Blue-light generation by quasi-phase-matched frequency doubling in thermally poled optical fibers

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Periodically patterned second-order nonlinearities are created in optical fibers by thermal poling in vacuo. Cw quasi-phase-matched frequency conversion to the blue is demonstrated. The measured bandwidth, 0.78 nm cm, is close to the theoretical prediction.

Following the initial report by Myers et al. of second-order nonlinearities of the order of 1 pm/V in thermally poled silica, there has been considerable interest in the possibility of efficient second-harmonic (S.H.) conversion in glass waveguides and fibers. A problem encountered in thermal poling is the spreading out of poled regions that occurs when one attempts to create high-resolution $\chi^{(2)}$ structures by using a patterned electrode. The elimination of this spreading is extremely important for the creation of quasi-phase-matched $\chi^{(2)}$ gratings and more-complicated patterns for efficient parametric frequency conversion (in glass and also perhaps in ferroelectric crystals such as LiNbO$_3$ and LiTaO$_3$) and for thermal poling of optical fibers. Although we have indicated that this problem can be sidestepped by selective erasure of a uniformly poled sample by use of a focused electron beam, we recently discovered that the spreading is caused by electrical breakdown in the air. This means that the problem can easily be eliminated by poling in vacuum. The spreading effect probably was one of the reasons for the small value of the nonlinear coefficient (of the order of $5 \times 10^{-3}$ pm/V) in the first demonstration of periodic thermal poling of fused silica.

In this Letter we report cw quasi-phase-matched frequency conversion to the blue in thermally poled silica fibers.

Before working on fibers, we started poling a flame-fused-silica glass (HeraSIL 1) sample $\sim 2$ mm thick. A periodic chromium anode of 20-μm pitch and total length 0.7 mm, with a 1:1 ratio of individual electrode width to electrode spacing, was photolithographically produced upon a silica (Suprasil) substrate. The anode and a uniform stainless-steel cathode were pressed to opposite sides of each silica glass sample. Thermal poling was carried out at 4.3 kV and 280 °C for 15 min in both an evacuated chamber at $1.2 \times 10^{-5}$ mbars and in air.

After poling the samples in vacuum and in air, we tested them for evidence of S.H. generation, using a mode-locked and Q-switched Nd:YAG laser operating at 1064 nm. Near-field S.H. patterns were imaged by a CCD camera connected to a TV monitor. As is evident from the photographs in Fig. 1, poling in vacuum gives a much better on/off ratio than in air.

The two fibers used in the poling experiments had Ge-doped silica cores and fused-silica cladding. The numerical aperture in each case was 0.09, and the core and the outer diameters were 16 and 125 μm for the first fiber (A) and 6 and 90 μm for the second fiber (B). The cutoff wavelengths were 690 and 1880 nm in fibers A and B, respectively. The OH concentration was 80 and 150 parts in 10$^6$, respectively, in the core and in the cladding (formed from HeraSIL 1). Regions $\sim 8$ mm long were side polished to within 1 μm of the core by a wheel polishing technique. The side-polished fibers were placed on top of 2-mm-thick silica substrates, manufactured by the same method as the starting tubes (HeraSIL 1), and the final assembly was sandwiched between two electrodes with the anode on top of the polished fiber surface. Thermal poling and vacuum parameters were the same as for bulk glass substrates.

We used a cw Ti:sapphire laser with a tuning range from of 780–880 nm to test periodically poled fibers. To achieve quasi-phase-matching in the optical fiber approximately in the middle of the tuning range of Ti:sapphire laser it was necessary to create a pitch of $\sim 28$ μm. Such a pitch may be achieved with a 20-μm periodic anode by a tilt through 45°. In this way we periodically poled $\sim 1$ mm of fiber A.

Fig. 1. Near-field second-harmonic patterns in thermally poled fused silica with a periodic anode and a planar cathode in vacuum (left-hand side) and with air (right-hand side). Note the greatly improved contrast (which is not clear) for poling in vacuum.

