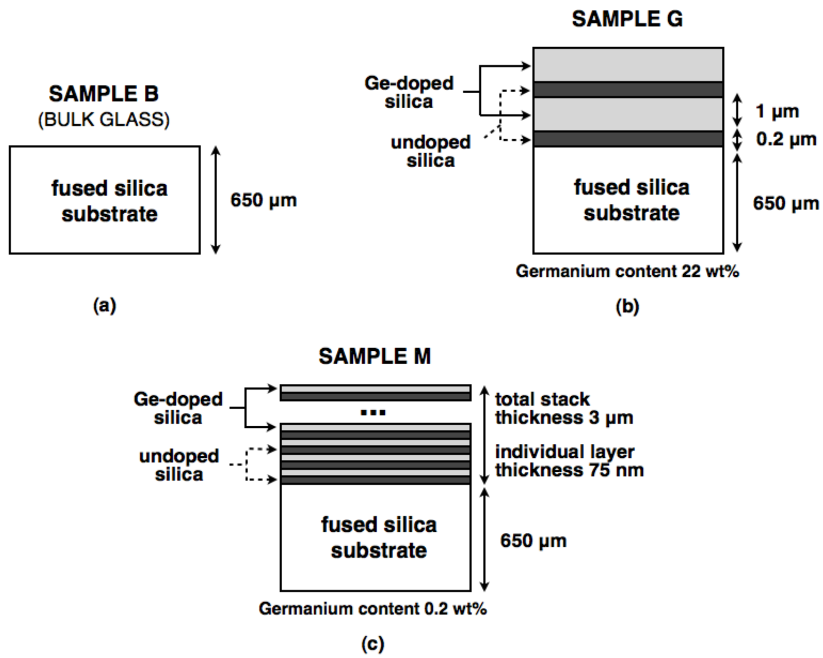

Fig. 2. Thermally poled silica-based samples studied in this work. (a) A bulk glass sample. (b) A four-layered stack of alternating undoped and germanium-doped layers. (c) A multilayered stack consisting of a large number of alternating undoped and germanium-doped nanolayers.

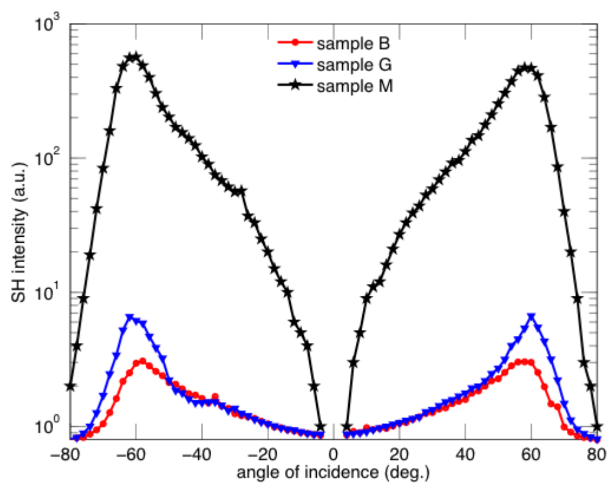

Fig. 3. SHG in thermally poled bulk silica glass and our silica-based multilayered structures. A two-fold SHG enhancement is obtained in sample G compared to sample B, where the two samples were poled under identical conditions. A 204-fold enhancement is obtained in sample M, which consisted of a 3 μm-thick stack with a large number of sub-100 nm-thick nanolayers.

In the second part of our research, we studied multilayered samples that had a total stack thickness of 1.5 to 5 μm, and consisted of a large number of 75 nm-thick silica layers. Although PECVD is generally not designed for the deposition of very thin layers because of its relatively high deposition rate, our system was capable of depositing sub-100 nm-thick alternating layers of undoped and lightly germanium-doped silica. When a sample with 40 nanolayers and 3 μm total stack thickness (see sample M in Fig. 2) was poled, a 204-fold

#157875 - $15.00 USD Received 8 Nov 2011; revised 30 Nov 2011; accepted 1 Dec 2011; published 16 Dec 2011

improvement in the generated second harmonic signal compared to poled bulk glass was observed (Fig. 3).

