Self-Reconstructing All Optical Poling in Polymer Fibers

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Self-sustained all-optical poling (AOP) second harmonic generation (SHG) experiments are conducted in single and multi-core dye-doped poly(methyl methacrylate) optical fibers. By tuning the polarization of the fundamental beam, the SHG signal is degraded and is reconstructed spontaneously up to its initial level. We found a new situation in which the photo-induced self-organization of azo-polymers creates a well-ordered periodic structure.

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Several methods have been proposed to overcome the lack of second-order nonlinearity in amorphous organic materials such as polymers, which are centrosymmetric by fabrication. Most methods were based on the molecular orientation of the material (poling) under external fields [1,2]. Among these methods, all-optical poling (AOP) has been developed as a self-induced second harmonic generation process [3]. Early experiments carried by Osterberg and Margulis in glass optical fibers evidenced that the induction of a stable second harmonic generation (SHG) was possible using a pulsed laser [4]. Despite a huge activity dedicated to the study of AOP in polymer materials, the stable induction of a second harmonic (SH) signal was never conclusively demonstrated [5]. The reason explaining these results is that the same light that can induce a second harmonic response can also destroy it, as earlier authors reported [6].

AOP is a photoinduced SHG method, which consists in a simultaneous irradiation of materials with a coherent superposition of fundamental and second-harmonic lights. It results in a polar orientation of the molecules inside the material [6]. Studies on the dependence of the photoinduced SHG in azobenzene side-chain polymers revealed a quadratic increase of the SHG with film thickness [7-9]. However, under continuous illumination with the fundamental light, SHG decays on a few minute time-scale, whereas with the use of a 100 µm-thick film, the decay is quantitatively slowed down.

Temporal stability of the polar orientation in AOP is one of the major drawbacks of the method. Previous results indicated that the limitation could be overcome by inserting the dye-doped polymer into a waveguide [10]. Optical waveguides create an ideal situation for nonlinear interactions as they provide a strong beam confinement over long propagation distances [11]. We report in this paper on the evaluation of the SHG photoinduced in different types of fibers. We show that after degrading the photoinduced SHG by rotating the polarisation of the fundamental laser beam, the photoinduced SHG grows again till its initial value or above. This gives the evidence that self-sustained SHG is achieved in polymers optical fibers.

Single and multi-core poly(methyl methacrylate) (PMMA)-based optical fibers were fabricated according to the procedure described in ref. [12]. The fiber cores were doped with the nonlinear disperser-1 dye (DR1). Glass transition temperature of the polymer material was about 105 °C. The method produced graded index preform profiles with index difference Δn = 4.2 x 10⁻⁵ at 632.8 nm wavelength. The end faces of the fibers were polished with sand paper and fine lapping paper. Measured propagation losses in the fibers were close to 20 dB/cm at 650 nm and 1 dB/cm at 1300 nm wavelength. Measured insertion losses of the laser beams into the fibers under study were about 30 %. The multi-core fiber contains three spatially separated cores and has an external diameter of 0.86 mm. The single-core fiber has an external diameter of 0.96 mm. The potential benefit of using a multi-core fiber in the context of AOP is to reduce the propagation losses in the harmonic beam, which are also a major limitation to the efficiency of the process [13].

The beam source was an actively Q-switched Nd:YAG laser (Thales Diva II model) delivering 25 ns pulses with 10 Hz repetition rate at λ = 1320 nm infrared (IR) wavelength. An external cavity type II phase matched KTP crystal was used for partial frequency doubling of the fundamental beam. The resulting so-called seeding beams are a coherent superposition of two co-propagating collinear beams: the SH beam at wavelength λ/2 and the fundamental IR at wavelength λ. The two output beams delivered by the laser had crossed polarizations. The intensity
of the laser (fundamental and harmonic beams) was controlled by the power supply of the laser. Rotation of half wave plate at 650 nm placed after the laser and followed by a polarizer was used to control the intensity ratio between the two fundamental and harmonic seeding beams. Both beams after the polarizer had parallel polarizations before injection into the fiber. The power ratio between fundamental and SH seeding beams was optimized using a reference DR1 thin film in order to approach maximum SHG efficiency.

