Supporting Information:

Controlling the Infrared Dielectric Function through Atomic-Scale Heterostructures

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S1. Polar material dielectric function and calculated reflectance

In order to calculate the reflectance of bulk AlN, GaN, and SiC as shown in Fig. 1b of the paper, we used an analytical expression for the dielectric function of each material. Each material belongs to the hexagonal crystal system, and is therefore birefringent, having an out-of-plane dielectric function $\varepsilon_\parallel$ for light polarized along the c-axis of the lattice (extraordinary) and another in-plane dielectric function $\varepsilon_\perp$ for light polarized perpendicular to the c-axis (ordinary). In the mid-infrared (IR), the dielectric function for a uniaxial polar, undoped semiconductor is given by the following formula:

$$\varepsilon_j(\omega) = \varepsilon_\infty,j \left( 1 + \frac{\omega_{LO,j}^2 - \omega_{TO,j}^2}{\omega_{TO,j}^2 - \omega^2 - i\omega\gamma_j} \right) \tag{S1}$$

where $j = \parallel$ or $\perp$ for the permittivity parallel or perpendicular to the c-axis. The parameter $\varepsilon_\infty$ is the high frequency dielectric constant and $\omega_{LO}$ and $\omega_{TO}$ are the LO and TO phonon frequencies, with a damping parameter of $\gamma$. The $\omega_{TO,\parallel}$ and $\omega_{LO,\parallel}$ phonon modes oscillate parallel to the c-axis and correspond to $A_1$ symmetry modes, while the $\omega_{TO,\perp}$ and $\omega_{LO,\perp}$ phonons oscillate perpendicular to the c-axis and correspond to $E_1$ symmetry modes. In Table S1, we list the appropriate LO and TO phonon frequencies for AlN, GaN, and SiC taken from literature.

| Material | $\varepsilon_\infty,\perp$ | $\varepsilon_\infty,\parallel$ | $\omega_{TO,\perp}$ [cm$^{-1}$] | $\omega_{LO,\perp}$ [cm$^{-1}$] | $\omega_{TO,\parallel}$ [cm$^{-1}$] | $\omega_{LO,\parallel}$ [cm$^{-1}$] |
|---------|----------------|----------------|----------------|----------------|----------------|----------------|
| AlN     | 4.3            | 4.0            | 673            | 916            | 614            | 893            |
| GaN     | 5.3            | 5.3            | 561            | 743            | 533            | 735            |
| SiC     | 6.5            | 6.7            | 798            | 966            | 783            | 964            |

**Table S1** Parameter values used for computing the bulk dielectric function of AlN, GaN, and SiC. The phonon frequencies were reported in Ref. 1, 2, and 3, for AlN, GaN, and SiC, respectively.

We calculated the reflectance in Fig. 1 using Fresnel equations for normal incidence, with the dielectric function of each material given by Eq. (S1) with the appropriate values from Table S1.
For the calculation, we let $\gamma_\perp = \gamma_\parallel = 5$ cm$^{-1}$ for each material. To calculate the reflectance of the SL structures shown in Fig. 1e and f, we used the ellipsometric software WVASE. This software uses the transfer-matrix method to calculate the reflectance from the SL structures. For the GaN and AlN layers and the SiC substrate, we again used Eq. (S1) for the dielectric function of each material. For sample A, we calculated the reflectance from 50 alternating layers of GaN and AlN layers, each with a thickness of 2.2 nm, on top of a 52 nm AlN buffer layer/350 µm thick SiC substrate. For sample B, we used 500 alternating layers of 1.4 nm thick GaN and 1.2 nm AlN on top of a 350 µm thick SiC substrate, with no buffer layer to match the sample.