The preceding discussion compares the SHG in our multilayered samples with the SHG in Suprasil glass. Although previous reports showed that the SHG in Suprasil is one order of magnitude lower than in other silica glasses [4], it is important to note that the SHG in our multilayered stacks is still more than an order of magnitude higher than in even the best bulk fused silica glasses. This result clearly indicates that the idea of poled multilayers can be used as a design strategy towards the realization of a thick silica-based nonlinear region. Further improvements in the induced nonlinearity are expected by optimizing design variables such as the layer deposition technique, the parameters of the multilayered stack, and the poling conditions. For instance, we studied variations of sample M with multilayered stacks of varying thicknesses (Fig. 4), and the results indicate that the large SHG in these structures can be optimized through the design of the multilayered region.

To study the spatial extent of the nonlinearity in sample M as a function of depth, we performed etching experiments in dilute hydrofluoric acid on localized areas of the sample. After each etch, the SHG in the remaining thickness of the sample was remeasured, with the results summarized in Fig. 5. The nonlinearity is located predominantly (>95%) in the multilayered stack, which confirms our hypothesis that silica layers can be used to achieve a high degree of control over the thickness and the location of the $\chi^{(2)}$. We also expect the PECVD process to offer tighter control over impurities in the deposited layers than the flame hydrolysis process that is used to manufacture synthetic silica glass, where the impurity levels may vary significantly between different ingots [15]. Therefore, the amount of impurities that participate in the poling process in our multilayered structures is expected to be consistent between devices that are fabricated at different times, which leads to improved reproducibility, and allows a controlled introduction of dopants into the multilayered structure as a means of enhancing the induced nonlinearity.

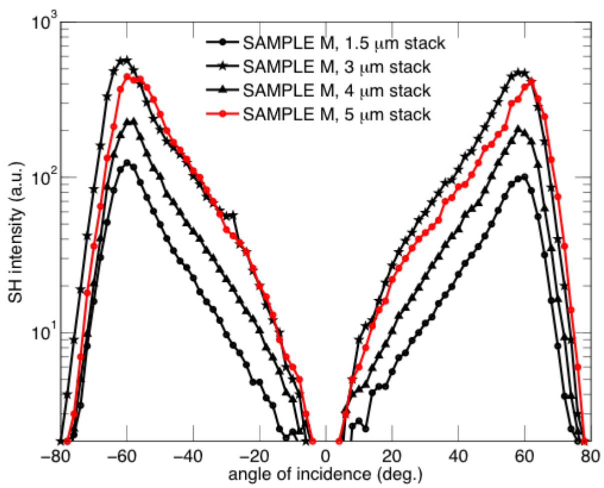

Fig. 4. SHG in thermally poled multilayered structures with varying total stack thicknesses ranging from 1.5 to 5 μm (with the individual layer thicknesses remaining at 75 nm for all samples).

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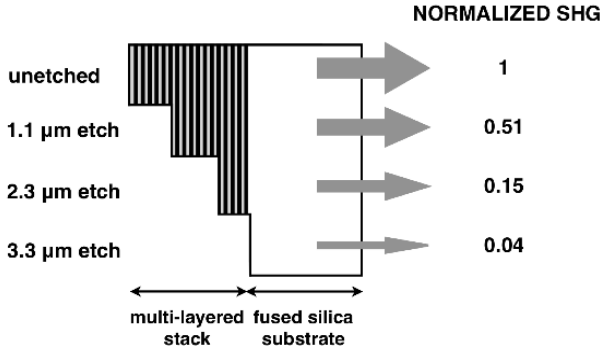

Fig. 5. Remaining SHG in etched thermally poled multilayered structure. Over 95% of the SHG in sample M is concentrated in the 3 μm-thick multilayered stack.

