

# Unravelling Nonlinear Spectral Evolution Using Nanoscale Photonic Near-Field Point-to-Point Measurements

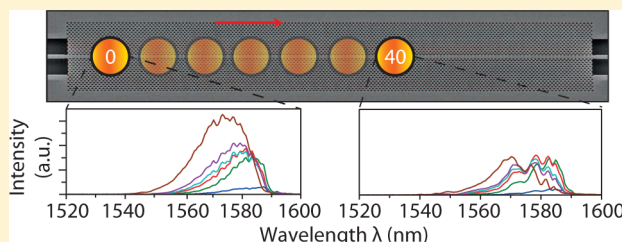
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**S** Supporting Information

**ABSTRACT:** We demonstrate nanoscale photonic point-to-point measurements characterizing a single component inside an all-optical signal-processing chip. We perform spectrally resolved near-field scanning optical microscopy on ultrashort pulses propagating inside a slow light photonic crystal waveguide, which is part of a composite sample. A power study reveals a reshaping of the pulse's spectral density, which we model using the nonlinear Schrödinger equation. With the model, we are able to identify the various physical processes governing the nonlinear pulse propagation. Finally, we contrast the near-field measurements with transmission measurements of the complete composite sample to elucidate the importance of gaining local information about the evolution of the spectral density.

**KEYWORDS:** Near-field spectroscopy, integrated nanophotonics, nonlinear optics, slow light, photonic crystal



The ever increasing demands for speed in telecommunication has led researchers to develop a broad variety of all-optical signal-processing elements such as switches,<sup>1</sup> demultiplexers,<sup>2</sup> buffer lines,<sup>3,4</sup> dispersion compensators,<sup>5</sup> or signal regenerators.<sup>6,7</sup> Many of these devices were demonstrated with photonic crystal (PhC) waveguides,<sup>8</sup> which have emerged as a promising platform for integrated photonic circuits<sup>9</sup> due to their high mode confinement<sup>8</sup> and the freedom to design the dispersion of the guided mode.<sup>10,11</sup> Furthermore, these structures can support a guided mode featuring a high group index that has been shown to enhance light-matter interaction<sup>12–15</sup> and hence enabling more compact device designs.

Nonlinear optical effects, which allow for the control of light with light,<sup>16</sup> are crucial for all-optical signal-processing. Conventionally, nonlinear optical properties of a device (e.g., a waveguide) are experimentally determined by analyzing the properties of a transmitted laser pulse for different input powers.<sup>17–20</sup> In contrast to the linear optics regime, where only the characteristics of the different devices inside the circuit matter, in the nonlinear regime the order in which the different components are arranged is also crucial. Thus, it is a challenging task to extract the characteristics of individual components of an integrated photonic circuit from a single transmission measurement. What is missing is a photonic analogue to electronic point-to-point measurements,<sup>21</sup> which would characterize the performance of each component within a nanophotonic chip, instead of measuring all effects accumulated in the sample at the same time.

Near-field microscopy, which allows for local optical measurements with a resolution beyond the diffraction limit<sup>22</sup> has the potential to address this need. The characterization of

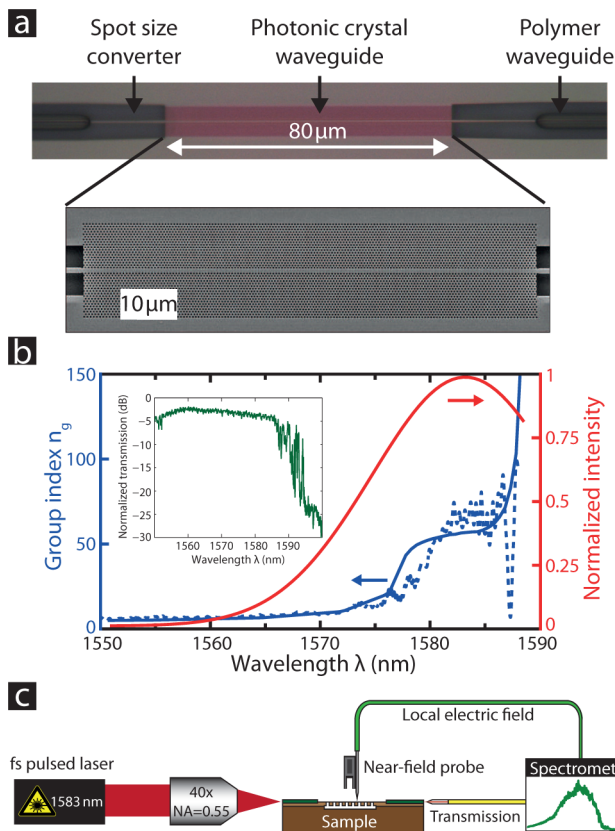
integrated photonic devices by near-field scanning optical microscopy (NSOM) involved mapping the nanoscopic electric field profile of modes in waveguiding structures.<sup>23,24</sup> Furthermore, NSOM measurements have visualized low intensity pulses propagating inside photonic structures, both in the time domain<sup>25,26</sup> and spectrally<sup>27</sup> in the linear optical regime. Alternatively, near-field microscopy was used to gain local temporal or spectral information of processes such as photoluminescence from J-aggregates,<sup>28</sup> quantum wires,<sup>29</sup> or quantum wells.<sup>30</sup> However, local spectral point-to-point measurements on a single component on a photonic chip that, for example, characterize a high intensity ultrashort pulse propagating in the nonlinear regime, are missing so far.

In this Letter, we demonstrate how a NSOM technique can be used to track the spectral evolution of such a pulse inside a single component of a more complex photonic chip. We measure the changes of the spectral density as the ultrashort pulse propagates through a slow light PhC waveguide by monitoring the shape, peak amplitude, mean wavelength, and width of the laser spectrum. Numerical calculations reveal that the spectral reshaping is caused by the interplay between dispersion, nonlinear, and free-carrier effects. Finally, we show that the information gained by NSOM allows us to model the pulse propagation inside the PhC waveguide without the need to model the propagation in the rest of the sample, in contrast to the case when only regular transmission spectra are available.

As a model system for our experiment, we use a five component sample (Figure 1a). There are two polymer access

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**Figure 1.** Sample and setup. (a) Optical microscope picture of the central part of the photonic sample. The PhC waveguide is visible in the middle. At the sides, the silicon spot-size converters and part of the polymer access waveguides can be seen. The inset shows a scanning electron micrograph of the silicon PhC. (b) The measured (dashed) and calculated (solid) group index distribution as a function of wavelength (blue, left axes) and the spectral density of the laser coupled into the sample (red, right axis). The inset shows the measured linear transmission of the PhC waveguide normalized to the transmission of a SU8 polymer waveguide. (c) Schematic of our experimental setup.

