

Thermal analysis of GaN-based photonic membranes for optoelectronics

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(Received 16 October 2024; revised 15 October 2025; accepted 19 December 2025; published 9 February 2026)

Semiconductor membranes are widely used in research fields that target medical, biological, environmental, and optical applications. Often such membranes derive their functionality from a nanopatterning, which challenges the determination of their optical, electronic, mechanical, and thermal properties. In this work, we demonstrate the noninvasive, all-optical thermal characterization of approximately-800-nm-thick and approximately-150- μm -wide membranes that consist mainly of wurtzite GaN and a stack of $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ quantum wells as a built-in light source. Because of their application in photonics, e.g., for vertical-cavity surface-emitting lasers, such photonic membranes are bright light emitters, which challenges their thermal characterization by optical means. We combine top-view two-laser Raman thermometry (2LRT) with time-resolved photoluminescence spectroscopy to extract the in-plane thermal conductivity $\kappa_{\text{in plane}}$ of these membranes, which represents a notable difference from previous studies on epitaxial GaN films. Thus, we can disentangle the entire laser-induced power balance. Thermal imaging by Raman spectroscopy yields $\kappa_{\text{in plane}} = 165^{+16}_{-14} \text{ W m}^{-1} \text{ K}^{-1}$ for the best membrane. This result compares well with $\kappa_{\text{in plane}} = 177 \text{ W m}^{-1} \text{ K}^{-1}$ obtained by *ab initio* simulations based on a solution of the linearized phonon Boltzmann transport equation, including three- and four-phonon scattering, as well as phonon-isotope and phonon-boundary scattering. Furthermore, we study how $\kappa_{\text{in plane}}$ is affected by a roughening of the membrane's back side and additional semiconductor layers. For the membrane with the roughest back side, we observe a reduction of $\kappa_{\text{in plane}}$ by almost 40%, which is accompanied by an anisotropy of $\kappa_{\text{in plane}}$ due to etch channel formation. Thanks to the 2LRT approach, such variations and anisotropies of $\kappa_{\text{in plane}}$ become accessible to the experimentalist via highly spatially resolved temperature maps.

DOI: [10.1103/PhysRevApplied.25.024028](https://doi.org/10.1103/PhysRevApplied.25.024028)

I. INTRODUCTION

The field of semiconductor membranes (SMs) is fueled by emerging materials that are promising for a wide range

of pioneering applications. The most established membrane materials are amorphous SiN [1,2] and crystalline Si [3,4]. However, recently, SiC [5], diamond [6], and Ge [7] have also become routinely available as membrane materials. In addition, many other semiconductor materials, such as MoS₂ [8], BN [9], group-III arsenides [10–12], and group-III nitrides [13,14], have also been used as free-standing membranes, sparking research related to a wide range of applications. In this work, we classify extrinsically nanostructured SMs (e.g., by processing hole lattices or via surface engineering [15]) as type I. However, SMs can also comprise further intrinsic structures such as quantum wells (QWs) or quantum dots (QDs), which are key

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to the desired functionality. We classify the latter SMs as type II.

For type-I SMs it is often the permeability for liquids or gases that motivates their use, which ranges from particle nanofiltration [9] to electrochemical separation [6] and ionic sieving [8]. SMs are promising not only for environmental applications such as wastewater treatment and water desalination [9] but also for medical applications such as hemodialysis [6]. Often the permeability of SMs is accompanied by a transmittance for x-rays and electron beams, which renders SM materials such as SiN and SiC ideal as sample holders for various related microscopic techniques [16]. In addition to such microscopy applications, further nanopatterning of semiconductor membranes can enable the fabrication of x-ray optics [17–19].

In the field of optics, one can already find SMs of type II with potential applications in quantum communication and quantum computing. The first structures were based on group-III arsenide SMs comprising (In,Ga)As QDs as light emitters [11]. QD structures are also used in membrane-based quantum transport devices [12]. Often a certain tuneability of the electronic structure of such QDs by, for example, external fields is a key feature to the desired functionality. Tuning of the optical and/or electronic properties of QDs by strain fields [11] or electric fields [12] can be significantly simplified when SMs are used due to their reduced thickness. In addition to their use in the quantum realm, SMs of type II can also be key to macroscopic devices, as detailed below.

For the realization of flexible electronics it is often necessary to transfer type-II SMs onto flexible substrates [20], because any direct material deposition is hindered by the low chemical and temperature robustness of such substrates. Similar challenges are faced for photonic structures based on group-III nitrides, e.g., when one is aiming for ultraviolet (UV) light-emitting diodes and laser diodes. In the latter case, vertical-cavity surface-emitting lasers (VCSELs) based on type-II SMs have been demonstrated for the blue [21,22] and UV [23–25] spectral ranges. However, the development of UV VCSELs [25,26] poses several challenges if one uses only a single deposition technique, such as metal-organic vapor phase epitaxy (MOVPE). The fabrication of monolithic structures that comprise two distributed Bragg reflectors (DBRs) and an optically active material region [e.g., GaN/(Al,Ga)N QW stacks] becomes increasingly challenging for shorter emission wavelengths. The problem stems from the in-plane lattice mismatch and the achievable (Al,Ga)N compositions, which challenge the development of deep-UV DBRs with the required reflectivities (greater than 99%) [23]. Alternatively, the bottom DBR structure can be fabricated by sputtering, followed by the transfer of type-II SMs [27]. Subsequently, the top DBR structure can be obtained by an additional growth step. The need for these successive steps in the UV VCSEL fabrication process has boosted

interest in *photonic* type-II SMs, which are analyzed in this work.

The analysis of the optical, electronic, mechanical, and thermal properties of such type-II membranes constitutes a challenge requiring joint material characterization efforts. In this work, we demonstrate an optical and thermal analysis of photonic membranes made of group-III nitrides. We combine two-laser Raman thermometry (2LRT) with time-resolved photoluminescence (TRPL) spectroscopy to obtain the in-plane thermal conductivity $\kappa_{\text{in plane}}$ of SMs with different back-side roughnesses and layer sequences. We control the SM back-side roughening during the fabrication process on the basis of electrochemical etching of a highly-*n*-type-doped sacrificial GaN layer [28]. Controlling the surface roughness allows us to tailor the optical and thermal properties of these SMs, which is needed, for example, for VCSELs that can face severe thermal limitations. Any thermal characterization of such SMs must be noninvasive and compatible with their size, ranging from a few micrometers to hundreds of micrometers. This motivates the use of Raman thermometry, which we call “one-laser Raman thermometry” (1LRT). However, recently, it was shown that 1LRT is often not suited for measuring $\kappa_{\text{in plane}}$ of SMs made of group-III nitrides [13]. A single laser heats the sample during 1LRT while simultaneously providing the temperature probing via the (resonant) Raman signal. Thus, laser-induced heating is monitored only at the heat spot, which is a drawback of 1LRT. Although the absorbed laser power can be obtained from the analysis of 1LRT data, it remains an open question over which *volume* heating occurs. Simple measurements of the laser spot size are not sufficient because the mean free paths of phonons l_{MFP} , which significantly contribute to thermal transport, must be considered. Such phonons are frequently named “thermal phonons.” However, the impact of thermal phonons on the thermal conductivity κ for some l_{MFP} values is often hard to access experimentally. Interestingly, 2LRT measurements allow us to bypass some of these fundamental challenges, which is also accompanied by other experimental advantages over 1LRT as highlighted in this work. Primarily, thermal imaging by 2LRT gives access to temperature distributions (main quantity measured), which can be used to determine $\kappa_{\text{in plane}}$ (derived quantity) on the basis of a suitable thermal model.

This work focuses on thermal imaging by 2LRT and not just on the determination of $\kappa_{\text{in plane}}$, which can also be achieved by other techniques. A seminal study regarding the thermal characterization of GaN films by time-domain thermoreflectance was conducted by Beechem *et al.* [29], who analyzed the effect of different film thicknesses, dislocation densities, and impurity concentrations by measuring κ . The present work does not focus on such GaN films, but instead focuses on SMs in which thermal transport is governed mainly by $\kappa_{\text{in plane}}$. In a previous

study [13], we focused on a SM made of GaN grown on silicon with limited $\kappa_{\text{in plane}}$ values of $95^{+11}_{-7} \text{ W m}^{-1} \text{ K}^{-1}$ (see Sec. III A regarding the asymmetric error interval) and size (less than $10 \mu\text{m}$). In this work we investigate much larger GaN-based SMs of higher quality due to epitaxy on sapphire. As a result, we can reach $\kappa_{\text{in plane}}$ values up to $165^{+16}_{-14} \text{ W m}^{-1} \text{ K}^{-1}$ for an approximately-830-nm-thick structure. Such $\kappa_{\text{in plane}}$ values are in line with the requirements for VCSELs to ensure efficient heat dissipation.

