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THE BEHAVIOUR OF $\chi^{(2)}$ GRATING ENCODING FUNCTION

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A new phenomenological model of self-organized second-harmonic generation is proposed describing the photoinduced $\chi^{(2)}$ grating formation as a cumulative third-order nonlinearity via a complex $\chi^{(2)}$ encoding function being dependent upon the light intensities of fundamental and second-harmonic writing radiations. The theoretical results attained are confronted with recent experimental measurements by Lambelet and Feinberg and the $\chi^{(2)}$ encoding function is constructed for this special case.

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The discovery of light-induced second-harmonic generation (SHG) in doped glass fibers [1] has inspired many efforts to explain the origin of self-organized growth of second-order susceptibility ($\chi^{(2)}$) grating (for review, see Refs. [2, 3]).

The first theoretical model of self-organized SHG was outlined by Stolen and Tom [4], who proposed that an internal dc electric field is produced in the fiber owing to the non-degenerate third-order optical rectification. The photoinduced dc electric field deprives the centrosymmetric structure of isotropic medium and enables an effective SHG [5]. This idea gave rise to a group of the orientational models [6–8] that assume the formation of electric-dipole-related asymmetry or microscopic charge separation. The macroscopic dc electric-dipole polarization is believed to be the result of the vector sum of oriented or induced microdipoles.

Another model explaining the $\chi^{(2)}$ grating formation by the macroscopic charge separation due to the photovoltaic effect was proposed by Dianov et al. [3, 9, 10]. An ionizing interference of two pump photons and one SH photon is assumed to cause the ejection of the electron from existing (photoinduced) defect centre in a preferred direction, which is trapped at the boundary of illuminated area. The macroscopic charge redistribution induces then the spatially periodic $\chi^{(2)}$ grating.

The experiments performed purported in favour of the photovoltaic model [11]. However, the photovoltaic model does not provide correct predictions concerning the saturation [12, 13].

The saturation of self-organized SHG was originally predicted on the basis of the directional photoionization model by Anderson et al. [12]. It has been predicted that the ratio $I_{2\omega}/I_\omega^2$ (I_ω and $I_{2\omega}$ being the light intensities of fundamental and SH radiation, respectively) saturates independently of the fiber length, and the theoretically predicted saturation value is $(I_{2\omega}/I_\omega^2)_{\rm sat} \lesssim 10^{-16}$ m^2/W [12]. Evaluating a number of published experiments we have found that the experimentally determined value of $(I_{2\omega}/I_\omega^2)_{\rm sat}$ ranges from about 0.2×10^{-16} to 2×10^{-16} m^2/W [13]. The uncertainty within one order is caused by the inaccuracy of the experimental data reported.

Another important feature of self-organized SHG is represented by the mutual phase shift of the SH radiation being generated at the photoinduced $\chi^{(2)}$ grating and the SH seeding (input) radiation. The mutual phase shift of the SH radiation generated and the SH seed was measured by Margulis et al. [14] for the first time. This measurement yielded the phase shift of about 90°. The SH seeding intensity was relatively strong and approached to the saturation value in this case in my opinion. Note that the $\pi/2$ phase shift is precisely the wrong value to allow a weak seeding light to grow in the course of SHG preparation process. However, for somewhat lower SH seeding intensities other values of mutual phase shift were reported as well [15, 16].

Most of the authors have believed that the mutual phase shift of the SH light generated at self-organized $\chi^{(2)}$ grating and the SH seed, $\Delta\Theta = \varphi_{2\omega}^{seed} - \varphi_{2\omega}^{gen}$, represents a universal constant.

In 1996 Lambelet and Feinberg [17] published a unique experiment in which they measured the power and phase of SH output radiation emerging from a short 25 mm long germanium-doped glass fiber being conditioned to SHG in the course of the whole preparation process. Realizing that the complex field amplitude of the total SH output radiation is the sum of the complex field amplitudes of the SH seed and the SH radiation generated in the fiber, $|A_{2\omega}^{\rm output}|\exp({\rm i}\varphi_{2\omega}^{\rm output}) = |A_{2\omega}^{\rm seed}|\exp({\rm i}\varphi_{2\omega}^{\rm seed}) + |A_{2\omega}^{\rm gen}|\exp({\rm i}\varphi_{2\omega}^{\rm gen})$, I have calculated the mutual phase shift of the SH seeding radiation and the SH radiation generated in the fiber to be

