We develop a nanoscopy method with in-depth resolution for layered photonic devices. Photonics often requires tailored light field distributions for the optical modes used, and an exact knowledge of the geometry of a device is crucial to assess its performance. The presented acousto-optical nanoscopy method is based on the uniqueness of the light field distributions in photonic devices: for a given wavelength, we record the reflectivity modulation during the transit of a picosecond acoustic pulse. The temporal profile obtained can be linked to the internal light field distribution. From this information, a reverse-engineering procedure allows us to reconstruct the light field and the underlying photonic structure very precisely. We apply this method to the slow light mode of an AlAs/GaAs micropillar resonator and show its validity for the tailored experimental conditions.

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OCIS codes: (140.3490) Lasers, distributed-feedback; (060.2420) Fibers, polarization-maintaining; (060.3735) Fiber Bragg gratings.

https://doi.org/10.1364/OPTICA.4.000588

1. INTRODUCTION

Photonic nanostructures, in which light can be guided or confined, are fundamental for a wide range of applications ranging from information communication to cavity quantum electrodynamics and optomechanics. The geometry and the constituting materials of the structure define the optical resonances and the spatial profile of the associated photonic modes. In the case of optical cavities, for example, photons are slowed down or trapped in localized modes to obtain lifetimes of up to milliseconds [1]. The light–matter interaction with an optically active material is drastically enhanced, provided that it is placed at a maximum of the light field distribution of such a long-lived mode. There are manifold challenging applications in the strong coupling regime such as bright single-photon sources [2], entangled photons [3], or polariton lasing [4], for which the exact knowledge of the resonator’s geometry and photonic field inside the nanostructure are crucial. Information about the surface of the fabricated structures is usually obtained with a high resolution by scanning electron microscopy (SEM) or atomic force microscopy. When in-depth information is desired, transmission electron microscopy and x-ray diffraction are used. The first one is destructive due to the required cross-sectional cuts. Often these techniques are not easy to access or to handle. In this case an established technique is to record the optical reflectivity spectrum and fit simulations based on a transfer matrix approach to the experimental data [5]. The drawback of this method in complicated multilayer structures is that it offers ambiguous in-depth information, since the reflectivity spectrum is an integrated measure determined by the whole structure.

For nanoscopy of buried films and interfaces, picosecond ultrasonics provides a suitable tool [6–9]. A picosecond acoustic pulse is optically generated and detected in a photonic nanostructure and allows one to derive information about the position of the internal interfaces. The success of this technique is due to the short wavelengths (down to 10 nm) of the coherent phonons forming the acoustic pulse, which have made it possible to investigate buried optical microcavities [10], superlattices [11,12], hetero-interfaces [13], quantum wells [14], and defects in contacts [15]. Most of the picosecond acoustic experiments in photonic devices were carried out to study specific phononic features arising from the acoustic mismatch between the constituting materials, while their optical properties were considered mainly for the purpose of understanding the generation or detection of terahertz and subterahertz phonons [7,16,17].

The efficiency of picosecond ultrasonics relies on a mismatch of the acoustic properties of the constituting materials, which is not necessarily given. However, the proposed method of this work is based on obtaining information about the unique light field distribution in the device under study. It only requires contrasting...
dielectric functions, which is an intrinsic feature of most photonic devices. The difficulty arises from the complex interplay of a picosecond acoustic pulse and the light field in a photonic device. This interplay has been comprehensively described in a number of publications [18,19]. It turns out that the properties of (i) the acoustic pulse, namely its duration, the phonon spectrum, and the phonon dispersion in the photonic device, the (ii) light field distribution, and (iii) the different mechanisms of light–matter interaction, like phonon–photon and phonon–electron scattering, need to be considered for a complete understanding. Previous works have so far not been aimed at deducing unambiguous information about the light field from the overall response [17,20]. The way to achieve this is to know all other parameters exactly so that they can be eliminated from the response. How this condition can be fulfilled for a practical photonic device by the proper design of the acoustic pulse and choice of the studied light field is discussed in the present paper.