Our study is structured as follows: In Sec. II we describe our samples (Sec. II A) and experimental techniques (Sec. II B), which form the basis for thermal imaging. In Sec. III, we demonstrate thermal imaging based on 2LRT map scans. We demonstrate the extraction of $\kappa_{\text{in plane}}$ for different in-plane directions of the SMs by a large-scale 2LRT map scan, as shown in Sec. III A. Subsequently, we emphasize the quantitative determination of the absorbed laser power that leads to local heating of the samples. Thus, through a combination of 2LRT and TRPL, we determine

the laser-induced heating power in Sec. III B. After having established the experimental basis for performing all-optical thermometry, we proceed by analyzing the impact of back-side roughness variations and etch channels on $\kappa_{\text{in plane}}$ in the SMs (Sec. III C). This is followed by a comparison with *ab initio* modeling in Sec. III D. By comparing experimental and theoretical results, we discuss the role of the phonon mean free paths on thermal transport in GaN in Sec. III E. Finally, the main conclusions are given in Sec. IV. Additional considerations are provided in Supplemental Material [30], which includes Refs. [13,23,27,28,31–33].

II. EXPERIMENTAL SECTION

A. Sample details

Figure 1(a) shows the group-III nitride layer stack grown by MOVPE that forms the basis for obtaining SMs. Details about the layer stack deposition and SM fabrication can be found in Sec. I in Supplemental Material [30]. All

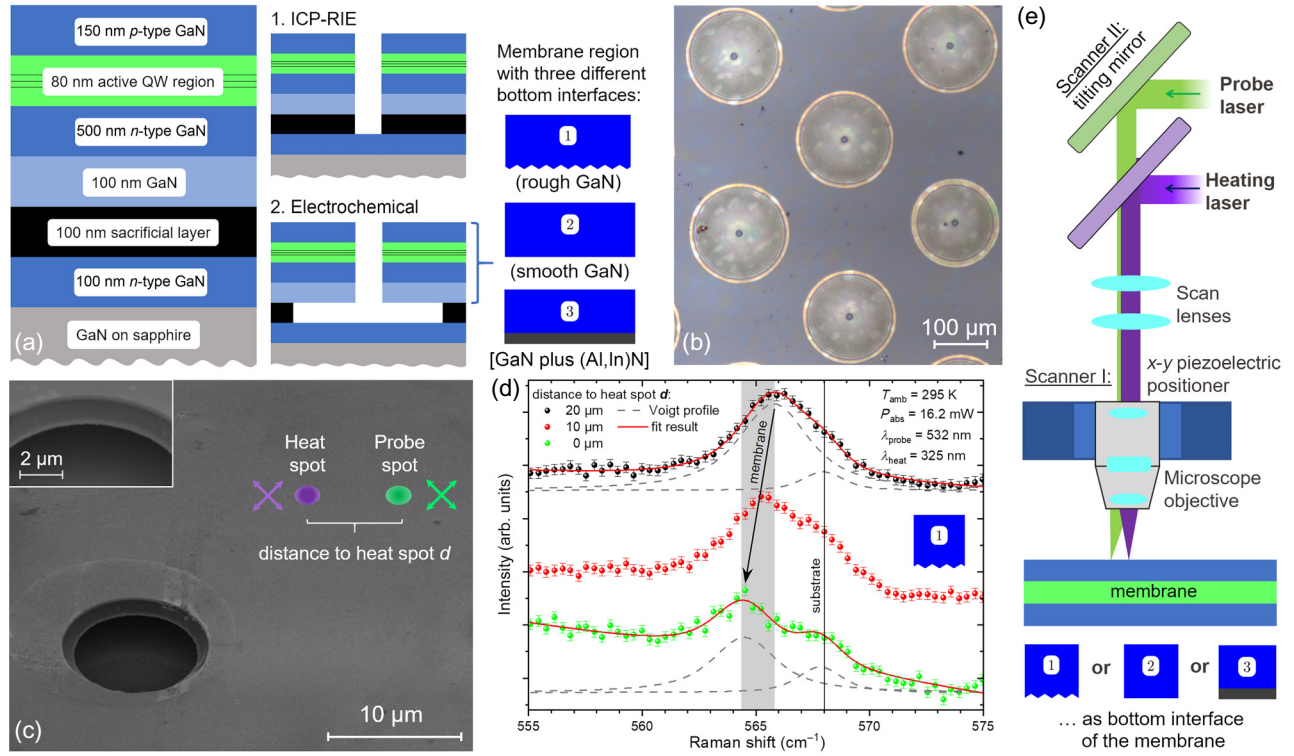


FIG. 1. (a) Layer sequence of the group-III nitride samples grown on sapphire, enabling the fabrication of photonic membranes by removal of a sacrificial layer. The three horizontal solid black lines indicate the location of embedded $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ QWs. Three samples (1–3) are obtained that each exhibit a different back-side roughness (ICP-RIE, inductively coupled plasma–reactive-ion etching). (b) Top-view optical microscopy image of a photonic membrane (sample 1) that consists mainly of *c*-plane GaN. (c) Side-view SEM image of sample 1, showing an etched hole and the membrane surface. The inset illustrates the smooth sidewall of the GaN membrane. The purple and green spots sketch the two laser spots that scan the membrane surface for the thermometry measurements based on Raman spectroscopy. (d) Raman spectra showing the E_2^{high} modes of the GaN membrane and the underlying GaN layer separated by an air gap for different distances between the heating ($\lambda_{\text{heat}} = 325 \text{ nm}$) and probe ($\lambda_{\text{probe}} = 532 \text{ nm}$) laser spots. (e) Sketch of the top-view 2LRT setup that allows the laser spots to scan the sample surface, leading to temperature maps that enable the determination of the in-plane thermal conductivity $\kappa_{\text{in plane}}$.

parameters of this layer stack (doping levels, layer thicknesses and sequence, alloy compositions, etc.), which is made mainly of wurtzite GaN, ensure bright light emission in the case of its use in a VCSEL [28,34]. $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ QWs form the active region that appears as horizontal black lines in Fig. 1(a). The layer stack comprises a sacrificial layer, which is needed to form a membrane. As shown in Fig. 1(a), first, holes are etched into the layer stack by inductively coupled plasma–reactive-ion etching (step 1). As a result, the etchant can reach the sacrificial layer during the electrochemical etching process (step 2) and a membrane can be formed. Note that the SM is not fully released, which ensures its thermal contact with the substrate. Depending on the voltages used during the electrochemical etching process, one can obtain either a rough GaN bottom interface (sample 1) or a much smoother bottom morphology (sample 2), as illustrated by the blue sample symbols. As sketched in Fig. 1(a), sample 3 comprises an additional etch stop layer made mainly of $\text{Al}_{0.82}\text{In}_{0.18}\text{N}$ (non-intentionally doped, *nid*), which enables the smoothest back-side morphology in this study. Samples 1–3 have the same top surface roughness and a close-to-identical $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ QW region.

Figure 1(b) depicts an optical microscope image of sample 1, showing the approximately-150- μm -wide SMs, which consist mainly of GaN. The optical image contrast arises from the approximately-100-nm-wide gap below the SMs (image details are explained in Sec. I in Supplemental Material [30]). A side-view scanning electron microscopy (SEM) image of the front side of sample 1 is shown in Fig. 1(c). The inset in Fig. 1(c) exemplifies the smooth edge of the central hole in the SM. Furthermore, two laser spots are sketched on the SM shown in Fig. 1(c). This double laser excitation scheme forms the basis for the optical and thermal analysis of all samples by 2LRT, which is explained in Sec. II B.

