Time-resolved resonance and linewidth of an ultrafast switched GaAs/AlAs microcavity

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Abstract

We explore a planar GaAs/AlAs photonic microcavity using pump-probe spectroscopy. Free carriers are excited in the GaAs with short pump pulses. The time-resolved reflectivity is spectrally resolved short probe pulses. We show experimentally that the cavity resonance and its width depend on the dynamic refractive index of both the \( \lambda \)-slab and the \( \lambda/4 \) GaAs mirrors. We clearly observe a double exponential relaxation of both the cavity resonance and its width, which is due to the different recombination timescales in the \( \lambda \)-slab and the mirrors. In particular, the relaxation time due to the GaAs mirrors approaches the photon storage time of the cavity, a regime for which nonlinear effects have been predicted. The strongly non-single exponential behavior of the resonance and the width is in excellent agreement to a transfer-matrix model taking into account two recombination times. The change in width leads to a change in reflectivity modulation depth. The model predicts an optimal cavity \( Q \) for any given induced carrier density, if the modulation depth is to be maximized.

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I. INTRODUCTION

Recently, there has been a fast-growing interest to switch cavities on ultrafast timescales. Switching cavities is important for dynamical control of light, such as optical wavelength modulation, bandwidth conversion, density of states switching, and trapping and releasing photons [1-5]. Micro- and nanocavities are particularly attractive in this context, as they are amenable to be integrated in next generation all-optical networks. Cavity switching becomes especially interesting when the photon storage time $\tau_{\text{ph}}$ becomes comparable to the timescales on which the cavity resonance changes [6]. In this regime, strong pulse chirping and frequency conversion effects have been observed [7, 8].

The availability of strong, short pulses, the good reproducibility, and the possibility of integration in all-optical networks has furthered the popularity of free-carrier switching of resonators or cavities, see, e.g., Refs. [3, 9, 10]. The dispersion of these carriers result in a change of both real ($n'$) and imaginary ($n''$) part of the refractive index that is proportional to the induced carrier density $N$. The carriers recombine within a typical timescale ranging from ps to ns, and the cavity resonance relaxes to its unswitched value. Surprisingly, these relaxation and broadening mechanisms have hardly been discussed, mostly due to the lack of frequency- and time-resolved data. While other groups have limited their studies to either the reflectivity at two frequencies [3, 11-14] or to the reflectivity at two probe delays [15], we present here a systematic study at all frequencies and delays which allows us to investigate the dynamic resonance and lineshape. In our earlier contribution, insufficient spectral resolution had prevented us from investigating these broadening mechanisms [9]. Here, we study both the dynamic shifting and broadening of a cavity resonance by performing time- and frequency resolved pump-probe spectroscopy on a planar GaAs/AlAs microcavity, with a frequency resolution much higher than the dynamic cavity linewidth.

The most general cavity is a slab of a dielectric, bounded by two mirrors, see Fig. II(a). This well-known Fabry-Pérot cavity serves as the basis for the understanding of 1-, 2- and 3D cavities [16]. Many important physical properties, such as storage time, bandwidth, mode volume and reflectivity depend on the optical properties of both the slab and the mirrors, and can be calculated analytically for such a model system [17]. Therefore, to obtain a physical understanding of switching of advanced photonic crystal micro- and nanocavities, it is advantageous to consider first the Fabry-Pérot cavity. One of such processes is switching

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by exciting free carriers in the mirrors and the $\lambda$-slab with a strong pump pulse, whereby the cavity resonance changes rapidly within $\tau_{on}$, and relaxes back to its original state within $\tau_{off}$. Previously, nonlinear effects observed by Refs. [7, 8] were obtained during the notoriously uncontrollable rapid up-change time of the cavity resonance, as $\tau_{on}$ depends on material parameters. Therefore, many efforts are devoted to decreasing the timescale of $\tau_{off}$ [10, 11, 18, 19]. Here, we show how the relaxation time can quite easily approach the storage time of the cavity, and show how this can be explained in terms of the basic Fabry-Pérot cavity. Nonlinear effects for optimized planar microcavity samples are expected during $\tau_{off}$. Switching of planar microcavities is thus expected to be a step towards achieving nonlinear pulse phenomena.

