SECOND-HARMONIC GENERATION IN OPTICAL FIBERS

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INTRODUCTION

Second-harmonic generation (SHG) in optical fibers was first reported in 1981 [1]. Since optical fibers are made out of glass, which has inversion symmetry, the SH light was believed to be caused by higher order effects such as electric quadrupole and magnetic dipole interactions. The low conversion efficiency ($\approx 10^{-5}$) reported in these early experiments made SHG in optical fibers a not very useful or interesting effect. This was, however, changed in October of 1985, when efficient SHG was observed in commercial single-mode telecommunication fibers (conversion efficiency $\approx$ 5%, input peak power $\approx$ 20 kW) [2]. The amount of green light generated from frequency-doubling a Nd:YAG laser in a 1-m long piece of fiber was sufficient to pump a dye laser [3]. Since then, even higher conversion efficiency has been reported [4].

The most noticeable characteristic of efficient SHG in optical fibers is that it does not appear immediately upon illumination, but grows exponentially with time during 1-10 hours depending on how the fiber is illuminated. This period of time is referred to as the preparation time. Typically, the initially very weak SH light grows 8-10 orders of magnitude before it saturates. The preparation of the fiber can be done in many different ways: one, by using intense fundamental light only, internal seeding ($P_{th} > 0.5$ kW, $T_{prep} \approx$ 2-10 hours) [3]; two, using fundamental and harmonic light, external seeding ($P_{th} > 100$ W, $T_{prep} \approx$ 1-2 hours) [5]; and three, using fundamental light and a dc-electric poling field [6]. In this paper, we will focus on the first two methods of preparation. The preparation alters the properties of the glass so that efficient SHG can take place. This alteration is not completely permanent; if the fibers are left in darkness for about 6 months the conversion efficiency drops by a factor of two. However, it takes only minutes of preparation to regain the loss in conversion efficiency.

Laser Optics of Condensed Matter, Vol. 2
Edited by E. Garmire *et al.*, Plenum Press, New York, 1991

It is the aim of this paper to present some of the various models put forward to date explaining the highly surprising observation of efficient SHG in optical fibers. The paper is organized in the following way: first, three different physical models for explaining a growing second-order susceptibility in an optical glass fiber will be discussed. Polarization and modal properties of the induced SH light is then treated, followed by some new ideas about the anomalous length dependence of the frequency-doubled light. Finally, we are also mentioning the unexpected observation of growing third-harmonic light in an optical fiber.

MODELS FOR A GROWING SECOND-ORDER SUSCEPTIBILITY

The first model proposed [7] is based upon a weak non-phase-matched SH signal, created via the quadrupole polarization, to initiate the self-writing of an axially periodic pattern of colour centres which are then postulated to lead to the growth of a $\chi^{(2)}$ grating. Phasematching, in other words, is obtained through a periodic $\chi^{(2)}$. The preparation time is accounted for by the slow formation of the colour centres at 532 nm. The concept of aligning colour centres of defects in glass by intense polarized light has been experimentally observed [8] and therefore this part of the model is plausible. However, the phase-matching scheme is not valid. The reason is that the non-phase-matched SH light has a spatial modulation $I_{SH} \approx \cos^{(2)}\Delta kz$, $\Delta k = k(2\omega) - 2k(\omega)$, leading to an induced $\chi^{(2)} \approx \exp[i(\Delta k/2)z]$, which will give no net transfer of energy from the fundamental light to the SH light over the length of the fiber.

This problem was overcome in the next model proposed [5]. Here it is proposed that the alignment of defects be accomplished by a dc-field induced through the interaction between fundamental and SH light in the fiber via the third-order nonlinear susceptibility.

$$P_{dc} \approx \chi^{(3)}(0,\omega,\omega,-2\omega)\, E_\omega E_\omega E^*_{2\omega} \cos(\Delta kz) \qquad (1)$$

It was then postulated that this dc-field would break the inversion symmetry of the glass and produce an effective second-order susceptibility,

$$\chi^{(2)} \approx \gamma P_{dc} \qquad (2)$$

where γ is an imaginary constant. This $\chi^{(2)}$ will produce quasi phase-matched SH light in the fiber. The main problem with this model was soon realized to be the magnitude of the induced dc-field [9]. For most experiments the induced dc-field would be on the order of 1-1000 mV/cm. These dc-fields exert smaller forces on the glass molecules than the randomizing forces from thermal fluctuations at room temperatures. Direct experimental tests of this model [10] later confirmed that it could not explain growth of SHG in optical fibers.