B. Experimental details and challenges

An example of typical Raman spectra that illustrate the transition from a heated to an unheated situation in the SMs is shown in Fig. 1(d). The Raman selection rules are strictly obeyed for these *c*-plane SMs consisting mainly of GaN, which means that for Raman measurements with the probe laser in an unpolarized, backscattering configuration $z(\cdot, \cdot)\bar{z}$, only the E_2^{low} and E_2^{high} modes appear as Raman-active modes. For the following Raman thermometry based on laser-induced heating, it is an advantage to rely on such nonpolar E_2 modes, because no coupling to free carriers occurs [35]. Such free carriers could be induced by doping, defects, or even the laser excitation itself. Among the two main Raman modes, E_2^{low} is known for its particularly small mode shifts under the impact of stress [36] and temperature [37]. Thus, the focus is on the E_2^{high} mode of GaN situated at approximately 567 cm^{-1} in

high-quality GaN [38,39] at $T_{\text{amb}} = 295\text{ K}$ as depicted in Fig. 1(d). Interestingly, in Fig. 1(d) we observe two E_2^{high} modes in close energetic vicinity. The first E_2^{high} mode at lower wave numbers belongs to the SMs, while the second E_2^{high} mode, situated at approximately 568 cm^{-1} , is linked to the *n*-type GaN beneath the sacrificial layer [Fig. 1(a)]. The wave-number position of the second E_2^{high} mode is typical for the growth of GaN on *c*-plane sapphire [40]. This small E_2^{high} mode separation constitutes an experimental challenge.

Our Raman mode assignment is further justified by comparing Raman spectra recorded at different distances to the laser-induced heat spot. We can control this distance d between the heating and the probe laser focus spots [see Fig. 1(c)] based on our experimental setup sketched in Fig. 1(e). In case of sufficient laser-induced heating, the spectral splitting between the two aforementioned E_2^{high} modes can be increased by decreasing d from $20\text{ }\mu\text{m}$ to $10\text{ }\mu\text{m}$ and finally to $0\text{ }\mu\text{m}$, as shown in Fig. 1(d). Only the first E_2^{high} mode shifts with decreasing values of d , while the second E_2^{high} mode position remains constant, as shown in Fig. 1(d). Thus, when the laser focus is set to the top surface of the SMs, the power of the heating laser does not suffice to heat the bulk material under the gap. The 2LRT measurements always take place in a vacuum, rendering this a vacuum gap or just a gap for the sake of simplicity. In contrast, SMs can be heated, which enable the acquisition of temperature maps as shown in Fig. 2(a). Note that the present custom-made optical setup requires only front-side access to the samples, which is not trivial and sets it apart from other setups reported in the literature [41].

C. Raman and photoluminescence detection

Laser-induced heating is obtained via a continuous-wave (cw) UV laser beam ($\lambda_{\text{heat}} = 325\text{ nm}$), which is guided toward the sample via a beam splitter, a $4f$ lens system (labeled “scan lenses”), and a $50\times$ UV microscope objective with a numerical aperture of 0.40 as depicted in Fig. 1(e). Details regarding the focusing procedure are provided in Sec. II in Supplemental Material [30]. The corresponding heating laser focus spot can be moved over the sample via a closed-loop *x-y* piezoelectric positioner labeled “scanner I.” The sample itself is placed in a vacuum chamber with a base pressure of approximately $1 \times 10^{-6}\text{ mbar}$ to avoid any convection cooling by gases. The Raman signal required for the temperature probe is generated via a cw probe laser beam ($\lambda_{\text{probe}} = 532\text{ nm}$) that is guided toward the sample via a two-axis piezoelectric-based tilting mirror (“scanner II”) before following the beam path of the heating laser [Fig. 1(e)]. Tilting the probe laser beam via scanner II enables scanning of the sample surface by the focused probe laser spot, while the heating laser focus spot remains fixed in a well-defined position.

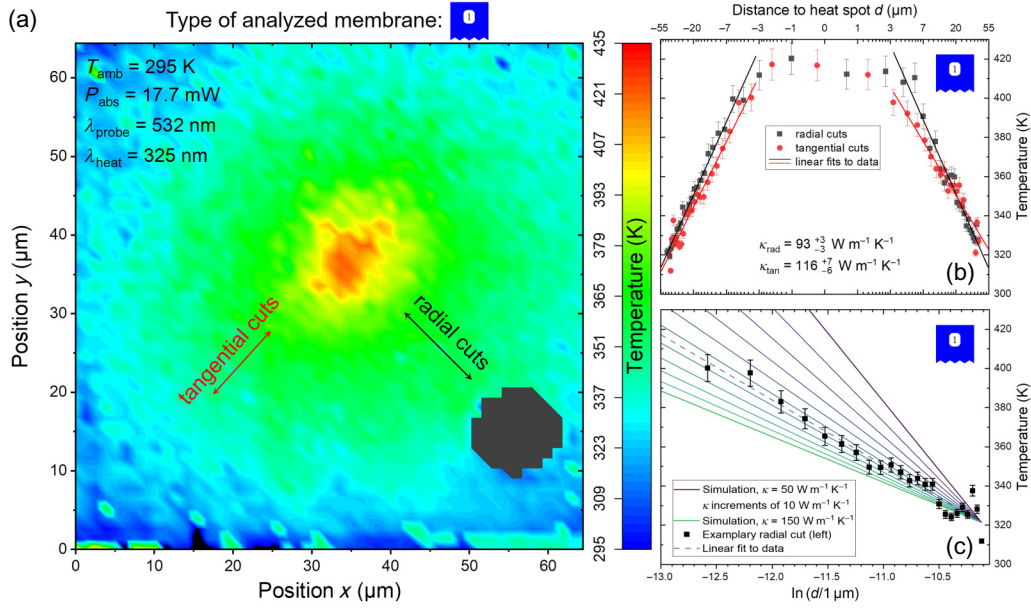


FIG. 2. Thermal analysis of sample 1 by 2LRT. (a) Temperature map obtained from the temperature-induced Raman shift. The hole in the membrane (dark gray area indicates the absence of temperature values) and the position of the laser-induced heat spot ($\lambda_{\text{heat}} = 325 \text{ nm}$) are visible. (b) Temperature profiles across the heat spot for radial (black) and tangential (red) directions. The symmetric double-logarithmic scale of the axis shows the distance to the heat spot d . For display reasons, this axis is linearized for $|d| \leq 1 \text{ μm}$. The solid lines represent linear fits to the data, indicating an in-plane thermal anisotropy. (c) Comparison of one side of a measured radial temperature profile (symbols) with temperature gradients simulated for different κ values (solid lines). As a result, we obtain $\kappa_{\text{in plane}}$ and its error interval from 2LRT.

As no tilting of the heating laser beam is introduced via scanner II, the $4f$ lens system simply focuses and collimates the heating laser beam passing through its center. As a result, the heating and probe laser focus spots can independently be moved across the surface of the samples, which enables the recording of Raman map scans for different heating laser powers based on the top-view configuration shown in Fig. 1(e). The corresponding absorbed power of the heating laser is denoted by P_{abs} in Fig. 1(d). The power of the probe laser is always low enough to avoid any unwanted heating. The detection beam path for the Raman signal or the photoluminescence (PL) signal is not sketched in Fig. 1(d) for the sake of simplicity. For detection, we used a single triple-grating monochromator equipped with filters and a charge-coupled device, providing a spectral resolution of 1 cm^{-1} or less at approximately 532 nm (grating with 1800 lines/mm), while for overview PL spectra we used a grating with 150 lines/mm . Details regarding TRPL measurements can be found in Sec. II in Supplemental Material [30].

