Free carrier absorption for high-resolution mode mapping in photonic crystal nanocavities

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We demonstrate a nonlinear photomodulation spectroscopy method to image the mode profile of a high-Q photonic crystal resonator (PhCR). This is done by scanning the PhCR surface with a 405 nm pump beam that modulates the refractive index by local thermal tuning, while probing the response of the resonance. We increase resolution by probing at high power, using the thermo-optical nonlinear response of the PhCR. Spatial resolution of the thermo-optical effect is typically constrained by the broad thermal profile of the optical pump. Here we increase the resolution and show that we can approach the diffraction limit of the pump light. This is due to free carrier absorption that heats up the PhCR only when there is overlap between the optical pump spot and the optical mode profile. This is supported with a thermo-optical model that reproduces the high-resolution mode mapping. Results reveal that the observed enhanced resolution is reached for surprisingly low carrier density.

I. INTRODUCTION

Photonic crystals (PhC) have raised interest from fundamental research [1, 2], due to their enhanced light-matter interaction, but maybe even more so from the field of optical signal processing and computing [3, 4]. They are prime candidates for optical integrated-circuit applications [5–7]. One reason for this is the possibility to design a defect on the PhC platform, creating a PhC resonator (PhCR) that typically has small mode volume and high quality factor (Q) [8] which maximizes the optical nonlinearity crucial to active photonic devices. Two-dimensional PhCs have been shown to be suitable for numerous applications such as fast optical switching [9, 10], reconfigurable circuits [11], optical memories [12] and optical parametric oscillation [13].

Investigating the optical mode profile of the PhCR is vital in all of these functionalities. Calculations to predict resonator modes, like FDTD simulation, do not provide complete information as PhCR modes are very sensitive to nanometer-scale imperfections arising from the fabrication process [14]. Therefore, high resolution imaging of PhCR modes is a necessary step in research and application development [15].

A benchmark method to characterize optical mode profiles is near-field scanning optical microscopy (NSOM), where a probe tip scans the surface of the PhC. High spectral and deep subwavelength spatial resolution imaging have been demonstrated [15–17], as well as phase contrast imaging [18] and imaging of the electric and magnetic field [19, 20]. However, the requirement of nanometer-distances between the tip and the sample can be demanding, and makes this method unsuitable for devices that are covered by a top cladding. Additionally, perturbation by the tip will affect the Q-factor of the cavity [21] making it challenging to image high-Q cavities of order $10^4 - 10^5$ without destroying the confinement [22].

Photomodulation spectroscopy (PMS) techniques introduce an excitation beam normal to the PhC surface that scans the PhC surface and perturbs the optical field by modulating the refractive index. The highest spatial resolution is achieved with electron or ion beam PMS scans, either with pulsed [23–25] or continuous wave (cw) [26] sources probing the resonance. It should be noted that exposure to ion and electron beams lowers the Q of the resonator, and in fact this technique is also being purposefully used to irreversibly alter or tune PhCs.

Ultrafast PMS experiments were demonstrated by Bruck et al. [11, 27], where the mode profile is imaged (and tuned) with an ultraviolet beam. Their method is rooted in a shift of the refractive index due to the Drude-like dispersion of a dense free-carrier plasma. This type of direct index perturbation needs a high concentration of carriers, and thus high-power optical excitation that can only be maintained on the picosecond scale due to risk of damage by photo-oxidation. The advantage then is the information in the time domain. However, picosecond-pulse excitation is not suitable for high-Q resonators, where the photon lifetime exceeds the lifetime of the carriers.

Thermal PMS is a method that is attractive due to its universality and ease of use. A pump beam is used to locally heat the PhC, and the resulting thermal change in refractive index leads to a shift in resonance frequency. Previously, our group has shown that with local thermal tuning the mode profile of a high-Q cavity can be recovered with a resolution that is limited by thermal diffusion [28, 30]. The effective spot size depends on the surrounding medium and it is generally much larger than the optical pump spot, so that even after deconvolution a moderate resolution remains [31].