The last model we will describe is based upon photo-induced charge redistribution [11]. The hypothesis for this model assumes that the nonlinear polarization, producing SH light, arises from a third-order nonlinear susceptibility mixing the pump field with a static electric field E_{dc}.

$$P_{2\omega} \approx \chi^{(3)}(2\omega,\omega,\omega,0)\, E_\omega E_\omega E_{dc} \qquad (3)$$

The dc-field is produced by the migration of electrical charges as defects in the glass are ionized by the intense light in the fiber. In ref. 11 an expression for the induced dc-field is obtained by using the "hopping" model, well known in photorefractive work [12],

$$E_{dc} = -\frac{kT}{e}\frac{\nabla I}{I} \qquad (4)$$

where I is the total light intensity in the fiber. DC-fields as large as 1000 V/cm have been predicted in the fiber core from this model. The dc-field does not have this magnitude from the beginning, however, but grows with time, as more light pulses pass through the fiber displacing more and more electrical charges. So far, no experiments have been performed to test all aspects of this new model.

Common for all these models is the assumption of defects being present in the fiber. Electron spin resonance (ESR) measurements in fibers before and after preparation indicate that a defect called Ge E´ is important for the growth of the SH signal [13]. An interesting observation in these ESR measurements was that Ge E´ defects already present in the fiber before illumination (due to mechanical and chemical processes) did not contribute to the SH signal, but only those defects generated by the light itself.

POLARIZATION AND MODAL PROPERTIES

Polarization measurements of the intrinsic and induced SH light are important for determining the tensor elements of the involved second-order nonlinear susceptibilities. In performing these measurements it is important to remember that even if care is taken to use a fiber that preserves the polarization well and the incident light is linearly polarized, the light propagating in the fiber core will inevitably have three polarizations due to the properties of the waveguide. Typically, if x is the direction of the incident light, one finds that the ratio E_x/E_y can vary between 0.0001 and 0.01, and E_x/E_z between 0.01 and 0.1. In addition to this, there are intensity-dependent contributions to the various polarizations. Taking all this into account we find, e.g. [14],

$$\frac{\chi^{(2)}_{xxx}}{\chi^{(2)}_{yxx}} = 30 \pm 10 \quad \text{and} \quad \frac{\Gamma^{(2)}_{xxxx}}{\Gamma^{(2)}_{yxyx}} = 2.4 \pm 1.2 \tag{5}$$

where $\chi^{(2)}$ refers to the induced second-order susceptibility and $\Gamma^{(2)}$ to the intrinsic. For the induced $\chi^{(2)}$ the fiber was prepared with both the fundamental photons in the x-direction. $\Gamma^{(2)}$ originates from a non-local nonlinearity, phenomenologically described by a term like:

$$P_i = \Gamma^{(2)}_{ijkl} E_j \nabla_k E_l \tag{6}$$

There are a couple of interesting observations to be seen in eq. 5. The ratios of the induced $\chi^{(2)}$ are obtained for many different pieces of fiber and at different power levels of preparation. No correlation was seen between the measured $\chi^{(2)}$ ratio and whether the fiber had been prepared at a high or low power level or if the fiber was highly polarization preserving or not (could vary over two orders of magnitude). Furthermore, for these particular fibers prepared in the x-direction (being polarization preserving to better than 1000:1), the SH light in the x-direction grew 8 orders of magnitude and at the same time the SH light in the y-direction grew just less than 7 orders of magnitude, showing the complex tensor nature of the growth process. During these measurements it was also shown that a fiber could be prepared at first in the x-direction and thereafter in the y-direction (with the same conversion efficiency for the SH light in the y-direction as in the x-direction) without affecting the conversion efficiency in the x-direction. The conclusion from this is that saturation of the overall conversion efficiency for SH light in an optical fiber is not due to lack of defects to interact with during the preparation process.