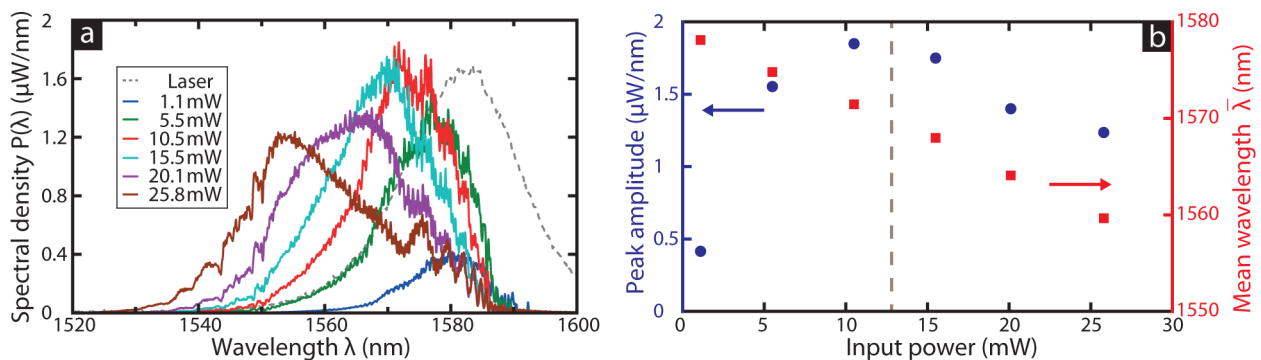
79 waveguides, one for each facet, and the silicon PhC that is  
 80 connected to the access waveguides with two silicon spot-size  
 81 converters. The polymer waveguides consist of SU-8 photo-  
 82 resist, are a few millimeters long, and have a large cross-section,  
 83 a width and a height of roughly 5 and 2  $\mu\text{m}$ , respectively, to

enhance the efficiency of the light incoupling.<sup>31</sup> Each spot-size  
 84 converter is a 50  $\mu\text{m}$  long inverse taper, whose purpose is to  
 85 funnel the light from the polymer access waveguides to the PhC  
 86 waveguide. The photonic crystal waveguide has a length of 80  
 87  $\mu\text{m}$  and is therefore at least 10 times shorter than each of the  
 88 access polymer waveguides. Details about the design and the  
 89 fabrication of the PhC waveguide can be found in the  
 Supporting Information. 91

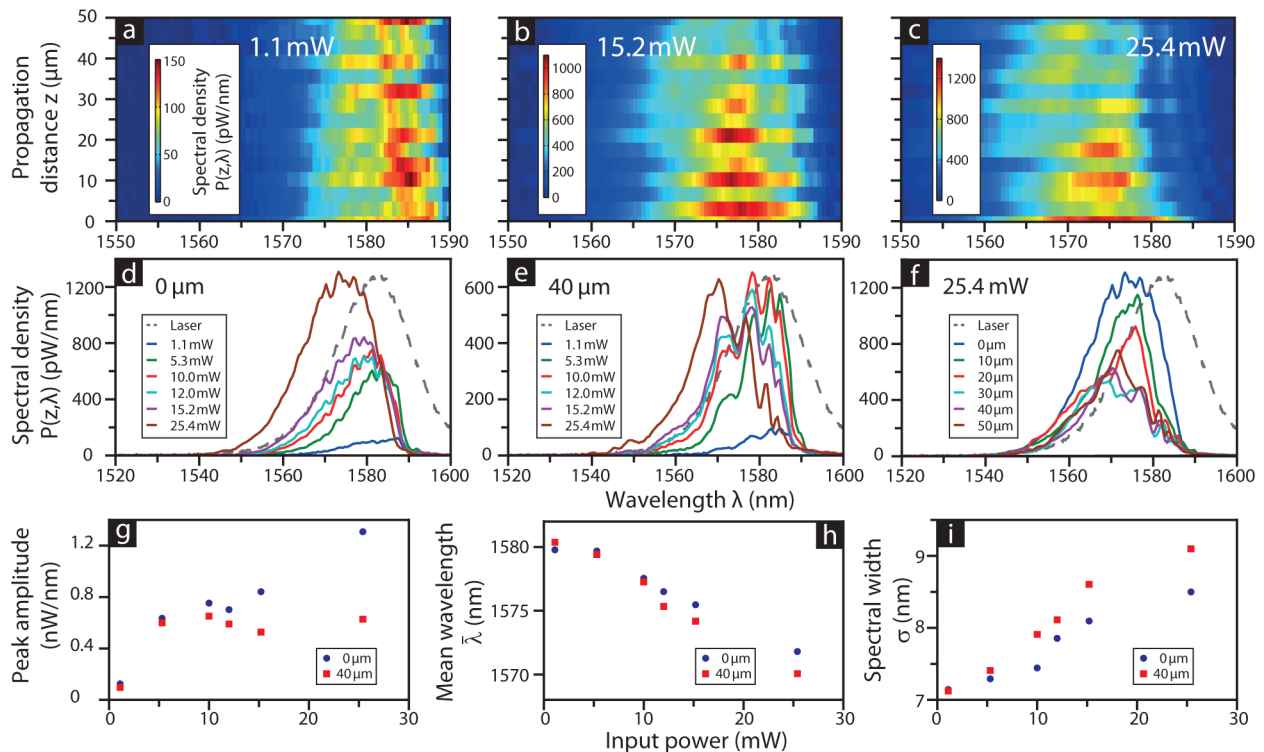
We measure the group index  $n_g$  characteristics by means of  
 92 Fourier transform spectral interferometry.<sup>32</sup> The result,  
 93 presented in Figure 1b, indicates that there is a plateau of  $n_g$   
 94  $\sim 60$  between 1577 and 1587 nm. The group index diverges for  
 95 longer wavelength as the cutoff is approached. For wavelengths  
 96 shorter than 1577 nm, the group index tends to a value of 5.  
 97 We also calculate the dispersion relation of the slow light PhC  
 98 waveguide using MIT Photonics Band (MPB) package.<sup>33</sup> The  
 99 extracted group index is in good agreement with the MPB  
 100 calculation (dashed and solid blue curve in Figure 1b,  
 101 respectively), indicating the high quality of fabrication and  
 102 characterization of the sample. 103

Finally, we also characterize the linear response of our sample  
 104 by measuring transmission at low intensities (inset in Figure  
 105 1b). Up to a wavelength of 1587 nm the transmission is  
 106 basically flat at  $-5$  dB. For longer wavelength, the transmission  
 107 drops rapidly, up to  $-30$  dB at 1600 nm, due to the slowdown  
 108 of the guided mode, which increases the backscattering and the  
 109 out-of-plane scattering<sup>34</sup> and the approaching cutoff. The  
 110 contribution of the linear material absorption to this loss is  
 111 negligible since single photons at telecom wavelengths do not  
 112 possess enough energy to bridge the electronic band gap. 113

