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Surface-emitted green light generated in Langmuir–Blodgett film waveguides

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We demonstrate second-harmonic generation due to counterpropagating beams in planar waveguides of 2-docosylamino-5-nitropyridine (DCANP). The DCANP molecules were deposited by Langmuir–Blodgett techniques and have a preferred alignment within the substrate plane. Four-layer waveguide structures were used to optimize the trade-off between propagation loss and efficient surface-emitted green light.

Langmuir–Blodgett (LB) films have many attractive features for applications to nonlinear optics. Since they can be deposited one monolayer at a time with a net second-order susceptibility, precise film thickness (and therefore effective index in waveguides) control for second-harmonic generation (SHG) is possible.[1–6] However, until recently the waveguide propagation losses have been too high for the efficient SHG needed for efficient blue sources. The development of low loss LB guiding films has now led to the demonstration of efficient collinear SHG.[1–6] With the flexibility of the deposition technique it is possible to reverse the direction of the nonlinearity within a film. This should ultimately allow the large diagonal elements of the nonlinear susceptibility tensor to be used for efficient SHG with fundamental and doubled guided waves with different mode numbers.

SHG normal to the waveguide surfaces by the mixing of oppositely propagating guided waves has features and applications different from the usual copropagating SHG, e.g., wavelength demultiplexing, picosecond signal processing, etc.[7–11] Recently the work in this field has concentrated on GaAs-based waveguide structures.[11–16] Due to their large nonlinear optical coefficients organic materials provide a promising alternative to semiconductor devices. Efficient surface emission of SHG has already been demonstrated in poled polymers of 4-dimethylamino-4′-nitrostilbene side-chain channel waveguides.[17] In this letter we report the first experiments on an alternative approach to using organic materials, planar waveguides made from a LB film, 2-docosylamino-5-nitropyridine (DCANP).[1–5]

LB films offer new opportunities for increasing the efficiency of this process. Since the output signal only traverses a few optical wavelengths of the sample, the large resonant nonlinearities of organics can be used. Furthermore, although this interaction is not subject to the stringent limitations of wave vector matching encountered in copropagating SHG, wave vector matching in the waveguide in the direction normal to the surface can considerably enhance the efficiency. In semiconductors this has been achieved by using molecular beam epitaxy growth techniques to modulate the magnitude of the nonlinearity.[11] Using LB films desired thicknesses can be fabricated to an accuracy of about 4 nm and the direction of the nonlinearity can be periodically reversed.[18] In the current case we do not reverse the nonlinearity but do demonstrate a four-layer geometry which includes an inactive layer to improve the propagation loss characteristics of the waveguides.

The linear and nonlinear optical properties of DCANP are well established and both Cerenkov and phase-matched, copropagating SHG using modal conversion in waveguide configuration have been reported.[2–5] The LB film fabrication procedure of DCANP induces a preferred alignment of the molecules along the dipping direction and films which are stable over years. This leads to a net dipole moment within the substrate plane and to a Y-type "herringbonelike" structure [Fig. 1(a)].

The intensity for SHG by counterpropagating beams can be described by $I(2\omega)=A^{\mathrm{NL}}I_{+}(\omega)I_{-}(\omega)$ where $I_{\pm}(\omega)$ are the line intensities (in W/m) of the fundamental beams and A^{NL} is the nonlinear cross section.[7] Here

$$A^{\mathrm{NL}}=\epsilon_0\omega k_0(2\omega)n_c(2\omega)|C|^4|S(\infty)|^2\ . \tag{1}$$

A^{NL} is strongly dependent on the superposition integral

$$S(\infty)=\int_{-\infty}^{+\infty}\frac{d_{33}(y')f^2(y')}{n_f(2\omega)}\exp[ik_0(2\omega)n_f(2\omega)y']dy'\ , \tag{2}$$

where $k_0(2\omega)=2\omega/c$, and $n_c(2\omega)$ and $n_f(2\omega)$ are the complex refractive indices of the cover and the film, respectively. $f(y')$ is the guided mode profile and C is the power normalization constant of the guided mode. Figure 2 shows the theoretical nonlinear cross section versus thickness for three- and four-layer geometries of DCANP. The substrate is either a high-index glass (AF45) or Pyrex. For the four-layer case an optically inactive TiO_2/SiO_2 film was used. The following parameters were utilized for the calculation: wavelength $\lambda=1064$ nm, nonlinear optical coefficient $d_{33}=7.8$ pm/V ($\lambda_{\max}=375$ nm), thickness of linear TiO_2/SiO_2 layer $t_{\mathrm{LIN}}=145$ nm, refractive indices at frequency ω: $n_{\mathrm{LIN}}=1.75$, $n_S=1.516$ (or $n_S=1.4667$ for Pyrex), $n_{\mathrm{DCANP}}=1.573$, refrac-