The basic experimental scheme of the method is presented in Fig. 1(a): a laser beam is directed onto the nanostructure and builds up the light field sketched in red. The device under investigation imprints a unique field distribution, which is exploited for its characterization. A picosecond acoustic pulse is injected into the structure from its back side and propagates with the sound velocity along the z axis toward the surface of the nanostructure. The temporal displacement profile \( u(t) \) can often be modeled by a Gaussian [21] with a duration that depends on the experimental conditions (excitation energy, temperature, non-linear phenomena) and ranges from several picoseconds up to \(~100~\text{ps}~\) [21]. In our model we use an amplitude of several tens of picometers and a full width at half-maximum (FWHM) of about 90 ps, as shown in Fig. 1(b) by the solid line. The derivative of the displacement is associated with the strain \( \eta(t) \) and is shown in Fig. 1(b) by the dashed line. The presence of the acoustic pulse slightly perturbs the light field in the structure. For the incident laser beam, this local perturbation leads to a change in the total reflectivity that depends on the light field intensity at position \( z \) of the acoustic pulse. The result for the simplest case of an optically homogeneous material is well known and is described by the so-called coherent Brillouin oscillations originating from the interference of two optical beams, one being reflected from the surface and the other one in the depths of the material, where the acoustic pulse dynamically modifies the optical properties due to the photoelastic effect [22]. For periodic multilayered structures, like distributed Bragg reflectors (DBRs), the oscillation spectrum is more complicated [16,23] and is defined by the photon–phonon momentum conservation, which also includes folded Brillouin oscillations, which involve phonon Umklapp processes [12,24].

Our aim is to record the reflectivity modulation for more complex photonic nanostructures, when the analysis cannot be based on the momentum conservation only. In the paper, we first present an analytical equation, which allows us to link the reflectivity modulation to the internal light field distribution along the propagation direction of the acoustic pulse. Afterward we demonstrate that the information obtained can be used to reconstruct the light field, which then in turn is exploited for the characterization of a model multilayer system. In the experimental part of the paper, we apply this technique to a slow photonic mode in a GaAs/AlAs micropillar resonator that falls into the high-energy flank of the first optical stop band. We show that the experimentally measured temporal evolution of the reflectivity allows us to precisely calculate the underlying light field distribution and to determine the geometry of the photonic resonator with in-depth resolution and an accuracy of a few nanometers.

2. THEORETICAL BACKGROUND

The interaction of picosecond acoustics perturbation with the light field in a photonic nanostructure is due to two fundamentally different contributions, namely, (i) the photoelastic effect and (ii) the interface displacement [18]. The photoelastic effect, on one hand, is based on the fact that the strain \( \eta(z, t) \) leads to a local change in the refractive index \( \Delta n \). In semiconductors, for photon energies not far from the bandgap \( E_G \), \( \Delta n \) can be approximated by

\[
\Delta n = \frac{\partial n}{\partial E_G} \frac{\partial E_G}{\partial \eta} \Delta \eta.
\]

It is dependent on the dispersion of the refractive index and on the deformation potential constant linking the shift of the energy gap \( E_G \) to the applied strain [12,18]. On the other hand, the interface displacement effect is related to the reflections originating from the surface and the internal material interfaces of the photonic nanostructure. The acoustic pulse modifies their interference terms, since it moves such an interface when passing it, thereby leading to a phase shift of the reflection originating from there. For the description of the acoustic pulse, we consider a one-dimensional case. Furthermore, we assume that the phonon dispersion is not affected by the nanostructure and may be described by a linear relation between the phonon frequency \( \omega \) and wavevector \( q_z \).
\[ \omega = \nu q z \]  

with the slope given by the longitudinal sound velocity \( \nu \). In onedimensional periodic nanostructures with a total length much longer than the phonon wavelength, both contributions to the light–matter interaction result in an optical reflectivity modulation whose spectrum is governed by photon–phonon momentum conservation, including Umklapp processes:

\[ q = 2[k \pm mG]. \]

Here, \( k \) is the photon wave vector, \( G \) is the reciprocal lattice vector, and \( m \) is an integer. In most periodic photonic nanostructures for light in the visible to near-IR range, this corresponds to phonon frequencies from zero to several tens of gigahertz, depending on \( k \) [12].

For nonperiodic multilayer structures, Eq. (3) is not valid and transfer matrix calculations may be considered for numerical simulations. Our aim is to find experimental conditions under which the treatment of the light–matter interaction is as simple as possible, e.g., by making one of the two interaction mechanisms negligible. The photoelastic contribution can be turned off by choosing a wavelength where the photoelastic constants are small [25]. Another possibility in the case of a bipolar strain profile [see Fig. 1(b)], like that typically generated in picosecond ultrasonics, is to tailor the acoustic spectrum such that the spectral amplitude for high-frequency phonons, which provide the largest contribution to the photoelastic effect, is small enough to consider only interface displacements [22].