$$\Delta\Theta(t) = \varphi_{2\omega}^{\rm seed} - \varphi_{2\omega}^{\rm gen} = {\rm arctg}\left[\frac{\left(\frac{I_{2\omega}^{\rm output}(t)}{I_{2\omega}^{\rm seed}}\right)^{1/2}\sin[\Theta(t)]}{\left(\frac{I_{2\omega}^{\rm output}(t)}{I_{2\omega}^{\rm seed}}\right)^{1/2}\cos[\Theta(t)]-1}\right], \tag{1}$$

where $I_{2\omega}^{\rm output}(t)$ and $I_{2\omega}^{\rm seed}$ are the light intensities of the total SH output radiation and the SH input seeding radiation, respectively, and $\Theta = \varphi_{2\omega}^{\rm seed} - \varphi_{2\omega}^{\rm output}(t)$.

Using the data of the Lambelet and Feinberg [17] experiment the time evolution of $\Delta\Theta$ in the one-hour preparation time interval was computed by means of Eq. (1). The results of the computation are shown in Fig. 1.

It is evident from Fig. 1 that the mutual phase shift between the SH seed and the SH light generated in the fiber $\Delta\Theta(t)$ monotonously decreases from its initial value $\Delta\Theta(0) \approx -70°$, which is believed to be dependent upon the ratio $I_{2\omega}^{\rm seed}/I_\omega^2$, and approaches $-95°$ in the saturation state. Note that the uncertainty of the determined value of Θ near by the saturation state in Ref. [17] is larger than 10% and, consequently, the determined saturation value of $\Delta\Theta$ must be loaded with an inaccuracy of about 5° at least.

Generalizing the above results we can conclude that the phase behaviour of self-organized SHG depends crucially upon the light intensities of pump and SH

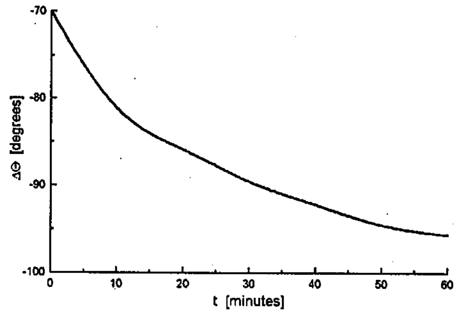

Fig. 1. Time evolution of the mutual phase shift between the SH radiation generated in the fiber and the SH seeding radiation, $\Delta\Theta = \varphi_{2\omega}^{\text{seed}} - \varphi_{2\omega}^{\text{gen}}$, using the data of Lambelet–Feinberg experiment [17].

writing radiations. It seems very likely that for the self-seeding procedure or for a very weak SH seeding, if it holds that $I_{2\omega}^{\text{write}}/I_{\omega}^2 \ll (I_{2\omega}/I_{\omega}^2)_{\text{sat}}$, both the SH seed and the SH light generated are in the phase and $\Delta\Theta = 0$. On the other hand, if the SH writing radiation approaches its saturation value, $I_{2\omega}^{\text{write}}/I_{\omega}^2 \lesssim (I_{2\omega}/I_{\omega}^2)_{\text{sat}}$, the $\pi/2$ phase shift occurs, i.e. $\Delta\Theta_{\text{sat}} = -90°$.

The lowest-order nonlinear term providing the right spatial periodicity of self-organized $\chi^{(2)}$ grating must be proportional to $E_{\omega}^2 E_{2\omega}^*$ and its complex compound value [2–4]. However, regarding the above discussed phase behaviour of self-organized SHG and the saturation, it is necessary to assume that the $\chi^{(2)}$ encoding function is complex and it is dependent upon the light intensities of writing radiations that vary along the fiber. Considering the interaction of monochromatic co-polarized plane fundamental and SH waves, $\boldsymbol{E}_{\omega}(z,t) = \boldsymbol{e}A_{\omega}(z,t) \times \exp[\mathrm{i}(\omega t - k_{\omega}z)]$ and $\boldsymbol{E}_{2\omega}(z,t) = \boldsymbol{e}A_{2\omega}(z,t)\exp[\mathrm{i}(2\omega t - k_{2\omega}z)]$, we can frame the following formula for the effective $\chi^{(2)}$ grating at a distance z after a preparation time t:

$$\chi^{(2)}(z,t) = -\exp(\mathrm{i}\Delta kz)\int_0^t \mathcal{F}\Big(I_{\omega}(z,t-\tau), I_{2\omega}(z,t-\tau), \tau\Big)$$
$$\times A_{\omega}^2(z,t-\tau)A_{2\omega}^*(z,t-\tau)\mathrm{d}\tau + \text{c.c.}, \tag{2}$$

where $\Delta k = k_{2\omega} - 2k_{\omega}$ is the phase-mismatch factor including also the Kerr nonlinearities and $\mathcal{F}\Big(I_{\omega}(z,t-\tau), I_{2\omega}(z,t-\tau), \tau\Big)$ represents the complex $\chi^{(2)}$ encoding function.

Typical times of $\chi^{(2)}$ grating formation are minutes or hours. The times of passing light through 10–40 cm long fibers are nanoseconds. Therefore, if considering the interaction of monochromatic continuous waves, the evolution of fundamental and SH field amplitudes can be described in a good approach by means of simple coupled first-order differential equations [18],

$$\frac{\delta A_{\omega}(z,t)}{\delta z} = -\mathrm{i}\left(\frac{\mu_0}{\varepsilon_0}\right)^{1/2}\frac{\omega}{n_{\omega}}\chi^{(2)}(z,t)A_{2\omega}(z,t)A_{\omega}^*(z,t)\exp(-\mathrm{i}\Delta kz), \tag{3a}$$

$$\frac{\delta A_{2\omega}(z,t)}{\delta z} = = \mathrm{i}\left(\frac{\mu_0}{\varepsilon_0}\right)^{1/2}\frac{\omega}{n_{2\omega}}\chi^{(2)}(z,t)A_{\omega}^2(z,t)\exp(\mathrm{i}\Delta kz), \tag{3b}$$

where ε_0 is the electric permittivity, μ_0 is the magnetic permeability of vacuum in SI units, n_ω and $n_{2\omega}$ are the effective indexes of refraction.

Usually, the nondepleted pump approximation is considered. For such a case Eq. (3b) is considered only and the pump light intensity I_ω and the complex field amplitude A_ω in Eq. (2) are taken as constants.

Equations (2) and (3) can be used for the description of interaction of radiation fiber modes as well if including proper overlap integrals into the effective quadratic susceptibility and $\chi^{(2)}$ encoding function [2].

The description of $\chi^{(2)}$ grating formation by means of Eq. (2) is quite general and, particularly, it involves the ideas of both above-mentioned previous models.

The great difficulty of the description of $\chi^{(2)}$ grating formation by means of Eq. (2) consists in the fact that the explicit form of the complex $\chi^{(2)}$ encoding function is unknown. For determining $\mathcal{F}\Big(I_\omega, I_{2\omega}(z, t-\tau), \tau\Big)$ by means of experimental way it would be necessary to measure both the SH light intensity and the phase at each distance z during the whole preparation process. Of course, such a measurement is impossible. However, the measurement of temporal evolution of the output SH amplitude and phase was performed [17], which provides a unique opportunity to determine the temporal behaviour of the spatial-average encoding function of the whole sample being conditioned to SHG, as it shall be demonstrated in the next treatment.

In order to be able to describe the evolution of SH field in the course of $\chi^{(2)}$ grating formation for long preparation times, we shall divide the time scale into small intervals Δt so that it holds $t = m\Delta t$. The complex $\chi^{(2)}$ encoding function in each time interval $[(j-1)\Delta t, j\Delta t]$ will be substituted by its spatial-time-average value $\overline{\mathcal{F}}_j = |\overline{\mathcal{F}}_j| \exp(\mathrm{i}\overline{\vartheta}_j)$.