In this paper we demonstrate that thermal photomod-
FIG. 1. Schematic of the GaInP PhC membrane, with lattice constant $a = 485$ nm, air hole radius of 136 nm, and membrane thickness of 180 nm. The mode-gap cavity consists of a broadened waveguide: the air holes lining the waveguide are shifted outwards from their lattice position (see colored air holes). We coupled NIR light into from the side into the injection waveguide, after which it evanescently couples to the barrier waveguide.

ulation spectroscopy in the nonlinear regime (NPMS) offers enhanced spatial resolution, due to the effect of a free-carrier absorption (FCA) term that only occurs when the pump and probe fields overlap spatially.

Our method reaches excellent sensitivity even with continuous wave excitation, because the high-Q resonator enhances both the probe field that heats the free carriers and the sensitivity with which the resulting thermal index shift is detected, both by a factor of order $Q > 10000$. In this way, the nonlocal thermo-optical effect amplifies a very small and local absorption term. This intrinsic amplification mechanism, leveraging the high $Q$ of the resonator, makes it possible to operate at a very low pump power, thus avoiding damage to the sample.

We reproduce our results using a thermo-optical (TO) model that takes into account all TO sources, showing that a significant effect already occurs when only a few hundred free carriers are present in the resonator.

II. EXPERIMENT AND THEORY

In Fig. 1 we show a schematic of the PhC, consisting of a 180 nm-thick GaInP slab with air holes in a triangular lattice and a lattice constant of $a = 485$ nm. To achieve a nanocavity with a high $Q$ and small mode volume, waveguide width modulation is used to create a mode-gap cavity [32, 33]. NIR probe light is coupled into the injection waveguide with a C-band wavelength-swept laser and then evanescently couples through to the barrier waveguide. A small fraction of light inside the cavity is scattered out of plane and collected with a 0.4 NA objective onto a cooled photodiode. The same objective focuses out-of-plane pump light onto the sample. The 405 nm pump light is steered by a fast scanning mirror, providing a localized heat source with which we scan the surface of the membrane.

In Fig. 2 we show a schematic of the three TO sources in the crystal. In Fig. 2(a) we depict the processes leading to direct probe light absorption in the PhC. Since the bulk bandgap exceeds the probe photon energy by more than a factor two this takes place via surface states, impurities, free electrons in the semiconductor, or a combination of these processes. This probe absorption is the origin of the TO nonlinearity which causes the well-known hysteresis behavior shown in Fig. 2(b) [30, 34]. Secondly, Fig. 2(c,d) shows how direct heating by the pump causes a redshift of the cold cavity resonance $\lambda_0$ when the thermal profile overlaps with the optical mode profile of the resonance [28]. In Fig. 2(e,f) a third source of heat presents itself when the free carriers generated by the pump absorb a probe photon, i.e., FCA. These hot carriers heat the lattice and increase the TO redshift, causing an increase in the hysteresis. The FCA process occurs only in the region where the pump spot and optical mode profile overlap, and is proportional to the product of pump and probe energy densities.

We model the system by solving the coupled optical resonance condition and the partial differential equation for thermal diffusion for all three TO sources. We solve the differential equation numerically using the Sturm-Liouville theory, propagating the modes in an orthogonal base [35]. See Appendix A for a full treatment of the theoretical model.

A qualitatively similar model by Iadanza et al. [34] differs from ours as it approximates the temperature distribution with elliptical regions. However, to interpret mode profile scans we need to specifically take into account the spatial degrees of freedom of the PhCR mode and use it to evaluate the temperature distribution in time and space on a high resolution grid.

III. RESULTS

We perform NPMS measurements on a high-Q mode-gap cavity and image the optical mode profile. The procedure consists of a pump line scan: probing at consecutive pump positions in a horizontal or vertical line. At each pump position we take a probe wavelength sweep (in the direction of increasing $\lambda$) to obtain a cross section of the mode profile. First we obtain both the theoretical and experimental responses of the known mode profile of the fundamental mode-gap resonance and validate the TO model. Secondly, we demonstrate that the increased pump sensitivity due to FCA resolves the first higher-order (HO) mode of the mode-gap resonance in the nonlinear regime.
FIG. 2. Schematic of the TO sources in the PhC. (a) NIR probe absorption by defect states causes (b) hysteresis behavior at high probe power (red lineshape). The blue lineshape illustrates the low-power, linear response. Dashed lines are instable branches. (c) Interband pump absorption of the 405 nm excitation laser generates a (d) shift of the resonance, i.e., a redshift of the complete lineshape, here depicted for the low-power lineshape. (e) Pump absorption creates free carriers that absorb NIR probe light, i.e., FCA (purple arrow), this causes an (f) increase in the hysteresis in the nonlinear lineshape (red and purple).