As soon as the photoelastic contribution is minimized by pursuing one or both of the proposed routes, the displacement effect becomes the dominant contribution. In this case, the reflectivity change for a fixed wavelength \( \lambda \) may be expressed analytically as

\[ \frac{\Delta R(t)}{R_0} = -k_0 \text{Im} \left[ \sum_i \delta \epsilon(z_i) \frac{E^2(z_i)}{r} u(z_i, t) \right], \]

where \( k_0 \) is the vacuum photon wave vector and \( r \) is the complex reflection coefficient of the whole structure (a similar expression was derived in Ref. [18]). The sum runs over all interfaces denoted by \( i \), where \( z_i \) marks the position of the interface, \( \delta \epsilon(z_i) \) is the difference of the dielectric constants at the interfaces, \( u(z_i, t) \) represents the temporal and spatial displacement profile, and \( E(z) \) is the normalized, dimensionless, and complex electric field distribution of the considered photonic mode. The electric field has the form \( E(z) = \exp(ikz) + r \exp(-ikz) \) at the front surface of the structure at \( z = 0 \), which determines its phase and amplitude [18]. From this starting point, the distribution \( E(z) \) in the whole structure is calculated by a transfer matrix that also yields the reflection coefficient \( r \) [5]. For nonabsorbing materials, \( \delta \epsilon(z_i) \) is real, and one can see that the reflectivity modulation is given by the convolution of the displacement profile with a quantity closely related to the photonic field, \( \rho(z) = \text{Im}(E^2(z)/r) \). This convolution is evaluated at discrete sampling points, namely the positions of the interfaces, with a weight determined by the optical contrast.

3. MODEL CALCULATIONS

First we discuss simulations performed for a planar microcavity structure as an intuitive example of the potential of our method. The bottom and top DBRs consist of 30 periods of alternating AlAs and GaAs layers with nominal thicknesses of 78 and 66 nm, respectively, designed for a center stop band wavelength of 925 nm at cryogenic temperatures. Between the two DBRs a \( \lambda \) cavity GaAs spacer is sandwiched. To show the sensitivity of the proposed method to the geometry of the photonic device, additional scaling factors of \((1 - x)\) and \((1 + x)\) are then applied to the initial nominal layer thicknesses of the bottom and top DBRs, respectively. In Fig. 2(a) the calculated reflectivity spectra for two microcavities with rather similar layer thicknesses in the bottom and top DBRs are shown (\( x = \pm 0.03 \)). For a wavelength of \( \lambda = 830 \) nm close to the stop band edge [indicated by the arrow in Fig. 2(a)], the different layer thicknesses give rise to a significantly different photonic field distribution \( |E(z)|^2 \), plotted in Fig. 2(b). Two features can be seen there: one is a fast oscillatory component determined by the optical wavelength, and another is a slowly varying envelope function. While the period of the fast oscillations is almost the same in the bottom and top DBRs, the periods for the slow ones are different: in the thicker DBR, the envelope function varies on a longer length scale and with a higher relative amplitude than in the thinner DBR. Such striking differences cannot easily be recognized in the reflectivity.

![Fig. 2. Simulations of two differently scaled planar microcavities with \( x = -0.03 \) (blue curves) and \( x = +0.03 \) (red curves). (a) Calculated reflectivity spectra; the arrow marks the wavelength studied in the following parts. The modeled structure and the influence of \( x \) are illustrated in the inset. (b) Light field distributions \( |E(z)|^2 \) in the two cavities for the wavelength indicated in (a); (c) temporal reflectivity change during the transit of a Gaussian displacement pulse with a FWHM of 90 ps through the device.](image-url)
spectrum shown in Fig. 2(a). One sees that the central stop band looks almost the same, independent of whether the bottom ($x < 0$, blue curve) or the top ($x > 0$, red curve) DBR is the thicker one. Only in the sidebands do slight deviations occur, so that it is hard to distinguish the two different structures from their reflectivity spectra alone.