As shown in Figure 1c, we end-fire couple femtosecond  
 114 pulses centered at 1583 nm, which are generated by an optical  
 115 parametric oscillator (Spectra Physics OPAL) into our sample  
 116 using a 40x aspheric lens ( $\text{NA} = 0.55$ ). At the end facet of the  
 117 sample, a lensed fiber collects the transmitted light, which is  
 118 then spectrally analyzed. The laser delivers bandwidth-limited  
 119 pulses of a temporal length of 180 fs and a concomitant  
 120 intensity bandwidth of 20 nm (fwhm). To investigate nonlinear  
 121 optical effects the laser power coupled into the sample can be  
 122 varied by a neutral density filter. The values of the input power  
 123 mentioned in this Letter are all determined before the  
 124 incoupling lens. A maximum limit for the input power of 26  
 125 mW is chosen for all experiments to avoid damage to our  
 126 sample, which was found to occur at  $\sim 30$  mW. 127



**Figure 2.** Spectral transmission measurements of the whole sample. (a) Transmission spectra for different average input powers and the spectrum of the laser before the sample, for comparison. (b) The dependence of the peak amplitude and the mean wavelength of the transmission spectra on the input power. The gray dashed line marks the boundary between the regions where the peak amplitude increases, and where it decreases with input power.



**Figure 3.** Spectrally resolved near-field measurements along the PhC waveguide. Three near-field measurement series showing the evolution of the spectral density of the pulse for average input powers of (a) 1.1, (b) 15.2, and (c) 25.4 mW. A series of spectra taken at the beginning (d) and after 40  $\mu\text{m}$  propagation (e) in the slow light PhC waveguide for different average input powers. (f) A series of spectra taken in 10  $\mu\text{m}$  steps at an average input power of 25.4 mW. Additionally, the laser spectrum is included in (d–f). Peak amplitude (g), mean wavelength (h), and spectral width (i) of the spectra series shown in (d,e).

128 Near-field measurements are performed by means of a home-  
 129 built NSOM. By placing the near-field probe at different  
 130 locations on the sample and spectrally analyzing the detected  
 131 light, the evolution of the spectral density can be tracked. At  
 132 each point the spectrum is collected while scanning the near-  
 133 field probe transversally over the waveguide in order to average  
 134 out the complex lateral field distribution of the PhC Bloch  
 135 modes.<sup>35</sup> Note that the resolution is limited by the aperture  
 136 size, which is in our case  $\sim 250$  nm. In principle, the resolution  
 137 could be pushed further into the nanoscale by decreasing the  
 138 aperture size at the cost of less collected light.

139 First, transmission measurements are performed to character-  
 140 ize the nonlinear response of the complete sample. Figure 2a  
 141 shows spectra measured for six different average input powers  
 142 ranging from 1.1 to 25.8 mW. The dependence of the shape of  
 143 the transmitted spectrum on the input power is a clear  
 144 signature of the presence of nonlinear effects.<sup>18</sup> The most  
 145 obvious feature is that the peak amplitude does not scale  
 146 linearly with the input power as shown in Figure 2b. The  
 147 maximum value of the spectrum increases until 10.5 mW. For  
 148 higher input powers the peak amplitude declines.

149 The second feature that can be observed is a blueshift of the  
 150 spectral density  $P(\lambda)$ , which we quantify by calculating the  
 151 mean wavelength  $\bar{\lambda}$  of each spectrum

$$\bar{\lambda} = \frac{\int \lambda P(\lambda) d\lambda}{\int P(\lambda) d\lambda} \quad (1)$$

153 Figure 2b depicts the dependence of  $\bar{\lambda}$  on input power. It is  
 154 clear that the mean wavelength blueshifts at higher powers. In  
 155 the linear optical regime, we expect to observe a transmission

spectrum resembling that of the input laser but cut off around  
 156 1587 nm. This spectral reshaping, which is indeed observed for  
 157 low input power, is due to the increased linear scattering loss at  
 158 longer wavelengths and the approaching cutoff. However, for  
 159 increasing input power the spectrum changes in shape. In  
 160 addition to the observed blueshift at higher input powers, the  
 161 spectral density broadens (a quantified measure of this  
 162 broadening can be found in the Supporting Information). We  
 163 note that the broadening and blueshift cannot be due to the  
 164 linear scattering loss in the PhC waveguide, as these changes  
 165 occur mostly at wavelengths below 1585 nm, where this linear  
 166 loss is constant (inset in Figure 1b). In summary, the power  
 167 dependence of the transmission spectra is clearly indicative of  
 168 nonlinear pulse propagation.

The transmission spectra represent the response of the  
 170 complete composite sample. The question that now arises is  
 171 which of the observed spectral changes occur in the PhC  
 172 waveguide and which are due to the other four components of  
 173 our sample. In order to answer this question, we perform in situ  
 174 NSOM measurements of the pulse propagating inside the  
 175 photonic crystal waveguide. Each of the measurements,   
 176 presented in Figure 3a–c, consists of 19 spectra collected at  
 177 positions evenly distributed over a distance of 50  $\mu\text{m}$  along the  
 178 PhC for the different input powers. These measurements show  
 179 that we can determine the spectra of the propagating pulse as a  
 180 function of position and thereby gain access to its spectral  
 181 evolution at the nanoscale.

For all input powers, we observe various abrupt amplitude  
 183 changes between the spectra taken at successive propagation  
 184 distances. This modulation in the amplitude is an artifact  
 185 caused by the Bloch nature of the photonic modes in the 186

187 waveguide. That is, the locally measured electric field amplitude  
188 is strongly dependent on the exact position of the probe within  
189 a single unit cell<sup>24</sup> and the positional accuracy is mainly limited  
190 by the thermal drift and the size of the investigated area of  
191 several tens of  $\mu\text{m}^2$ . However, the shape of the spectral density  
192 does not depend on the probe position within a unit cell. While  
193 it is tempting to normalize the data, in doing so we would lose  
194 any information about a possible amplitude drop due to  
195 absorption and so we analyze the unnormalized spectra.

196 Beside these abrupt amplitude variations there is no  
197 significant drop in amplitude of the spectral density with  
198 propagation visible for an input power of 1.1 mW (cf. Figure  
199 3a). That is, there is no significant absorption occurring inside  
200 the PhC waveguide at this low input power. From this and the  
201 fact the shape of the spectrum of the pulse is the same  
202 throughout the waveguide, we conclude that nonlinear effects  
203 are negligible here. At 15.2 mW the evolution of the spectral  
204 density starts to show evidence of nonlinear effects (cf. Figure  
205 3b), since the peak amplitude decays by 50% in the first 50  $\mu\text{m}$ .  
206 For the highest investigated input power, close to the damage  
207 threshold, shown in Figure 3c, the amplitude decay of the  
208 spectral density occurs even faster. The peak amplitude of the  
209 spectral density decreases by 50% in the first 30  $\mu\text{m}$ . Thus, this  
210 series of spectra demonstrate that the pulse experiences power-  
211 dependent losses during propagation inside the PhC wave-  
212 guide.