The ratios between the different tensor elements in eq. 5 cannot be measured directly. The reason is that the SH light can be in different modes for different polarization directions, which affects the conversion efficiency. The most general expression for the ratio between two tensor elements is

$$\frac{\chi^{(2)}_{ijk}}{\chi^{(2)}_{lmn}} = \frac{f_{lmn}}{f_{ijk}} \left[\frac{I^{\omega}_m \cdot I^{\omega}_n}{I^{\omega}_j \cdot I^{\omega}_k} \right]^{1/2} \left[\frac{I^{2\omega}_i}{I^{2w}_l} \right]^{1/2} \tag{7}$$

where

$$f_{ijk} = \frac{1}{N} \int_{A_\infty} P^{2\omega} \times H^{2\omega} \, dxdy \tag{8}$$

and H is the magnetic field for the SH light. So, the overlap integral f has to be calculated for each ratio to be determined. Significant for SHG in optical fibers is that in most reported cases the saturated SH light is not in the same mode as the initial SH light.

The particular mode the SH light is in, as can be seen from eq. 8, gives information about what type of nonlinearity could have caused it. All fibers reported so far have had the initial SH light in an asymmetric mode, either LP31 or LP11. This means that by examining the angular part of the overlap integral in eq. 8, the initial SH light has to come from either a quadrupole interaction (bulk and interface) or an electric dipole interaction (interface). It is, however, extremely difficult to separate the quadrupole contribution from the electric-dipole contribution [15]. Experiments on fibers with different compositions indicate that the intrinsic SH light is from quadrupole interactions at the core-cladding interface [14]. For the saturated SH light, it has been reported to occur in asymmetric as well as symmetric modes. This can only be explained by electric-dipole interactions being the source for the saturated SH light.

LENGTH DEPENDENCE-ABSORPTION

The fact that the mode for the SH light changes during the growth process indicates that the refracting index is being altered. We propose that this is related to a significant change in the absorption for the SH light.

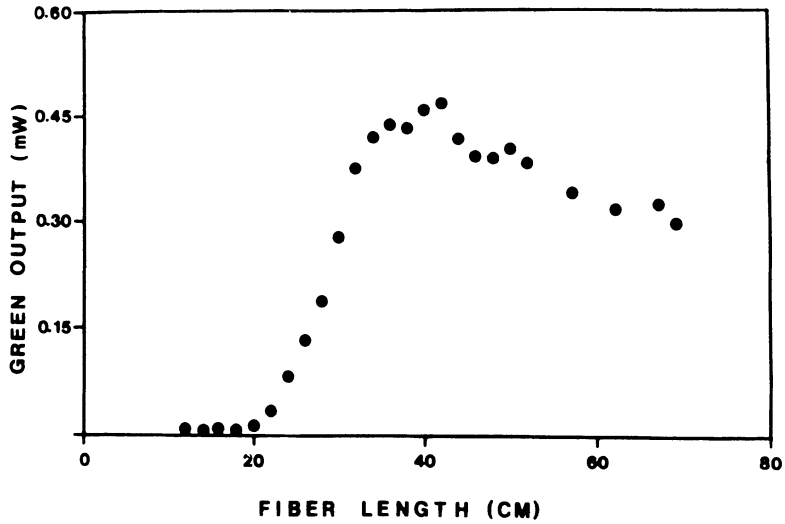

Fig. 1. Second-harmonic light as a function of distance into the fiber [16].