The specific features of the photonic field, which are not recognized in the reflectivity spectrum, become visible when calculating the reflectivity change induced by an acoustic pulse according to Eq. (4). In Fig. 2(c) the temporal traces for $\Delta R(t)/R_0$ are presented for a Gaussian displacement pulse with a FWHM of 90 ps propagating in the structure with a mean sound velocity of $\bar{v} = 5230$ m/s. Here, the time $t = 0$ ns corresponds to the injection of the acoustic pulse into the bottom DBR and the time $t \approx 1.75$ ns to the time when the acoustic pulse reaches the top surface of the top DBR. Both curves show oscillatory behavior whose periods resemble the variations of the light field’s envelope function: when the acoustic pulse propagates through the DBR with the faster (slower) changing light field envelope, we observe a faster (slower) reflectivity modulation. With these findings one can clearly distinguish between the two differently scaled microcavities.

Note that the oscillations in the reflectivity modulation are not directly proportional to the light field intensity. Although the periods of the oscillations in the thinner and thicker DBRs match the periods of the corresponding envelope functions very well, which is also important information, there is a nontrivial phase difference determined by the complex reflection coefficient $r$, the width of the displacement pulse, and for absorbing media the dielectric contrast $\delta e(e(z))$ also. Furthermore, it is necessary to select a probe wavelength $\lambda$ whose light field distribution is strongly controlled by the parameters of the photonic device such that small variations in these parameters lead to a strong change in the reflectivity modulation. For the microcavity presented here, this means, for example, a probe wavelength close to the optical stop band.

We have shown by transfer matrix calculations that for an acoustic pulse with a width of $\sim 100$ ps, the contribution of the photoelastic effect is negligible and the interface displacement effect according to Eq. (4) governs the temporal evolution of the reflectivity changes (see Supplement 1). Due to the relatively long acoustic pulse, the small-period oscillating field does not contribute to $\Delta R(t)/R_0$. Moreover, the wavelength $\lambda$ of the light field considered is chosen to be in the transparency region of AlAs and GaAs (at low temperatures), where the refractive index dispersion is flat and the photoelastic effect is consequently rather inefficient [26]. Although reflections of the acoustic pulse at internal interfaces have been included in the calculations, they do not play an important role here, since the phonon spectrum of the modeled acoustic pulse does not reach frequencies as high as the first photonic stop band at around 18 GHz [10].

4. EXPERIMENT

Experiments to demonstrate the validity of the method in practical photonic devices are performed on micropillar lasers with two DBRs consisting of 33 and 26 periods of alternating AlAs and GaAs layers for the bottom and the top mirror, respectively. The nominal thicknesses of the AlAs and GaAs layers are 74 and 69 nm, respectively. In between the two DBRs a 266 nm thick Al$_{0.09}$Ga$_{0.55}$In$_{0.36}$ spacer is sandwiched such that the nominal total thickness of the structure is about 8.7 $\mu$m. An ensemble of Al$_{0.09}$Ga$_{0.55}$In$_{0.36}$ quantum dots is placed in the cavity layer center and serves as the active medium of the laser. Micropillars with different radii ranging from 1.5 to 7.5 $\mu$m are studied, because they offer stronger light field confinement than planar structures [27]. We use a time-resolved pump–probe setup to generate the acoustic pulse with the pump and detect the reflectivity modulation with the probe laser beam. Both originate from a pulsed laser with a central wavelength of 800 nm and a spectral width of 10 nm, which falls into the high-energy flank of the sample’s first optical stop band [see Fig. 3(a)]. The acoustic pulse is generated on the back side of the sample and injected from there into the micropillars [28]. To ensure a lossless transfer through the 220 $\mu$m thick GaAs substrate [29], the sample is placed in a flow cryostat and attached to a cold finger kept at a temperature of 8 K. More details about the sample and the experimental setup can be found in Supplement 1.
In Fig. 3(b) the measured reflectivity change $\Delta R(\tau)/R_0$ obtained for a micropillar with a radius of 7.5 $\mu$m is presented as a black curve. At a delay of $\tau_0$ the acoustic pulse reaches the foot of the micropillar. Here we record a peak and a subsequent dip, since the beam spot is approximately larger than the micropillar’s diameter, and we also collect light that is reflected from the surface next to the micropillar. For increasing delays $\tau$, the acoustic pulse advances toward the micropillar’s top surface and is located in the bottom DBR for $\tau_0 < \tau < \tau_1$. As for the curves in Fig. 2, oscillatory features are recorded. The temporal separation between the two peaks appearing right before $\tau_1$ is $220 \pm 6$ ps. After leaving the bottom DBR at $\tau_1$, the acoustic pulse passes through the cavity layer within about 50 ps and enters the top DBR. We observe a phase jump in the signal, and afterward the reactivity clearly starts to oscillate with a stronger amplitude. The temporal interval between neighboring peaks in the oscillations is $230 \pm 6$ ps here. These oscillations persist, until the acoustic pulse reaches the top surface of the micropillar at $\tau_2$. Taking the temporal difference between $\tau_2$ and $\tau_0$ of 1.47 ns and the averaged sound velocity in the layered structure of $v = 5211$ m/s, we conclude that the micropillar has a height of about $h = 7.7$ $\mu$m, which corresponds to several double layers not etched away in the bottom DBR [see Fig. 3(a)]. At the top of the device, the acoustic pulse is reflected and redirected into the micropillar. In the temporal reflectivity trace this leads to a phase jump at $\tau = \tau_2$. Following the reflection of the acoustic pulse, the reverse sequence is recorded, including the second transit through the cavity layer at $\tau_3$ and so forth. Finally, the acoustic pulse leaves the micropillar at $\tau_4$. The signal at delays $\tau > \tau_4$ is assigned to reflections of the acoustic pulse inside the layered structure and is not considered further.