213 Interestingly, we observe a power dependence of the spectral  
214 density at the beginning of the PhC waveguide. For example, a  
215 blueshift due to increased input power becomes obvious by  
216 comparing these spectra to the input laser spectrum (Figure  
217 3d). To quantify the spectral changes that occurred before the  
218 PhC, we investigate a series of spectra taken at the start of the  
219 PhC for different input powers, shown in Figure 3d. There are  
220 four different features that can be analyzed: shape, peak  
221 amplitude, mean wavelength, and spectral width. First, the  
222 shape of spectra at the input of the PhC remains the same for  
223 all input powers, that is, a Gaussian cutoff at longer wavelength.  
224 Second, the peak amplitude of the spectra does not scale  
225 linearly with input power (cf. blue dots in Figure 3g). Third,  
226 there is a power-dependent blueshift of the spectral density.  
227 This wavelength shift can be quantified using eq 1. As shown in  
228 Figure 3h, a change of the input power from 1.1 to 25.4 mW  
229 results in a blueshift of 8 nm. Fourth, the spectra broaden  
230 slightly with input power, that is, the spectral width (defined in  
231 the Supporting Information) increases by  $\sim 1.5$  nm between the  
232 spectra measured at 1.1 and 25.4 mW (cf. blue dots in Figure  
233 3i). These observations suggest that the intensity of the pulse is  
234 sufficiently high for nonlinear effects to play a role in the access  
235 waveguide or the silicon inverse taper.

236 To quantify the effect of the PhC waveguide on the changes  
237 in the laser spectrum, we measure a series of spectra after 40  
238  $\mu\text{m}$  propagation inside the slow light photonic crystal  
239 waveguide (Figure 3e) for the same range of input powers. A  
240 comparison of Figure 3d,e makes it obvious that the shape of  
241 the spectral density changes significantly with input power.  
242 Note that after 40  $\mu\text{m}$  inside the PhC, the spectra measured by  
243 NSOM begin to resemble those measured in transmission  
244 (Figure 2a). Hence, this observation together with the fact that  
245 the shape of the spectra at the beginning of the PhC is not  
246 power-dependent, shows that the reshaping of the spectral  
247 density happens inside the PhC waveguide.

248 The peak amplitude of the spectra at 40  $\mu\text{m}$  also scales in a  
249 nonlinear fashion with input power but in a more drastic

fashion than at the beginning of the PhC waveguide as shown 250  
in Figure 3g. At 10 mW, the peak amplitude reaches its 251  
maximum value after which it shows a small drop until finally 252  
recovering for 25.4 mW. This scaling of the peak amplitude 253  
shows qualitative agreement with the trend observed in the 254  
transmission spectra (Figure 2 b) other than at the highest 255  
input power. 256

In addition, a power-dependent blueshift can be again 257  
observed near the end of the PhC (cf. blue squares in Figure 258  
3h). At an input power of 1.1 mW the spectrum has a mean 259  
wavelength of 1580 nm that decreases to a value of 1570 nm 260  
when the input power is increased to 25.4 mW. Interestingly, 261  
this wavelength shift of 10 nm is only slightly larger than the 262  
shift observed at the beginning of the PhC waveguide. 263  
Moreover, since the transmission spectra exhibit a blueshift 264  
roughly twice as large, this suggests that most of the shift in 265  
wavelength of the spectral density occurs in the other 266  
components of the sample. 267

A similar observation can be made for the spectral 268  
broadening presented in Figure 3i. By comparing the spectral 269  
width of the two measurement series we conclude that only a 270  
small fraction of the spectral broadening occurs inside the PhC 271  
waveguide. The transmission spectra show a broadening of 5 272  
nm (cf. Supporting Information) which is three times larger 273  
than the broadening observed in the PhC. Thus, together with 274  
the blueshift the broadening of the spectral density occurs 275  
mainly in the other components of the sample, and not in the 276  
PhC waveguide. This may seem unexpected, particularly given 277  
the slow-light enhancement and the spatial confinement of the 278  
light in the PhC, yet it can be explained with two reasons. First, 279  
the pulse is temporally broadened due to dispersion before it 280  
enters the photonic crystal waveguide giving rise to a decreased 281  
peak amplitude. Second, the blueshift of the spectral density 282  
already before the photonic crystal waveguide causes the pulse 283  
to experience smaller group indices than would be a priori 284  
assumed. In conclusion, the slow light photonic crystal 285  
waveguide has a major effect on the reshaping and the scaling 286  
of the peak amplitude, but the blueshift and the broadening of 287  
the spectral density occurs mainly in the other components of 288  
the sample. 289

There remains the question about the cause of the spectral 290  
reshaping. To answer it we investigate the evolution of the 291  
spectral density inside the PhC waveguide at an input power of 292  
25.4 mW by comparing spectra at different locations. This is 293  
shown in Figure 3f for six spectra, taken in 10  $\mu\text{m}$  steps. This 294  
measurement suggests that there is no energy redistribution 295  
inside the spectrum, for example, energy transfer from the long 296  
wavelengths to the shorter ones, since the spectral density taken 297  
at the beginning of the PhC waveguide possesses the highest 298  
amplitude at each wavelength. Instead it seems more that the 299  
spectral reshaping originates from different decay behavior of 300  
different spectral components, that is, an increased amplitude 301  
drop at the long wavelength side. 302

In essence, the near-field measurements allow us to separate 303  
the spectral reshaping happening inside different devices of an 304  
integrated photonic circuit on the nanoscale that is not possible 305  
by means of transmission measurements. Additionally, the 306  
NSOM measurements show that although the polymer 307  
waveguides are at least one magnitude longer than the PhC 308  
waveguide, most of the spectral reshaping occurs inside the 309  
PhC. This observation confirms the potential of slow light for 310  
enhancing nonlinear optics. 311

To gain more insight into the physical processes playing a role in the reshaping of the spectral density we model pulse propagation by means of the nonlinear Schrödinger equation (NLSE),<sup>36</sup> which includes dispersion, various nonlinear optical effects and free-carrier dynamics. Because the spectrum of our pulse covers a broad range of different group indices we take the full dispersion relation  $\gamma(\omega)$  of the PhC waveguide into account. Therefore, we split the NLSE into two equations, one that deals with the nonlinear optical effects and the free-carrier dynamics, and one that considers the dispersion. Concurrently, we solve an auxiliary rate equation describing the temporal evolution of the free-carrier density.<sup>37</sup>

The first equation, which describes nonlinear and free-carriers effects during propagation, is most conveniently formulated in the time domain

$$\frac{\partial A}{\partial z} = -\frac{\alpha}{2}A + \left(-\frac{\beta}{2A_{\text{eff}}} + i\frac{2\pi}{\lambda A_{\text{eff}}}n_2\right)|A|^2A - \left(\frac{\sigma}{2} + i\frac{2\pi}{\lambda}k_c\right)N_c A \quad (2)$$

The meaning and the value of the parameters in eq 2 and subsequent equations, which are either material- or geometry-dependent and therefore taken from literature, can be found in Table 1.