The two beams with parallel polarizations were focused with a microscope objective (NA = 0.25) into the fiber. A XYZ-translation stage was used for optimal light injection into the fibers. The output beam was collected with a 10 mm focal length lens and passed through a prism to separate the IR from the SH beam. The waists of the IR and SH-seed beams at the laser output were about 3 mm. The average beam power was kept well below the damage threshold of the polymer. They were 10 mW in the IR and 42 μW in the SH.

The AOP process, which was described in [6] consists in two alternated periods: the writing phase where the fundamental laser beam is sent together with the SH seeding beam; and the readout phase where only the fundamental laser beam is sent to probe the SHG. The intensity of the SHG is measured using a photomultiplier tube (Hamamatsu H11462-11) by blocking the SH seeding beam with a RG1000 Schott glass placed at the laser output. During the writing process, the intensity of the SHG is sampled and averaged over 10 laser shots every 5 minutes. During the measurements, the monitored SHG signal has a fluctuation of ± 0.05 μW.

Figure 1 shows the evolution of the SH signal in the different fiber samples under study. The growth rates during seeding were fitted to an exponential \( I_{\text{SHG}} = I_{\text{SHG}}^{\text{Gast}} [1 - \exp(-t/t_{\text{G}})]^{2} \) where \( t_{\text{G}} \) is the growth time constant and \( I_{\text{SHG}}^{\text{Gast}} \) the SHG intensity at saturation of the growth. The growth time is substantially shorter in the multi-core for the longer fibers. SHG growth appears relatively slower that in previous publications on AOP in DR1-based materials [5]. This can be attributed to the rate of excitation, which is reduced in our experiment in which the laser is tuned away from resonance, increasing the growth time constant. The SHG values at saturation should increase quadratically with the fiber length. They do not owing to propagation losses (see also the insert in Fig.1b). They should actually increase like \( I_{\text{SHG}}^{\text{Gast}} \propto L^{2} \exp(-\alpha L) \) [6]. From the fitted \( I_{\text{SHG}}^{\text{Gast}} \) values, we can get an in-situ estimate the propagation losses of the mode at second harmonic frequency: \( \alpha = 4.2 \pm 0.4 \) cm\(^{-1}\) in the single core-core and \( \alpha = 3.5 \pm 0.4 \) cm\(^{-1}\) in the multi-core fiber. The maximum SHG achieved in the multi-core fiber is about 5.5 times and 7 times smaller than in the single-core fiber for \( L = 5.3 \) mm and \( L = 2.6 \) mm respectively. The difference between multi-core and single-core fibers may originate from the overlap of the optical mode with the DR1-doped cores. Close to saturation of the SHG, the seeding process was stopped (vertical arrows in figure 1) and the relaxation of the photoinduced SH nonlinearity was studied under IR excitation. After a brief transition (5 – 10 min), which manifests as a moderate loss of SHG, the SHG recovers and remains constant. The SHG is self-sustained without SH seeding laser beam. The transition may be attributed to the SH-active region in the fiber that migrates from the front side to the exit side of the fiber, which process happens with the same dynamics as the initial growth [14]. This effect happens because the magnitude of the SHG is comparable to the SH-seed beam intensity, when the 30% insertion losses are accounted for.