We compare the measured signal with the simulations according to Eq. (4). The spectral width of the laser pulse of 10 nm needs to be taken into account, which is done by calculating the field distributions for all wavelengths occurring in the laser emission and weighting them by their spectral amplitude [see Fig. 3(a)]. For the acoustic pulse a displacement input profile given by a Gaussian with a FWHM of 90 ps is used. The curve is fitted to the experimental data by scaling the layer thicknesses, and the best result is shown as the red dashed curve in Fig. 3(b). The excellent agreement between the simulated curve and the experiment underlines the validity of our approach. Deviations occur mainly when the acoustic pulse reaches the foot of the micropillar and the reflection from the area next to it is modulated ($\tau = \tau_0$), which is not included in the model. After the reflection of the acoustic pulse from the micropillar surface ($\tau > \tau_2$), we observe in the experiment an asymmetric profile around $\tau_2$, in contrast to the model expectations. This indicates a rough top surface resulting in diffuse scattering of the phonons in the acoustic wave packet. The field distributions for three wavelengths used in the reverse-engineering calculations are displayed in Fig. 3(c).

Ultimately, the simulation allows us an analysis of our photonic device. We consider three quantities to be extracted from the experimental data: the cavity layer thickness and the mean periodicity in the bottom and top DBRs. Each DBR is assumed to possess its own uniform periodicity, which for simplicity is obtained by rescaling the nominal parameters. The central cavity layer is found to be 2% (=5.3 nm) thicker than the nominal value of 266 nm by adjusting the relative phase of the oscillations in the top and in the bottom DBR. The periods of the oscillations in the bottom and top DBRs precisely provide the DBR periodicity. Each GaAs/AlAs DBR layer is found to be about 3.9% and 4.1% thinner in the bottom and in the top DBR, respectively. The precision of the found layer thicknesses corresponds to a spatial resolution of a few nanometers for single layers, like in the case of the cavity spacer, where a deviation of 5 nm from the nominal value is found. An even higher resolution is achieved for the mean period in periodic structures. Here, the possibility of resolving the different mean layer thicknesses in the bottom and top DBRs corresponds to a subnanometer resolution. In the experiments, the error for the period of the oscillations is 6 ps and is determined by the time resolution of the setup, which is governed by the step size of the mechanical delay line rather than by the shorter laser pulse duration. How this error limits the accuracy of acousto-optical nano-scopes depends strongly on the sensitivity of the probe light field to structural changes in the device; i.e., the accuracy is a function of the chosen probe wavelength, and it is hard to provide an analytical expression for the formal error of the individual layers. Figure 4(a) shows the simulated reflectivity change for different cavity layer thicknesses to get an impression of the method’s precision. The phase jump at delay $\tau_1$ indicates the passage of the acoustic pulse from the bottom DBR into the top DBR. While the reflectivity modulation for delays larger than $\tau_1$ is barely affected by the cavity layer thickness, pronounced phase shifts of the oscillations in the bottom DBR are observed. One can see that the chosen curve for a 2% thinner layer fits much better than the other ones. In the bottom DBR the width of the peaks is too narrow in all simulations. This issue cannot be solved by increasing the duration of the acoustic pulse. Anyway, since the agreement is much better for the top DBR, the width of the peaks Fig. 4. Error and validity of the method. (a) Simulations for the AlAs/GaAs micropillar for different cavity layer thicknesses. The experimental curve (black line) is vertically shifted for clarity. (b) Experimental measurement of the reflectivity change in a thin AlAs/GaAs micropillar with a radius of 1.5 $\mu$m (top) and beside a micropillar (bottom). Next to the micropillar clear coherent Brillouin oscillations are observed in the fast Fourier transform (FFT) (inset).
is obviously related to the DBR and not to the acoustic pulse. Maybe the quality of the bottom DBR is a bit lower and the thicknesses of the layers are not homogeneous across one oscillation period. Another possible explanation for the broader width of the experimental oscillations might be related to the A0.09Ga0.55In0.36. As quantum dots, whose interaction with the optical field is not included. We briefly discuss the mechanical eigenmodes supported by the micropillar resonator to exclude the possibility that they lead to the observed reflectivity modulation. The pillar's extensional ground mode can be calculated from the height h and the mean sound velocity v and is found to have a frequency of 0.15 GHz [30]. The frequency of the radial modes scales inversely with the radius [31]. Such a dependency of the modulation frequency on the radius was not found when a second micropillar with a smaller radius of 1.5 µm was studied [see the top curve in Fig. 4(b)]. The basic shape of the reflectivity modulation does not depend on the radius of the micropillar; however, the smaller the radius, the higher the noise and the more pronounced the peak at τ0 associated with the sample surface surrounding the micropillar. Finally, calculations show the first phononic stop band arising from the DBRs at 18 GHz [32], so we conclude that the observed modulation is not related to any mechanical resonance.