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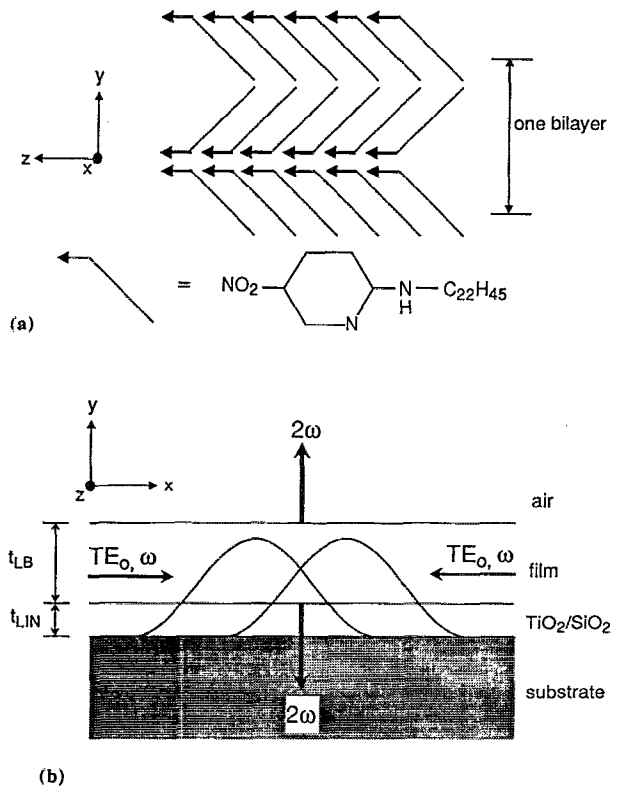

FIG. 1. (a) Orientation of the DCANP molecules in the LB film. The dipping procedure induces the noncentrosymmetric Y-type structure. The arrows indicate the direction of the dipoles. (b) Sample geometry and schematic diagram of the counterpropagation experiment.

tive indices at frequency 2ω: $n_{\mathrm{LIN}}=1.85$, $n_S=1.531$ (or $n_S=1.4743$ for Pyrex), $n_{\mathrm{DCANP}}=1.624$. Typically A^{NL} is larger for emission towards the substrate than towards the air due to the asymmetry of the electric fields in the waveguide.

In our experiments we used the four-layer structures mainly for two reasons. First, the attenuation losses of the waveguide can be strongly decreased for LB films as has been demonstrated in Ref. 4. Second, fewer layers of DCANP molecules have to be deposited to obtain similar nonlinear cross sections as for usual three-layer structures. This is clear since optimum conversion efficiencies are obtained for film thicknesses which are separated by $\lambda_0/[2n(2\omega)]$. For a TiO_2/SiO_2 film of thickness $t_{\mathrm{LIN}}=145$ nm, the first peak in A^{NL} accessible above waveguide cutoff for the fundamental occurs for a LB thickness of only 230 nm. Note, however, that this first peak for a single layer film may not always be the largest since the input field confinement may be small if it occurs near cutoff.

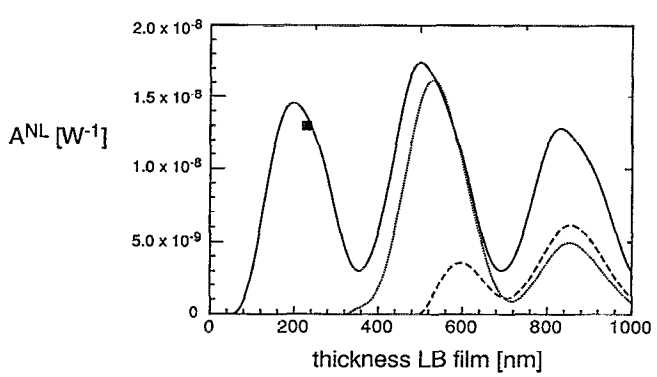

FIG. 2. The nonlinear cross section A^{NL} vs film thickness for three- and four-layer waveguide geometries of DCANP. The lines represent the calculated values, and the (■) is the measured point. The solid curve is for the four-layer structure. The dotted and dashed curves represent the three-layer structure with Pyrex and AF45 as the substrate, respectively.