II. EXPERIMENTAL SETUP

Our sample is a planar microcavity consisting of a GaAs $\lambda$-slab sandwiched between two Bragg mirrors, which was also studied earlier [9]. The sample is schematically shown in Fig. 1(b). The slab has a thickness of $275.1 \pm 0.1$ nm, and the Bragg mirrors consist of 12 and 16 pairs of $\lambda/4$ thick layers of nominally pure GaAs or AlAs. The sample gives rise to a cavity resonance at $E_{cav} = 1.2783$ eV, well below the bandgap of GaAs ($E_g = 1.44$ eV) to avoid intrinsic absorption at the cavity resonance. The sample is grown with molecular beam epitaxy at $550^\circ C$ to optimize the optical quality, but no effort was put into reducing the recombination time. A slight variation of a few % in stopgap and cavity resonance over the sample allowed us to verify the experimental observations for different resonance frequencies.

We performed two separate studies which yielded consistent results. For experiments outside the present scope the sample was doped with $10^{10}$ cm$^{-2}$ InGaAs/GaAs quantum dots, which hardly influence our experiment [20]. The experiments were conducted at room temperature. Continuous-wave (cw) reflectivity was measured with a broad band white light setup with a spectral resolution of $\sim 0.25$ meV.

Our switching setup consists of two optical parametric amplifiers (OPA, Topas), that are the sources of the pump and probe beams [21]. The frequency of both OPAs are computer controlled and have a continuously tunable output frequency between 0.44 and 2.4 eV. The repetition rate is $f_{rep} = 1$ kHz. In order to verify that the cavity was not degrading due to heating effects, we simultaneously measured both switched and unswitched spectra by using
FIG. 1: (a) The model Fabry-Pérot system, consisting of a slab bounded by two mirrors. The resonance depends on both the optical properties of the mirrors and of the slab. (b) Schematic of sample. The GaAs is indicated by the shaded areas. The GaAs λ-slab is bounded by 12 and 16 pairs of λ/4 GaAs/AlAs pairs at the front and at the rear side, respectively. The intensity distribution is shown at the cavity resonance, for a unity field.

a chopper. This averaging caused the effective pump or probe rate to be \( f_{\text{rep}}/2 = 500 \text{ Hz} \). The alignment and synchronization of the pulses to the chopper was chosen so as to overlap only every second pump pulse with a probe pulse, so that the effective pump rate was \( f_{\text{rep}}/4 \) \[22\]. The pulse duration is \( \tau_p = 140 \pm 10 \text{ fs} \) (measured at \( E_{\text{Pump}} = 0.95 \text{ eV} \)) \[23\], and the spectral width is \( \Delta E/E_0 = 1.33\% \). A 40 cm long delay stage controls the probe delay with a time resolution of \( \Delta t = 10 \text{ fs} \). The pump beam has a much larger Gaussian focus of 113 μm FWHM than the probe beam (28 μm), ensuring that only the central flat part of the pump focus is probed. The probe fluence of \( I_{\text{Probe}} = 1 \pm 0.3 \text{ mJcm}^{-2} \) is kept well below the pump fluence of \( I_{\text{Pump}} = 30 \pm 3 \text{ mJcm}^{-2} \) to prevent inadvertent pumping by the probe pulses. These high pump fluences, which shift the cavity resonance by several linewidths, facilitate the analysis of the dynamic behavior. Free carriers are excited in the GaAs by two-photon absorption at \( E_{\text{pump}} = 0.73 \text{ eV} \) to obtain a spatially homogeneous distribution of carriers \[24\].

In order to resolve the dynamic lineshape of the microcavity, the reflected probe light was analyzed with a spectrometer. This spectrometer consists of a PI/Acton SP-2558 spectrograph, using a 1024 channel InGaAs detector (OMA-V), yielding a resolution of 0.12 meV at 1.24 eV, much higher than the unswitched cavity linewidth of \( \Delta_0 = 1.03 \text{ meV} \). To average over pulse-to-pulse variations of the OPAs, the spectrograph was operated in free-running mode for a duration of 1 s, acquiring 500 probe pulses, of which which half of them are
incident together with a pump pulse.

