Grating-assisted phase-matched second-harmonic generation from a polymer waveguide

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Diffraction gratings can be used to achieve phase matching between the fundamental modes of a slab or confined waveguide. Compared with the usual techniques such as quasi-, Čerenkov, and intermodal phase matching, the matching method used here involves spatial harmonics of the guided electromagnetic field that are generated by the corrugated grating. This grating acts simultaneously as a linear waveguide coupler at both the pump and harmonic frequencies. Using the third spatial harmonic, we report what we believe to be the first observation of grating-assisted phase-matched second-harmonic generation between counterpropagating TM_0 modes of an organic waveguide.

The development of compact blue laser sources has attracted great interest because of their potential use for high-density optical recording, laser printers, etc. For this reason there is great activity in the area of frequency doubling of laser diodes [(Al,Ga)As]. High conversion efficiencies have been reported in inorganic nonlinear (NL) waveguides[1] (KTP, $LiTaO_3$, $LiNbO_3$) that take advantage of strong confinement and interaction lengths of the order of centimeters. The phase mismatch between the fundamental and the second-harmonic (SH) waves, caused by chromatic dispersion, is compensated for by use of different phase-matching schemes.

Polymer waveguides provide new perspectives in the development of efficient frequency-doubling devices. The key advantage of using polymers as NL media is their ease of fabrication and their large nonresonant optical nonlinearity. But there still exist some important technical problems that limit the performance of the SH-generation devices. Phase matching over long distances requires high precision on the layer thickness and homogeneous polarization, which is difficult to achieve by standard techniques (e.g., spin coating, corona polarization).[2,3] In several materials some additional problems occur, including residual absorption in the blue region and difficult end-fire coupling to the polymer layer. Because of these problems it can be advantageous to use grating couplers and to apply phase-matching schemes such as the Čerenkov technique,[4] distributed outcoupling,[5] or phase matching based on the use of spatial harmonics.[6] The aim of this Letter is to report what is to our knowledge the first experimental demonstration of SH generation in a poled polymer slab waveguide, in which the phase-matching process involves only spatial harmonics of the guided-field wave vectors. This is what we call grating-assisted phase matching.

Phase matching in waveguides with periodic corrugation was first proposed by Somekh and Yariv.[6] SH generation by use of artificial periodic structures was further analyzed theoretically by Tang and Bey.[7] When a plane wave is falling on a periodic corrugated waveguide, a rigorous electromagnetic theory[8] or a method combining a linear theory of diffraction with a coupled-mode formalism[9] can be used to study the SH generation process. The SH device presented here is a simple corrugated grating that operates as an incoupler and an outcoupler for the slab waveguide (Fig. 1). For simplicity we consider here only the case for which the polarization of the light at both ω and 2ω is transverse magnetic (TM). When a mode of the structure is excited, the guided light at the pump frequency can be written as[10]

$$H_z^{\omega}(x,y) = \sum_p H_{z,p}^{\omega}(y)\exp\left[i\left(k_0^{\omega} n_{\text{eff}}^{\omega} + p\,\frac{2\pi}{d}\right)x \right], \quad (1)$$

where d is the grating period, $k_0^{\omega} = \omega/cn_{\text{eff}}^{\omega}$ is the wave vector of the guided mode, and p labels the order of diffraction. Thus the guided field is the sum of a fundamental component $p = 0$ and many spatial harmonics with $p \neq 0$. The field generated at 2ω is

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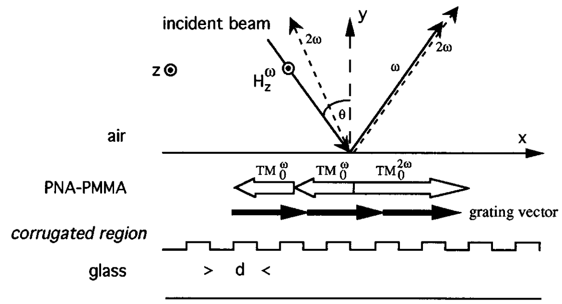

Fig. 1. Schematic of the device with the corrugated grating on the glass substrate and *para*-nitroaniline/poly(methyl methacrylate) (PNA-PMMA) as the guiding NL layer. The arrows in the air designate the incident beam and the reflected orders. The arrows in the guiding layer correspond to the TM_0 modes at the fundamental and harmonic wavelengths phase matched through the wave vector associated with the grating.

proportional to the square of the pump field:

$$H_z^{2\omega}(x,y) \propto \sum_{p,q} H_{z,p}^{\omega}(y) H_{z,q}^{\omega}(y) \times \exp\left\{ i\left[2k_0^{\omega} n_{\mathrm{eff}}^{\omega} + (p+q)\frac{2\pi}{d}\right] x\right\}. \quad (2)$$

Moreover, any guided field at frequency 2ω obeys a relation similar to Eq. (1):

$$H_z^{2\omega}(x,y) = \sum_m H_{z,m}^{2\omega}(y)\exp\left[i\left(k_0^{2\omega} n_{\mathrm{eff}}^{2\omega} + m\frac{2\pi}{d}\right) x\right]. \quad (3)$$