When germanium is introduced into the silica glass matrix, it substitutes for Si^{4+} in the SiO_4 tetrahedra. Unlike other dopants, germanium incorporates into the lattice without any charge-compensating ions because it has the same number of valence electrons as silicon [20]. The enhanced nonlinearity in samples G and M is therefore likely due to a combination of a few factors. First, defects at the interfaces between the germanium-doped and undoped silica act as barriers against the migration of the positive charges, an effect that is consistent with previous observations in poled fibers [21]. These barriers can be advantageous in one of two ways: (1) the accumulation of alkali impurity ions in the vicinity of the interfaces may create strong localized nonlinearity peaks; or (2) the barriers prevent the detrimental positive ions that are injected from the anode from moving further into the sample. We suspect that the two pairs of layers in sample G do not block charge migration as well as the multiple interfaces in sample M [22], which may explain the shape difference (in addition to the magnitude difference) between the Maker fringe patterns of these two samples in Fig. 3. Second, experiments have shown that silica glass with germanium content less than 0.1 mole% (as is the case in sample M) has a lower ionic conductivity compared to undoped silica [23], which furthers slow down any positive charges that may have overcome the interface barrier. And third, when doped with germanium, the refractive index of silica glass increases, thus raising its $\chi^{(3)}$ and in turn the induced effective $\chi^{(2)}$ [22]. A further increase in the $\chi^{(3)}$ of germanium-doped layers may also occur during the poling process [24].

Is the large nonlinearity enhancement observed here limited to layers with germanium-doped silica? We do not believe that this is the case. Our results suggest that the presence of interfaces is the most important factor, and thus it is likely that an enhancement in the SHG will also be observed in multilayered structures with different dopants. In our previous work on corona poled structures with thick layers of phosphorus- and boron-doped silica [25], we showed that a 14-fold increase was obtained in the SHG in phosphorus-doped structures but very little improvement was observed in similar boron-doped structures, which clearly indicates that design optimizations are possible. The use of multilayered structures with glasses that have a large intrinsic $\chi^{(3)}$ (for instance, lead glass [16]) may also yield interesting results. However, to observe the enhanced effect in germanium-doped glass is encouraging since it is the dominant material for the fabrication of low-loss waveguides and fibers. Moreover, the higher refractive index of germanium-doped films raises the average refractive index of the stack, which creates a planar waveguide structure in which an optical mode could be guided, achieving maximum overlap with the nonlinear region that is localized within the stack. Although here we study only planar structures, the same idea can be extended to cylindrical waveguiding structures. An optical fiber with a multilayered silica-based core, where the multilayered design is realized during the fiber preform preparation process, can be thermally poled as illustrated in Fig. 6 by making use of in-fiber electrodes running parallel to the core [26, 27], thereby realizing a second-order nonlinear fiber.

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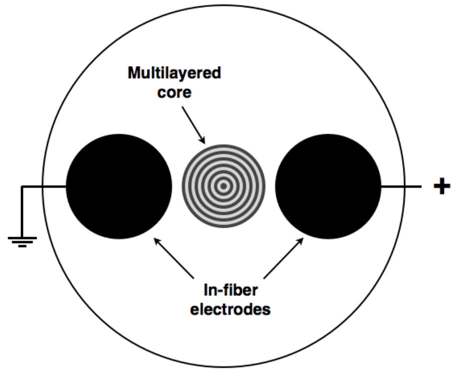

Fig. 6. A conceptual setup for the realization of an efficient second-order nonlinear fiber by thermal poling of a multilayered silica-based core.

7. Conclusion

In conclusion, thermal poling of multilayered silica-based structures was shown to produce a 204-fold enhancement in the induced nonlinearity compared to poled bulk silica glass. This new scheme to create relatively thick, well-controlled nonlinear regions may be further enhanced by the optimization of the thermal poling conditions for the multilayered glass structures. This approach has far-reaching implications for the design of practical silica-based devices with second-order optical nonlinearities, where the doped layers are used as building blocks to create highly customizable artificially created nonlinear materials. Some of the most prominent applications that could benefit from such engineered silica glass nonlinear materials include: (1) all-silica monolithic integration of electro-optic switches and modulators in the PLC platform (as an alternative for the multi-material systems that are in use today); (2) frequency doubling of rare-earth doped fiber lasers to create rugged and inexpensive lasers in the visible spectral region; (3) frequency doubling of high-power pulsed lasers (without concern for damage owing to the high optical damage threshold of silica); and (4) realization of an all-fiber monolithic single-photon source for quantum cryptography applications. The results presented in this paper give a strong indication that the multilayered approach is the key to overcoming the existing challenges of poled glass devices, thus opening the door to practical implementations of efficient active devices in silica glass.

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