III. LINEAR REFLECTIVITY

Fig. 2(a) shows a white light reflectivity spectrum of the planar photonic microcavity at normal incidence. The high peak between 1.193 and 1.376 eV is due to the stopgap of the Bragg stacks. The stopband has a broad width of 0.183 eV (14.3% relative bandwidth), which confirms the high photonic strength. Near 1.279 eV we observe a sharp resonance caused by the λ-slab in the structure. This resonance has a width of 1.7 meV that is limited by wavevector spreading caused by the finite numerical aperture of 0.12. A transfer matrix (TM) calculation was performed for the reflectivity. In the calculation, we included the dispersion and absorption of GaAs [25] and AlAs [26]. The only free parameters in the model were the thicknesses of the GaAs ($d_{GaAs} = 68.78 \pm 0.03$ nm) and AlAs ($d_{AlAs} = 81.90 \pm 0.03$ nm). The results reproduce the experimental resonance, stopband, and Fabry-Pérot fringes well.

We made use of the large probe bandwidth compared to the cavity linewidth to spectrally resolve the microcavity with the spectrometer (Fig. 2(b)): The spectral width of the cavity at the resonance $E_{cav} = 1.2783$ eV is $\Delta_0 = 1.03$ meV, and corresponds to an unswitched quality factor of $Q_0 = 1242$ and thus a photon storage time of $\tau_{ph} = 640$ fs. This larger quality factor with respect to the cw measurement is attributed to the reduced wavevector spreading due to the lower numerical aperture (NA = 0.02) which is confirmed by the higher modulation depth. The large probe bandwidth in combination with the high spectral resolution allows us to probe not only the cavity resonance as in [9] but also the lineshape, even when the cavity resonance has shifted by several linewidths.

IV. RESULTS AND DISCUSSIONS

We dynamically probe the excited cavity by time- and frequency resolved pump-probe reflectivity. Figure 3(a) shows the transient reflectivity spectrum as a function of probe delay. The unswitched cavity resonance is clearly seen at 1.2783 eV. The width of the unswitched cavity resonance does not increase in the slightest, as shown by the half maxima. We therefore conclude that there are no permanent damage or heating effects to the sample.
FIG. 2: (a) Continuous wave reflectivity spectrum of the planar microcavity (open circles). The cavity resonance can clearly be seen at $E_{\text{cav}} = 1.279$ eV. The solid curve is a transfer matrix calculation that includes the dispersion and absorption of GaAs and AlAs. (b) Reflectance spectrum in analog to digital units (adu) per pixel per second from the same microcavity (circles) measured with the pulsed probe beam ($\tau_P = 140 \pm 10$ fs, $\Delta \omega_P = 1.35\%$), and from a reference (squares).

In Fig. 3(a), the second trough emerging at 1.2898 eV, at $\Delta t = 3$ ps, is the switched cavity resonance. Up to probe delays $\Delta t = \tau_{\text{on}} = 3$ ps, the cavity resonance shifts by as much as $\Delta E_{\text{cav}} = 11.5$ meV, or 11 cold cavity linewidths. This switching-on time is in good agreement to the thermalization time of free carriers in GaAs [27, 28]. Both the resonance energy and linewidth relax to their initial values within $\Delta t = 100$ ps. Both the switched resonance’s half maxima as well as their resonances are indicated. We observe that the dynamic resonance is not centered between the half maxima, but is located much closer to the red edge. The asymmetric shape of the line indicates that resonance is shifting appreciably during the dynamic photon storage time, an effect which we coin ‘kinetic broadening’. Therefore, the broad width of $\Delta = 6$ meV at $\tau_{\text{on}}$ is partially caused by this artificial kinetic effect. The real width is expected to be significantly lower.