**S2. AlN/GaN SL vs. AlGaN IR responses**

We briefly note an important point that the IR response of a SL of two materials is, in general, distinct from an alloy of the same two materials. In Fig. S1, we overlay the reflectivity of AlGaN thin film (500 nm thick Al$_{0.42}$Ga$_{0.58}$N on SiC wafer) and Sample B for comparison. The difference in IR response is because the two materials have distinct sets of phonon modes. AlGaN displays two-mode and one-mode behavior for zone-center TO and LO phonons, respectively, with the phonon frequencies depending on the AlGaN composition. In contrast, the AlN/GaN SL displays multiple TO and LO phonon modes. The SL phonon density of states depends on the structural geometry, and the phonon modes correspond to confined and interface-like modes.
Figure S1 Comparison of the reflectivity of an Al$_{0.42}$Ga$_{0.58}$N alloy thin film (500 nm on SiC wafer) and AlN/GaN SL Sample B from manuscript.

S3. Chemical composition and structure analysis of Sample B

We used electron energy loss spectroscopy (EELS) in order to characterize the chemical structure of the SL. Figure S2a and b show a high-angle annular dark-field imaging (HAADF) image of Sample B and EELS data acquired along the red dashed line in the STEM image. From the oscillating Z-contrast observed in the HAADF image and line profile, there is obvious chemical segregation between the layers. The EELS results show that there is a relatively high concentration of Ga everywhere on the sample, which likely results from residual Ga used in the milling process to prepare the cross-sectional sample. Even so, there appears to be a significant amount of Al in the Ga layers, which suggests some intermixing between the layers. The effect is seen more clearly in Figure S2c where we compare the EELS spectra from the Ga- and Al-rich layers. Here, it can be seen that both Al and Ga are present in each layer, but that the Al almost disappears in the Ga rich layer, while the Ga peak only reduces slightly in the Al layer indicating that much of the Ga signal is due to Ga implantation from the FIB and not from layer intermixing.
Figure S2 (a) Cross-sectional HAADF-STEM image of Sample B, with line scan region marked by the dashed red line. (b) EELS line profile HAADF intensity (black) for Ga L Edge (red) and Al K edge (blue) along the red dashed line in the left panel. (c) EEL spectra from the Ga rich and Al rich layers highlighted in (b).

In addition to the EELS results, intermixing between the layers is also suggested by the sinusoidal-like HAADF intensity along the SL growth direction. This is shown in Fig. S3, in which the HAADF intensity is vertically binned along the red rectangle in the STEM image (Fig. S3a) of Sample B to look at the atomic scale intensity. In Fig. S3b, the total integrated HAADF intensity profile is plotted, and in Fig. S3c the intensity profile is filtered using a Gaussian to remove the atomic-level effects. The HAADF intensity changes between the layers are not sharp at an interface but sinusoidal-like, further indicating some intermixing between the layers. We estimated the composition the layers by fitting satellite peaks in XRD scans. The best fits indicate that the Ga-rich (Al-rich) layer has a composition of \( \sim \text{Al}_{0.2}\text{Ga}_{0.8}\text{N} \) (\( \text{Al}_{0.9}\text{Ga}_{0.1}\text{N} \)). However, despite such intermixing, it is clear from the deviations of Sample B from the \( \text{Al}_{0.42}\text{Ga}_{0.58}\text{N} \) sample in Fig. S1 that such intermixing is not the origin of the changing dielectric function.
S4. Determination of optic phonon modes using IR second harmonic generation

In addition to the SHG spectra shown in the main text of this work, here we provide the complete data set measured on Sample A (see Fig. S4a). This sample exhibits a spatial gradient in both the AlN and GaN layer thicknesses across the wafer. Therefore, performing the measurement on different locations on the sample corresponds to varying absolute and relative AlN and GaN layer thicknesses. Here, the different locations on the sample are labelled 1 to 10 as shown in the inset of Fig. S4c.

Clearly, the observation that the bulk resonances from the SiC substrate and the AlN buffer layer are not affected by the SL thickness holds true for all 10 measurements. In contrast, the SL phonon resonances exhibit a gradual modulation of both the frequency and amplitude as the AlN and GaN layer thicknesses vary. While the exact dependence between layer thickness (ratio) and frequency/amplitude appears non-trivial, the full data set further confirms the previous observation in demonstrating the tunability of the phonon modes in the XH.
The same behavior is observed in the corresponding reflectivity spectra (see Fig. S4b), even though it is much harder to assess as the resonances appear as edges rather than peaks as they are observed in the SHG data. The latter have been analyzed using least-squares fitting of a Lorentzian line shape function. The resulting peak shifts and amplitudes are shown in Fig. S4c-d.