D. Acquisition of temperature maps

Typically, the Raman mode shift, the Raman mode broadening, or the Raman mode intensity can be used as a thermometer based on suitable calibration. For the third of these, it is often convenient to use the intensity ratio

of a selected Raman mode on the Stokes side and the anti-Stokes side of the Raman spectrum. However, the resulting temperature resolution is often limited [42], which in the present case is further worsened by a strong background PL that disturbs the precise determination of the Raman mode intensities. This phenomenon is especially troublesome for 2LRT measurements based on a heating laser and a probe laser. The heating laser generates a strong PL signal that is composed of GaN band edge, QW, and defect luminescence traces distributed over a broad spectral range (details follow in the context of Fig. 3). Thus, in contrast to conventional Raman spectroscopy, the Stokes side and the anti-Stokes side are often impacted by background PL, which can be avoided only (partially) by a careful selection of λ_{probe} . However, flexibility regarding the choice of λ_{probe} is limited by the available microscope objectives, which face the challenge of focusing the heating laser and the probe laser simultaneously. In general, the Raman mode broadening represents a more suitable temperature gauge, because any impact of temperature-induced stress is minimized, in contrast to the case of the Raman mode shift [13]. However, the available spectral resolution often sets the limit for the applicability of the Raman mode broadening. Thus, for limited temperature increases, it is often practical to rely on the Raman mode shift for 2LRT measurements, as discussed in our previous study [13]. Accordingly, we also used this approach in the present work. Our procedure

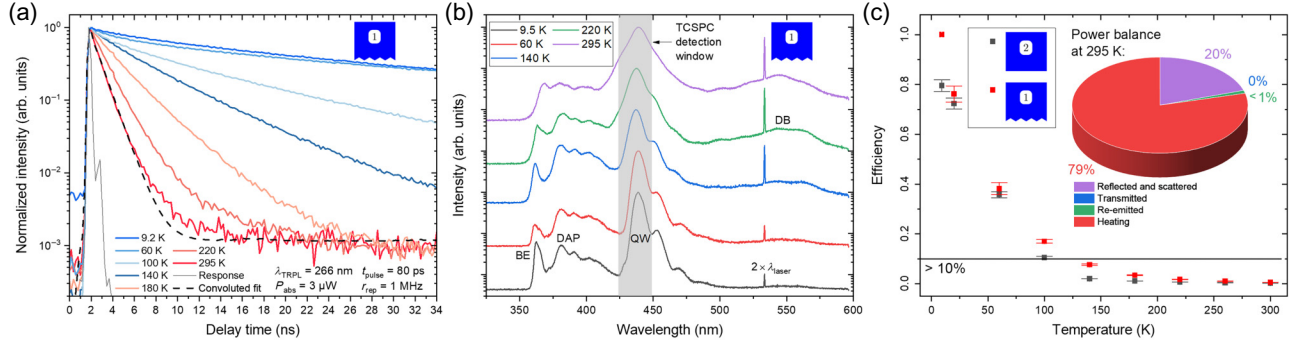


FIG. 3. Photoluminescence characterization: (a) TRPL analysis of sample 1 showing transients related to the QW emission at various temperatures (9.2–300 K). The impact of nonradiative channels lowers the measured lifetimes, yielding, so-called effective lifetimes τ_{eff} . In addition, the temporal system response (solid gray line) and an example for the convoluted fit (dashed black line) are shown. (b) Corresponding PL spectra show the GaN BE, QW, and DB emission of sample 1 for selected temperatures. In addition, the DAP emission is observed at 9.5 K, which is taken over by band-acceptor emission for temperatures larger than 60 K (see main text). The gray-shaded area illustrates the detection window that formed the basis for the recorded transients (TCSPC, time-correlated single-photon counting). (c) Estimation of the PL efficiency of samples 1 and 2 for temperatures up to 295 K under excitation with a pulsed laser with emission at $\lambda_{\text{TRPL}} = 266 \text{ nm}$. This wavelength λ_{TRPL} provides an excitation of GaN above its band gap similar to that provided by the heating laser used for 2LRT measurements. The pie chart summarizes the laser-induced power balance.

for measuring temperature maps by 2LRT is described in Sec. II in Supplemental Material [30], and further details can be found in Ref. [13].

We performed the required local temperature calibration on sample 1 and cross-checked the result with the other samples. For this calibration, we recorded Raman spectra for a fixed distance to the hole in the SMs ($d = 20 \text{ }\mu\text{m}$) with a Raman system almost identical to the one used for the 2LRT measurements. The only, minor difference is an increased spectral resolution of approximately 0.6 cm^{-1} at approximately 532 nm. Note that for the temperature calibration, the sample was placed in a vacuum chamber provided by a custom-made heat stage (HCS421VXY) made by Instec. From such a local temperature calibration, we obtain the spectral position of the E_2^{high} mode for different temperatures. After this trend is inverted, it is used as a thermometer for all 2LRT experiments.

III. RESULTS AND DISCUSSION

A. Temperature maps with high spatial resolution

Figure 2(a) shows a 2LRT temperature map of sample 1 over an area of $60 \times 60 \text{ }\mu\text{m}^2$ at $T_{\text{amb}} = 295 \text{ K}$. This map results from 2×2601 Raman spectra with an integration time of 10 s each, resulting in a total measurement time of approximately 15 h (which includes scanner displacement and settling times). We always record a heated and an unheated Raman map to remove any minor impact of strain fluctuations on the recorded results (see Sec. II in Supplemental Material [30]). Recording of such a map requires a high position stability of the two focus spots of the heating and probe lasers, but also

of the sample itself. The subtraction of the heated and unheated maps would otherwise yield a nonsymmetric temperature map due to spatial drift. During recordings, we only observed spatial drifts of 300 nm or less, which is significantly smaller than the step resolution of approximately $1.3 \text{ }\mu\text{m}$ in Fig. 2(a). This confirms the good mechanical stability of our custom-made setup. Thus, a subpixel drift compensation did not prove necessary for the present work, which studies only SM structures with rather large features on the scale of micrometers. However, we note that the reported 2LRT results also relied on a temperature stability in the laboratory of $\pm 1 \text{ }^\circ\text{C}$ and an active temperature stabilization of the main components of the setup (e.g., breadboard for all optics) of better than $\pm 0.1 \text{ }^\circ\text{C}$.

From the temperature map shown in Fig. 2(a), one can extract temperature gradients for any in-plane direction. We decided to plot the temperature gradients for the radial and tangential directions with respect to the central hole in the SM. This hole is indicated by the dark gray pattern in Fig. 2(a), which indicates the absence of any temperature values in this region of the SM. In the hole, only the E_2^{high} mode of the substrate appears as introduced in Fig. 1(d), which does not show any heating. Temperature cuts are shown in a symmetric double-logarithmic-scale plot in Fig. 2(b), which exhibits a linearized inner region ($|d| \leq 1 \text{ }\mu\text{m}$) for display reasons. From a simple analytical model based on Fourier's law, we can derive a basic understanding of the temperature T cuts shown in Fig. 2(b). For a thin membrane of thickness t with full radial symmetry and a pointlike heat source that injects the heating power P_{heat} , one obtains the following expression for the thermal

conductivity κ [3,43]:

$$\kappa = -\frac{P_{\text{heat}}}{2\pi t} \left(\frac{dT}{d \ln(d/1 \mu\text{m})} \right)^{-1}, \quad (1)$$

where d is the distance to the central heat source in micrometers and κ is assumed to be independent of T for sufficiently small values of dT . The measured temperature cuts plotted over d should be linear in the logarithmic part of a double-logarithmic-scale plot. Thus, the slope of the plotted temperature gradients is inversely proportional to κ . Interestingly, we find different slopes for the radial (black symbols) and tangential (red symbols) temperature cuts. The overall experimental situation requires a more sophisticated numerical modeling, which takes the three-dimensional (3D) sample geometry, the light penetration depths of the lasers, and the corresponding beam profiles into account. Details regarding the numerical model that was implemented in COMSOL MULTIPHYSICS based on a full 3D solution of the heat transport equation can be found in the supplemental material for Ref. [13]. The light penetration depths at λ_{heat} and λ_{probe} are based on absorption values given in the literature [44]. On the basis of this numerical model, we can derive temperature gradients for a range of κ values, as depicted in Fig. 2(c), to obtain a direct comparison with experimental data. Comparison of a linear fit [dashed gray line in Fig. 2(c)] with the data points and the corresponding set of numerical temperature gradients allows the determination of κ for the radial and tangential directions. Note that the κ increase in the numerical simulations is finer by a factor of 4 than that shown in Fig. 2(c) for display reasons.

1. Quantification of uncertainties

Equation (1) shows the dependence of κ on P_{heat} , t , T , and d . The errors of these quantities propagate into the (asymmetric) error of κ . The uncertainty of P_{heat} is closely related to the determination of P_{abs} , which we explain in Sec. III B. The membrane thickness t is not a significant source of uncertainty, because it can be determined with sub-10-nm-precision by spatially resolved reflectometry and spectroscopic ellipsometry (not shown). Thus, we have to focus on the uncertainties of T and d .