**Table 1. The Names and Values of the Parameters Used in the Numerical Calculations and the References from Which They Were Taken<sup>a</sup>**

parameter	name	value	slowdown scaling	reference
$A_{\text{eff}}$	effective mode area	$0.66 \mu\text{m}^2$		
$\alpha$	linear loss coefficient	20 dB/cm	$S$	38
$\beta$	TPA coefficient	1 cm/GW	$S^2$	39
$n_2$	Kerr coefficient	$6 \times 10^{-14} \text{ cm}^2/\text{W}$	$S^2$	39
$\sigma$	FCA cross-section	$1.45 \times 10^{-21} \text{ m}^2$	$S$	40
$k_c$	FCD coefficient	$1.35 \times 10^{-27} \text{ m}^3$	$S$	41
$\tau_{\text{rec}}$	free-carrier recombination time	450 ps		1

<sup>a</sup>The effective mode area was extracted from MPB simulations and the slowdown scaling factor for each parameter is given, if applicable. The linear loss coefficient  $\alpha$  accounts only for scattering loss and its dependence on the slowdown is explained in more detail in the Supporting Information.

Equation 2 describes how the slowly varying temporal envelope  $A(z,t)$  of the pulse, defined by  $A(z,t) = (I(z,t) \cdot A_{\text{eff}})^{1/2}$ , where  $I(z,t)$  is pulse intensity, is influenced by third-order nonlinear effects, that is, two-photon absorption (TPA) and self-phase modulation (SPM) due to the Kerr effect, and by free-carriers dynamics, that is, free-carrier absorption (FCA) and free-carrier dispersion (FCD).

The temporal evolution of the free-carrier density  $N_c(t)$  is governed by the following rate equation<sup>38</sup>

$$\frac{dN_c}{dt} = \frac{\pi\beta}{h\omega_0 A_{\text{eff}}^2} |A|^4 - \frac{N_c}{\tau_{\text{rec}}} \quad (3)$$

where  $h$  is the Planck constant and  $\omega_0$  the central angular frequency of the pulse. In this equation, we see that free-carriers

are only created by two-photon absorption (first term) and that they recombine with a lifetime  $\tau_{\text{rec}}$  (second term). The free-carrier lifetime has a value of a few hundred picoseconds in silicon structures featuring a large surface area.<sup>1</sup> Since the laser pulse needs less than 16 ps to propagate through the PhC waveguide, the amount of carrier recombination during pulse propagation is negligible. Moreover, free-carrier accumulation between pulses is disregarded since the time between subsequent laser pulses is 12.5 ns and therefore much longer than the carrier lifetime.

In contrast to the nonlinear effects and the free-carrier dynamics that are dealt with in the time domain, it is more beneficial to treat dispersion in the frequency domain. Here the temporal broadening of the pulse envelope can be described as a simple differential equation for the spectral density  $\hat{A}(z)$ , which is the temporal Fourier transform of  $A(z,t)$ :

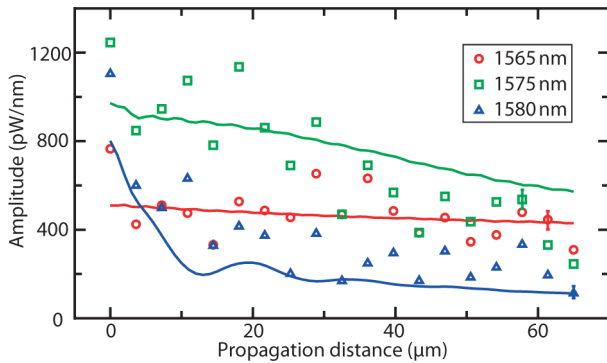
$$\frac{\partial \hat{A}}{\partial z} = i\gamma(\omega - \omega_0)\hat{A} \quad (4)$$

Details of how the system of equations is solved and fitted to our experimental data can be found in the Supporting Information.

In addition, it is important to take the slow group velocity of the light inside the PhC waveguide into account, because it has been shown that slow light propagation enhances nonlinear effects as well as interactions of the pulse with free carriers.<sup>38</sup> Two-photon absorption and the Kerr effect, which leads to self-phase modulation, scale quadratically with the so-called slowdown factor  $S$  which is defined by  $S = n_g/n_{\text{sil}}$ .<sup>42</sup> This quadratic enhancement arises because<sup>43</sup> (I) a slower pulse has more time to interact with matter, and (II) the pulse is spatially compressed upon entering into the slow light waveguide, leading to an increase in its intensity. In contrast, effects which are not influenced by the pulse intensity, that is, linear loss, free-carrier absorption, and free-carrier dispersion scale only linearly with  $S$ . Thus, the material parameters describing these effects are replaced by effective parameters to emulate the slow light enhancement. These effective parameters consist of the values listed in Table 1, properly scaled with the slowdown factor. Further, we note that the effective mode area  $A_{\text{eff}}$  is related to the group index in a nontrivial fashion. However, for most of the spectral regime in our experiment the variation of  $A_{\text{eff}}$  is minor. Thus, the effective mode area is taken to be constant in our simulations.

It is crucial to account for the frequency dependence of the slowdown factor  $S$ , which is mimicked by solving our model several times, once per investigated wavelength, and taking the group index valid at that wavelength into account for the slow light enhancement. Thus, for the sake of simplicity our approach inherently assumes that  $S$  is constant and we only investigate how one specific spectral component evolves with propagation. If we wanted to investigate the evolution of the complete spectrum of the pulse, we would have to slice the spectral density and calculate the evolution of these slices by solving the model repeatedly. To minimize the computation effort and still show that this approach delivers accurate results, we investigate only three different wavelengths, namely 1565, 1575, and 1580 nm, which feature different group indices with values of 7, 18 and 53, respectively.

Figure 4 presents a comparison between typical modeling results and data taken from the near-field measurements (Figure 3c). The figure shows how the three spectral components decay with propagation inside the PhC waveguide.



**Figure 4.** Decay behavior of different spectral components during propagation inside the PhC waveguide. The experimentally (points) and theoretically (solid curves) determined spectral amplitude as a function of propagation distance at three different wavelengths. The experimental data is taken from Figure 3c where the average input power is 25.4 mW. For each wavelength an error bar is depicted on the right side of the figure.