![Figure S4](image-url)  

**Figure S4** Full data set of the SHG and reflectivity spectra. a) and b) show, respectively, the SHG and reflectivity spectra in false colors at each of the labelled positions on the SL sample corresponding to different GaN and AlN layer thicknesses. c) and d) show, respectively, the spectral positions and peak amplitudes of the two hybrid TO phonon modes, H1 (red) and H2 (green), also marked in a.

**S5. XH dielectric function**

We used ellipsometry to determine the mid-IR permittivity of the SL structures. Ellipsometry measures the change in the polarization of reflected light from the surface of a sample and is generally reported with the parameters $\Psi$ and $\Delta$. These parameters are related to the ratio of the Fresnel reflection coefficients for $p$- and $s$-polarized light, $r_p$ and $r_s$, respectively, by

$$\frac{r_p}{r_s} = \tan(\Psi)e^{i\Delta}.$$
The SL permittivity was determined by modeling the permittivity with an analytic formula and then fitting the ellipsometric data by using the permittivity model to compute the Fresnel reflection coefficients. Since ellipsometric measurements are determined from the ratio of two measured quantities, the derived permittivity values are very precise. We used the ellipsometric software WVASE to fit the ellipsometric data and extract the permittivity.

**Figure S5** Ellipsometry parameters $\Psi$ and $\Delta$ of sample A and sample B. The black curves are the measured ellipsometry data. The red curves are the calculated $\Psi$ and $\Delta$ for each sample assuming that the permittivity of the individual SL layers can be described by their bulk permittivity values. The blue curve is a fit to the data which uses Eq. (2). Due to the strong overlap between the measured and multiphonon calculation, the blue and black curves appear nominally indistinguishable over much of the reported spectral range. This model treats the SL as a single multiple polar phonon mode material and assumes a single dielectric function for the entire SL.

We acquired ellipsometric measurements from both samples A and B for incident angles ranging from 35° – 75° in 10° increments. The black lines in panel a) of Fig. S5 show the measured $\Psi$ and
Δ for sample A at an incident angle of 65º. Phonon modes of the sample can be correlated with spectral peaks and dips within the Ψ spectrum. We include in Fig. S5a the calculated values of Ψ and Δ (red lines) for sample A, assuming that the permittivity of the individual SL layers can be described by Eq. (S1). These calculated values of Ψ and Δ were determined using the transfer-matrix method for the SL layers and include the effect of 52 nm AlN buffer layer and the SiC substrate. From the calculation, we can see that Ψ peaks (dips) for the E₁(TO) (A₁(LO)) modes of GaN and AlN at about 560 cm⁻¹ (740 cm⁻¹) and 670 cm⁻¹ (890 cm⁻¹), respectively. In contrast, for the measured Ψ, three spectral peaks are observed at 585, 640, and 670 cm⁻¹ and four spectral dips at 738, 828, 864, and 890 cm⁻¹. The phonon modes at 670 cm⁻¹ and 890 cm⁻¹ match the bulk values of AlN, however, as shown from the SHG data, these features originate from the AlN buffer layer phonons and not the SL. Overall, it is apparent that the bulk model calculation does not accurately describe the ellipsometric data, as many of the phonon modes are spectrally shifted from the bulk values. Aside from the apparent shift in the phonon frequencies, the bulk model is inadequate for fitting the ellipsometric data because it only includes two A₁(LO) phonons for the SL, but we observe three (neglecting the A₁(LO) phonon of the buffer layer). Similar disagreement is observed between the measured (black curve) and bulk calculation (red curve) of Ψ and Δ for Sample B shown in panel b of Fig. S5. Note for Sample B, there is no buffer layer so there are no additional peaks complicating the interpretation. Again, the bulk model predicts two dips in Ψ, but we observe three.

