

## Nonlinear optical spectroscopy in one-dimensional photonic crystals

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We have conducted a spectroscopic investigation of the enhancement of nonlinear optical effects around a defect in a one-dimensional photonic crystal. Degenerate four-wave mixing studies were performed on a dielectric stack that contained a polymer thin-film defect layer doped with a nonlinear organic dye. This sample exhibited a large nonlinear response at a resonant defect frequency. Nonlinear spectroscopy was performed around the defect resonance and at frequencies well away from resonance. We have shown that the four-wave mixing signal exhibits extremely high-quality resonance, consistent with the expected cubic dependence on the calculated intensity within the defect layer. © 2003 American Institute of Physics. [DOI: 10.1063/1.1636249]

Photonic crystals (PhCs) are expected to play an important role in the development of new optical devices.<sup>1,2</sup> They consist of a periodically arranged composite of dielectric materials, which influences the propagation of light in a manner analogous to the way crystalline solids influence the transport of electrons.<sup>3</sup> Defects in PhCs can be engineered to further tailor the optical properties of devices. For example, one well-known property of defects in PhCs is the localization of electromagnetic energy near the defect for certain frequencies that are forbidden to propagate within the bulk of the PhC, enabling the creation of microcavity resonators and waveguides.<sup>4–6</sup>

The device possibilities for PhCs will be greatly enhanced by the addition of optical nonlinearities; such possibilities include ultrafast all-optical switching for communications, and potentially even optical computing.<sup>7–13</sup> Correspondingly, the influence of the photonic bandgap can be exploited to enhance or modify the behavior of nonlinear optical materials. For example, the flattening of the photon dispersion relation near a photonic bandgap has been demonstrated to ease phase-matching constraints leading to enhanced second- and third-harmonic generation,<sup>14,15</sup> and localization associated with defects has been shown to enhance a variety of third-order nonlinear processes in one-dimensional (1D) PhCs.<sup>16–20</sup>

In particular, Tsurumachi and co-workers<sup>17–19</sup> have demonstrated that nonlinear optical effects can be greatly enhanced by the localization of the optical field within a defect in a 1D PhC. Their work focused on attaining the largest possible enhancement of the nonlinear effects and understanding the magnitude of the enhancement. In contrast, in this letter, we present a *spectroscopic* study of enhanced nonlinear effects in the case of relatively weak enhancement. Spectroscopic characterization of such effects is essential to the design of devices which utilize them, especially bandwidth-sensitive devices such as those used in telecommunications applications. To this end, we have performed

nonlinear optical spectroscopy on a dye-doped defect layer embedded within a 1D PhC.

The samples used in this study consisted of a pair of dielectric quarter-wave stacks surrounding a polyvinyl alcohol (PVA) defect layer. The quarter-wave stacks were obtained from a commercial supplier;<sup>21</sup> each was comprised of 12 periods of alternating TiO<sub>2</sub> and HfO<sub>2</sub> layers with optical thickness of (600 nm)/4. This combination of materials possesses a low index contrast of 1.11, and was chosen because the resulting rejection band is deep and narrow, while the defect peaks are relatively broad, and thus could be probed across the rejection band edge and through the band using a tunable picosecond R6G dye laser. The defect layer was created by spin-coating a water-based solution of PVA onto a pair of the quarter-wave stacks and squeezing them together in a vise. The PVA solution was mixed with a solution of ethanol and an infrared laser dye possessing strong optical nonlinearities. Two reference samples were also prepared that were identical to the primary sample except that one contained no dye in the polymer layer, and one was made using blank glass substrates in place of the quarter-wave stacks. A transmission scan from the primary sample is shown in Fig. 1. Inset in the figure is a diagram of the sample structure. The two defect peaks that appear within the rejection band represent consecutive cavity modes, a fact that can be used, along with calculated transmission spectra, to calculate the thickness of the defect layer; it was determined to be 2.87 μm.

The dye chosen for use in this study was Styryl-9M. Nonlinear optical characterization of this dye was performed using the Z-scan method.<sup>22</sup> These measurements confirmed that Styryl-9M possesses a large, fast-responding, complex third-order nonlinear susceptibility  $\chi^{(3)}$ . The Z-scan measurements were taken using the reference sample that contained the dye-doped polymer layer between blank glass substrates, utilizing a picosecond R6G dye laser with a pulse width of 8 ps and a pulse energy of 20 nJ (which was also used for the four-wave mixing measurements to be described later). Scans were taken over a range of wavelengths to confirm that the nonlinear properties did not change significantly over the tuning range of the laser. Pulse repetition rates were low enough ( $\approx 100$  Hz) to ensure that thermal effects were