Phase matching is achieved when the constants of propagation of the generated space harmonics of the fundamental and the harmonic waveguide modes equalize:

$$2k_0^{\omega}\ \mathrm{Re}(n_{\mathrm{eff}}^{\omega}) \pm k_0^{2\omega}\ \mathrm{Re}(n_{\mathrm{eff}}^{2\omega}) + (p_\omega + q_\omega + m_{2\omega})(2\pi/d) = 0. \quad (4)$$

The real part of the effective index is chosen to be positive. The negative sign in Eq. (4) must be used in the copropagating scheme (fundamental and harmonic waves propagating in the same direction), and the positive sign must be used for the counterpropagating scheme. Here we chose the counterpropagating scheme because it requires short periodic gratings, thereby avoiding an important electromagnetic leakage. In the reported experiment, phase matching is obtained for $(p_\omega + q_\omega + m_{2\omega}) = -3$.

The grating is fabricated by an electron-beam pattern generator on a poly(methyl methacrylate)-coated glass substrate (Corning Chemcor 0317) followed by neutral ion-beam etching. The grating dimensions are 5 mm × 5 mm. The groove profile is nearly rectangular, with a periodicity of $d = 500$ nm and a groove depth of 120 nm. The line-space ratio is 1:1. A thin film of poly(methyl methacrylate)/*para*-nitroaniline side-chain polymer is deposited on the glass substrate by spin coating. The film is poled by a corona discharge through the glass substrate. The linear characteristics of the device are analyzed with a cw Nd:YAG laser and the 514-nm beam of an argon laser. The effective index of the TM_0 mode at 1064 nm is found to be $n_{\mathrm{eff}} = 1.51 + i8.25 \times 10^{-6}$ by use of the M-line technique. Using the same method, we obtain $n_{\mathrm{eff}} = 1.64 + i5.4 \times 10^{-4}$ for the TM_0 mode at 514 nm. The waveguide thickness is $w = 580$ nm. NL optical measurements are carried out with a passively Q-switched Nd:YAG laser at 1.064 μm and with an optical-parametric-oscillator laser pumped by the third harmonic of a seeded Nd:YAG laser. Both lasers have a pulse duration of 10 ns and a repetition rate of 10 Hz. The optical parametric oscillator is used to approach the phase-matching condition by tuning the pump wavelength. The NL coefficient of the poled sample is measured with the Nd:YAG laser at 1064 nm. A comparison of the SH signal from the *para*-nitroaniline layer with the signal obtained in a Maker fringe experiment with a quartz slab yields the absolute value of the NL coefficient, $\chi_{33} = 7.3$ pm/V.

All experiments (linear and NL) are in TM polarization for both pump and SH beams. A monochromator and a 4-mm-thick KG5 filter are placed in front of a photomultiplier tube to detect the selected SH light. A small part of the fundamental beam is split off and frequency doubled in a KDP single crystal. This signal serves as a reference to compensate for intensity fluctuations of the laser. The device is placed on a rotation stage, and the detection system, placed in the reflected beam, can rotate independently around the same axis as the grating. The SH signals from the sample and the reference are sent to a boxcar averager. Finally, a ratio of both is taken with a computer. The efficiency is defined as the ratio between the SH power density and the square of the pump power density. The incident beam has a peak power of 56.3 kW, with a spot size of 4 mm × 0.5 mm.

In the experiment we first measure the SH efficiency away from the phase-matching condition. The wavelength is fixed, and the angle of incidence θ is varied. When the projection of the wave vector at the pump frequency is equal to that of a space harmonic of the waveguide mode at ω or 2ω, resonance occurs. This results in an enhancement of the SH efficiency. According to Ref. 9, the pertinent equations are

$$-k_0^{\omega}\ \mathrm{Re}(n_{\mathrm{eff}}^{TM_0^{\omega}}) - l\frac{2\pi}{d} = k_0^{\omega}\sin\theta_1, \quad (5)$$

$$\frac{1}{2}\left[k_0^{2\omega}\ \mathrm{Re}(n_{\mathrm{eff}}^{TM_0^{2\omega}}) - l'\frac{2\pi}{d}\right] = k_0^{\omega}\sin\theta_2. \quad (6)$$

In Eqs. (5) and (6), θ_1 and θ_2 denote the value of the angle of incidence θ of the pump beam (Fig. 1) for which resonance occurs at ω ($\theta = \theta_1$) and at 2ω ($\theta = \theta_2$). The TM_0 mode at the pump frequency is coupled backward by the first order $l = -1$, whereas the TM_0 mode at the harmonic frequency is coupled forward by the first order $l' = +1$. The phase-matching condition [Eq. (4)] is fulfilled when $\theta_1 = \theta_2$. We can achieve this in the experiment by tuning the wavelength.