To account for the additional observed phonon modes, we instead model the entire AlN-GaN SL as a single material with multiple phonon modes using Eq. 2. The best fit for each sample is shown in blue in Fig. S5. For Sample A, a good fit was achieved for \( k = 2 \) and \( k = 3 \) for the in-plane and out-of-plane dielectric function, respectively. The resulting dielectric function is shown in Fig. S6.
We found that the same number of phonon modes could be used to achieve a reasonable fit for Sample B, however, including an additional phonon mode for the out-of-plane dielectric function significantly improved the fit. For the Sample B fit shown in Fig. 3a-b, we used $k = 2$ and $k = 4$ for the in-plane and out-of-plane dielectric function, respectively. The derived dielectric function from the fit is plotted in Fig. 3 of the manuscript. Table S2 summarizes the best-fit parameters for both samples.

![Graph of dielectric functions](image)

**Fig. S6** Measured dielectric function of Sample A AlN/GaN SL.
### Table S2 Phonon mode best-fit parameters of ellipsometry data using Eq. (2).

| Sample | Mode | $\varepsilon_{\infty}$ | $\omega_{TO}$ [cm$^{-1}$] | $\omega_{LO}$ [cm$^{-1}$] | $\gamma$ |
|--------|------|------------------------|-------------------------|-------------------------|---------|
| $A$    | $(E_1)_1$ | 4.7 | 584.3 | 608.3 | 6.0 |
|        | $(E_1)_2$ | - | 640.2 | 879.0 | 8.3 |
|        | $(A_1)_1$ | 8.3 | 560.0 | 736.8 | 8.6 |
|        | $(A_1)_2$ | - | 818 | 828 | 15 |
|        | $(A_1)_3$ | - | 852 | 863.3 | 10.1 |
| $B$    | $(E_1)_1$ | 4.8 | 572 | 599 | 14.2 |
|        | $(E_1)_2$ | - | 629 | 808 | 18.1 |
|        | $(A_1)_1$ | 5.2 | 552 | 725 | 9.8 |
|        | $(A_1)_2$ | - | 727 | 740 | 15.8 |
|        | $(A_1)_3$ | - | 766 | 798 | 27.9 |
|        | $(A_1)_4$ | - | 829 | 859 | 6.1 |

### S6. Birefringence of Sample B

Fig. S7 Birefringence, $\Delta n = n_e - n_o$, of Sample B AlN/GaN SL.

### S7. Computational results of phonon modes for AlN$_4$/GaN$_5$ SL

Density-functional perturbation theory (DFPT) was used to compute the phonon modes of an infinite AlN/GaN SL in the $[0001]$ direction. The calculation was performed for 4 monolayers and 5 monolayers of AlN and GaN, respectively, as informed by the layer thicknesses for Sample B. The SL was fully relaxed before calculation of the phonon modes. An illustration of the simulation
unit cell is shown in Fig. S8. Because the different constituent layers have opposite parity for the number of monolayers, it was necessary for the simulation unit cell to contain two layers of each material to achieve appropriate bonding at the AlN-GaN interface. Therefore, the unit cell contained \( N = 36 \) atoms, which resulted in \( 3N - 3 = 105 \) phonon modes. Of these modes, 35 are double degenerate E-symmetry modes and 35 are A-symmetry modes.

![Figure S8 Illustration of DFPT simulation unit cell. Blue, orange, and gray spheres denote Ga, Al, and N, respectively.](image)

Table S3 below lists the resultant phonon modes from the DFPT calculation. Modes 1-17 are folded acoustic modes, while modes 18 – 35 (highlighted in gray) are optic phonon modes. The IR dielectric function of the SL can be calculated from these optic phonon modes using Eq. 2. For this calculation, the E-symmetry (A-symmetry) modes were used for \( \varepsilon_\perp (\varepsilon_\parallel) \). Figure 3c-d of the manuscript show the result of this calculation using an arbitrary damping term \( \gamma = 10 \text{ cm}^{-1} \) for all phonon modes. Note that many of the modes do not contribute to the dielectric function
because of negligible TO-LO splitting. Additionally, certain modes have nearly identical TO and LO frequencies, for instance, $\omega_{LO} = 816.40 \text{ cm}^{-1}$ for mode 32 and $\omega_{TO} = 816.42 \text{ cm}^{-1}$ for mode 34, and are not optically distinct modes. As a result, these modes with overlapping TO and LO frequencies generate only a single pole and zero crossing.