The error bars that we depict for T in, for example, Figs. 2(b) and 2(c) originate from the fit uncertainty of the Voigt peak positions [see Fig. 1(d)]. Commonly, our setup can achieve even lower error bars for T . However, the spectral vicinity of two Raman modes [see Fig. 1(d)] causes the fit uncertainties of the peak positions to be larger than the uncertainties caused by the long-term (more than 12 h) spectral instability of the setup. To quantify the 2LRT setup spectral stability, we performed stability measurements similar to those shown in Refs. [45,46]. Long-term

spectral instabilities are often related to temperature variations of the laser and the spectrometer, which we minimize by active temperature stabilization. As a result, we can obtain 2LRT imaging with even sub-1-K temperature resolution, which we recently demonstrated for bulk Si in the supplemental material for Ref. [47]. In Fig. 2(b) the error bars for d are smaller than the symbol size. Finally, the error interval for all κ values is obtained by comparison of the slope uncertainty of the linear fit (dashed gray line) with the modeled temperature gradients illustrated in Fig. 2(c). In this way, for example, the error of T (and d) is considered for the derivation of κ , which mimics an error propagation analysis. More details regarding this approach are given in Ref. [13] (see Fig. 7 therein).

For the radial and tangential directions indicated in Fig. 2(a), we obtain $\kappa_{\text{rad}} = 93_{-3}^{+3} \text{ W m}^{-1} \text{ K}^{-1}$ and $\kappa_{\text{tan}} = 116_{-6}^{+7} \text{ W m}^{-1} \text{ K}^{-1}$. This anisotropy for $\kappa_{\text{in plane}}$ is related to the direction of the etching process during fabrication. The etch front propagates in radial directions, which can lead to the formation of radial etch channels (see Sec. III C 2). All our κ values derived by 2LRT correspond to in-plane values ($\kappa_{\text{in plane}}$) for c -plane SMs. Averaging over the two temperature gradients for each temperature cut orientation allows us to further reduce the error intervals. The subscript and the superscript for the $\kappa_{\text{in plane}}$ values denote the lower bound and the upper bound of the asymmetric error interval. This asymmetry on the error interval can be understood on the basis of on Eq. (1), that connects κ with the inverse of $dT/d \ln(d/1 \mu\text{m})$. Thus, κ values obtained by 2LRT have asymmetric error bars, which is also true for 1LRT [13].

2. Effect of thermally induced stress

The maximal temperature rise T_{rise} amounts to approximately 100 K in Fig. 2(b). Remaining at such comparably low T_{rise} values ensures a negligible impact of thermally induced stress. In Ref. [13] we showed that thermally induced stress can alter the determination of temperatures by Raman mode shifts in contrast to the case of Raman mode broadenings. To first-order, the Raman mode broadenings are stress independent, in contrast to the Raman mode shifts [36]. As a result, starting at $T_{\text{rise}} \approx 150 \text{ K}$ in GaN, one can observe a deviation of the temperatures extracted by both methods. The local thermal expansion in the focus spot of the heating laser evokes a local compression of the sample due to thermal expansion, which commonly leads to a shift of the Raman modes to higher energies [36]. This shift toward higher energies can be compensated by the shift of the Raman modes to lower energies due to heating. Therefore, we can observe artificially lowered T_{rise} values in, for example, GaN for $T_{\text{rise}} \gtrsim 150 \text{ K}$, leading to an overestimation of $\kappa_{\text{in plane}}$ [Eq. (1)]. In this work, we circumvent such overestimations by imposing the aforementioned limit on T_{rise} .

B. Determination of the absorbed power P_{abs}

The absorbed laser power P_{abs} is a critical parameter for the determination of $\kappa_{\text{in plane}}$. It remains to be clarified how much of the absorbed laser power leads to the heating given by P_{heat} that we observe in 2LRT maps. The laser power P_{laser} that impinges on the samples contains the following contributions:

$$P_{\text{laser}} = P_{\text{abs}} + P_{\text{ref}} + P_{\text{trans}} + P_{\text{scat}}. \quad (2)$$

A certain fraction of the laser power P_{laser} is reflected (P_{ref}) or transmitted (P_{trans}), while pronounced light scattering with power P_{scat} can also occur. In general, P_{laser} can be measured simply with a conventional laser power meter, which also allows measurement of the transmission of all optical components in the laser beam path. P_{ref} can also be measured for every value of P_{laser} at λ_{heat} . For all P_{laser} values used in this work, we always observed a linear scaling of P_{ref} , strongly suggesting that this observation is also valid for P_{abs} . In addition, the measured value for P_{ref} at λ_{heat} agrees with reflectivity measurements taken with a UV-visible spectrophotometer and calculations of the reflectivity via the Fresnel equations for an angle distribution given by the numerical aperture of the microscope objective (see Sec. II C). As a result, we obtain a reflectivity of approximately 20% at $\lambda_{\text{heat}} = 325$ nm.

The total thickness of the SMs amounts to 830 nm; see Fig. 1(a). For determination of P_{trans} it is a good approximation to assume we have a pure GaN SM, which yields transmission of approximately 0.001% at $\lambda_{\text{heat}} = 325$ nm [44]. Thus, because of the high absorption of GaN at 325 nm, one can safely ignore P_{trans} for the given thickness. Consequently, any interference effects in the SM [13] can be ignored, because the high absorption coefficient of GaN at 325 nm ($\alpha \approx 135\,000\text{ cm}^{-1}$) translates into a light penetration depth of just 74 nm [44]. Thus, any corrections related to P_{trans} are negligible compared with the uncertainty of the $\kappa_{\text{in plane}}$ values on the order of 5%–15% (see Sec. III A).

A similar statement is valid for P_{scat} . For the measurements of P_{ref} we probe the specular reflection of the heating laser from the SM, which does not show any pronounced contribution of diffusely scattered light at deviating angles. We assign this observation to the monocrystallinity of the samples, their low front-side roughness (see Sec. III C 1), and a comparably low density of structural defects (e.g., the dislocation density is on the order of a few 10^8 cm^{-2}) [48]. Thus, we can ignore any contributions of P_{scat} , which simplifies Eq. (2) to

$$P_{\text{laser}} \approx P_{\text{abs}} + P_{\text{ref}} = P_{\text{heat}} + P_{\text{rad}} + P_{\text{ref}}. \quad (3)$$

Consequently, we have to determine the fraction of the laser power that is re-emitted by the samples, which we

denote by P_{rad} . Because of our comparably low values of T_{rise} and T_{amb} and the rather large gap of approximately 100 nm between the SM and the underlying nitride material, we can ignore thermal radiation. However, the SMs are photonic membranes, and their internal structure based on a stack of QWs is made to emit light. The main part of the related PL arises from a temperature-dependent balance of band-to-band transitions and excitonic transitions in the QWs, which are accompanied by additional sources of PL in group-III nitrides; see Fig. 3(b). Therefore, to estimate the total re-emitted power P_{rad} , we performed time-resolved PL spectroscopy, with results as summarized in Fig. 3.

The most pronounced source of PL in these samples is the embedded $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ QWs. Figure 3(a) shows the decay dynamics of the QW emission for temperatures from 9.2 to 295 K (see Sec. II in Supplemental Material [30] for experimental details). We observe a strong decrease of the measured lifetimes with increasing temperature due to a rising contribution of nonradiative channels. We use a convoluted fitting routine for all PL transients, which is exemplified in Fig. 3(a) for $T_{\text{amb}} = 295$ K by the dashed black line. For this fitting procedure, we convolute the measured temporal response of the detection system [solid gray line in Fig. 3(a)] with a single-exponential decay. The resulting dataset is fitted to the PL decay curves by altering the parameters of the exponential function, such as the effective decay time τ_{eff} , which represents a standard procedure [49]. The spectral window that was covered during the recording of the transients is illustrated by the gray-shaded area in Fig. 3(b). Here, we display the entire PL spectrum of sample 1 from the UV to the visible spectral range for some selected temperatures. At a temperature of 9.2 K, the PL spectrum starts (from left to right) with the band-edge (BE) emission and continues with the donor-acceptor-pair emission (DAP) and the main QW emission with its longitudinal optical phonon replicas, before a broad defect luminescence band (DB) appears at approximately 500–600 nm. All these sources of PL are well-documented in the literature for *p*-type GaN [50], which forms the top layer of the layer stack [Fig. 1(a)]. It should be noted that the DAP emission gradually transitions to a band-acceptor emission starting at a temperature of approximately 60 K due to the increasing ionization of all involved donor states. [50] For the following analysis of Eq. (3), we focus on the dominant QW emission that most prominently affects P_{rad} .