FIG. 3. Input mode for the theoretical NPMS vertical line scan. (a) FDTD calculated mode profile of a fundamental mode-gap resonance, with a cross section through the center of the mode plotted in gray. (b) Overlay of the input mode on top of a CCD image of the PhC waveguide. Orange dashed line and arrow indicate line scan and scan direction.

A. Fundamental mode-gap resonance: vertical cross section

Fig. 3(a) shows the optical mode profile of the 3D FDTD-simulated mode-gap resonance that we use to generate the theoretical NPMS obtained profile. The vertical line scan goes through the center of the mode-gap resonator as depicted in Fig. 3(b).

In Fig. 4 we show the experimental and theoretical NPMS vertical line scan of the mode-gap resonance profile, performed with a 0.5 μm FWHM pump spot. This was done for three increasing probe energies to investigate the response of the mode in both the linear and nonlinear regime, effectively increasing FCA. We observe a nearly perfect agreement between the experimentally measured and numerically calculated profiles. A full list of the fit parameters and constants used in the model can be found in Appendix B. All fits are found by manually adjusting four fit parameters, as an automated fit procedure failed to converge in a reasonable number of iterations, especially as each iteration requires a time-intensive calculation.

In Figs. 4(a,d), at low probe power, pump absorption is the only significant TO effect. The resonance is redshifted by the pump where the mode profile and pump thermal profile overlap, i.e., when the pump is close to the cavity. A dip in the redshift when the pump hits the center of the waveguide is explained by considering the thermal conductance of the PhC. On the waveguide there are no air holes, causing a higher thermal conductance. Additionally, in the model we account for the observation that the pump is partially reflected by the waveguide (it...
FIG. 4. NPMS vertical cross section of the mode profile of a mode-gap cavity, experimentally measured via the out of plane scattered light at (a) the linear response regime at low input power, at (b) increasing input power and (c) high input power deep into the nonlinear response regime. The simulated NPMS mode profile in (d), (e) and (f) for increasing input power left to right, generated with the input mode of Fig. 3, matching the corresponding experimental results. See Appendix B for fit details. (g,h,i) The isolines of the profiles, corresponding to the white isolines in (b,c,e,f). A cross section of the FDTD profile (corresponding to Fig. 3(b)) is indicated with a shaded area (gray).

shows up bright at the pump wavelength) diminishing the absorbed-pump power by 20% when the pump beam is on the waveguide.

In Figs. 4(b,e), at increasing probe power, the TO effect causes an asymmetric lineshape (see Figs. 2(b,f)) that maximally detunes the resonance when pump and probe heat up the mode cumulatively.

In Figs. 4(c,f) we probe deep into the non-linear regime. The high-energy-density areas of the mode profile become even more pronounced, as the TO detuning reaches further into the long wavelengths due to FCA. It is by these shapes that we scale the FCA effect to match the experiment and find an agreement with a carrier lifetime of $\tau_c \approx 0.4$ ns for all simulations throughout this article. The strength of the FCA source is related to the product of the carrier lifetime and the effective cross section, see Appendix A for details.

The carrier lifetime is the parameter with the largest a priori uncertainty since no measurements in these type of GaInP slabs exist to the best of our knowledge. Therefore we use it as a fit parameter. Our fit results should not be regarded as a measurement of the carrier lifetime, though it falls within the expected range for a GaInP PhC considering small volume to surface ratio [36–39].

The lifetime translates to a carrier density of $\rho_c \approx 1 \times 10^{16}$ cm$^{-3}$ or equivalently not more than $\sim 500$ carriers in the cavity at any time.

