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Simultaneous corona poling of multiple glass layers for enhanced effective second-order optical nonlinearities

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Silica-based thin-film multilayers are investigated as a means to enhance the effective second-order nonlinearity induced in silica glass structures by corona poling. Structures consisting of phosphorus-doped and undoped silica glass layers exhibit second harmonic generation (SHG) that is higher by an order of magnitude compared to the SHG in bulk silica glass poled under the same conditions. When the poled structure consists of two multilayered stacks separated in space, the stacks exhibit comparable poling-induced nonlinearities. This result suggests that the poling voltage is divided between the two stacks such that simultaneous poling of multiple regions within the sample is realized. © *2011 American Institute of Physics*. [doi:10.1063/1.3614435]

Electro-optic modulation and frequency doubling are some of the most prominent applications that would greatly benefit from silica possessing second-order nonlinear properties. Silica glass is an amorphous material with intrinsic inversion symmetry, and as such it cannot have any even-order nonlinearities. However, poling techniques can be used to break the symmetry, realizing an artificially created second-order nonlinear material.

Migration of impurity ions within the glass network is believed to be the main mechanism behind poling-induced nonlinearity. In a typical poling process, a voltage of a few kilovolts is applied across the glass at an elevated temperature, and the sample is allowed to cool to room temperature with the voltage still applied. Once the voltage is switched off, a stable nonlinear region is induced near the anodic surface of the glass, with a typical second-order susceptibility coefficient $\chi^{(2)} \sim 1$ pm/V.[1] The main mechanism believed to be responsible for this nonlinearity involves the migration of impurity alkali ions (mainly sodium), whose mobility increases at the poling temperature, away from the anodic surface under the influence of the applied electric field.[2] This migration results in a negatively charged depletion region within a few micrometers of the anode. In addition to this negatively charged region, a positive charge layer is formed at the anodic surface due to ionization occurring at the glass-anode interface.[3] The charge separation resulting from this positive charge layer and the negatively charged depletion region establishes a strong electric field $E \sim 10^7$ V/cm within the depletion region, which remains frozen in the glass after it is cooled. This frozen-in electric field couples with the third-order nonlinear susceptibility $\chi^{(3)}$ (which is present in all materials), resulting in an effective second-order susceptibility that is proportional to $\chi^{(3)}E$.

The main reason why practical nonlinear devices have not yet been realized with poled glasses is the very limited extent of the induced nonlinear region, whose typical thickness is only $\sim 5\ \mu$m.[1,4] This results in a poor overlap between the nonlinearity and the optical field propagating through the region, and thus inefficient nonlinear interaction.[5] Poling for longer durations cannot be used to enhance this interaction, as substantial injection of positive charges from the anodic surface destroys the depletion region,[6] thus decreasing the magnitude of the induced nonlinearity.

The work presented here is a step towards increasing the extent of the nonlinear region by using multilayered silica-based structures. Evidence of enhanced $\chi^{(2)}$ near interfaces[7,8] suggests that interfaces between different glasses play an important role during poling. In this work, we take this idea further by designing multilayered structures that specifically take advantage of these unique nonlinear properties at interfaces. All the layers explored here are silica-based thin films deposited using standard microfabrication techniques. The results in this paper show that thin-film multilayered silica structures can be used to increase the effective nonlinearity upon poling and that silica dopants have a strong effect on the dynamics of the poling process across interfaces and thus on the induced nonlinearity.

The multilayered silica-based structures were deposited by low-pressure chemical vapor deposition (LPCVD) at 420 °C on double-side polished synthetic fused silica substrates (ES grade by Tosoh Quartz). All the substrates used in these experiments were cut from the same ingot to ensure that the alkali impurity levels remain the same. Because of the vertical positioning of the substrates in the deposition chamber, equivalent stacks of layers were deposited on both sides of the substrate.

In order to break the symmetry of the glass structures and induce second-order nonlinearities, the samples were poled using the corona poling technique.[9] The sample was heated to 300 °C, and a positive voltage between 6 and 9 kV was applied between a tungsten needle held 8 mm above the sample and a pressed-on n-type silicon electrode in contact with the back of the sample. After 8 min, the heater was switched off, and the sample was allowed to cool to room temperature before the voltage was removed.

In order to characterize the poling-induced nonlinearity, we measured the second harmonic generated (SHG) in the samples using the Maker fringe method. A mode-locked Ti:Sapphire laser operating at 800 nm, 80 MHz repetition rate, and pulse width < 70 fs was used as the fundamental

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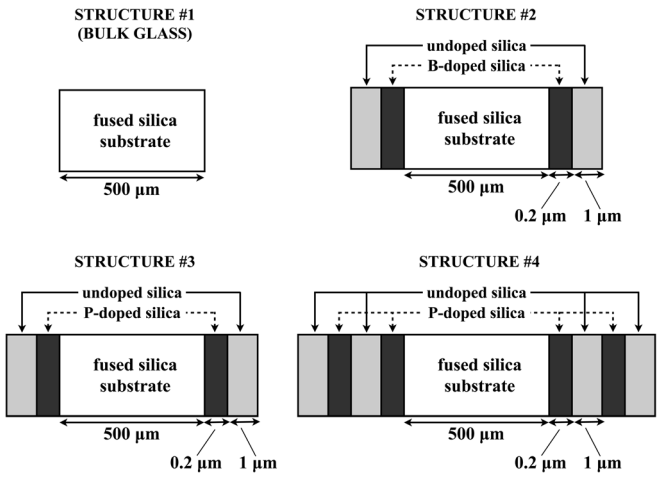