Following a procedure described by Langer *et al.* [51], one can estimate the internal quantum efficiency (IQE) of a QW for all temperatures by measuring the temperature dependence of $\tau_{\text{eff}}(T)$ and the initial QW PL intensity $I_0(T)$ at the beginning of the temporal decay depicted in Fig. 3(a). Typically, resonant or quasi-resonant excitation [52,53] of the QW emission is required for this determination of the IQE, which aims to prevent any

significant light absorption in the GaN matrix material. However, such sample excitation below the absorption edge of GaN is not compatible with the case of laser-induced heating with $\lambda_{\text{heat}} = 325$ nm. Thus, we performed the TRPL analysis with a wavelength of 266 nm, resulting in a light penetration depth of approximately 45 nm in GaN [44]. Consequently, carrier transfer from the 150-nm-thick *p*-type GaN cap layer of the sample to the QWs becomes essential. Thus, the TRPL analysis is influenced by nonradiative loss mechanisms occurring in the matrix material (GaN) and the QW. This is especially relevant, because the top *p*-type GaN layer is thicker than the light penetration depth of the heating laser (74 nm at $\lambda_{\text{heat}} = 325$ nm) and the laser used for TRPL measurements (approximately 45 nm at $\lambda_{\text{TRPL}} = 266$ nm). Thus, these measurements result not in an estimate of the sample IQE but in an “efficiency” of the dominant emission for the given excitation conditions required for sample heating. The results for samples 1 and 2 are plotted in Fig. 3(c), which represents an upper boundary estimate of P_{rad} , because $\tau_{\text{eff}}(T)$ and $I_0(T)$ were measured only for the dominant QW emission. Consideration of some of the other sources of PL (BE emission, DAP emission, and band-acceptor emission) would yield even lower efficiency values, because the QW emission proves to be more robust with increasing temperature. Here, only the DB emission is an exception, because it is activated with increasing temperature; see Fig. 3(b). However, its overall intensity remains small compared with the emission of the QWs. In general, all these additional PL contributions yield only minor corrections to the estimate of the sample efficiency and therefore P_{rad} , because the QW emission dominates the PL spectra at all relevant temperatures up to 295 K.

Thus, any relevant contributions of P_{rad} to the determination of $\kappa_{\text{in plane}}$ occur only for $T_{\text{amb}} \leq 100$ K, as shown in Fig. 2(c). For all more elevated temperatures the corrections given by P_{rad} compare well with the error of $\kappa_{\text{in plane}}$ on the order of 5–15% [13]. Nevertheless, for the present study at $T_{\text{amb}} = 295$ K, we still consider all corrections to P_{abs} by P_{rad} according to Eq. (3). However, because P_{rad} amounts to only approximately 0.5% of P_{laser} , which is also small compared with P_{heat} , one could safely simplify the right-hand side of Eq. (3). We obtain $P_{\text{abs}} \approx P_{\text{heat}}$ for the samples as long as $T_{\text{amb}} > 100$ K holds. Thus, the consideration of P_{rad} will play a dominant role for thermal transport studies on GaN membranes by 2LRT only at cryogenic temperatures. In addition, the consideration of P_{rad} may be needed even at higher temperatures depending on the sample quality, which can be further improved by, for example, transitioning to native GaN substrates and/or the application of so-called indium-containing underlayers that can boost the IQE of (In,Ga)N QWs [48]. Especially for thermometry experiments conducted on photonic membranes it remains essential to carefully check the

weight of P_{rad} to achieve a meaningful determination of P_{heat} .

Finally, the entire power balance of P_{ref} , P_{scat} , P_{trans} , P_{rad} , and P_{heat} at $T_{\text{amb}} = 295$ K can be plotted for the samples, as shown in the inset in Fig. 3(c). Almost 80% of the incident laser power leads to a heating of the samples, which is key to the determination of $\kappa_{\text{in plane}}$. Any 2LRT analysis of even brighter samples with higher P_{rad} at $T_{\text{amb}} = 295$ K would further be challenged by the PL background in the Raman spectra, as exemplified in Fig. 1(d). At the given high values of P_{abs} that are required for 2LRT measurements (see Fig. 2), one can observe a broadening of the QW emission [54], which leads to a PL tail that can appear as a background in the Raman spectra. In addition, the thermally activated defect band appears at $\lambda_{\text{probe}} \approx 532$ nm, as shown in Fig. 1(b), which further troubles the acquisition of Raman spectra. Therefore, it would be beneficial to shift λ_{probe} above 600 nm. However, the commercially available UV-visible microscope objectives limit such possibilities. It also remains a future task to vary P_{abs} during the TRPL measurements to check whether an increase of the PL efficiency can be achieved by saturation of nonradiative defects in GaN that hinder the carrier transfer to the QWs. Nevertheless, the interpretation of such measurements will prove challenging due to the fundamentally different excitation schemes during 2LRT and TRPL measurements, which rely on a cw laser and a pulsed laser, respectively. Furthermore, a partial screening of the built-in electric fields will also occur with increasing P_{abs} in the $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}/\text{GaN}$ heterostructure of the SMs [55], which further complicates the analysis. In a best-case scenario, the determination of the sample PL efficiency would be performed with the same laser that is used for 2LRT. Thus, efficiency measurements in an integration sphere over wide temperature ranges (from cryogenic temperatures up to the highest temperatures achieved during 2LRT) are promising for future work.

C. Temperature imaging by 2LRT

2LRT imaging of SMs can not only provide access to temperature maps that can yield $\kappa_{\text{in plane}}$ for all in-plane directions; large differences of $\kappa_{\text{in plane}}$ can also be made directly visible, as shown by comparison of Figs. 4(a) and 4(b).

1. Impact of the back-side roughness

Sufficiently large $\kappa_{\text{in plane}}$ differences can be generated by tuning the bottom surface roughness of the SMs while the top surface roughness remains unchanged. So far only sample 1 was analyzed in Fig. 2(a), which shows the roughest bottom surface morphology due to a processing bias voltage of 10 V (see Sec. I in Supplemental Material [30]). A corresponding AFM image is shown in the inset in Fig. 4(c), indicating a root-mean-square (rms) surface

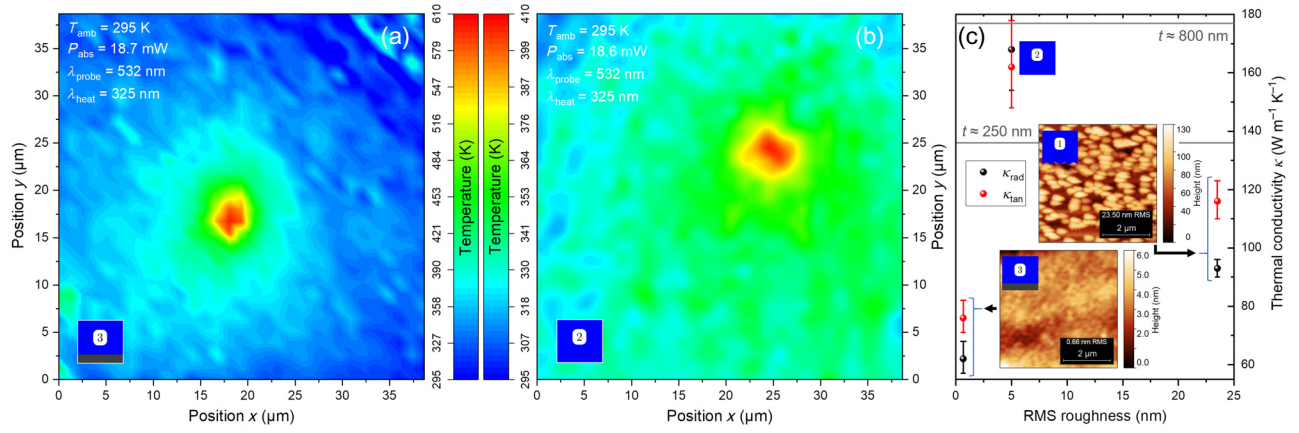


FIG. 4. Impact of the SMs' bottom surface roughness on $\kappa_{\text{in plane}}$. The different $\kappa_{\text{in plane}}$ values of (a) sample 3 and (b) sample 2 become directly visible in the temperature maps when one compares the maximal temperature and the overall temperature distribution (localized vs extended heating). (c) Summary of all radial (κ_{rad}) and tangential (κ_{tan}) values. The rms values for the bottom surface roughness are supported by two AFM images linked to samples 1 and 3. The horizontal gray lines show the theoretical κ values for two membrane thicknesses t .