In Figs. 4(g-i) low- and mid-level isolines of the experimental and theoretical profiles are shown. Since the high intensity edge of the high-power profiles marks the transition to an unstable state (see lineshapes of Fig. 2(b,f)) the edge is very sensitive to any effect that might disturb the balance at the edge of the stable energy branch, such as a nearby dark mode, variation in the incoupling efficiency, or disorder in the PhC. Therefore, comparison of isolines that are situated in the stable region of the profile is more reliable. In Appendix C we present another mode-gap resonance that has a suppressed high energy density peak of the mode profile, but nonetheless shows excellent agreement with the model in the stable regimes of the mode profile.

Fig. 4 demonstrates that imaging of the PhCR mode improves at high probe power since the highly broadened FDTD profile in Fig. 4(g) gains resolution in Fig. 4(i).
FIG. 5. Input mode for the theoretical NPMS horizontal line scan. (a) Render of the first higher-order Hermite-Gaussian function that is used as the input mode of the theoretical NPMS profile. A horizontal cross section of the input mode is plotted in gray. (b) Overlay of the input function on top of a CCD image of the PhC waveguide. The orange dashed line and arrow indicate the horizontal line scan and scan direction.

FIG. 6. NPMS horizontal line scan of the first higher-order mode-gap resonance, yielding a cross section of the optical mode profile. (a) The profile is experimentally measured via out-of-plane scattered light at low probe power and (b) the theoretical profile is generated with the input mode from Fig. 5 at low probe power. Additionally, (c) the experimentally measured profile at high probe power and (d) the theoretical profile at high probe power. For the theoretical profiles, a horizontal cross section of the input mode is plotted in white.

B. Higher order resonance: horizontal cross section

Fig. 5(a) depicts the first HO mode of a Hermite-Gaussian function by which we approximate the first HO mode of the mode-gap resonance. We use this HO mode to generate the theoretical NPMS profile by performing a horizontal scan over the center of the waveguide as shown in Fig. 5(b).

Fig. 6 shows the experimentally and theoretically obtained mode profile of the HO mode-gap resonance in both the low and high probe power regime. The low-probe power, experimental profile in Fig. 6(a) shows no details that would point to the measurement of a HO mode or any other mode that would have multiple peaks.

In correspondence, the low-power theoretical profile in Fig. 6(b) shows the same single broad-peaked shape.

However, at higher probe power the NPMS experimental mode profile in Fig. 6(c) reveals a centered double peak that highly resembles the double peak of the HO mode-gap profile modeled in Fig. 6(d). We see a very good agreement between experiment and theory is obtained by assuming this resonance is the first HO mode of the mode-gap resonance. We note that the characteristics of this HO mode can only be observed in the nonlinear regime at high probe power, where FCA is significant, proving again that our method enables high-resolution mode mapping of the resonance.

The distance between the peaks in Fig. 6(d) is fitted to match Fig. 6(c) by setting the waist of the HO Hermite-
Gaussian mode to $w = 2.3\mu m$. We remark that the waist of the HO mode is of the same order as the fundamental mode, see Fig. 3.

We use the same fit method and parameters in the model as before in Fig. 4 including the same carrier lifetime that scales the FCA source strength. See Appendix B for a full list of constants and parameters.

IV. DISCUSSION

In our current experimental setup we do not reach the resolution limit of the imaging method. The most important limiting factor for the resolution is the pump focus. Therefore we simulate with the realistically smallest pump spot of 213 nm (obtainable with an objective of NA = 0.95) and find a resolution limit of 240 nm using Sparrow’s criterion. This suggests we can indeed approach the diffraction limit of the visible light pump. The model does not include carrier diffusion, which might become significant at the nanometer scale, but this resolution limit opens up prospects of measuring the standing waves of the optical mode. See Appendix B for details on the resolution limit simulation.

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In this section we formulate a thermo-optical (TO) model by solving the optical resonance condition coupled to the thermal diffusion equations for all three TO sources. This means we will find an expression for the TO sources and work towards the full heat equation of the system, taking into account the optical mode in the waveguide, the thermal properties of the PhC and the optical pump.