FIG. 1. Samples studied in this work – bulk silica glass (structure #1) and multilayered silica-based structures (#2–#4).

source, and the second harmonic (SH) was detected by a photomultiplier tube. Because of a reduced interference between the bound and free second harmonic waves that occurs with an ultrafast laser source, a comparative measurements of the induced $\chi^{(2)}$ coefficient with a known reference is difficult,[10] thus our characterization focused on determining the relative enhancement in the SHG in the poled multilayered structures compared to the SHG in poled bulk silica substrate.

Three sets of experiments were designed to study the induced nonlinearity in the poled multilayered structures. The goals of the first set of experiments were (1) to determine whether poling of multilayered silica-based structures increases the induced nonlinearity compared to the nonlinearity induced in poled bulk silica glass and (2) to study whether the choice of glass dopants (phosphorus or boron) has an effect on the induced nonlinearity. To this end, the four structures illustrated in Figure 1 were poled, and the second harmonic generated in the samples was measured, with the results summarized in Figure 2. The experimental results show that the second harmonic (and thus the induced effective nonlinearity) is always higher in the multilayered structures than in bulk silica. While the increase is modest for the structure with boron-doped layers (structure #2), the enhancement is larger than one order of magnitude in structures with phosphorus doping (#3 and #4). Note that the SHG in the multilayered structure that did not undergo poling is insignificant, thus the enhanced nonlinearity can be attributed to the effect of the multilayered structures on the poling process.

A comparison of the poled multilayered structures in Figure 2 indicates that the doping of the layers is crucial to achieving a significant enhancement in the induced nonlinearity. We believe that the effectiveness of the samples with phosphorus-doped layer is related to the way phosphorus incorporates into the silicon dioxide network. Both phosphorus and boron substitute for Si^{4+} in the SiO_4 tetrahedra and form oxides themselves (primarily P_2O_5 and B_2O_3, respectively). However, because phosphorus has five valence electrons (compared to four in silicon), non-bridging oxygen ions become associated with the phosphorus sites. These oxygens have local negative charges, which trap any positive ions drifting through the lattice through a coulombic interaction. This property has been extensively used to limit migration of sodium impurity ions to stabilize the behavior of early MOSFET (metal-oxide-semiconductor field-effect transistor) devices.[11] Unlike phosphorus-doped silica, which essentially forms a trapping layer for positive ions, boron is incorporated into the glass network in a trivalent form, thus creating electron-deficient centers that have no affinity for positive ions.

Based on the encouraging results with phosphorus-doped layers, the rest of the experiments dealt exclusively with structure #4. In the second set of experiments, we studied the dependency of the induced nonlinearity on the poling voltage, with the summary of the results presented in Figure 3. Although a comprehensive study of the effect of poling voltage has not been done, the results presented in Figure 3 show that using a higher poling voltage results in an increased nonlinearity, and the SHG in the poled multilayered samples is consistently about an order of magnitude higher than in bulk silica glass poled under the same conditions.

The goal of the last set of experiments was to study the distribution of the nonlinearity in our multilayered structures. In the case of bulk glass poling, as positive impurity ions move away from the anode, the applied voltage drops primarily across the established depletion region. However, as shown above, the use of doped silica layers allows a greater control over the migration of charges during the poling process. This observation gives rise to an intriguing idea – can

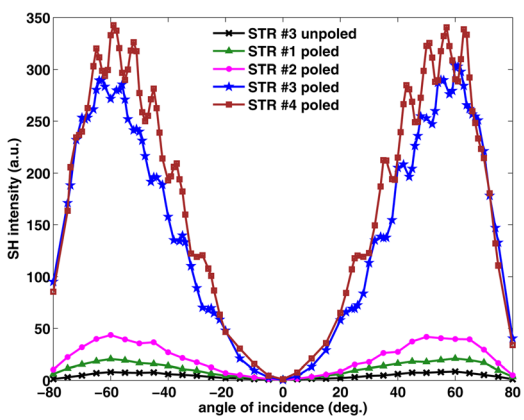

FIG. 2. (Color online) Comparison of the SHG in poled bulk silica glass (structure #1) and multilayered silica-based structures (#2–#4).

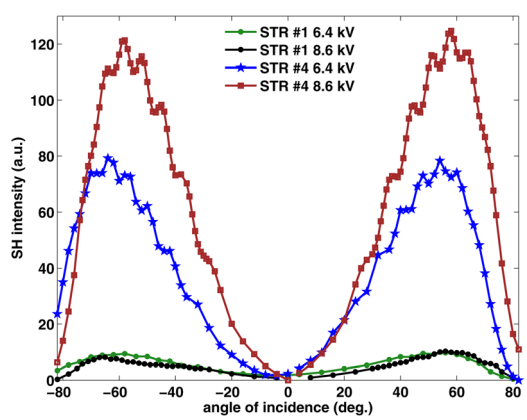

FIG. 3. (Color online) Comparison of the SHG in bulk silica glass (structure #1) and multilayered sample (structure #4) for different poling voltages.