Comparison of the experimental results with the simulations shows that our model is valid for probing interfaces in photonic structures with acoustic pulses. The response can be modeled solely by the interface displacement effect according to Eq. (4) for a relatively long acoustic pulse. Shortening the strain pulse obviously brings high-frequency phonons into play, and the photoelastic effect should be taken into account, too. When the reflectivity modulation in the bulk GaAs beside the micropillar is recorded, 42 GHz Brillouin oscillation is clearly observed [see the inset in Fig. 4(b)]. However, no oscillations with frequencies corresponding to the GaAs/AlAs DBRs, which should be about 41 GHz, are observed when the micropillar is probed. This indicates that there are no high-frequency phonon components in the acoustic pulse propagating in the micropillar. A possible explanation might be a strong scattering of high-frequency phonons at imperfections in the micropillar walls and its foot. The reflectivity modulation does not depend strongly on the shape and the duration of the modeled acoustic pulse. The simulations are independent of whether a Gaussian displacement profile or a shockwave acoustic pulse, whose high-frequency components have been filtered by the micropillar, is assumed (see Supplement 1).

The proposed method is applicable to manifold kinds of multilayered photonic devices, where in-depth information is of interest. If a transparent probe wavelength is chosen, the maximum available depth is limited only by the penetration depth of the acoustic pulse. This length is governed by the acoustic mismatch at the internal interfaces, if the structure is cooled down and photon attenuation becomes negligible. It is favorable to work with long acoustic pulses containing no high-frequency phonons, which lead to coherent Brillouin oscillations. Unlike micropillars, most photonic devices do not suppress these phonons, and a more sophisticated generation process is necessary. One might employ a pump wavelength with a longer absorption length, for example.

In conclusion, we presented a method for the characterization of a photonic device based on in-depth sensing of the internal light field distribution with a picosecond acoustic pulse. We have established experimental conditions under which we can treat the response of a given photonic mode to the acoustic pulse in a simplified and analytical way. From the recorded reflectivity modulation for the selected optical wavelength, we can deduce information on the internal light field distribution. In the final step, we reconstructed the original photonic device in a reverse-engineering procedure from the information we obtained about the light field. The technique was discussed in simulations of a planar microcavity as an intuitive model structure and afterwards validated by experiments performed on an AlAs/ GaAs micropillar resonator. From the simulation we were able to determine the layer thicknesses with a high precision of a few nanometers.

**Funding.** Alexander von Humboldt-Stiftung; Ministry of Education and Science of the Russian Federation (Mинобрнауки) (14.Z50.31.0021); Bayerisches Staatsministerium für Wissenschaft, Forschung und Kunst (StMWFK); Deutsche Forschungsgemeinschaft (DFG) (TRR 160 Project B6, TRR 142 Project A6).

**Acknowledgment.** A. V. A. acknowledges the Alexander von Humboldt Foundation. M. B. acknowledges partial financial support from the Russian Ministry of Science and Education.

See Supplement 1 for supporting content.

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