### a. Carrier absorption

First we will find an expression for the heat produced by free carrier absorption (FCA) as depicted in Fig. 2(e), i.e., free carriers produced by the pump light absorb probe light. To find the thermal dissipation from the FCA source we multiply the optical absorption coefficient, \( c \) is the speed of light, \( n \) the refractive index, \( E_{pr}(t) \) is the input probe light energy, \( U_{pr}(x,y) \) the optical mode profile of the probe field, and \( I_{pr}(x,y,t) \) is the spectral lineshape of the resonance. The probe energy is determined by the input probe power \( P_{pr} \) and a coupling parameter \( \tau_{pr} \) accounting for the incoupling efficiency of the probe light to the cavity, which means \( E_{pr}(t) = \tau_{pr}P_{pr}(t) \). The absorption coefficient is related to the effective FCA cross section \( \sigma_{FCA} \) and the carrier density \( \rho_{c}(x,y) \) by \( \alpha_{FCA}(x,y,t) = \sigma_{FCA}(x,y,t) \rho_{c}(x,y,t) \). Where the effective absorption cross section is the summed cross sections of all carrier types, where for each type \( \sigma_{i} = e^{3}A_{FCA}^{2}/4\pi^{2}c^{3}(m_{i}^{*})^{2}\mu_{i}\epsilon_{0} \epsilon_{r} \) is the absorption cross section of a single carrier. Here \( e \) is the electron charge, \( \lambda_{pr} \) the probe wavelength, \( m^{*} \) the effective carrier mass, \( \mu \) is the carrier mobility and \( \epsilon_{0} \) the vacuum permittivity.

The carriers are created by the pump light. We assume the carriers stay localized on the optical timescale.

\[
\alpha_{FCA}(x,y,t) \times \left( \frac{1}{n} E_{pr}(t) U_{pr}(x,y) I_{pr}(x,y,t) \right),
\]

where \( \alpha_{FCA}(x,y) \) is the probe absorption coefficient, \( c \) is the speed of light, \( n \) the refractive index, \( E_{pr}(t) \) is the input probe light energy, \( U_{pr}(x,y) \) the optical mode profile of the probe field, and \( I_{pr}(x,y,t) \) is the spectral lineshape of the resonance.
which means the carrier density is directly proportional to the local production rate of carriers and their lifetime, such that

$$\rho_c(x, y, t) = \frac{\tau_c}{\hbar \omega_{pu}} P_{pu}(t) U_{pu}(x, y), \quad (A2)$$

with $\tau_c$, the average lifetime of a carrier, $d$ the thickness of the membrane, $\omega_{pu}$ the pump light frequency, $P_{pu}(t)$ the pump power, and $U_{pu}(x, y)$ the optical mode profile of the pump. This finally leads to the expression

$$S_{\text{FCA}} = \sigma_{\text{FCA}} \left( \frac{\tau_c}{\hbar \omega_{pu}} P_{pu}(t) U_{pu}(x, y) \right) \times \left( \frac{C_{\text{pr}}}{n} P_{pr}(t) U_{pr}(x, y) I_{pr}(x, y, t) \right). \quad (A3)$$

b. Heat Equation

The heat equation, taking into account all TO sources (shown in Fig. 2), is a partial differential equation which we solve using a stable spectral method on a high-resolution space-time grid. We combine the in-plane cooling through dissipation in the PhC membrane, out-of-plane cooling via the surrounding gas layer and substrate, and all heating terms, including the FCA term of Eq. (A3), into the heat equation

$$C_{2D} \frac{\partial T(x, y, t)}{\partial t} =$$

$$-K_{2D} \nabla^2 T(x, y, t)$$

$$- K_{\text{gas}} (T(x, y, t) - T_0)$$

$$+ \alpha_{\text{prA}} P_{\text{pr}}(t) U_{\text{pr}}(x, y)$$

$$+ \beta_{\text{puA}} P_{\text{pu}}(t) U_{\text{pu}}(x, y)$$

$$+ \sigma_{\text{FCA}} \frac{\tau_c}{\hbar \omega_{pu}} P_{\text{pu}}(t) U_{\text{pu}}(x, y)$$

$$\times \left( \frac{C_{\text{pr}}}{n} P_{\text{pr}}(t) U_{\text{pr}}(x, y) I_{\text{pr}}(x, y, t) \right),$$

