Second-harmonic generation in Langmuir–Blodgett waveguide overlays on single-mode optical fiber

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Second-harmonic radiation has been obtained from Langmuir–Blodgett films of *E*-*N*-octadecyl-4-[2-(4-dibutylaminophenyl)ethenyl]quinolinium octadecylsulfate, deposited as a waveguide overlay upon optical fiber that is single mode at the pump wavelength ($\lambda = 1064$ nm). A quadratic relationship between the pump power and second-harmonic intensity was observed. © 1999 Optical Society of America

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Organic materials have larger hyperpolarizabilities than their inorganic counterparts, and therefore various donor–(π-bridge)–acceptor materials have been investigated for nonlinear optical processes, in particular, for second-harmonic generation (SHG).[1] The molecules tend to adopt an antiparallel arrangement but, by use of Langmuir–Blodgett (LB) deposition techniques, the required noncentrosymmetric alignment can be achieved to thicknesses suitable for waveguiding.[2–4] Efficient SHG requires the phases of the interacting beams to be matched and, among the various methods available for LB waveguides, Čerenkov-type phase matching requires the least stringent control of the parameters for frequency conversion.[4–7] Thus, even though most donor–(π-bridge)–acceptor materials are not transparent at visible wavelengths, this is not a problem because the second harmonic propagates in the substrate and not in the film. In this regime the fundamental beam is guided in the LB film and the generated second harmonic is radiated into the substrate, as shown in Fig. 1

Grating and prism couplings have been used with LB waveguides, but the former can suffer from low coupling efficiency and the latter can easily damage the fragile organic overlay. Fiber-optic coupling may be preferable, but, in a previous study, Selfridge *et al.*[8] reported a low conversion efficiency for hemicyanine LB films deposited cylindrically about a 600-μm-diameter core of a multimode optical fiber. This may be attributed, in part, to the fact that only a small proportion of the pump power is guided in propagating fiber modes phase matched to the film and, in part, to the film's being structurally disordered. The film is predominantly noncentrosymmetric (Z type) but, as deposition on the upstroke was accompanied by deposition, albeit slight, on each subsequent downstroke, regions of the film must have a centrosymmetric Y-type character.

We report a configuration in which the light guided by a single-mode optical fiber is evanescently coupled into a LB overlay waveguide and the second harmonic is generated in the form of Čerenkov radiation. A refined deposition technique ensures noncentrosymmetric Z-type alignment of molecules.

The device is based on an in-line fiber-optic channel-dropping filter and consists of a planar LB waveguide upon a side-polished single-mode optical fiber,[9–11] the two being evanescently coupled (see Fig. 2). Optical power is coupled from the optical fiber into the LB film at a particular wavelength λ_m when the phase-matching condition between the fiber and the overlay mode is satisfied, i.e., when the effective refractive indices of the waveguides are equal. Insertion of a phase-matching condition into the eigenvalue equation for the zero-order mode of the three-layer, asymmetric, uniaxial waveguide[12] describes the dependence of λ_{m} on the thickness and the refractive index of the planar waveguide:

$$\frac{2\pi d}{\lambda_m}\sqrt{{n_o}^2 - {n_{\mathrm{eff}}}^2} = \Phi_a + \Phi_{\mathrm{cl}}\,, \tag{1}$$

where n_{eff} is the effective refractive index of the fiber mode ($n_{\mathrm{eff}} = \beta_m 2\pi/\lambda$, where β_m is the mode propagation constant); d and n_o are the thickness and the refractive index of the overlay and Φ_a and Φ_{cl} are polarization-dependent phase-change terms at the overlay–superstrate and overlay–fiber cladding boundaries, respectively. TE and TM polarized light would be expected to couple to the overlay at different wavelengths. Furthermore, λ_m depends on the overlay thickness, and the LB technique permits control to within one molecular length (~3 nm), thus permitting the overlay waveguide characteristics to be defined such that phase matching occurs at the desired pump wavelength. This, coupled with the fact

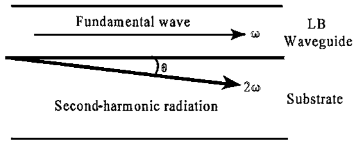

Fig. 1. Schematic diagram of the Čerenkov-type configuration.

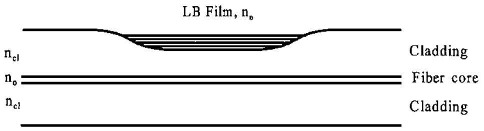

Fig. 2. Schematic diagram of the fiber-optic device.

that some materials, albeit very few, deposit in a noncentrosymmetric Z-type manner, makes the LB deposition technique ideal for the fabrication of such devices. Although it is not apparent from Eq. (1), λ_m also depends on temperature by virtue of the large $\mathrm{d}n/\mathrm{d}T$ characteristics of the overlay and can be thermally tuned. For the LB film-forming material used, $\lambda_m/\Delta T = -0.6$ nm/°C.