Using the iterative method for the solution of Eqs. (2) and (3b) and taking the sampling of preparation time sufficiently fine so that it hold $4(\mu_0/\varepsilon_0)^{3/2}|\overline{\mathcal{F}}_j| I_\omega^2 \Delta t z / n_\omega^2 n_{2\omega} \ll 1$, for the ratio of output SH complex field amplitudes in two successive time intervals the following approximate formula was obtained:

$$\frac{A_{2\omega,j}^{\text{output}}}{A_{2\omega,j-1}^{\text{output}}} \approx \exp\left(|\gamma_j| \Delta t z \sin \overline{\vartheta}_j\right) \exp\left(\mathrm{i}|\gamma_j| \Delta t z \cos \overline{\vartheta}_j\right), \tag{4}$$

where $\gamma_j = 4(\mu_0/\varepsilon_0)^{3/2}|\overline{\mathcal{F}}_j| I_\omega^2 / n_\omega^2 n_{2\omega}$.

Considering the original definition of $\mathcal{F}$, Eq. (2), and Eq. (4), it is easily to find the expressions for the absolute value and the angle of rotation in the complex plane of the spatial average $\chi^{(2)}$ complex encoding function $\overline{\mathcal{F}}_j$ at the preparation time t_j,

$$|\overline{\mathcal{F}}_j| \approx \left(\frac{\varepsilon_0}{\mu_0}\right)^{3/2} \frac{n_\omega^2 n_{2\omega}}{4\omega I_\omega^2 \Delta t z} \left\{ \left[\frac{1}{2} \ln \left(\frac{I_{2\omega,j}^{\text{output}}}{I_{2\omega,j-1}^{\text{output}}}\right)\right]^2 + \left(\Delta\varphi_{2\omega,j}^{\text{output}}\right)^2 \right\}^{1/2} \tag{5}$$

and

$$\overline{\vartheta}_j \approx \operatorname{arctg} \left[\frac{\ln\left(I_{2\omega,j}^{\text{output}} \big/ I_{2\omega,j-1}^{\text{output}}\right)}{2\Delta\varphi_{2\omega,j}^{\text{output}}} \right], \tag{6}$$

where $\Delta\varphi_{2\omega,j}^{\text{output}} = \varphi_{2\omega,j}^{\text{output}} - \varphi_{2\omega,j-1}^{\text{output}} = -(\Theta_j - \Theta_{j-1})$. Of course, $\Delta\varphi_{2\omega,j}^{\text{output}}$ must be taken in radians.

We have taken advantage of the measurement of phase and power of the SH radiation emanating from the fiber being conditioned to SHG [17] for determining the behaviour of spatial-average complex $\chi^{(2)}$ encoding function $\overline{\mathcal{F}}(\tau)$ in the course of $\chi^{(2)}$ grating formation by means of Eqs. (5) and (6). The spline results of the computation are shown in Fig. 2. Of course, as there was a large spread in the measured values of phase and power (intensity) of the output radiation, there must be a sizable inaccuracy in the construction of $\overline{\mathcal{F}}(\tau)$. Notwithstanding, we are of the opinion that Fig. 2 provides a satisfactorily true picture of the behaviour of $\chi^{(2)}$ encoding function in the real SHG preparation process.

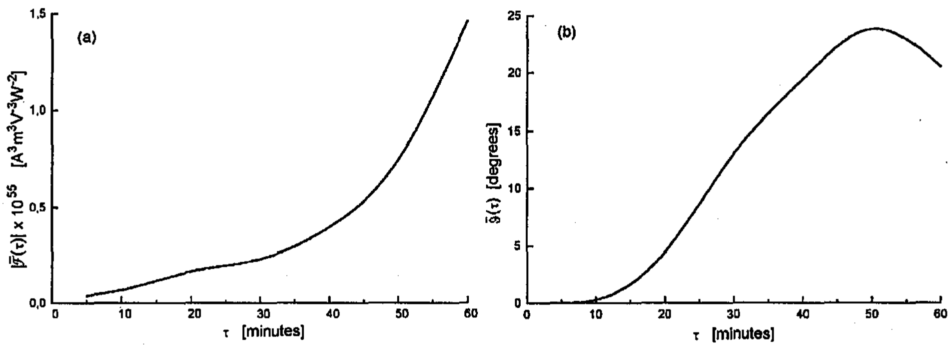

Fig. 2. The absolute value (a) and the angle of rotation in the complex plane (b) of the space-average $\chi^{(2)}$ encoding function $\overline{\mathcal{F}}(\tau) = |\overline{\mathcal{F}}(\tau)| \exp[i\overline{\vartheta}(\tau)]$ versus the integral time variable τ for the 25 mm long Ge-doped fiber using the data of Lambelet–Feinberg experiment [17].