where $C_{2D}$ is the 2D specific heat of the PhC membrane, $K_{2D}$ the thermal conductivity of the PhC membrane, and $K_{\text{gas}}$ the thermal conductance of the gas layer between the air-suspended membrane and substrate. At the boundary we have $T = T_0$ the temperature at the edge of the photonic crystal where it meets the substrate. Out-of-plane cooling via the other side of the PhC membrane into free space is neglected, as this contributes less then one percent of the heat loss. For the heat produced via probe absorption we take into account the absorption fraction $\alpha_{\text{prA}}$, i.e., the probability a cavity photon is absorbed rather than scattered or leaked out of the cavity. Direct pump heating is related to $\beta_{\text{puA}}$, the absorption fraction of the pump light.

The resonance condition is hidden in the optical energy density of the probe field that is related to the spectral lineshape of the resonance

$$I_{\text{pr}}(x, y, t) = \frac{\Gamma^2}{\Gamma^2 + (\Delta - \delta_{\text{th}}(x, y, t))^2}, \quad (A5)$$

where $\Gamma$ is the resonance linewidth, $\Delta \equiv \lambda_{\text{pr}} - \lambda_0$ is the detuning, and $\delta_{\text{th}}(t)$ is the thermal resonance shift that causes a dynamic detuning expressed by

$$\delta_{\text{th}}(x, y, t) = \eta \int U_{\text{pr}}(x, y) T(x, y, t) dx dy. \quad (A6)$$

Here, $\eta$ is the thermo-optical coefficient of the semiconductor material.

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Appendix B: Simulation parameters

Table IV shows the parameters and constants used in the model to generate Figs. 4(a,d,f), 6(b,d), and 7(d,e,f).

The on-chip pump power cannot be directly measured. We measure the upper limit of the on-chip pump power, scale to fit the redshift of the cold resonance observed in the experiment, and then fixed for all simulations. The shift of the cold resonance (in the linear response regime of the cavity) is directly related to the pump power and has no crosstalk with the other fit parameters.

The probe energy $E_{\text{pr}}$ determines the detuning at which the resonance drops out of stability, i.e., the high intensity edge. Therefore, the probe energy is a fit parameter to match the experimental hysteresis edge at pump position $\pm 10 \mu m$ where the pump has negligible effect on the optical mode. The spectrum of the input probe energy is not entirely flat, therefore we normalize it using the back reflected light measured through the injection waveguide.

The diminished pump absorption on the waveguide (caused by partial reflection on the waveguide) is determined in the vertical scan by fitting the fringe on top of the waveguide, see Fig. 4(a,d).

The fourth and last fit parameter is related to the strength of the FCA term, i.e., the amount of heat produced by FCA. Looking at Eq. (A3) the FCA source is proportionate to the product $\sigma_{\text{FCA}} \tau_c$, the effective FCA cross section and the carrier lifetime. We use the effective carrier mass and mobility to calculate the effective cross section and use the carrier lifetime as fit parameter to match the resolution of the experimental mode profile. A small carrier lifetime leads to a broadened profile with less sharp peaks and vice versa.

Using the values from Table IV the fit yields a carrier lifetime of $\tau_c \approx 0.4 \text{ ns}$. This is an approximated value, as
| symbol   | value       | source |
|----------|-------------|--------|
| refractive index | n | 3.06 | [44] |
| thermal conductivity of GaInP | κ_{GaInP} | 4.9 W/m.K | [45] |
| thermal conductivity of N$_2$ | κ$_{N_2}$ | 0.024 W/m.K | [46] |
| specific heat of GaInP | C$_{sp}$ | 310 J/K.kg | [47] |
| density of GaInP | ρ | 4810 kg/m$^3$ | [48] |
| thickness of the PhC membrane | h | 180 nm | | |
| filling fraction | φ$(x, y)$ | 0.714 (PhC membrane) | 1.0 (waveguide, bulk) | |
| 2D specific heat | C$_{2D}$(φ$(x, y)$) | hφ$(x, y)$ρC$_{sp}$ | | |
| thermo-optical coefficient | η | $-2 \times 10^{-4}$ K$^{-1}$ | | |
| on-chip upper limit of pump power | P$_{lim}$ | 1.56 μW | | |
| on-chip pump power | P$_{pu}$ | 0.35 × P$_{lim}$ | | |
| absorption fraction of probe | α$_{PrA}$ | 0.05 | | |
| absorption fraction of pump | β$_{PuA}$ | 1.0 (PhC membrane) | 0.2 (waveguide) | |
| electron effective mass | m$_e^*$ | 0.088m$_0$ | | |
| light hole effective mass | m$_{lh}^*$ | 0.12m$_0$ | | |
| heavy hole effective mass | m$_{hh}^*$ | 0.7m$_0$ | | |
| electron mobility | μ$_e$ | 1000 cm$^2$/Vs | | |
| hole mobility | μ$_h$ | 40 cm$^2$/Vs | | |
| effective FCA cross section | σ$_{FCA}$ | σ$_e$ + 1/2(σ$_{lh}$ + σ$_{hh}$) | | |