roughness of 23.5 nm for a $5 \times 5 \mu\text{m}^2$ area. Decrease of this processing voltage to 6 V reduces the rms surface roughness to 5.0 nm for sample 2 (same area). The corresponding AFM image of the bottom surface of sample 2 can be found in Sec. III in Supplemental Material [30]. Any further decrease in the bottom surface roughness of the samples could be achieved only by adding an etch stop layer made of AlN/(Al,In)N [1 nm of AlN (nid) and 20 nm of Al_{0.82}In_{0.18}N] [28]; see Sec. I in Supplemental Material [30]. Such a variation of the bottom interface quality does not affect the core region of the SM where the QW is positioned. In other words, the approximation of P_{rad} from Sec. III B remains valid for samples 1–3. For sample 3, a bottom rms surface roughness of just 0.66 nm is achieved, as shown in the inset in Fig. 4(c), which is almost identical to the top rms surface roughness (0.67 nm for samples 1 and 2, and 0.76 nm for sample 3) [28].

However, the addition of an AlN/(Al,In)N interlayer to sample 3 comes at the cost of strongly reduced $\kappa_{\text{in plane}}$ values, as summarized in Fig. 4(c) for samples 1–3 for all radial (κ_{rad}) and tangential (κ_{tan}) in-plane directions with respect to the hole in the SMs. The values of κ_{rad} and κ_{tan} for sample 3 are more than halved compared with the values for sample 2, which exhibits a bottom rms surface roughness more than 5 times higher. Thus, the bottom surface termination of sample 3 with AlN/Al_{0.82}In_{0.18}N is beneficial from a thermal point of view as phonon scattering is strengthened. The AlN/Al_{0.82}In_{0.18}N provides a large acoustic contrast compared with GaN, yielding a higher thermal resistance due to the mismatch of the phonon dispersion relations. Thus, the omission of such additional etch stop layers in, for example, sample 2 is favorable for $\kappa_{\text{in plane}}$.

The pronounced $\kappa_{\text{in plane}}$ difference between samples 2 and 3 is visible from comparison of Figs. 4(a) and 4(b).

For almost identical values of P_{abs} , maximal temperatures T_{max} are reached at the central heat spot that differ by approximately 200 K. Visual inspection of the two thermal images in Fig. 4 reveals that the heat spot for sample 3 is more localized compared with the heat spot for sample 2, which is a sign of lower $\kappa_{\text{in plane}}$ values. It should be noted that the different temperature ranges in Figs. 4(a) and 4(b) are required for illustration of the thermal images due to the strongly deviating T_{max} values. 2LRT measurements become more challenging for high values of $\kappa_{\text{in plane}}$, because the heated area begins to extend over several hundred square micrometers. In general, it is not required to enclose the entire temperature gradient between T_{max} and T_{amb} in a 2LRT image, because the determination of $\kappa_{\text{in plane}}$ is based only on the temperature differences given by T_{rise} . Furthermore, higher values of $\kappa_{\text{in plane}}$ go hand in hand with lower values of T_{rise} , which are increasingly difficult to resolve by Raman thermometry [13]. In addition, as mentioned in Sec. III A, we aim to limit the T_{rise} values to mitigate the impact of thermally induced stress.

2. Impact of etch channels

We do not expect an intrinsic reason for pronounced thermal anisotropies in the c plane of the present SMs made mainly of wurtzite GaN. Hence, we assume that the lower value of κ_{rad} compared with κ_{tan} (a difference of approximately 20% for sample 1) is related to radial etch channels that can form in the SMs due to electrochemical etching. Such channels can form at the perimeter of the central hole in the SM and progress from there on radially in n -type GaN. As a result, the density in the membranes is no longer isotropic, which causes an anisotropy of $\kappa_{\text{in plane}}$. To picture such etch channels, we provide further backside and cross-section SEM images of the membranes in

Sec. III in Supplemental Material [30]. In Fig. 4(c) we summarize the 2LRT results for samples 1–3 by plotting κ_{rad} and κ_{tan} . We can resolve in-plane thermal anisotropy for samples 1 and 3, while large error bars prevent any firm conclusion for sample 2. A simple visual inspection of the temperature profile shown in Fig. 2(a) enables the observation of a thermal anisotropy (as visible from the extent of the temperature profile in the tangential direction with respect to the radial direction away from the hole). This observation directly shows a key benefit of 2LRT compared with 1LRT.

Natural diamond can exhibit similar etch channels [56]. Recently, Zhang *et al.* [57] also reported on etch channels in GaN due to electrochemical etching. Any modeling of anisotropic $\kappa_{\text{in plane}}$ values is hindered by the small diameters of the etch channels, which approach the mean free path of thermal phonons in GaN (see Figs. 2 and 3 in Supplemental Material [30]). Consequently, the thermal transport cannot be modeled by, for example, COMSOL MULTIPHYSICS, and more realistic modeling of nondiffusive phonon transport, for example, by OpenBTE [58], will be required.

D. *Ab initio* modeling of the thermal conductivity κ

$\kappa_{\text{in plane}}$ was computed via an *ab initio* solution of the linearized phonon Boltzmann transport equation (BTE) as implemented in the ELPHBOLT package [59]. The workflow based on density functional theory is detailed in Ref. [13]. From the phonon-mode-resolved solution of the BTE, we can, for example, calculate the phonon mean free path l_{MFP} distribution and the thermal conductivity tensor of wurtzite GaN. The phonon modes are identified by the band s and the wave vector $\mathbf{q}$. We consider both three-phonon scattering and four-phonon scattering. The latter proves to be relevant for wurtzite GaN due to a pronounced gap separating the high-lying optic sector from the rest in the phonon band structure, which causes a dip in the three-phonon scattering phase space [13]. In addition, the phonon-isotope scattering is considered within the Tamura model [60], while the mode-resolved phonon-boundary scattering rates are computed with use of the following empirical expression:

$$\frac{1}{\tau_{\text{phonon-boundary}}^{\text{sq}}} = \frac{2v_{\text{sq}}^{\perp}}{t}, \quad (4)$$

with $v_{\text{sq}}^{\perp}$ the mode-resolved phonon group-velocity components perpendicular to the plane of the SM and t the membrane thickness. Equation (4) considers fully diffusive scattering of phonons at the top and bottom of the photonic SMs. We obtain $\kappa_{\text{in plane}} = 177 \text{ W m}^{-1} \text{ K}^{-1}$ for an 800-nm-thick pure GaN membrane, which compares well with the results obtained for sample 2, yielding $\kappa_{\text{rad}} = 168_{-14}^{+15} \text{ W m}^{-1} \text{ K}^{-1}$ and $\kappa_{\text{tan}} = 162_{-14}^{+16} \text{ W m}^{-1} \text{ K}^{-1}$.

The corresponding theoretical cumulative $\kappa_{\text{in plane}}$ and its dependence on l_{MFP} is plotted in Sec. IV in Supplemental Material [30] for two t values. We also discuss the applied averaging process (isotropic vs pure in-plane averages) to determine the theoretical κ values in Sec. IV in Supplemental Material. Figure 4(c) includes two horizontal gray lines that summarize our modeling results for t of approximately 250 nm and approximately 800 nm.

The impact of the different layers in the layer stack of samples 1 and 2 (e.g., GaN layers with different doping, QWs) appears almost negligible compared with the impact of the addition of the etch stop layers made of AlN and $\text{Al}_{0.82}\text{In}_{0.18}\text{N}$ for sample 3. A brief inspection of the layer stack related to samples 1 and 2 [see Fig. 1(a)] shows that almost the entire photonic SM is made of GaN, in agreement with the numerical simulations. The n -type and p -type doping of GaN up to $3 \times 10^{18} \text{ cm}^{-3}$ (n_{Si}) and $(1\text{--}5) \times 10^{19} \text{ cm}^{-3}$ (n_{Mg}) does not significantly reduce $\kappa_{\text{in plane}}$ [13] due to the low atomic fraction. The main acoustic contrast in the material stack of samples 1 and 2 arises from the multi-QW stack. However, this stack comprises only three 2.8-nm-thick $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ QWs. As a result, only 1% of the photonic SM consists of material that is not GaN, and even in this case the atomic fraction of foreign atoms is small, at just 15%. This justifies the theoretical approach and further explains the good agreement between the theoretical and experimental results for sample 2.