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The resulting power-versus-wavelength behavior is shown in Fig. 2. Several phase-matching peaks are apparent, corresponding to different second-order nonlinear interactions among different-order modes. Keeping the wavelength constant on one of the peaks and varying the power, we verified the expected frequency-doubling quadratic relationship. The maximum blue light detected was 20 pW, corresponding to ~200 mW of infrared pump in the fiber core.

We applied the same periodic poling technique to single-mode fiber B. A mask of pitch 28 µm and 6 mm long was used to create a grating suitable for frequency doubling near 870 nm. The S.H. power as a function of the fundamental wavelength is shown in Fig. 3. This dependence shows a well-defined main peak at 860 nm of ~1.3-nm bandwidth, together with weak side peaks at longer wavelengths. The small discrepancy between experimental and predicted phase-matching wavelengths is due to the uncertainty in the Sellmeier coefficients that we used in defining the dependence of the refractive index on the wavelength. We related the presence of side peaks at longer wavelengths to a chirp in the grating. In Fig. 4 the measured square-law dependence of the second-harmonic signal on the fundamental power in the single-mode fiber is shown. The maximum blue-light power detected was ~400 pW, corresponding to a fundamental power in the fiber of 100 mW. An increase of a factor of ~40 in the conversion efficiency in comparison with that of multimode fiber has thus been obtained.

However, the effective nonlinear coefficient for the grating structure is still more than 1 order of magnitude lower than the value expected in a poled fiber structure with a nonlinearity of 1 pm/V. We believe that this reduction is related to a combination of grating imperfections (slight chirping and randomness in the length of the individual poled sections) and of inexact overlap between the poled layer and the fiber core. The presence of a parabolic chirp is suggested by the side peaks in the phase-matching curve, all at longer wavelengths with respect to the main peak. The physical reason for this chirp could be related to the polishing technique, which involved the use of a wheel and may introduce some symmetrical change in the side-polished fiber thickness. Indeed, a calculated phase-matching curve obtained for a grating of 6-mm total length with a given parabolic chirp of the length of the individual poled sections (inset in Fig. 5) and Gaussian random fluctuations in the position of the walls between poled regions of standard deviation ~6.7 µm (Fig. 5) is very close to the experimental wavelength dependence (Fig. 3). The chirp introduces side peaks at longer wavelength, and the strongest ones are at ~862 and ~865 nm. However, the magnitude of this given chirp is not great enough to modify substantially the bandwidth of the main peak at 860 nm (the calculated value of the bandwidth of the main peak for a perfect grating of 6 mm length is ~1.3 nm), although it lowers the corresponding conversion efficiency (the bandwidth scales inversely with interaction length, and the conversion efficiency scales quadratically with interaction length). The random fluctuation implies dependent poled section lengths but does not substantially change the shape of the phase-matching curve because it does not introduce regions with a different period. As a result of these imperfections the conversion efficiency is reduced by slightly more than 1 order of magnitude with respect to the perfect grating. But the main reason for the small conver-
sion efficiency, especially in the single-mode fiber, is the low value of the overlap integral between the fiber mode and the poled region. The estimated value of the effective nonlinear coefficient, including the overlap factor, in the multimode fiber was ~4 times higher than the same value in the single-mode fiber—probably because of the better overlap between the modes and the \( \chi^{(2)} \) grating. At present it is difficult to estimate the value of the overlap factor because of the uncertainty in the location of the poled layer in the fiber. Nevertheless it is possible to predict that this factor may be increased by optimization of the distance between the flat fiber surface and the fiber mode and by optimization of poling parameters, e.g., poling time.

It is worth noting that the experimental bandwidth of phase-matched S.H. conversion in poled silica fiber, which is very close to the theoretical value (0.79 nm cm), is an order of magnitude larger than in an equal length of the periodically poled bulk lithium niobate (0.06 nm cm). Moreover, the group-velocity mismatches are ~130 fs/mm and ~1.8 ps/mm, respectively, at 860 nm. This may be of importance in short-pulse work, where large acceptance bandwidths and long interaction lengths are desirable.

Considerable improvements are expected by optimization of the poling process, in particular by improvement of the overlap between the fundamental and S.H. modes with the poled region.

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