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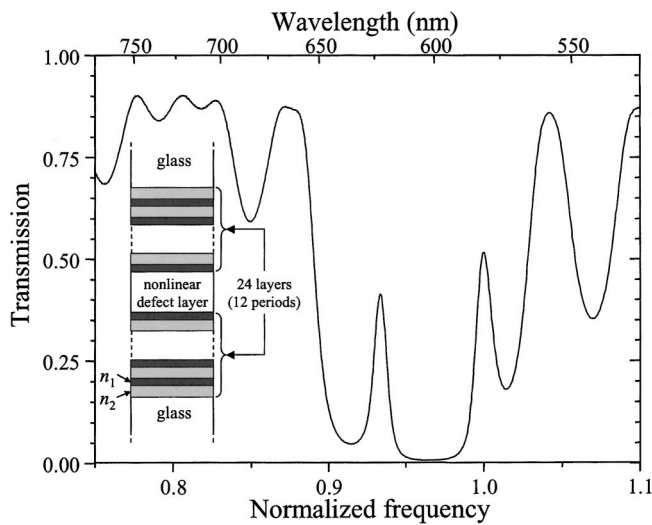


FIG. 1. Transmission scan of the 1D PhC sample. Two defect modes are apparent. The frequency axis has been normalized to unity at the frequency of the higher defect mode. Wavelength is indicated along the top axis. Inset is a schematic indicating the sample structure.

not responsible for the apparent nonlinearity. The dye-doped polymer layer was determined to possess a nonlinear refractive index  $\gamma = 1.25 \times 10^{-15} \text{ m}^2/\text{W}$ , and nonlinear absorption  $\beta = -4.5 \times 10^{-9} \text{ m/W}$  by fitting the Z-scan data, following the method of Ref. 22.

Nonlinear spectroscopy of the dye-doped defect layer embedded within a 1D PhC could not be accomplished using the Z-scan method because the defect mode is very sensitive to the thickness of the defect layer, which was not perfectly uniform over the area of the sample.<sup>23</sup> Therefore, a degenerate four-wave mixing (DFWM) experiment was performed.<sup>24,25</sup>

The origin of the nonlinear enhancement can be understood from Fig. 2, which depicts the calculated steady-state electric field amplitude (normalized to input) within our 1D PhC sample at both the defect mode wavelength and a wavelength near the band edge. Notice that, although the amplitude at the output is approximately the same, the field within

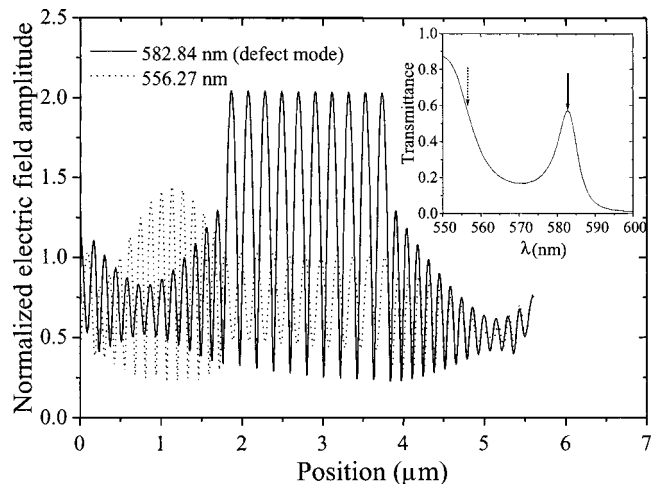


FIG. 2. Calculated electric-field amplitude within the PhC structure at the defect mode wavelength (solid curve) and the band edge (dotted curve). The net transmittance at both wavelengths is the same. The inset contains the calculated transmittance versus wavelength; the values where the field profiles were calculated are indicated by arrows.

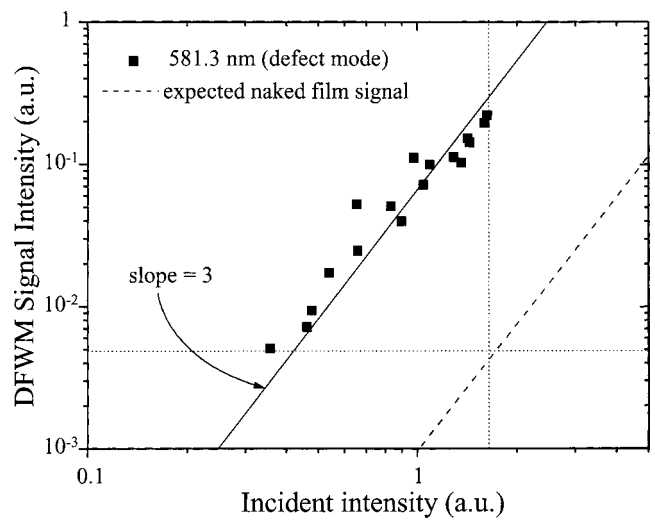


FIG. 3. DFWM signal from 1D PhC sample with nonlinear dye-doped defect layer. The dashed line indicates the signal that would be expected from a naked dye-doped film. The dotted lines indicate the limits of the experiment's sensitivity.