405 Clearly, the longest wavelength experiences the fastest  
 406 amplitude decay. In contrast, the amplitude of the shortest  
 407 wavelength stays constant. Thus, we find an excellent  
 408 quantitative agreement between the modeling and the  
 409 experimental results for all spectral components considered,  
 410 using material parameters found in literature. Only the decay  
 411 for the wavelength of 1575 nm seems to be slightly  
 412 underestimated, suggesting that some of the optical properties  
 413 (e.g., TPA or linear loss coefficient) of our sample differ from  
 414 those reported in the literature. This observation illustrates the  
 415 potential of our method to investigate the local effective optical  
 416 properties inside an integrated photonic circuit. Over the small  
 417 wavelength range that our pulses span the material parameters  
 418 of the PhC are practically constant while the group index varies  
 419 greatly (cf. Figure 1b). Hence, different wavelengths of light  
 420 within the PhC experience vastly different slowdown factors,  
 421 and it is this dispersion of the slow light enhancement that  
 422 causes the observed spectral density reshaping.  
 423 The oscillations visible in the experimental data originate  
 424 from the positioning uncertainty of the probe inside the  
 425 photonic crystal waveguide unit cell mentioned before.  
 426 Consequently, the error bars represent the standard variation  
 427 of the intensity distribution of the guided mode inside a unit  
 428 cell of the PhC, except for the longest wavelength where a 5%  
 429 noise level is assumed. In the slow light regime, the light will be  
 430 more sensitive to many different effects, which therefore  
 431 justifies a larger error bar than the spatial mode profile would  
 432 suggest. Details about the calculation of the error bars can be  
 433 found in the Supporting Information.

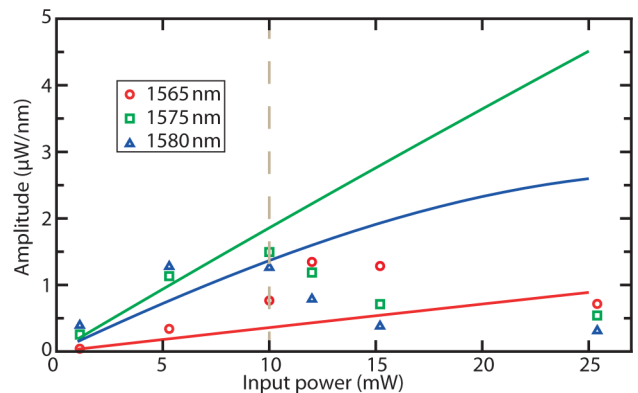
434 We now turn to discuss the influence of the different physical  
 435 effects that occur during the pulse propagation, which can be  
 436 separated in two categories: (I) those that lead to losses and  
 437 (II) those that change the shape of the spectrum. There are  
 438 three processes that may lead to an amplitude decay with  
 439 propagation, scattering, TPA, and FCA. The linear losses due to  
 440 scattering are too small to have a significant impact over a 50  
 441  $\mu\text{m}$  distance which we also observe in our measurements, since  
 442 there is no absorption visible at the lowest input power (cf.  
 443 Figure 3a). Thus, the measured amplitude decay is mainly  
 444 caused by TPA and FCA.

445 SPM and FCD are usually considered to be the source for  
 446 spectral reshaping such as a blueshift.<sup>40</sup> However, in our

447 experiment both processes do not have a significant influence  
 448 on the spectral shape, as shown in Figure 3f. Instead, the  
 449 reshaping is caused by dispersion of the PhC waveguide, which  
 450 in general has a 3-fold influence. First, a large dispersion  
 451 stretches the pulse temporally and therefore limiting its peak  
 452 intensity. Second, a wavelength dependence of the effective  
 453 material parameters as discussed above is created. Third, it also  
 454 causes a wavelength dependence of the free-carrier effects  
 455 leading to an enhanced decay of the slower propagating spectral  
 456 components, that is, longer wavelengths. This is explained by  
 457 the build-up of the free-carrier density in time between the  
 458 rising and trailing edge of a single pulse so that the trailing edge,  
 459 containing longer wavelengths, will experience enhanced FCA  
 460 and FCD.<sup>44</sup> In essence, the spectral reshaping stems from the  
 461 wavelength-dependent slowdown factor  $S$  and an interplay of  
 462 dispersion and the temporal dynamics of the free carriers.

463 Next, we demonstrate that a similar approach, which uses the  
 464 transmission spectra from the entire composite sample (cf.  
 465 Figure 2), does not accurately predict the spectral evolution in  
 466 the PhC waveguide. We again solve eqs 2–4 using the same  
 467 parameter values and assume that there are no nonlinear effects  
 468 inside the polymer access waveguide and spot-size converter.  
 469 Only the temporal broadening that occurs before the PhC  
 470 waveguide is taken into account.

471 We try to model the evolution with input power of the same  
 472 three wavelengths as above and compare it with experimental  
 473 data taken from the transmission measurements, as shown in  
 474 Figure 5. There is a clear discrepancy between the model and  
 475



**Figure 5.** Scaling behavior of different wavelengths with power in the transmission spectra. The experimentally (points) and theoretically (solid curves) determined spectral amplitude as a function of average input power for three different wavelengths. The experimental data is taken from Figure 2a. The gray dotted line marks the input power from which on the evolution of the spectral density differs significantly.

475 the experiment. First, the model no longer reproduces the  
 476 scaling of the amplitudes with input power. The measured  
 477 amplitudes of all spectral components show a decrease with  
 478 input power for very high input powers, whereas the modeling  
 479 results feature a steady increase. Second, the blueshift visible  
 480 in the measurement is not reproduced by the model. In the  
 481 measured data, at the lowest input powers the longest  
 482 wavelength exhibits the highest amplitude, whereas for high  
 483 input powers the shortest wavelength has the highest  
 484 amplitude, while the model does not exhibit this behavior.  
 485 Hence, the model can reproduce neither the peak amplitude  
 486 scaling nor the power-dependent spectral reshaping visible in  
 487 the transmission measurements.

Overall, it seems that the discrepancy between our transmission measurements and the model can be explained by a power-dependent absorption process, which is missing or underestimated in the modeling approach at higher input powers. We understand this discrepancy in terms of the blueshift that occurs before the PhC waveguide. The blueshift transfers energy to a spectral region with higher dispersion, so that the spectral density covers a larger range of group indices. This leads to a dramatic enhancement of the reshaping, as explained above, for increasing input power.

This interpretation is also supported by our experimental observations. For input powers higher than 15.2 mW, we observed a change in the spectral reshaping in the transmission spectra (Figure 2a) as well as in the near-field measurements (Figure 3e). The measured spectral density is no longer centered on the flat dispersion band at 1580 nm, but shifted toward 1570 nm, where the group index changes significantly with wavelength. Thus, the strong reshaping goes hand in hand with and is actually influenced by the blueshift in front of the PhC waveguide. In essence, the failure of the model to reproduce the evolution of the transmission spectra of the composite structure shows that obtaining the nonlinear optical behavior of the PhC waveguide that is part of a composite structure is far from trivial, since the other components may have a significant influence.