The reason for incorporating absorption into the analysis of SHG in optical fibers is related to the anomalous length dependence, fig. 1 [16]. The curve in fig. 1 was measured for the saturated SH light after ~10 hours of internal seeding. The late onset (~23 cm) of strong growth of the SH signal is characteristic for internal seeding. A thorough study of the length dependence [17] shows that the onset of strong growth of the SH signal is a function of the amount of SH light used to prepare the optical fiber. The more SH light used in the preparation, the earlier the induced SH light starts to grow strongly. All these observations can be accounted for by incorporating two-photon absorption into the equation for the length-dependence,

$$\frac{dE_{2\omega}^{(z)}}{dz} = i \cdot \gamma_1 \cdot \chi_{eff}^{(2)} \cdot E_\omega \cdot E_\omega - \gamma_2 \chi^{(3)} |E\omega|^2 E_{2\omega} \qquad (9)$$

γ_1 and γ_2 are well-known constants for second- and third-order processes, $\chi_{eff}^{(2)}$ has the overlap integral factor incorporated into it, and $\chi^{(3)}$ is the imaginary part of the third-order nonlinear susceptibility.

The origin of the two-photon absorption is surmised to be due to a resonance in the GeO molecules in the core of the fiber. These molecules have a resonant transition at 364 nm ($\omega + 2\omega \rightarrow 355$ nm) which is two-photon allowed [18]. Integrating eq. 9 we can obtain an expression for the distance it takes for the SH light to grow to 1/10 of its maximum value,

$$Z_{1/10} = \frac{0.38}{A} - B \cdot E_{2\omega}(0) \qquad (10)$$

where z is measured in meters and $A = \frac{3\pi}{4\lambda n} \chi^{(3)} |E\omega|^2$ and $B \approx 10^{-7} - 10^{-8}$, depending on the fiber. From fig. 1, where $z_{1/10} \approx 23$ cm, we can calculate the two-photon absorption coefficient $\beta \approx 0.3$ cm/GW. This is a small but realistic value [19].

Experimentally, what we find is that the distance $z_{1/10}$ is constant during the whole growth process, but that the slope of the exponentially growing region increases with the growth of the SH light [17]. As mentioned earlier we suggest that this is due to the "linear" absorption increasing with increasing visible SH light in the fiber. Evidence in favour of this can be seen from fig. 1. For the saturated regime ($z \gtrsim 40$ cm) the SH light should be constant ($\alpha_{loss} \approx 0.016$ dB/m) but is instead seen to decrease surprisingly rapidly ($\alpha_{loss} \approx 9$ dB/m), indicating a much larger linear absorption for a prepared fiber than for an unprepared fiber.

In order to match our model with the experimental data in fig. 1 we can deduce that the linear absorption coefficient α has to depend on the SH light intensity to the power of 3 $\left(\alpha_{2\omega} \propto I_{2\omega}^3\right)$. The validity of this has to be studied more extensively.

In eq. 9 we have written down the length dependence as if it were phase-matched. The justification for this is that the model is based upon a two-photon resonant transition between the fundamental and harmonic light, which will lock their phases together [20]. A consequence of this is that the coherence length is no longer important; instead the optimum length for our nonlinear process is a function of the induced absorption.

The assumption that the resonant transition provides full or partial locking of all the involved phases does not rule out a periodic $\chi^{(2)}$ for additional phase-matching. As a matter of fact, it has been experimentally shown that indeed a periodic $\chi^{(2)}$ is induced in the fiber [21].

CONCLUSION

We have tried to give a brief account for the models proposed to date of explaining SHG in optical glass fibers. Based on experiments and calculations, we have argued that SHG in optical fibers starts from electric quadrupole interactions at the core-cladding interface but that the induced SH light is from electric dipole interactions. We have also proposed a new model, based on absorption, for explaining the anomalous length dependence and mode-coupling. The question of what makes the $\chi^{(2)}$ grow in an optical glass fiber still remains to be properly explained, though.

To further confuse the issue, we would like to end our conclusion by mentioning the recent discovery of a growing third-harmonic signal in an optical fiber [22]. It was observed that the third-harmonic signal could grow exponentially with time, at the expense of the SH signal, almost 2 orders of magnitude.

It is obvious that the observations of growing harmonic signals in optical fibers is very interesting and that the physics behind it is far from understood.

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