As discussed in the main text, $\varepsilon_{\perp}$ exhibits two Reststrahlen band regions: one narrow region around 560 cm$^{-1}$ and another broad region from about 630 cm$^{-1}$ to 800 cm$^{-1}$. While $\varepsilon_{\perp}$ in Fig. 3c-d was calculated from many E-symmetry modes, only two of the modes (mode # 27 and 34 in Table S3) have significant TO-LO splitting, and these modes largely define the Reststrahlen regions. From the atomic displacements associated with these two modes (Fig. S9), it is immediately clear that mode # 27 resembles a GaN-like confined mode, while mode # 34 appears to be mostly an interface phonon in the AlN layers.

**Fig. S9** Vibrational patterns for the E-symmetry modes a) 27 ($\omega_{TO} = 559.96 \text{ cm}^{-1}$, $\omega_{LO} = 582.7 \text{ cm}^{-1}$) and b) 34 ($\omega_{TO} = 615 \text{ cm}^{-1}$, $\omega_{LO} = 784.2 \text{ cm}^{-1}$).
In contrast to $\varepsilon_\perp$, a number of A-symmetry modes have significant TO-LO splitting and contribute to $\varepsilon_\parallel$. Figure S10 shows the vibrational pattern of the 4 A-symmetry modes with the largest TO-LO splitting. Overall, these modes are not tightly confined to one material. The out-of-plane dielectric function in Fig. 3c-d has a broad Reststrahlen region from around 540 – 700 cm$^{-1}$. From Fig. S10, mode #18, which contributes to the permittivity in this region does not appear to be confined to either layer. Within the 740 – 830 cm$^{-1}$, $\varepsilon_\parallel$ exhibits somewhat complex behavior which appears to arise from a number of phonon modes. These modes are weakly localized to the AlN layer.

![Vibrational patterns of A-symmetry modes.](Image)

**Fig. S10** Vibrational patterns of A-symmetry modes.
| Mode # | $\omega_T$ [cm$^{-1}$] | $\omega_L$ [cm$^{-1}$] | $\omega_T$ [cm$^{-1}$] | $\omega_L$ [cm$^{-1}$] |
|--------|-----------------|-----------------|-----------------|-----------------|
| 1      | 16.22           | 16.22           | 47.07           | 47.07           |
| 2      | 48.59           | 48.59           | 63.95           | 63.95           |
| 3      | 61.32           | 61.32           | 112.97          | 113.55          |
| 4      | 68.4            | 68.41           | 126.53          | 126.74          |
| 5      | 94.43           | 94.43           | 181.27          | 181.27          |
| 6      | 97.93           | 97.93           | 183.75          | 183.75          |
| 7      | 115.82          | 115.83          | 226.06          | 226.25          |
| 8      | 121.36          | 121.36          | 242.84          | 243.09          |
| 9      | 133.91          | 133.91          | 271.08          | 271.08          |
| 10     | 140.98          | 140.98          | 291.13          | 291.13          |
| 11     | 146.25          | 146.25          | 306.54          | 306.55          |
| 12     | 159.74          | 159.74          | 320.59          | 320.65          |
| 13     | 160.62          | 160.62          | 327.04          | 327.04          |
| 14     | 196.8           | 196.8           | 407.39          | 408.67          |
| 15     | 197.04          | 197.05          | 408.88          | 408.88          |
| 16     | 225.23          | 225.23          | 490.68          | 491.17          |
| 17     | 225.25          | 225.25          | 491.15          | 491.15          |