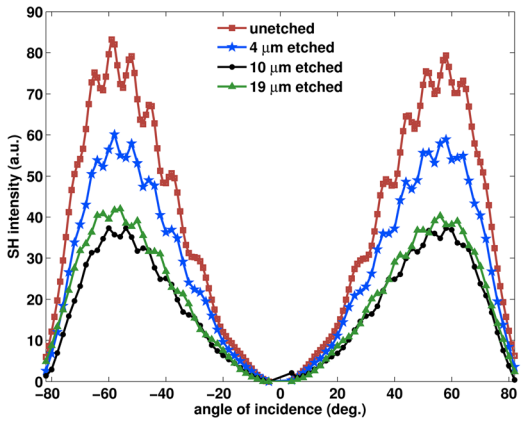

FIG. 4. (Color online) The second harmonic signal generated in structure #4 as the anodic side is etched.

the layers be designed such that the entire structure behaves as a few charge-accumulating capacitors[12] in series? If this is the case, then the applied voltage could be divided between the "capacitors" such that a few regions of glass are poled simultaneously. Since our samples have multilayered stacks on both sides of the substrate, the merit of this idea can be tested by investigating whether the poling-induced nonlinearity is concentrated only near the anode side of the sample (as in the bulk glass case) or is divided between the two multilayered stacks (which would indicate simultaneous poling of the two regions).

To study the distribution of the nonlinearity, localized regions on the poled multilayered sample were etched in dilute hydrofluoric acid, and the SHG in the remaining thickness was measured. As can be seen in Figure 4, when the anodic side of the poled sample is etched, the SH signal gradually decreases. This trend stops at etch depths greater than 10 μm, at which point the SH signal remains the same (within the experimental error). When the same experiment was repeated on the cathodic side of the sample, a very similar behavior was observed, leading to the conclusion that comparable nonlinearities were induced by poling on both sides of the sample (both spanning between 6 and 10 μm). An important implication of this is that the same idea can be used to bring the two stacks closer and use the simultaneous poling of more than one region to realize a thick nonlinear region that is compatible with standard waveguide or fiber dimensions.

Note that in a typical Maker fringe experiment, two comparable nonlinearities on both sides of the sample would result in deep ($\sim$100%) modulation fringes. However, this is not the case here because of the ultrafast laser source used in our experiments. The spatial extent of the femtosecond pulses is much smaller than the thickness of the sample, thus when the bound and the free second harmonic waves move at different group velocities, their spatial overlap decreases drastically by the time they reach the back of the sample. Therefore, the decreased interaction between these two waves results in severely damped fringes in the Maker fringe envelope.[10]

In the case of bulk glass, previous studies reported no nonlinearity peak at the cathode[13] or estimated it to be extremely thin.[14] In our case, however, the nonlinearity at the cathode is comparable in magnitude and thickness to the nonlinearity induced at the anode side of the sample. In order to verify that this is a consequence of the multilayered design and not of our specific poling process, we compared the SHG in structure #4 as shown in Figure 1 ("double-stack sample") to the SHG in an identical structure but with the backside layers removed prior to poling ("single-stack sample"). Unlike in the double-stack sample case, the Maker fringe measurement for the single-stack sample did not exhibit any modulation fringes, indicating that the remaining nonlinearity extends over a region shorter than the coherence length in silica (11.9 μm at 800 nm). When the cathodic side of the poled single-stack sample was etched, no change in the SH was observed. This confirms that the single-stack sample has only a single nonlinearity peak at the anode, and thus the cathodic nonlinearity peak in the double-stack sample is a direct result of the backside multilayers.

In summary, multilayered thin-film structures can be used to enhance the effective second-order nonlinearity induced by corona poling in silica-based devices by an order of magnitude compared to poling of bulk silica glass. Phosphorus-doped silica layers deposited by LPCVD act as trapping layers for positive charges that migrate during the poling process. While in a previous study of corona poling of bulk silica glasses it was concluded that continuous injection of positive charges from the anode surface destroys the formation of a depletion region,[9] we believe that the trapping of positive charges by the phosphorus-doped silica layers is responsible for the improvement in the effective second-order nonlinearity induced in the multilayered structures.

In addition, our experiments show that when the poled structure consists of multiple multilayered sections, the poling voltage is divided between them, realizing simultaneous poling of multiple glass regions. This ability is unique to the multilayered thin-film structures and is an important step towards enhancing the effective poling-induced nonlinearity by increasing the total thickness over which the nonlinearity occurs. Extending this idea to larger multilayered stacks, with optimized dimensions and doping levels, is expected to result in even stronger enhancement, paving the way for a practical electro-optic modulator in silica glass that is compatible with standard planar lightwave circuit technology.

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