We plot the transient reflectivity at both the unswitched and the switched cavity resonance in Fig. 3(b). At the unswitched cavity resonance, the reflectivity increases from 10% at $\Delta t = -3$ ps to just over 50% at $\Delta t = 3$ ps. This corresponds to a modulation depth of $2(50\% - 10\%) = 80\%$, where the factor 2 corrects for averaging over switched and unswitched spectra. At the switched cavity resonance however, the modulation depth is $2(100\% - 75\%) = 50\%$. We can thus see that broadening and the concomitant decrease in
FIG. 3: (a) Time- and frequency resolved reflectivity spectra of both switched and unswitched cavity vs. probe delay, measured alternatingly. The white squares indicate the frequency of the switched cavity resonances, the white curve is a guide to the eye. The half-minima of the unswitched cavity resonance are given by the grey squares. The half-minima of the switched cavity resonance are given by the black squares, and the pump and probe fluences are $I_{\text{Pump}} = 30 \pm 3 \text{ mJcm}^{-2}$ and $I_{\text{Probe}} = 1 \pm 0.3 \text{ mJcm}^{-2}$. (b) Ultrafast change in reflectivity at the unswitched (1.2783 eV) and at the switched (1.2898 eV) cavity resonance. The observed changes should be multiplied by a factor of 2 to correct for averaging the switched and unswitched spectra.

reflectivity leads to a significantly reduced signal modulation depth.

Before we quantitatively describe the behavior of the modulation depth, we will focus on the dynamics of the cavity resonance. Figure 4(a) shows the time-resolved frequency shift of the cavity resonance, taken from Fig. 3(a). Naively, one might assume that the behavior of the cavity resonance follows a single exponential decay [3, 9, 11, 15]. Interestingly, Fig. 4(a) shows a fit of a TM model to the resonances assuming one single recombination time for both the $\lambda$-slab and the GaAs mirrors, but the agreement is only moderate. Permitting the recombination times in the $\lambda$-slab to differ from those in the GaAs mirrors, the fit is much better (details below): Because the resonant mode extends into the Bragg mirrors
for a distance $L_B = 425$ nm, the cavity resonance and the width will be a function of the
dynamic refractive index in both the $\lambda$-slab and the Bragg mirrors. At pump incidence, the
carrier density and thus the refractive index will be the same everywhere. At probe delays
$\Delta t > \tau_{\text{on}}$, however, the carriers in the thin $\lambda/4$ Bragg recombine more rapidly than in the
thicker $\lambda$ slab. Therefore, the time dependent refractive indices will differ, and will cause
the resonance and width to behave as a double exponential. Returning to the analogy with
the Fabry-Pérot model, we identify the $\lambda$-slab of the microcavity with the slab, while the
Bragg mirrors are identified with the two mirrors. Because the Fabry-Pérot resonator is
the template for all cavities, we conclude that our observation of two separate relaxing sub-
systems will be valid for all micro- and nanocavities which consist of topologically separated
slab and mirrors.

V. MODEL AND INTERPRETATION

We will now quantitatively describe the relaxation of the cavity resonance. To a first
order approximation, the change in dynamic cavity resonance is a linear function of the
refractive index in the GaAs mirrors $n_{\text{gm}}$ and in the GaAs $\lambda$-slab $n_{\text{gc}}$, which are weighted
by field distribution coefficients $a$ and $b$, respectively [29]. If the field distribution does not
change significantly during the switch [30], we can write for the shift in cavity resonance:

$$\Delta E_{\text{cav}}(\Delta t) = a\Delta n'_{\text{gm}}(\Delta t) + b\Delta n'_{\text{gc}}(\Delta t).$$

(1)

Since the induced change in refractive index is proportional to the carrier density $N_0$ for
sufficiently small $N_0$, equation (1) can be written as

$$\Delta E_{\text{cav}}(\Delta t) = a'N_{\text{gm}}(\Delta t) + b'N_{\text{gc}}(\Delta t),$$

(2)

where $a'$ and $b'$ are field weighting factors in the mirrors and in the $\lambda$-slab, respectively. Here,
$a' = \frac{a''e^2}{2m^*_e\epsilon_0\epsilon_{\text{pr}}} \lambda^2$ and similarly for $b'$, where $m^*_e$ is the effective electron-hole mass in GaAs, $n_g$
the static refractive index of GaAs, $\epsilon_0$ the static dielectric constant of GaAs, and $\epsilon_{\text{pr}}$ the
probe frequency in rad/s. Often, the time evolution of $N_{\text{gm}}(t)$ and $N_{\text{gc}}(t)$ are considered to
be equal. Due to the different thicknesses, however, they are governed by different relaxation
constants $\tau_1$ and $\tau_2$. Therefore, the change in cavity resonance can be written as

$$\Delta E_{\text{cav}}(\Delta t) = a'N_0 \exp(-\Delta t/\tau_1) + b'N_0 \exp(-\Delta t/\tau_2),$$