Commercially available TiO_2/SiO_2 waveguides (substrate glass AF45) from ASI AG (Zurich, Switzerland) were used into which gratings with a period of 1000 mm^{-1} were fabricated by ion milling. Subsequently the surface was cleaned and the LB film was deposited. The exact preparation procedure is described in Ref. 3. Figure 1(b) shows the sample geometry together with a schematic diagram of the counterpropagating SHG process. For our experiment a sample with a linear layer thickness of $t_{\mathrm{LIN}}=145$ nm and a LB film thickness of $t_{\mathrm{LB}}=230$ nm (corresponding to 104 monolayers) was used.

First the attenuation of the waveguide was measured by imaging the streak of the guided wave with a camera (Electrophysics Micronviewer Model 7290) and by subsequent analysis of the streak. We obtained a value of about 5.5 dB/cm for our sample. Note that the loss would have been about 2.5 times larger (and the signal more than 2 times smaller) for operation on the first peak of a guiding film made solely from DCANP.

100 ps pulses from a Q-switched (500 Hz) and modelocked (76 MHz) Nd:YAG laser (Quantronix 416) were split in half and then coupled into both sides of the waveguide to excite TE_0 modes. The overlap in the waveguide was optimized by changing the optical delay in one arm. The coupling gratings were separated by 5.5 mm thus defining the interaction length of the device. The fundamental beams were focused on the grating using cylindrical lenses in order to optimize the coupling. Coupling efficiencies of 11% and 8% were estimated. The peak power of the fundamental in the waveguide was 74 W (average power of 70 μW) yielding a peak input line intensity of 3.46 kW/m. The generated second-harmonic light could be observed perpendicular to the sample surface easily by the eye in room light conditions. The measured second-harmonic power showed the expected quadratic behavior on the intensity of the fundamental. We measured a maximum average power of $P_{\mathrm{av}}(2\omega)=6$ nW corresponding to a peak power of $P_p(2\omega)$ of 12.6 mW. The full divergence angle of the green light was of the order of 0.036 rad. Therefore it could be expected that all the light generated was incident on our detector. From $P(\omega)$ and $P(2\omega)$ the nonlinear cross section A^{NL} could be calculated. Our measured value of 1.3×10^{-8} W^{-1} is in good agreement with the theoretical value of $A^{\mathrm{NL}}=1.4\times10^{-8}$ W^{-1} in Fig. 2 which also shows the measured point.

The signals measured here are adequate for the projected applications. However it is useful to project what the ultimate signal limits might be. In the LB film case, the superposition integral can be optimized by constructing LB layers with periodically reversed nonlinearities and optimized layer

thicknesses, increasing the cross section by about one order of magnitude. Since the generated second harmonic only needs to propagate in the nonlinear medium for a short distance (of the order of 600 nm for the periodically reversed layer case), working with resonantly enhanced nonlinearities is feasible.[17] Assuming operation at the communications wavelength of 1300 nm with different molecules with resonant nonlinearities, values of $d^{(2)}$ in excess of 300 pm/V are feasible and $A^{NL} \rightarrow 2 \times 10^{-4}$ W^{-1}, including enhancements due to periodically reversed layers.[19,20] Using the expression $P(2\omega) = A^{NL} P^2(\omega) L/W$ where L and W are the sample length and channel width, respectively, $P(2\omega)/P^2(\omega) \rightarrow 200\%/W$ for a 1-cm-long guide and a channel 1 μm wide. These values could even be competitive with copropagating devices, without their tight tolerances on phase matching.

In conclusion we have demonstrated efficient SHG due to mixing of counterpropagating beams in planar waveguides of a LB film for the first time. We have measured large nonlinear cross sections for single films of DCANP. Our four-layer structure (linear/nonlinear) waveguide allowed an advantageous trade-off between loss and conversion efficiency.

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