The device was fabricated by removal of a portion of cladding from a 30-mm section of fiber by an annular polishing process.[13] The cladding was removed to within approximately 2 μm of the core, monitored by an oil drop test,[14] with a polishing technique previously reported.[9] The optically nonlinear dye, E-N-octadecyl-4-[2-(4-dibutylaminophenyl)ethenyl]-quinolinium octadecylsulfate (Fig. 3), was spread from a dilute chloroform–methanol solution onto the pure water subphase of a Nima Technology LB trough, left for 5 min, and then compressed at 0.5 cm^2 s^{-1} (equivalent to 0.1% s^{-1} of total area). The LB film was transferred to the polished region of the fiber (Spectran SMC-AO1060B; cutoff wavelength 987 nm, core diameter 6.4 μm, cladding diameter 125 μm) by the modified deposition technique previously described, which prevents deposition on the downstroke.[15] Assuming complete deposition, a 184-layer film was obtained at a surface pressure of 35 mN m^{-1}.

Light from a quartz halogen lamp and a monochromator was coupled via a polarizer and a 20× microscope objective lens into the fiber. A fiber-optic state-of-polarization controller was used to define the polarization in the polished region, and a photodiode and a lock-in amplifier detected the output. This arrangement permitted continuous on-line monitoring of the optical fiber–LB overlay waveguide structure during deposition such that the operating wavelength λ_m could be accurately set. The resultant channel-dropping response shows λ_m centered on 1064 nm with a drop in transmission of ~13 dB (Fig. 4).

Čerenkov-radiation-type phase matching requires that the effective refractive index of the fundamental wave propagating in the LB waveguide be greater than the substrate's fundamental refractive index and less than its second-harmonic index; i.e., $n_s^{(2\omega)} > N_{\mathrm{eff}}^{(\omega)} > n_s^{(\omega)}$. The refractive indices of the cladding are $n_s^{(\omega)} = 1.4497$ and $n_s^{(2\omega)} = 1.460$, and, using an equivalent slab waveguide model of optical fibers,[16] we calculated an effective refractive index of 1.452 for the fundamental in the LB film. Light from a Q-switched Nd:YAG laser, operating at 1064 nm with a pulse duration of 10 ns and a repetition rate of 2 Hz, was coupled into the optical fiber, as shown in Fig. 5. The region of fiber coated with the LB film was mounted upon a Peltier thermoelectric element to permit thermal control of the resonant wavelength λ_m and hence the amount of light coupled into the overlay. The second-harmonic intensity relative to the signal from a Y-cut quartz reference was monitored by photomultiplier tubes, and the power of the guided fundamental exiting the fiber was continually measured by a powermeter.

The second-harmonic intensity from the LB overlay is plotted as a function of fiber transmission. We obtained the data shown in Fig. 6 by thermally varying λ_m and consequently the pump power coupled into

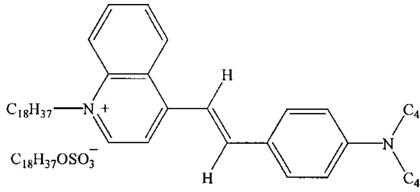

Fig. 3. Molecular structure of the quinolinium dye.

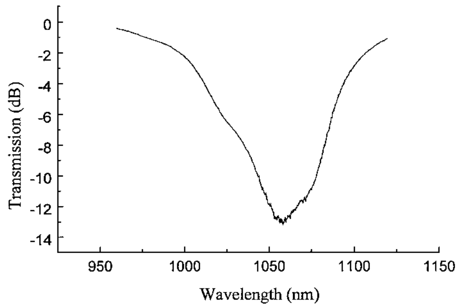

Fig. 4. Normalized spectral response from184 Z-type layers of the quinolinium dye deposited upon the side-polished single-mode optical fiber; maximum coupling to the film occurs at 1064 nm.

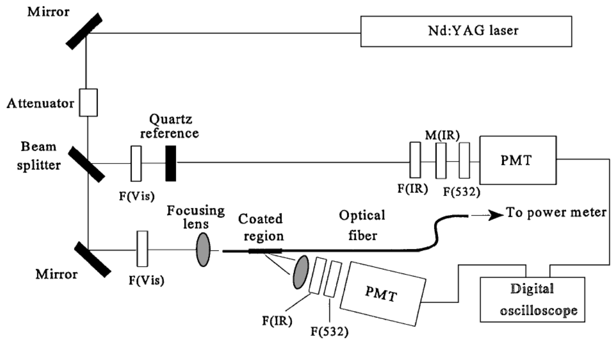

Fig. 5. Schematic diagram of the SHG apparatus: F(vis)'s, visible filters; F(IR)'s, IR filters; F(532)'s, 532-nm bandpass filters; M(IR), IR mirror; PMT's, photomultiplier tubes.