(3)
Because of homogeneous carrier excitation via two-photon absorption [24], the carrier density is initially constant. In the present experiment, non-radiative recombination of free carriers at GaAs/GaAlAs interfaces is expected to be the dominant recombination process [31]. This enhanced recombination which takes place at the planar surfaces, combined with carrier diffusion, and topologically separated λ-slab and mirrors resulting in a discontinuous carrier density distribution, gives rise to the double exponential behavior. The different rates are thus identified with the different recombination rates in the GaAs mirrors and in the λ-slab, due to the different thicknesses. From the fit, using 100 iterations with the Levenberg-Marquardt algorithm, and assuming \( N_0 = 2.58 \times 10^{25} \text{m}^{-3} \) (see below), we find \( \tau_1 = 5.5 \pm 1 \) ps and \( \tau_2 = 44.8 \pm 2 \) ps, while \( a' = 1.3 \pm 0.1 \times 10^{-28} \) and \( b' = 3.2 \pm 0.1 \times 10^{-28} \) eVm³. From the relative sizes of \( a' \) and \( b' \) we observe that the field is primarily located in the λ-slab. To calculate the recombination times instead of the relaxation times, which also takes into account the small change in field distribution, it is necessary to fit a series of TM spectra to our measured data. Here, we fit the initial carrier density \( N_0 \), and two recombination times \( \tau_{gm} \) and \( \tau_{gc} \) for the recombination times in the GaAs mirrors and the λ-slab, respectively. The fit gives \( N_0 = 2.58 \pm 0.01 \times 10^{19} \text{cm}^{-3} \), \( \tau_{gm} = 14.8 \pm 0.5 \) ps and \( \tau_{cav} = 62.9 \pm 1 \) ps, and is shown in Fig. 4(a). The sum of the differences squared is 44.6, in contrast to 166.1 if we assumed that the dynamic carrier density is the same in the mirrors and the λ-slab (\( \tau_{gc} = \tau_{gm} = 38 \pm 0.5 \) ps, dashed curve). Comparing \( \tau_{gm} \) and \( \tau_{gc} \) to \( \tau_1 \) and \( \tau_2 \), respectively, we find that the former are considerably higher, which we attribute to the change in field distribution. Note that the ratio of fitted recombination times \( \tau_{cav}/\tau_{gm} \) is in excellent agreement with the ratio of layer thicknesses \( \lambda/(\lambda/4) \), confirming the notion of the double exponential decay, due to the dominant recombination at the planar GaAs/AlAs interfaces.

We emphasize that these two different recombination rates can significantly change the analysis of the dynamic field behavior in the cavity: even though an effective single exponential decay time can be orders of magnitude above the cavity dwell time, the composite shorter recombination times will give rise to significantly different field dynamics [6, 7]. To gain access to timescales comparable to \( \tau_{ph} \), several groups made use of the fast upswitch time \( \tau_{on} \sim 1 \) ps [6, 8]. Here, we probe for the first time during the relaxation, whose timescale is controllable, and still comparable to \( \tau_{ph} \). The recombination rates in microcavities may further be increased controllably by growing samples with a larger number of recombination
centers at the GaAs/AlAs interfaces [19]. Interestingly, Tanabe et al. deduced a non-single exponential cavity resonance relaxation of a 2-D hexapole cavity by rigorously solving the diffusion equation combined with surface recombination [32]. For their system, in which slab and the mirrors are topologically connected, the different relaxation rates are related to the carrier diffusion and the surface recombination rates, respectively.

In order to quantify the width (Fig. 4(b)), which is crucial for understanding the behavior of the modulation depth, we calculate the dynamic width from the TM model using the carrier densities obtained previously. In addition to the time-dependent broadening by the free carriers, we include three additional broadening mechanisms inherent to our experiment: first, wavevector spreading because of the finite probe NA, and second, the spatially changing resonance at the focus waist. These two broadening mechanisms are static, and result in the calculated width approximately matching the measured width. Finally, we take into account kinetic broadening: the spectrometer measures all light exiting the shifting cavity. The total width including kinetic broadening is given by

\[ \Delta_{\text{kin}}(t) = \left( \Delta_{\text{inhom}}(t)^2 + \left[ \tau_{\text{ph}}(t) \frac{d}{dt} E_{\text{cav}}(t) \right]^2 \right)^{1/2} \]