In conclusion, we presented a near-field scanning optical microscopy technique that can be used to test and evaluate nonlinear pulse propagation inside single components of integrated photonic circuits on the nanoscale in analogy to the wafer testing applied in the electronic industry. This approach allowed us to gain information about the evolution of the spectral density inside the composite sample and to unravel the different physical occurring during ultrashort propagation. Specifically, the observed spectral reshaping is mainly caused by nonlinear optical effects inside a slow light photonic crystal waveguide and can be understood by an interplay between dispersion, free-carrier effects, and a wavelength-dependent slow light enhancement of various nonlinear effects. Reaching the same insight with only conventional transmission measurements, which reflect the behavior of the complete sample, is far from trivial, since the characteristics of the different components influence each other in a complex manner in the nonlinear optical regime.

## ASSOCIATED CONTENT

### Supporting Information

More details about sample design and fabrication, broadening of the transmission spectra, numerical calculations as well as error bars. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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### Notes

The authors declare no competing financial interest.

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# Supporting Information for Unravelling nonlinear spectral evolution using nanoscale photonic near-field point-to-point measurements

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## Sample details

The photonic crystal (PhC) consists of a 220 nm thick silicon membrane which is perforated with air holes. The holes have a radius of  $0.29a$  and are periodically arranged in a triangular lattice ( $a = 420$  nm). One row of holes is not present in the  $\Gamma - K$  crystal direction, creating the waveguide. The sample was fabricated by electron beam lithography, subsequent dry etching and sacrificial wet etching to release the membrane.<sup>1</sup> The two rows of holes directly adjacent to the waveguide are shifted by  $-0.11a$  and  $0.06a$ , respectively, to create a spectral slow light regime with low group velocity dispersion.<sup>2</sup> A negative shift is defined here as a displacement from the position dictated by the triangular lattice towards the waveguide and a positive shift as a displacement away from the waveguide.

The light injection from a ridge waveguide to a slow light photonic crystal can be very inefficient due to an increasing mode mismatch with increasing  $n_g$ . In our experiment, this mismatch

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would result in inefficient in- and outcoupling for wavelengths above 1577 nm. To overcome this problem we have changed the lattice constant of the first and last ten periods of our photonic crystal to a value of 440 nm which shifts the whole bandstructure to longer wavelengths. This shift creates a photonic crystal with  $n_g = 5$  across the entire spectrum of our laser, which acts as an interface between the ridge waveguide and the slow light PhC. It has been shown in literature that this gradual approach of first exciting a fast light mode, which then couples to a slow mode, is very efficient and can lead to injection efficiencies above 90 % for group indices up to 100.<sup>3</sup>

## Broadening of the transmission spectra

The width of a spectrum can be quantified by calculating its second moment, i.e. variance  $\sigma$ , which is defined by

$$\sigma^2 = \frac{\int (\bar{\lambda} - \lambda)^2 P(\lambda) d\lambda}{\int P(\lambda) d\lambda}. \quad (1)$$

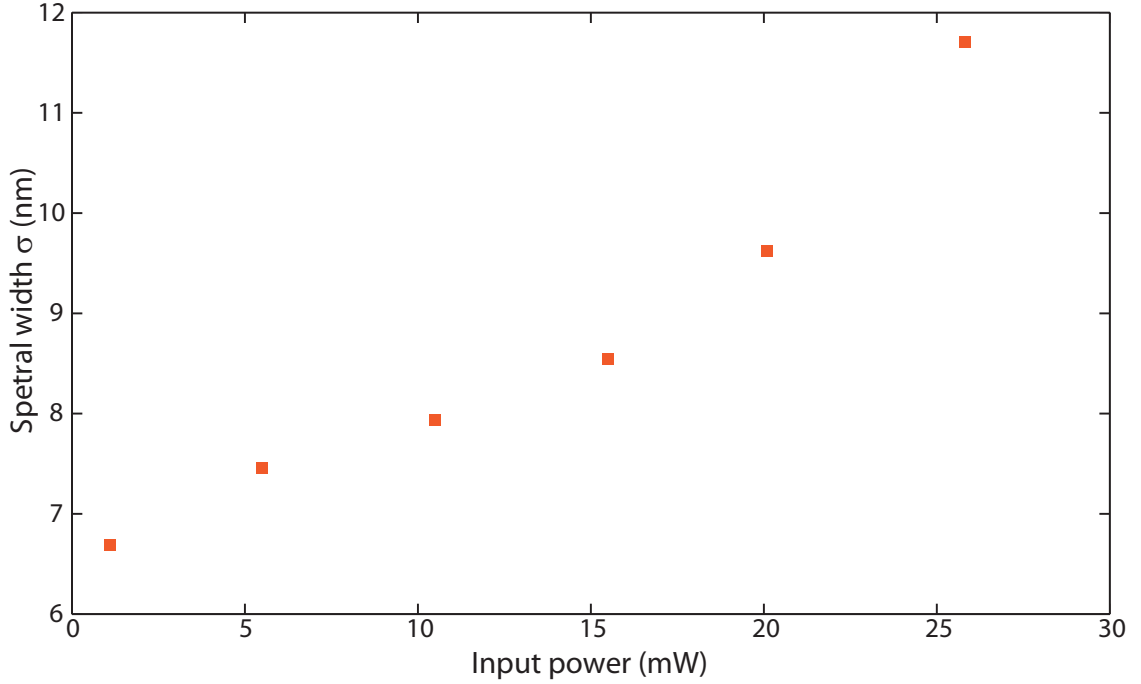
where  $\bar{\lambda}$  is the first moment, i.e. mean wavelength, as defined in Eq. (1) in the maintext.

The change of the spectral width, which is the square root of the variance, of the transmission spectra with input power is shown in Supporting Figure 1. It is obvious that the spectra get broader with increasing input power. By going from 1.1 mW to 25.8 mW the spectral width nearly doubles from 7 nm to 12 nm.

## Details of the modeling approach

To model the pulse propagation through the PhC waveguide Eq. (2) and Eq. (4) of the maintext are solved by means of the split-step Fourier method.<sup>4</sup> The temporal evolution of free-carrier density  $N_c$  is calculated by solving Eq. (3) of the maintext with a first-order finite-difference scheme.<sup>5</sup>

We fit our model to the measured pulse evolution by adjusting only two parameters: a simple scaling factor modeling the near-field probe pickup-efficiency, and a number that describes the



Supporting Figure 1: Spectral width of the transmission spectra shown in figure 2a in the maintext for different average input powers.

chirp the pulse experiences in the polymer access waveguide and the silicon spot-size converter. This latter parameter determines the temporal length of the pulse at the beginning of the PhC waveguide. As starting conditions the first spectrum taken at the beginning of the PhC waveguide and the free-carrier density  $N_c$  are set to zero. The best agreement between the model and the experimental results is achieved with a temporal length of 1.38 ps (FWHM). This amount of temporal broadening is reasonable considering the few millimeter distance that the pulse has to propagate through the sample before it enters the PhC waveguide. The blueshift of the spectral density, which occurs either inside the polymer access waveguide or the silicon spot-size converter, is taken into account in the modeling by shifting the centre frequency of the pulse in a one-time fit to the spectrum measured at the beginning of the PhC (cf. Figure 3d of the maintext).