the defect layer is much larger at the defect mode. Nonlinear optical effects are strongly dependent on the amplitude; third-order effects such as DFWM scale according to the cube of the intensity and hence the sixth power of the field amplitude. Therefore, the amplitude enhancement of slightly more than two times is expected to yield an enhancement in the DFWM signal of over 70 times.<sup>26</sup>

We performed DFWM power-scaling studies on our samples using the backward-wave geometry with our picosecond dye laser. The peak pump-beam intensity was  $\sim 15 \text{ MW/cm}^2$ ; the probe beam was 10% of the pumps. The results are plotted in Fig. 3 for the defect-mode wavelength. The expected cubic dependence is indicated by the solid fit line. A similar measurement was attempted using the reference samples; for the dye-free PhC sample, no signal was detected, as one would expect. The dye-doped polymer layer with no PhC also did not yield a detectable signal, confirming that the presence of the PhC does indeed greatly enhance the nonlinear effects. The expected enhancement factor of  $\sim 70$  predicts a reference signal indicated by the dashed line in Fig. 3, while the dotted lines indicate the range of experimental sensitivity as limited by the maximum laser output (vertical line) and the noise floor (horizontal line). The lack of a signal from the reference sample is consistent with these limits; as shown, the expected reference signal (dashed line) lies just outside the detectable region.

The results of our nonlinear spectroscopy measurements across the defect mode can be seen in Fig. 4. The plot has been scaled into units of the expected naked-film reference signal, and thus indicates how the DFWM enhancement factor changes with wavelength (note the logarithmic units). In order to obtain these data, DFWM measurements were performed repeatedly at several wavelengths; it should be noted that these data points have been scaled to compensate for unequal laser output power as the wavelength was tuned. The DFWM resonance is indicated in Fig. 4 alongside the measured transmittance and calculated electric field amplitude within the defect layer (both normalized to match the DFWM peak). The curves are similar because the transmiss-

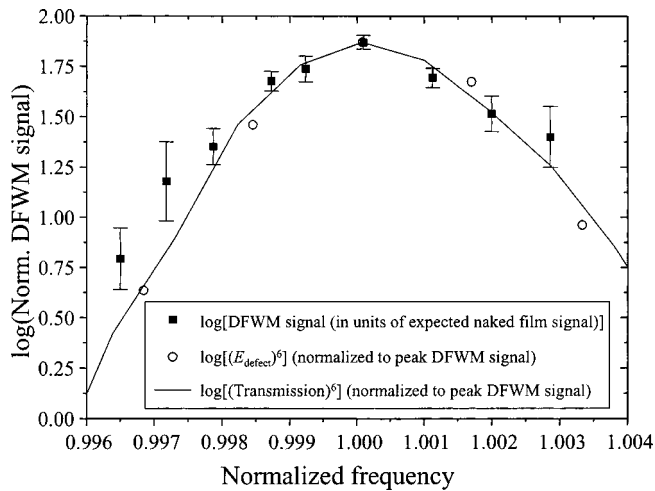


FIG. 4. Dependence of DFWM signal on normalized frequency near the defect mode. Error bars represent statistical uncertainties calculated as standard deviation of multiple repeated measurements. The signal has been normalized to the calculated signal level from a naked dye-doped film: note the logarithmic units. Also indicated are the calculated electric field (circles) and the measured transmission (curve), both raised to the sixth power (in normalized units).

sion and electric field have been raised to the sixth power, and the DFWM signal  $\propto I^3 \propto |E|^6$ . Hence, the DFWM signal peak is much sharper than that of the field amplitude and the transmission; the DFWM resonance quality factor, defined as  $f/\Delta f_{\text{half-max}}$ , is observed to be higher than that of the transmission by a factor of  $\sim 3$ .

In order to confirm that the resonance in the DFWM signal was indeed attributable to the presence of the 1D PhC surrounding the defect layer, and not to some resonant effect in the Styryl-9M dye, DFWM measurements were performed using a 1-mm-thick cell filled with the dye-doped PVA/water/ethanol solution. These measurements showed no significant variation as the wavelength was changed.

We have performed a study characterizing the nonlinear optical properties of 1D PhCs containing defect layers doped with the nonlinear dye Styryl-9M. Particular attention was paid to the enhancement of the localized electric-field amplitude within the defect layer and its influence on the nonlinear optical properties of the samples. We observed DFWM signals from our samples that were consistent with theoretically predicted enhanced amplitudes in the defect layer. Spectroscopic measurements demonstrated that the enhanced DFWM signal was sharply peaked around the defect mode in

a manner consistent with the wavelength variation in the field enhancement, while the bulk dye solution showed no wavelength dependence.

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- <sup>26</sup> Note that this enhancement is expressed relative to the signal that would be obtained from the *identical* nonlinear layer contained within the reference sample, not to that from a reference film with matched linear transmittance, as in Ref. 16.