(4)

\[ = \left( \Delta_{\text{inhom}}(t)^2 + \left[ \frac{\hbar N_0}{(e \Delta_0(t))} \left( a'/\tau_1 \exp(-\Delta t/\tau_1) + c'/\tau_2 \exp(-\Delta t/\tau_2) \right) \right]^2 \right)^{1/2} \]

(5)

Using \( \tau_{\text{gm}} \) and \( \tau_{\text{gc}} \) obtained from the fit in Fig. 4(a), and the relaxation parameters, we can calculate the dynamic width (Fig. 4(b), solid line). At \( \Delta t = 3 \text{ ps} \), the calculated width is only around half of the measured width. At larger probe delays \( \Delta t > 20 \text{ ps} \), the calculation agrees excellently with the measurement. The disagreement at short probe delays might stem from the fact that the storage time \( \tau_{\text{ph}}(t) \) stays constant throughout the excitation and the subsequent relaxation. In that case, the kinetic contribution might be considerably larger. To elaborate this point, we plot the dynamic shift ratioed by the dynamic width, shown in Fig. 4(c). At coincidence, the ratio is 2. It shows a maximum at \( \Delta t = 35 \text{ ps} \), before relaxing to 0 at longer probe delays. The agreement of the calculation to the data is excellent for \( \Delta t > 20 \text{ ps} \), but fails at short probe delays. We can exclude coherent transients and electron-electron thermalization due to the short times (in the order of \( \sim 3 \text{ ps} \) [28] and 100 fs [34], respectively). Transient heating can be ruled out because heating would also influence the cavity resonance, which we find to agree excellently to our model without heating. However, assuming a constant \( \tau_{\text{ph}}(t) \) (Fig. 4(c), dotted line), the agreement is
FIG. 4: All data extracted from Fig. (a) Time-resolved shift of the cavity resonance (open circles). The expected resonance from a TM model assuming equal recombination times in both the GaAs mirrors and the λ-slab $\tau_{gc} = \tau_{gm} = 38 \pm 0.5$ ps is given by the dashed curve. The solid curve is a TM calculation for $N_0 = 2.58 \times 10^{19}$ cm$^{-3}$, $\tau_{gm} = 14.7 \pm 0.5$ ps and $\tau_{cav} = 62.9 \pm 1$ ps. (b) Time-resolved width of the cavity (open squares). Here, the solid curve is a TM calculation including free-carrier absorption from the TM calculation, spatial inhomogeneity, wavevector spreading and kinetic broadening, which includes a time-dependent storage time $\tau_{ph}$ as shown in eq. (c) Open triangles: Measured shift of cavity resonance ratioed by the measured width. Solid curve: Calculated dynamic shift divided by dynamic width (open triangles) from (a) and (b), i.e., assuming a variable storage time $\tau_{ph}$. The dotted curve is a calculation of the shift-width ratio, where the storage time is constant in the calculation of the width.

excellent for both $\Delta t > 20$ ps and $\Delta t < 20$ ps, indicating that $\tau_{ph}$ might indeed remain constant. We note that this surprising finding should be explored further, as this opens the possibility of switching with only limited degradation of the storage time.

In order to show by how much the modulation depth is affected by free carrier broadening, we plot in Fig. the expected modulation depth at the switched cavity resonance at pump-probe coincidence for different initial $Q_0$. Here, the carrier density is kept constant at
\[ N_0 = 2.58 \times 10^{19} \text{ cm}^{-3}, \text{ as in the preceding experiment.} \]

At low values of \( Q_0 \), the modulation depth is shallow, as expected. It acquires a maximum at around \( Q_0 = 300 \). Up to this point, the modulation depth has been limited by the low \( Q_0 \) of the sample. For \( Q_0 > 300 \), the expected modulation depth decreases rapidly. In this regime, the linewidth of the resonance becomes more and more sensitive to free carrier absorption, and to the decrease of the mirrors’ reflectivity. The measured modulation depth of the present measurement is included. The discrepancy indicates that the modulation depth is not well understood for short probe delays. For a carrier density an order of magnitude lower, the maximum is reached for \( Q_0 = 5000 \). For \( Q_0 > 5000 \), the sensitivity of the modulation depth on \( Q_0 \) is less pronounced. From the calculations we conclude that there is a maximum attainable modulation depth for every initial linewidth and carrier density.