| resonance wavelength | λ$_0$ | 1547.960 nm | | |
| resonance linewidth | Γ | 5 pm | | |
| FWHM of the pump in (x,y)-direction | | | | |
| carrier lifetime | τ$_c$ | 0.4 ns | | |
| probe light energy | | | | |

| resonance wavelength | λ$_0$ | 1530.952 nm | | |
| resonance linewidth | Γ | 9 pm | | |
| FWHM of the pump in (x,y)-direction | | | | |
| carrier lifetime | τ$_c$ | 0.4 ns | | |
| probe light energy | | | | |

| resonance wavelength | λ$_0$ | 1536.106 nm | | |
| resonance linewidth | γ | 4 pm | | |
| FWHM of the pump in (x,y)-direction | | | | |
| carrier lifetime | τ$_c$ | 0.4 ns | | |
| probe light energy | | | | |

| TABLE I. Constants and parameters used to generate the theoretical mode profiles of Figs. 4(d,e,f), 6(b,d) and Fig. 7(d,e,f). |

the effective cross section has large uncertainty due to the fact that the population of heavy and light carrier holes is unknown. Additionally, the effective carrier mass and carrier mobility cannot directly be measured in our experimental setup, thus we use literature values with limited precision. With Eq. (A2) we find the carrier density created by the cw pump spot to be $ρ_c \approx τ_c P_{pu}/hω_{pu}dπr^2$, where $r$ is half the FWHM of the pump spot. This leads to the carrier density of $ρ_c \approx 1 \times 10^{16}$ cm$^{-3}$, or equivalently, not more than $\sim$500 carriers in the PhCR at any time.

Appendix C: Suppressed FCA response

At high optical probe energy the high energy branch of a bistable state can be susceptible to many disturbances that destabilize the branch. This is demonstrated in Fig. 7 where we show a resonance that is suppressed in its nonlinear behavior as we do not see states with large detuning when we have overlap between pump and mode, due to an instable high energy branch. Nevertheless, the isolines in the stable part of the high energy branch (at low and medium intracavity energy) still match the theory.
FIG. 7. NPMS vertical cross section of the mode-gap cavity profile, experimentally measured at (a) low input power, (b) increasing input power and (c) high input power. The simulated NPMS mode profile in (d), (e) and (f) for increasing input power, generated with the input mode of Fig. 3. (g,h,i) The isolines of the profiles.

FIG. 8. Simulation of the resolution limit using Sparrow’s criterion. Increasing distance between Gaussian peaks: (a) 220, (b) 240, (c) 260 nm. Input modes are plotted in white. At (b) 240 nm the two peaks can still be resolved.

Appendix D: Resolution limit simulation

The most important limiting factor for the resolution is the size of the pump beam. To calculate the resolution limit we use a very tight but realistic pump focus that can be obtained with an objective of NA = 0.95. We then generate the mode profile for two sharp, adjacent Gaussian modes with FWHM = 100 nm and varying distance on the x-axis between them, using our TO model.

Fig. 8 shows the simulation demonstrating that the resolution limit lies at 240 nm following Sparrow’s criterion. All constants and parameters are identical to the horizontal scan of Fig. 6 except for an adjusted pump FWHM of 213 nm in both the x and y direction (see Table I).