To grasp well the behaviour of $\chi^{(2)}$ encoding function in Fig. 2, it is necessary to realize its "reverse" significance. Namely, $\overline{\mathcal{F}}(\tau)$ describes the beginning of $\chi^{(2)}$ grating formation process for large values of τ ($\tau \to 60$ min) and the end of the process for small τ ($\tau \to 0$).

It is evident from Fig. 2a that the absolute value of $\overline{\mathcal{F}}(\tau)$ takes its maximum at the beginning of SHG preparation process and, later, it monotonously decreases and approaches zero in the saturation state.

The evolution of complex structure of $\overline{\mathcal{F}}(\tau)$ is apparent from Fig. 2. The ratio of imaginary and real parts of $\overline{\mathcal{F}}(\tau)$, $\text{Im}[\overline{\mathcal{F}}(\tau)]/\text{Re}[\overline{\mathcal{F}}(\tau)]$, slightly increases during the first 10 minutes of fiber preparation process, and then it decreases and approaches zero in the saturation state. We are of the opinion that the initial increase in $\overline{\vartheta}(\tau)$ ($\text{Im}[\overline{\mathcal{F}}(\tau)]/\text{Re}[\overline{\mathcal{F}}(\tau)]$) can be attributed to the creation of new excited dopant-defect centres owing to the multiphoton (two-, three-, and four-photon) absorptions and charge transport at the beginning of self-organized SHG.

As can be seen from Fig. 3b in Ref. [17] the major enhancement of SH writing radiation happens in the first 30 minutes of the preparation process, in which the

$\chi^{(2)}$ encoding function possesses a sizable imaginary part and also its absolute value is relatively large.

It is expected that general tendencies in the behaviour of $\overline{\mathcal{F}}\left(I_{\omega}, I_{2\omega}(t-\tau), \tau\right)$ will be similar to those shown in Fig. 2 for every experiment, but the complex structure of $\overline{\mathcal{F}}(\tau)$ in the most efficient stage of the preparation process will be very different in individual cases. In the case of a very weak SH seeding, $I_{2\omega}^{\text{seed}} \ll I_{2\omega\text{sat}}$, the imaginary part of $\overline{\mathcal{F}}(\tau)$ will strongly predominate the real one and, thus, the maximum value of $\overline{\vartheta}$ will approach 90° at the beginning of the efficient SHG preparation in this case. On the other hand, if using a strong SH seeding near-by the saturation level, $I_{2\omega}^{\text{seed}} \approx I_{2\omega\text{sat}}$, the encoding function $\overline{\mathcal{F}}(\tau)$ will possess merely small dominantly real values and, hence, $\overline{\vartheta}$ will be close to zero in the whole course of the preparation process. A relatively long precursory stage with zero or very small real encoding function $\overline{\mathcal{F}}(\tau)$ must occur if using the self-seeding procedure. This precursory stage should occur even in the experiments with external SH seeding but its duration is considerable shorter and can be out of time-scale of the measurement if the SH seeding radiation is sufficiently strong.

Note that it seems very probable that an efficient parametric down conversion (PDC) can occur if the SH seeding radiation is relatively strong compared with the saturation value, $I_{2\omega}^{\text{seed}} \gg 10^{-16} I_{\omega}^{2}$ (in SI units). The $\chi^{(2)}$ encoding function $\overline{\mathcal{F}}(\tau)$ is believed to be situated in the lower half of complex plane in this case. Even when the strong visible green/blue light alone causes the erasure of $\chi^{(2)}$ structure created, we are of the opinion that optimum pump and SH seeding light intensities can be found which are able to write very effectively stable $\chi^{(2)}$ gratings into doped glass using PDC processes. A successful and unambiguous experimental demonstration of very effective self-organized PDC proposed would be a goal that could radically reverse the further development of the research on photoinduced self-organized nonlinear optical phenomena in doped-glass waveguide structures. Many potential applications are envisioned, especially for new kinds of self-shaped amplifiers or frequency down converters.

It is expected that the further investigation of the $\chi^{(2)}$ encoding function will afford not only a more complete phenomenological picture of self-organized SHG, but it can also significantly contribute to a better understanding of the intrinsic physical mechanisms being responsible for the photoinduced $\chi^{(2)}$ grating formation.

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