VI. CONCLUSION

Using time- and frequency resolved pump-probe spectroscopy, we have experimentally demonstrated ultrafast switching of up to 11 linewidths within 3 ps of a microcavity using two-photon absorption. The dynamics of the cavity resonance is investigated in detail, where we find a double exponential behavior, which is shown to result from the different recombination rates in the \( \lambda \)-slab and the Bragg mirrors. We suggest that all photonic crystal cavities are subject to at least two different relaxation times, a fact which has not
been measured previously. Our observed distribution of recombination times is predicted to strongly modify the field dynamics with respect to a single-exponential decay. In particular, the regime for which the timescale of relaxation of the cavity resonance is comparable to the photon dwell time becomes accessible experimentally. From the behavior of the resonance, the time-resolved width can be deduced. We discuss broadening effects due to wavevector spreading, spatially varying resonances on the sample, and kinetic broadening. Surprisingly, the calculations, which assume a constant storage time $\tau_{ph}$ during the excitation and subsequent relaxation of the cavity resonance agree best to the data, a fact which has not been appreciated previously. The broadening mechanisms are shown to contribute to the final width at all probe delays. Finally, we discuss the effect of these broadening mechanisms on the modulation depth. From a calculation we are able to infer an optimum value of $Q_0$ for a given carrier density and a required modulation depth.

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[1] P. M. Johnson, A. F. Koenderink, and W. L. Vos, “Ultrafast switching of photonic density of states in photonic crystals,” Phys. Rev. B. 66, 081102(R) (2002).
[2] B. P. J. Bret, T. L. Sonnemans, and T. W. Hijmans, “Capturing a light pulse in a short high-finesse cavity,” Phys. Rev. A 68, 023807 (2003).
[3] V. R. Almeida, C. A. Barrios, R. R. Panepucci, and M. Lipson, “All-optical control of light on a silicon chip,” Nature (London) 431, 1081 – 1084 (2004).
[4] Q. Xu, P. Dong, and M. Lipson, “Breaking the delay-bandwidth limit in a photonic structure,” Nat. Phys. 31, 406–410 (2007).
[5] Y. Tanaka, J. Upham, T. Nagashima, T. Sugiya, T. Asano, and S. Noda, “Dynamic control of the q factor in a photonic crystal nanocavity,” Nat. Mater. 6, 862–865 (2007).

[6] M. Notomi and S. Mitsugi, “Wavelength conversion via dynamic refractive index tuning of a cavity,” Phys. Rev. A 73, 051803 (2006).

[7] A. M. Yacomotti, F. Raineri, C. Cojocaru, P. Monnier, J. Levenson, and R. Raj, “Nonadiabatic dynamics of the electromagnetic field and charge carriers in high-Q photonic crystal resonators,” Phys. Rev. Lett. 96, 093901 (2006).

[8] S. F. Preble, Q. Xu, and M. Lipson, “Changing the colour of light in a silicon resonator,” Nat. Phot. 1, 293–296 (2007).

[9] P. J. Harding, T. G. Euser, Y. R. Nowicki-Bringuier, J.-M. Gérard, and W. L. Vos, “Ultrafast optical switching of planar GaAs/AlAs photonic microcavities,” Appl. Phys. Lett. 91, 111103 (2007).

[10] M. Först, J. Niehusmann, T. Plötzing, J. Bolten, T. Wahlbrink, C. Moormann, and H. Kurz, “High-speed all-optical switching in ion-implanted silicon-on-insulator microring resonators,” Opt. Lett. 32, 2046–2048 (2007).

[11] T. Tanabe, K. Nishiguchi, A. Shinya, E. Kuramochi, H. Inokawa, and M. Notomi, “Fast all-optical switching using ion-implanted silicon photonic crystal nanocavities,” Appl. Phys. Lett. 90, 031115 (2007).

[12] H. M. Gibbs, T. N. C. Venkatesan, S. L. McCall, A. Passner, A. C. Gossard, and W. Wiegmann, “Optical modulation by optical tuning of a cavity,” Appl. Phys. Lett. 34, 511–514 (1979).