Using similar arguments, we can describe why adding the thin AlN/ $\text{Al}_{0.82}\text{In}_{0.18}\text{N}$ layer has such a strong impact on $\kappa_{\text{in plane}}$ for sample 3. Both layers are also thin (1 and 20 nm, respectively); however, their atomic composition deviates more strongly from the remaining part of the layer stack in the SM, leading to a larger acoustic contrast. It will be an interesting challenge to calculate the thermal impact of such additional layers that can not only serve as an etch stop but can also passivate surfaces [61]. However, the addition of AlN and $\text{Al}_{0.82}\text{In}_{0.18}\text{N}$ layers also changes the built-in electric field at the bottom region of the SMs. Alteration of such a built-in electric field changes the electrochemical underetching process and therefore the etch channel formation (see Sec. III in Supplemental Material [30]). A detailed microscopic analysis of such etch channels is beyond the scope of this work. Furthermore, a thermal characterization in the cross-plane direction would also be of interest, requiring alternative techniques such as frequency-domain thermorefectance measurements, time-domain thermorefectance measurements, or the square pulsed source method [62,63].

Our highest measured value for $\kappa_{\text{in plane}}$ of $168_{-14}^{+15} \text{ W m}^{-1} \text{ K}^{-1}$ (κ_{rad}) exceeds previously reported values for GaN films of similar thickness, but still remains approximately a factor of 2 below bulk GaN κ values [64]. The data summary given by Beechem *et al.* [29] reports maximum κ values of approximately $130 \text{ W m}^{-1} \text{ K}^{-1}$.

for approximately-1- μm -thick GaN films. However, no direct comparison can be made between SMs and GaN films, because of the different bottom interface (e.g., GaN/vacuum vs GaN/substrate). Interestingly, for sample 3 we measure $\kappa_{\text{in plane}}$ values close to the maximum reported by Beechem *et al.*, which suggests a link between the high acoustic contrast at the GaN/ $\text{Al}_{0.82}\text{In}_{0.18}\text{N}$ interface and the GaN/substrate interface found in films. The good agreement between the theoretical $\kappa_{\text{in plane}}$ value of $177 \text{ W m}^{-1} \text{ K}^{-1}$ and the highest experimental equivalent ($\kappa_{\text{rad}} = 168^{+15}_{-14} \text{ W m}^{-1} \text{ K}^{-1}$) also illustrates the relevance of the four-phonon-scattering process in GaN at $T_{\text{amb}} = 295 \text{ K}$. Furthermore, recently, we demonstrated in the supplemental material of Ref. [47] that 2LRT is capable of deriving realistic κ values for bulk Si, which supports the results in the present paper.

E. The role of the phonon mean free path

Despite the detailed determination of P_{abs} in Sec. III B, it is questionable over which volume heating occurs. Although the laser spot size can be measured precisely (see Sec. II in Supplemental Material [30]), the extent of the heated volume is unknown due to the distribution of l_{MFP} values. For instance, phonons with l_{MFP} values beyond even $1 \mu\text{m}$, for example, can significantly contribute to κ in bulk GaN. [13] Thus, for GaN SMs it is not sufficient to perform 1LRT measurements as soon as the laser spot diameter approaches the l_{MFP} values of phonons that exhibit a significant contribution to κ . This statement remains true also when, for example, relevant l_{MFP} values reach 10% of the laser spot diameter, because phonon escape from the temperature probe volume still occurs. As a result, 1LRT can yield too large κ values for high-quality samples [13]. A workaround is achieved by an increase of the laser spot diameter during 1LRT measurements [47], but this contradicts the aim of any local thermal characterization. In contrast, 2LRT is a powerful tool to measure temperature gradients up to a distance of several tens of micrometers from a heat spot [Fig. 2(b)]. In such regions our numerical modeling of the experimental data by COMSOL MULTIPHYSICS is more adequate. A next step would be the comparison of the present data with the output of a real-space solver for *ab initio* phonon transport such as OpenBTE [58]. However, solving the steady-state phonon BTE on the basis of input from *first-principles* calculations (e.g., based on the use of ELPHBOLT [59]) is a numerical challenge for our large structures, comprising boundaries and interfaces.

IV. CONCLUSIONS

In this work, we demonstrated the noninvasive thermal characterization of photonic SMs that can be applied, to example, to VCSELs. These SMs are made mostly of GaN and are free-standing, which distinguishes this

work from previous studies. We showed how to cope with highly emissive samples during an all-optical thermal characterization by considering the fraction of light that is re-emitted during the laser-induced heating. The reported TRPL analysis yielded a threshold for the thermometry of $T_{\text{amb}} \approx 100 \text{ K}$. Below this temperature, ignoring the QW-induced light emission leads to an erroneous determination of $\kappa_{\text{in plane}}$. This work also shows the relevance of a detailed optical precharacterization that is needed for an all-optical thermal characterization. We disentangled the laser-induced power balance during a 2LRT analysis by considering the power of the reflected, scattered, transmitted, and re-emitted light. As a result, we obtained an estimate of the fraction of the laser power that is converted into heat, which is a general challenge for optical thermometry.

One highlight of the present work is the thermal imaging by 2LRT over several hundred square micrometers, which allows us to overcome the limits of conventional Raman thermometry, which is not spatially resolved. Through this thermal imaging, we found an indication for a thermal anisotropy in the plane of photonic SMs, which is linked to extrinsic reasons such as the presence of etch channels. This shows the suitability of 2LRT for studying thermally anisotropic materials. Using COMSOL MULTIPHYSICS simulations based on a 3D solution of the heat transport equation, we demonstrated how to extract $\kappa_{\text{in plane}}$ values from 2LRT. In a best-case scenario, thermal imaging renders any differences in $\kappa_{\text{in plane}}$ directly accessible to the experimentalist. Such differences were linked to variations in the SMs' bottom surface roughness and the presence of additional layers. It was shown that, despite the presence of three QWs with a thickness of a few nanometers, the membranes can still be approximated as being made of pure GaN material. In contrast, the addition of interlayers made of AlN and $\text{Al}_{0.82}\text{In}_{0.18}\text{N}$ reduces $\kappa_{\text{in plane}}$ by a factor of more than 2 due to their high acoustic contrast in comparison with GaN. Finally, for an 800-nm-thick GaN membrane we simulated $\kappa_{\text{in plane}} = 177 \text{ W m}^{-1} \text{ K}^{-1}$ (*c* plane), which compares well with the experimental 2LRT results for the best sample with comparable thickness, yielding an averaged $\kappa_{\text{in plane}}$ of $165^{+16}_{-14} \text{ W m}^{-1} \text{ K}^{-1}$. The thermal analysis in this work should provide a pathway toward thermal imaging over wide temperature ranges, which is key to most direct experimental analyses of different phonon transport regimes or to probing phonon-polariton transport across interfaces.

ACKNOWLEDGMENTS

N.H.P. acknowledges funding from the Deutsche Forschungsgemeinschaft (DFG) for an Emmy Noether research grant (Grant No. 534386252). G.R. acknowledges funding from the MIT-IBM Watson AI Lab (Challenge No. 2415). G.R. and G.C. acknowledge funding from

MIT Global Seed Funds. M.E. and G.C. acknowledge funding from the Central Research Development Fund of the University of Bremen for the project “Joint optical and thermal designs for next generation nanophotonics.” The research of W.S., M.E., K.D., G.W., and G.C. was further funded by the Major Research Instrumentation Program of the DFG (Grant No. 511416444). G.C. further acknowledges the MAPEX-CF Grant for Correlated Workflows (Grant No. 40401080). J.C. and Å.H. acknowledge financial support from the European Research Council under the European Union’s Horizon 2020 research and innovation program (Grant Agreement No. 865622), and the Swedish Research Council (Grant No. 2018-00295). The samples were processed at Myfab Chalmers, Chalmers University of Technology.

DATA AVAILABILITY

The data that support the findings of this study are not publicly available. The data are available from the corresponding author upon reasonable request.

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