Figure 2 shows the SH signal versus the angle of incidence recorded for two different wavelengths. At the pump wavelength $\lambda = 1044$ nm the TM_0^{ω} and $TM_0^{2\omega}$

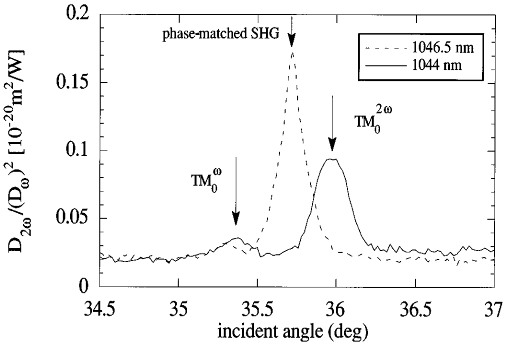

Fig. 2. SH efficiency measured as a function of the incident angle in the specular reflected order for two different wavelengths of the pump beam. The phase-matching condition is fulfilled at $\lambda = 1046.5$ nm and $\theta = 35.72°$. SHG, second-harmonic generation.

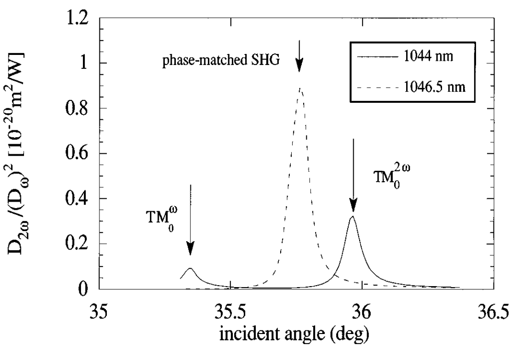

Fig. 3. Numerical calculations of the SH efficiency with the plane-wave assumption versus the incident angle corresponding to the experimental data shown in Fig. 2.

modes are resonantly excited at two different incident angles, far from the phase-matching condition. The angles corresponding to this excitation can be determined with Eqs. (5) and (6). An enhancement of the SH efficiency is clearly observed at the phase-matching condition ($\lambda = 1046.5$ nm, $\theta = 35.72°$). The corresponding effective indices of the fundamental modes at ω are $n_{\text{eff}} = 1.51$ and $n_{\text{eff}} = 1.63$ at 2ω.

The obtained resonance width and height of the measured curves must be seen in the context of the characteristics of the incident beam (optical parametric oscillator spectral width $\Delta\lambda = 0.5$ nm, beam divergence $\Delta\theta = 0.14°$). We obtain a much lower efficiency for the TM_0^{ω} resonance at 1.044 μm in Fig. 2 than for the $TM_0^{2\omega}$ resonance. We explain this in terms of the divergence of the incident beam, which does not fit the very narrow TM_0^{ω} resonance ($\Delta\theta = 0.01°$ in linear measurements). We also obtain thinner resonances and higher efficiencies [$\eta(TM_0^{\omega}) = 0.08 \times 10^{-20}$ m^2/W, $\eta(TM_0^{2\omega}) = 0.13 \times 10^{-20}$ m^2/W] when the Nd:YAG laser is used at 1.064 μm (beam divergence $\Delta\theta = 0.06°$) on the same device. This result shows that the measured efficiencies are determined mainly by the characteristics of the incident beam and not by eventual inhomogeneities in waveguide thickness, poling, or grating characteristics.

In a first attempt we simulate the experimental situation by numerical calculation. We use the rigorous method of Ref. 8 with the plane-wave assumption. We obtain good agreement with respect to the $TM_0^{2\omega}$ resonance, but the calculated resonance curve of the TM_0^{ω} mode is much higher than in the experiment. Obviously the spectral width of the beam, which is not included in the calculations, affects the TM_0^{ω} resonance much more than the $TM_0^{2\omega}$, as already discussed. Therefore we have artificially increased the imaginary part of the refractive index n^{ω} at the pump frequency, which increases the imaginary part of the mode propagation constant n_{eff}^{ω}. The exact value of $\text{Im}(n^{\omega}) = 10^{-3}$ was chosen to ensure a correspondence between the experimental and theoretical maxima (Fig. 3). This value can also be roughly estimated when the resonance width that is due to the beam divergence of the optical parametric oscillator is linked with n_{eff}^{ω} through $\text{Im}(n_{\text{eff}}^{w}) = \cos\theta\Delta\theta$. The complex value of n_{eff}^{ω} describes the resonant behavior of the device owing to waveguide losses, grating coupling, and excitation by the incident beam.

In conclusion, we have observed phase matching in a NL diffractive device, using spatial harmonics. The counterpropagating scheme is well adapted to this kind of frequency doubler. The rather low efficiency $\eta = 0.18 \times 10^{-20}$ m^2/W is due mainly to the large spectral width of the pump beam. Plane-wave calculations yield $\eta = 2.88 \times 10^{-14}$ m^2/W for the same device, assuming a monochromatic incident beam and including the propagation losses of the $TM_0^{2\omega}$ mode. Therefore the efficiency can be increased by use of a narrow-bandwidth laser with a large spot size. However, when a divergent pump laser with a large bandwidth must be used, a grating with deeper grooves would be better adapted. Moreover, the use of gratings with shorter periods would lead to phase matching involving only one times the grating vector. Because the lower-order space harmonics are more important this would lead to an additional increase of the SH signal. Unfortunately, gratings with shorter periods or deeper grooves are not yet available to us.

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