[13] T. Tanabe, M. Notomi, S. Mitsugi, A. Shinya, and E. Kuramochi, “All-optical switches on a silicon chip realized using photonic crystal nanocavities,” Appl. Phys. Lett. 87, 151112 (2005).

[14] Q. Xu, V. R. Almeida, and M. Lipson, “Micrometer-scale all-optical wavelength converter on silicon,” Opt. Lett. 30, 2733–2735 (2005).

[15] I. Fushman, E. Waks, D. Englund, N. Stoltz, P. Petroff, and J. Vučković, “Ultrafast nonlinear optical tuning of photonic crystal cavities,” Appl. Phys. Lett. 90, 091118 (2007).

[16] K. J. Vahala, “Optical microcavities,” Nature (London) 424, 839–846 (2003).

[17] M. Born and E. Wolf, Principles of Optics (Cambridge University Press, 1997).

[18] A. Chin, K. Y. Lee, B. C. Lin, and S. Horng, “Picosecond photoresponse of carriers in Si ion-implanted Si,” Appl. Phys. Lett. 69, 653655 (1996).
[19] G. Segschneider, T. Dekorsy, H. Kurz, R. Hey, and K. Ploog, “Energy resolved ultrafast relaxation dynamics close to the band edge of low-temperature grown GaAs,” Appl. Phys. Lett. 71, 2779 (1997).

[20] The maximum unsaturated unbroadened refractive index change of the dots amounts to only $10^{-8}$, while the absorption at resonance is less than 50 cm$^{-1}$.

[21] T. G. Euser, A. J. Molenaar, J. G. Fleming, B. Gralak, A. Polman, and W. L. Vos, “All-optical octave-broad ultrafast switching of Si woodpile photonic band gap crystals,” Phys. Rev. B 77, 115214 (2008).

[22] T. G. Euser, “Ultrafast optical switching of photonic crystals,” Ph.D. thesis, University of Twente, ISBN 978-90-365-2471-1, www.photonicbandgaps.com (2007).

[23] $\tau_p$ denotes the FWHM of the pulse intensity, and was measured in an autocorrelator.

[24] T. G. Euser and W. L. Vos, “Spatial homogeneity of optically switched semiconductor photonic crystals and of bulk semiconductors,” J. Appl. Phys. 97, 043102 (2005).

[25] J. S. Blakemore, “Semiconducting and other major properties of gallium arsenide,” J. Appl. Phys. 53, R123 – R181 (1982).

[26] R. E. Fern and A. Onton, “Refractive index of AlAs,” J. Appl. Phys. 42, 3499 – 3500 (1971). Note that the refractive index is given by Re($\epsilon^{1/2}$).

[27] S. G. Hense and M. Wegener, “Ultrafast switch-off of a vertical-cavity semiconductor laser,” Phys. Rev. B 55, 9255–9258 (1997).

[28] G. C. Cho, W. Kütt, and H. Kurz, “Subpicosecond time-resolved coherent-phonon oscillations in GaAs,” Phys. Rev. Lett. 65, 764–766 (1990).

[29] The relative error of this linearization is less than 2% for $N_0 < 3 \times 10^{25} \text{ m}^{-3}$, where $N_0$ is the initial carrier density.

[30] Independent calculations confirmed that the $L_B$ changes by less than 10% for our carrier densities.

[31] J. M. Gérard, B. Sermage, L. Bergomi, and J. Y. Marzin, “Differentiation of the non radiative recombination properties of the two interfaces of MBE grown GaAs-GaAlAs quantum wells,” Superlattices Microstruct. 8, 417–419 (1990).

[32] T. Tanabe, H. Taniyama, and M. Notomi, “Carrier diffusion and recombination in photonic crystal nanocavity optical switches,” J. Lightwave Tech. 26, 1396–1403 (2008).

[33] M. Notomi, E. Kuramochi, and T. Tanabe, “Large-scale arrays of ultrahigh-Q coupled
nanocavities,” Nat. Phot. 2, 741–747 (2008).

[34] A. J. Sabbah and D. M. Riffe, “Femtosecond pump-probe reflectivity study of silicon carrier dynamics,” Phys. Rev. B 66, 165217 (2002).