ are the wavevectors of fundamental and SH wave).

Equations (1) and (2) are solved in two separate cases. i) For the writing process we suppose that fundamental and SH radiations do not interact together by means of $\chi^{(2)}$ grating so that $\chi_{\mathrm{eff}}^{(2)}(t,z) = 0$ in Eq. (2), and we can obtain analytical solutions E_ω and $E_{2\omega}^{\mathrm{write}}$. ii) During the reading process we suppose time-independent $\chi^{(2)}$ grating, $\chi_{\mathrm{eff}}^{(2)}(t,z) = \chi_{\mathrm{eff}}^{(2)}(z)$. The solution of Eq. (1) is the same as in the previous case. The solution $E_{2\omega}^{\mathrm{read}}$ of Eq. (2) can be found numerically.

Now we need an equation describing the $\chi^{(2)}$ writing process. The usual approach is to suppose that $\chi^{(2)}$ is created by mixing fundamental and SH wave through the $\chi^{(3)}$ nonlinearity [11]

$$\chi^{(2)}_{\rm eff} \propto \chi^{(3)}_{\rm eff}(0 = \omega+\omega-2\omega)E_\omega E_\omega E_{2\omega}^{\rm write*}\exp(i\Delta kz)+\text{c.c.} \tag{3}$$

Then the resulting $\chi^{(2)}$ grating has the correct period for the quasi-phase matched process. Unfortunately, Eq. (3) does not describe the dynamics of the writing process and it cannot be used when dealing with pulse radiations. That is why we suppose a constant medium response to each couple of fundamental and SH pulses so that the effective quadratic susceptibility being formed by N couples of pulses is given by

$$\chi^{(2)}_{\rm eff}(z) = N\Gamma\exp(i\Delta kz)\int_{T_1}^{T_2} E_\omega(t,z)E_\omega(t,z)E_{2\omega}^{\rm write*}(t,z)dt+\text{c.c.} \tag{4}$$

where Γ is a one-pulse-couple response constant, and the integral limits T_1 and T_2 must be taken in such a way that the integration covers the whole pulse duration.

In most experiments, the fundamental radiation is produced by mode-locked Q-switched Nd:YAG laser so that the incident fundamental pulse can be assumed to have a Gaussian intensity envelope in time and the phase modulation proportional to the instantaneous intensity [12], [13]

$$E_\omega(t,z=0) = A_\omega\exp\left[-\frac{t^2}{2T_0^2}+i\Phi\exp\left(-\frac{t^2}{T_0^2}\right)\right] \tag{5}$$

where A_ω is complex constant being related with the total power, T_0 characterizes the pulse duration and Φ describes the peak modulation depth.

During the writing process the SH radiation is created in a nonlinear crystal which is placed between laser output and fibre input. In such a case we can use the approximate relation $E_{2\omega}^{\rm write} \propto E_\omega^2$ and the SH pulse launched into fibre given by

$$E_{2\omega}^{\rm write}(t,z=0) = A_{2\omega}\exp\left[-\frac{t^2}{T_0^2}+i2\Phi\exp\left(-\frac{t^2}{T_0^2}\right)\right] \tag{6}$$

where $A_{2\omega}$ is complex constant.

The reading of the $\chi^{(2)}$ grating created is assumed to be performed with the pumping radiation pulse alone so that

$$E_{2\omega}^{\rm read}(z=0) = 0. \tag{7}$$

Using the above equations we simulated the writing and reading process with data from some published experiments. The comparison of calculated and experimentally determined values of the effective fibre length is shown in the Table.

The results of our calculation for Österberg–Margulis' experiment [2] are shown in Figs. 1–3. It can be seen from Figs. 1 and 2 that the Kerr nonlinearities cause decreasing $\chi^{(2)}$ grating amplitude at first and phase becomes shifted up to $-\pi/2$ nearly at the effective interaction length, $l_{\rm eff}^{\rm theor} = 0.42$ m. The major SH energy is generated almost at the effective interaction length, and the contributions of SHG at the interaction lengths greater than l_{eff}^{theor} are negligible (see Fig. 2). The calculated effective interaction length is in excellent agreement with the experimentally determined value $l_{eff}^{exp} \approx 0.42$ m (see Fig. 1 in [2]).

Comparison between theoretical and experimental values of the effective fibre length for various cases

Experiment	l_{eff}^{theor} [cm]	l_{eff}^{exp} [cm]
ÖSTERBERG and MARGULIS [2]	42	42
BATDORF *et al.* [14]	~20	10–30
DEMOUCHY [7] A	8	9
DEMOUCHY [7] B	68	35
DEMOUCHY [7] C	240	17
DEMOUCHY [7] D	57	60