In order to assess the self-reconstructing SHG, the idea is to degrade the all-optical poling organization and let it reconstruct without assistance from the SH-seed beam. For that purpose, we need to keep some SHG efficiency, which will act as a self-seed beam. To realize it, we turned the polarization of the reading IR beam incrementally using a half-wave plate, after the interruption of the seeding process. 20° increments allow to monitor the progressive degradation of the poling as illustrated after the left-most vertical line in Fig.2. Except for the short single-core fiber for which maximum SHG was not yet achieved, the SHG degrades till the polarization was turned by 90°. We observe that the SHG grows again after stopping the rotation (at about 30 min-time in Fig.2). It recovers and exceeds the initial SHG intensity. After about 10 min, the reconstructed SHG remains strong and stable. Rotation of the polarization impedes more on the SHG of the longer fibers (Fig.2b), in which SHG almost vanishes at 30 min time, than on the shorter ones (Fig.2a). In the longer fibers, SHG dropped below 20% of its maximum, before reconstructing. This experiment evidences the efficiency of SHG self-reconstruction in plastic optical fibers. In short and/or multicore fibers, the SHG reconstructs into a significantly larger value (up to twice as much as after seeding). A slight increase of the SHG may simply be attributed to the balance between IR and SH-seeding beams, which was optimized at once in thin-films, but which depends on the particular transmission of each fiber at the harmonic mode [14]. The significant SHG growth observed after seeding may be interpreted qualitatively in terms of the three-dimensional pumping model developed by Dumont [15]. Indeed, after initial poling with horizontal polarizations, the horizontal orientation population of the azo-dyes is depleted, which increases the radial population around the s-direction. When the polarization is turned to p, the AOP process benefits from an increased initial pool of re-orientable species (a rough estimate would give 25% increase). This delivers a larger SHG, when the self-sustained process is fast enough to accommodate the change. After 75 min
(the last period in Fig.2), polarization was rotated abruptly by 90°. The SHG then drops significantly, down to the minimum value achieved at 30 min, before reconstructing again with the same rise time until the same plateau. The AOP process is self-recovering.

In conclusion, we have demonstrated efficient SHG in dye-doped single and multi-core PMMA optical fibers. Both fibers show self-sustained SHG, which is actually a self-seeding effect when the SH-seeding laser beam has been cut at saturation of the nonlinearity. Most importantly, SHG grows back to its initial value or above after degradation of the polar order resulting from polarization rotation of the IR reading beam, in a self-reconstructing AOP process. This new property may open new application areas to dyed plastic fibers [16]. That is a new manifestation of the photo-induced self-organization of azo-polymers [17] in which a well ordered phase-matched periodic structure is created.

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References
Fig. 1. Comparison of SHG intensity growth during and after the seeding period for two different fiber cores, (a) 2.6 mm fiber length, (b) 5.3 mm fiber length. Insert in part A is a microscope image of the multi-core fiber with 0.19 mm diameter and 0.86 mm cladding. Dark areas are the DR1-PMMA cores with $2r = 35 \pm 3$ µm and $d = 25 \pm 3$ µm. Insert in part B shows the SHG growth in a 14 mm long multi-core fiber. The arrows materialize the end of the seeding process. The growth process was fitted to an exponential law with $t_g = 30$ and 28 min for the 2.6 mm single-core and multi-core fibers, respectively; $t_g = 41$ and 20 min for the 5.3 mm single-core and multi-core fibers, respectively.

Fig. 2. Evolution of the self-reconstructing SHG upon rotation of the reading beam polarization at 1300 nm. Initial polarization was set $s$ (horizontal) during the seeding process, as indicated by an horizontal arrow. The seeding process was interrupted after 15 to 20 min, as pointed by the vertical arrows. Except for the last increment, rotation of the half-wave plate in front of the IR reading beam was carried by increments of 10° every 5 min, which corresponds to a 20° rotation of the polarization, till a rotation of exactly 45° (90° polarization $p$ indicated by central vertical arrow). The system was then left stationary during 35 min, at which time the polarization was turned by 90°, back to $s$ (horizontal arrow). The successive time periods are marked by the dashed horizontal arrows delimiting the polarization symbols. (a) 2.6 mm fiber length, (b) 5.3 mm fiber length. The same fibers as Fig. 1 were used.