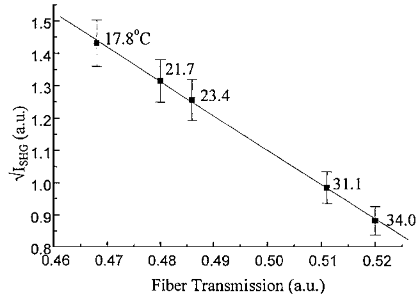

Fig. 6. SHG versus fiber transmission. Constant pump power coupled into the fiber with thermal tuning of λ_m causes the power coupled to the LB overlay to change.

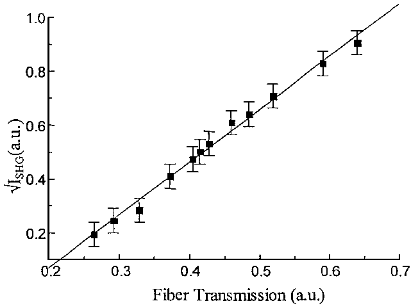

Fig. 7. SHG versus fiber coupled pump power at constant temperature, $T = 21.4\,°C$.

the overlay. In this case the increased transmission is indicative of reduced coupling into the overlay, and the second-harmonic intensity decreases as the square of the transmitted fundamental intensity. In contrast, we obtained the data shown in Fig. 7 by varying the light coupled into the fiber, and therefore into the overlay, at a constant temperature of 21.4 ± 0.1 °C. The slope is positive, and, as expected from theory, the second-harmonic intensity increases as the square of the fundamental intensity.

SHG from a LB waveguide overlay, coated onto a side-polished single-mode optical fiber, has been demonstrated for the first time to our knowledge. The second harmonic is generated as Čerenkov radiation. It is emitted into the area surrounding the LB overlay and is not guided by the LB film or fiber core. Preliminary results have shown the expected quadratic SHG dependence on pump power. Furthermore, as depositing the layers either on the upstroke, as described herein, or on the downstroke alters the orientation of the molecular dipole within the LB overlay, optimization of conversion efficiency can be realized by fabrication of susceptibility-inverted waveguide structures. These have been described for LB waveguides by use of conventional coupling techniques.[3,7]

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References

1. C. Bosshard, K. Sutter, P Prêtre, J. Hulliger, M. Flörsheimer, P. Kaatz, and P. Günter, *Organic Nonlinear Optical Materials*, Vol. 1 of Advances in Optics, A. F. Garito and F. Kajzar, eds. (Gordon & Breach, Basel, Switzerland, 1995).
2. G. J. Ashwell, "Langmuir–Blodgett films: molecular engineering of non-centrosymmetric structures for second-order nonlinear optical applications," J. Mater. Chem. (to be published).
3. T. L. Penner, H. R. Motschmann, N. J. Armstrong, M. C. Ezenyilimba, and D. J. Williams, Nature **367,** 49 (1994).
4. K. Clays, T. L. Penner, N. J. Armstrong, and D. R. Robello, Proc. SPIE **1775,** 326 (1992); T. L. Penner, N. J. Armstrong, C. S. Willand, J. S. Schildkraut, D. R. Robello, and A. Ulman, Nonlinear Opt. **4,** 191 (1993); K. Clays, N. J. Armstrong, and T. L. Penner, J. Opt. Soc. Am. B **10,** 886 (1993).
5. C. Bosshard, M. Flörsheimer, M. Küpfer, and P. Günter, Opt. Commun. **85,** 247 (1991).
6. W. Hickel, B. Menges, O. Althoff, D. Lupo, U. Falk, and U. Scheunemann, Thin Solid Films **244,** 966 (1994).
7. N. Asai, H. Tamada, I. Fujiwara, and J. Seto, J. Appl. Phys. **10,** 4521 (1992); I. Fujiwara, N. Asai, and V. Howarth, Thin Solid Films **221,** 285 (1992); N. Asai, I. Fujiwara, H. Tamada, and J. Seto, Mater. Res. Soc. Symp. Proc. **328,** 91 (1994).
8. R. H. Selfridge, T. K. Moon, P. Stroeve, J. Y. S. Lam, S. T. Kowel, and A. Knoesen, Proc. SPIE **971,** 197 (1988).
9. R. B. Charters, S. E. Staines, and R. P. Tatam, Opt. Lett. **19,** 2036 (1994).
10. D. Flannery, S. W. James, R. P. Tatam, and G. J. Ashwell, Opt. Lett. **22,** 567 (1997).
11. R. B. Charters, A. P. Kuczynski, S. E. Staines, R. P. Tatam, and G. J. Ashwell, Electron. Lett. **30,** 594 (1994).
12. V. Ramaswamy, Appl. Opt. **13,** 1363 (1974).
13. C. D. Hussey and J. D. Minelly, Electron Lett. **24,** 805 (1988).
14. O. G. Leminger and R. Zengerle, J. Lightwave Technol. **LT-3,** 864 (1985).
15. S. S. Johal, S. W. James, R. P. Tatam, and G. J. Ashwell, Meas. Sci. Technol. **10,** 60 (1999).
16. A. Sharma, J. Kompella, and P. K. Mishra, J. Lightwave Technol. **8,** 143 (1990).