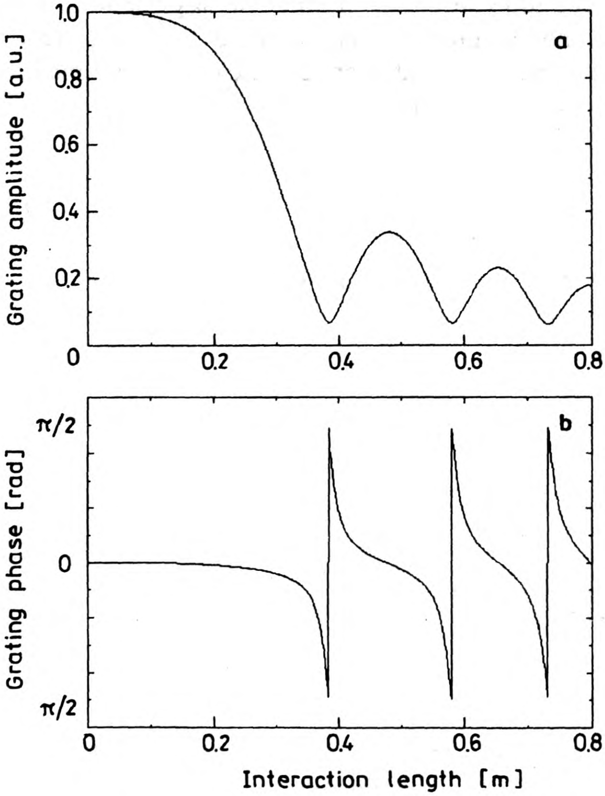

Fig. 1. Calculated length dependence of $\chi^{(2)}$ grating amplitude (a) and phase (b) using the data of Österberg–Margulis' experiment [2]

The behaviour of the amplitude and phase of $\chi^{(2)}$ grating being formed by optical pulses is to be attributed to the frequency dependent Kerr-induced phase mismatch of individual frequency components of the interacting field.

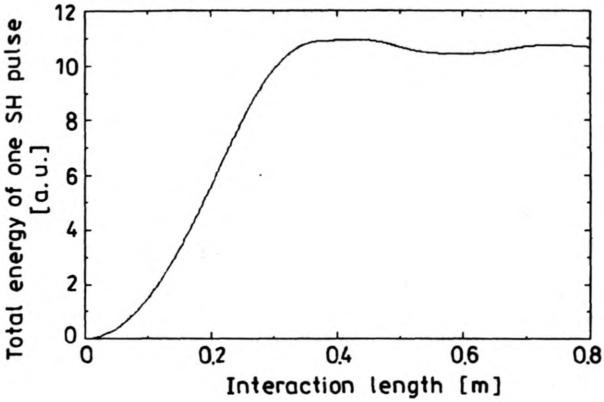

Fig. 2. Length dependence of the total energy of one SH pulse generated at the $\chi^{(2)}$ grating possessing the amplitude and phase shape as in Fig. 1

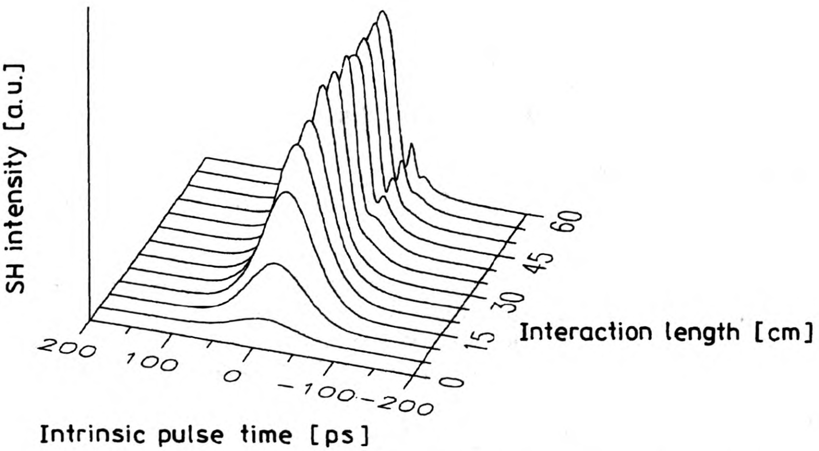

Fig. 3. Evolution of the SH pulse generated at the $\chi^{(2)}$ grating with the amplitude and phase shape shown in Fig. 1

The evolution of the SH pulse in the course of SHG at the $\chi^{(2)}$ grating being formed by optical pulses, including Kerr nonlinearities, is shown in Fig. 3. The side peaks, which begin to rise at distances being approximately equal to the effective interaction length, are the consequence of walk-off effect. This behaviour was observed in experiment, see Fig. 4b in [1].

Very instructive experiments were performed by BATDORF *et al.* [14]. It was found that the length dependence of SH radiation is related to the intensity of SH radiation during the writing process. This cannot be explained by our easy model because the material coefficient Γ in Eq. (4) does not depend on radiation intensities (in fact it is a constant). In any case we estimated (not all details needed for our calculation were published) the effective fibre length to be about 20 cm which is in reasonable agreement with the published values.

The influence of various factors on effective fibre length was investigated by DEMOUCHY [7] (the experiments are denoted as A, B, C, and D). In the first experiment A the short effective length is due to relative strong Kerr nonlinearities. The experiments B and C were performed with the aim to reduce the influence of Kerr nonlinearities. However, in these experiments the assumptions of our model were not fulfilled. The laser did not work in the usual Q-switched mode-locked mode. The input pulses did not have the Gaussian form [7] which may be the cause of the discrepancy between theoretically and experimentally determined value of effective fibre length in case B. In addition, the case C, the main limitation factor arose from fluctuations of laser frequency [7] which cannot be described by our model. In the last experiment (D), the record 60 cm effective fibre length was achieved. Our model gives value 57 cm which is in good agreement.

It follows that for most experiments on self-organized SHG the dominant factor limiting the effective fibre length is represented by the Kerr nonlinearities.

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