The effective values of all wavelength-dispersive parameters used for simulating the propagation of the different spectral components, shown in Figure 4 of the main text, are presented in Supporting Table 1. The intrinsic material dispersion is negligible over the considered wavelength range. Thus, the observed wavelength dependency arises solely from the photonic crystal induced

dispersion of the slowdown factor  $S$  and its influence on the various parameters (see Table 1 in the main text).

The scaling of the linear loss with the slowdown factor is nontrivial, because it consists of both backscattering and out-of-plane scattering, which have different  $S$  dependencies. In our modelling we have assumed that the linear losses are dominated by out-of-plane scattering, which is typically for moderate group indices (e.g.  $n_g < 50$ , depending on the PhC design).<sup>6</sup> Consequently,  $\alpha_{\text{eff}}$  increases linearly with  $S$ .

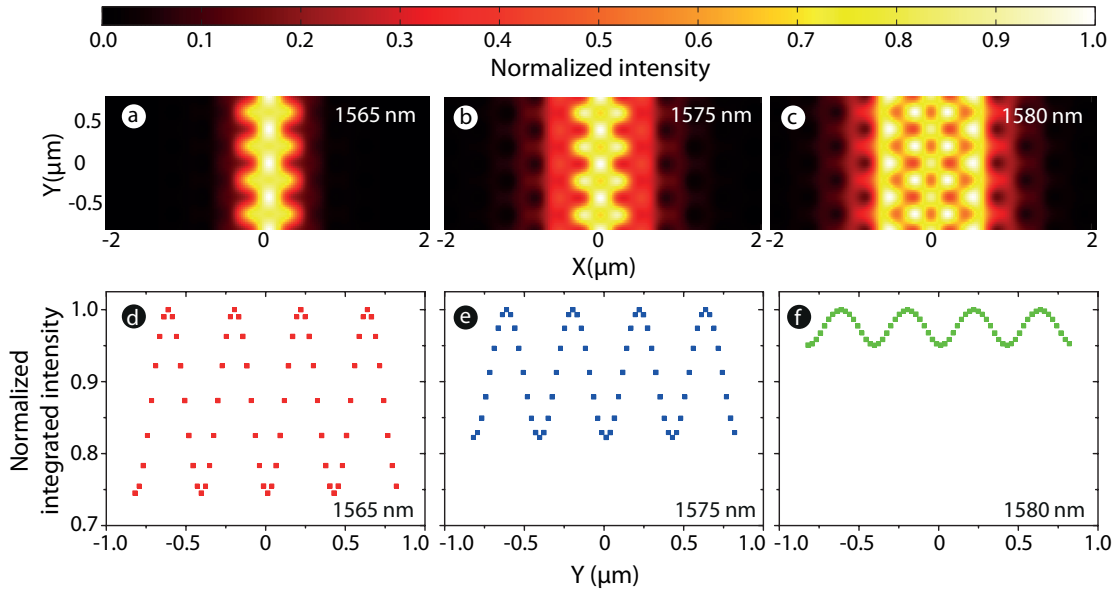
Supporting Table 1: The effective of all wavelength-dispersive parameters used in the simulation. These values are inclusive of the slowdown enhancement.

Free-space wavelength	$\alpha_{\text{eff}}(\text{dB}/\text{cm})$	$\beta_{\text{eff}}(\text{cm}/\text{GW})$	$n_{2,\text{eff}}(\text{cm}/\text{W})$	$\sigma_{\text{eff}}(\text{m}^2)$	$k_{\text{c,eff}}(\text{m}^3)$
1565 nm	22.93	3.97	$2.38 \cdot 10^{-13}$	$2.89 \cdot 10^{-21}$	$2.69 \cdot 10^{-27}$
1575 nm	27.09	26.24	$1.57 \cdot 10^{-12}$	$7.43 \cdot 10^{-21}$	$6.92 \cdot 10^{-27}$
1580 nm	31.78	227.48	$1.36 \cdot 10^{-11}$	$2.19 \cdot 10^{-20}$	$2.04 \cdot 10^{-26}$

## Error due to change of transversal field profile

As mentioned in the maintext there is a positional accuracy in placing the near-field probe always at a single position inside a unit cell of the PhC waveguide throughout the measurements. As a consequence, we observe an artificial amplitude modulation in the spectra taken by means of the SNOM. To quantify the possible variation of the signal, we simulate the intensity of the mode inside the PhC waveguide by using MIT Photonics Band Package (MPB).<sup>7</sup> We convolute the calculated intensity profile with a circle of 250 nm to mimic the effect of the aperture size of the near-field probe. The results for the three wavelengths, which are also investigated and modeled in the maintext, are shown in Supporting Figure 2a)-c).

Since we scan the near-field probe transversally over the PhC waveguide to counteract the different extension of the mode profiles visible in the MPB calculations, we integrate the simulated intensity distribution over the x-axis resulting in sinus curves with different modulation depths as shown in Supporting Figure 2d)-f). The error bars, used in Figure 4 in the maintext, are calculated



Supporting Figure 2: Calculated intensity distribution convoluted with a 250 nm diameter circle for a wavelength of (a)1565 nm, (b)1575 nm and (c)1580 nm for four unit cells of the PhC waveguide. Corresponding transversally integrated intensity curves.

by determining the ratio of the standard variation to the mean value of these integrated intensity curves, which are listed in Supporting Table 2. It is clear that the intensity profile at the shortest wavelength features the biggest modulation along the propagation direction leading to the largest error bar. For increasing wavelength, the modulation decreases.

Supporting Table 2: Table containing the mean value, standard variation and their ratio for the curves shown in Supporting Figure 2d)-f).

Wavelength	Mean	Standard variation	Ratio
1565 nm	0.87	0.09	10.38 %
1575 nm	0.91	0.063	6.93 %
1580 nm	0.98	0.018	1.8 %

As mentioned in the maintext, the 2 % error bar calculated for the wavelength 1580 nm is too small (cf. Figure 4 in maintext). We attribute this discrepancy to the slow group velocity that the mode inside the PhC waveguide experiences at this wavelength. The slow light enhancement leads to an increased sensitivity of the guided light to possible perturbations. Thus, we estimated an increased background noise level leading to an error bar of 5 %.

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