Table S3: Summary of the transverse ($\omega_T$) and longitudinal ($\omega_L$) phonon frequencies in AlN/GaN.
S8. XH polaritonic response

The theoretical reflectivity spectra in Fig. 4 of the main text are calculated using a 4 x 4 transfer matrix formalism. The incident medium is set to be KRS5 (n=2.4), thus providing the same optical response as obtained experimentally in the Otto geometry. The formalism allows for the calculation of the reflection and transmission coefficients at arbitrary incidence angles for any number of material layers, each described by either an isotropic or anisotropic dielectric tensor. For the simulation labeled ‘XH’ in Fig. 4 of the main text, the formalism was applied on a system of 4 layers (KRS5 / air (2.8 μm) / XH (585 nm) / 6H-SiC), giving rise to an excellent agreement with the experimental data. Here, the dielectric function extracted from ellipsometry shown in Fig. 3 of the main text was used in its parametrized form (Eq. 2) using the values reported in Table S2 to describe the XH layer. The spectra labeled ‘Macroscopic’ that fail to reproduce the experimental results were calculated accounting for each AlN and GaN layer individually, i.e. for a system of 503 layers (KRS5 / air (2.8 μm) / 250x (AlN (1.2 nm) / GaN (1.4 nm)) / 6H-SiC). Here, the AlN and GaN layers were described by their bulk dielectric functions using the parameters given in Table S1. All calculations explicitly account for the anisotropy of the dielectric response for the polar materials, i.e., SiC, AlN, GaN, XH.

To understand the nature of the different polariton modes observed as dips in the XH Otto geometry reflectivity data shown in Fig. 4 and as peaks in the Im(r_p) plots in Fig. 5 of the main text, one needs to consider multiple effects:

(i) Multimode: the XH is a multimode material, i.e. it can support surface polaritons in multiple branches for each TO-LO phonon pair and the respective Reststrahlen region.

(ii) Thin film: In the experiment, we use a 585 nm thick film of the XH material, which is more than 10 times smaller than the wavelengths employed and also on the length scale
of the absorption length on the strongest phonon resonances. Therefore, optical thin film effects are heavily affecting the optical response. Also, volume-confined hyperbolic modes only emerge in thin films.\(^6\)

(iii) Substrate: Since SiC can also support a surface polariton, and its dielectric function is strongly dispersive in the frequency range of interest, the substrate also strongly affects the polariton response of the XH thin film. Notably, the film thickness employed here is well beyond the strong coupling regime observed in previous work with AlN ultrathin films.\(^7\)

Qualitative understanding of these effects is gained by inspecting Fig. S11, where we show theoretical polariton dispersions of four different samples: a) a thick semi-infinite XH crystal/air interface, b) a thin slab of the XH interfaced with air on both sides and c) a thin slab of the XH on a SiC substrate interface to air, and d) a thick semi-infinite SiC/air interface. As in Fig. 5 a-b) of the main text, these plots show the imaginary part of the reflection coefficient as a function of in-plane momentum, which conveniently reveals all polariton modes.\(^6\) Calculations were performed with the same transfer matrix formalism as the reflectance calculations in Fig. 4 of the main text, but without explicitly treating the prism. Additionally, we also extracted the dispersion of all modes by tracing the peaks in similar calculations for the XH with reduced damping, \(i.e.\) by using Eq. 2 with parameters in Table S2, but with all phonon dampings \(\gamma\) divided by 5.

The mode assignment given in the main text is supported by analyzing the XH slab-thickness dependence of the polariton dispersion shown in Fig. S12, where the mode progression of the volume-confined hyperbolic modes\(^6,8\) is directly visible.
Figure S11 Polariton dispersion for a thick crystal a) of the XH and d) of SiC, as well as a 585 nm-thin XH slab that is b) free-standing and c) on a SiC substrate, all interfaced to air. For all dispersions, we plot the imaginary part of the reflection coefficient for p-polarized excitation. Notably, only the thin XH slabs exhibit dispersing modes for large momenta \((k/k_0 > 5)\). Thin film effects can be traced by comparing a) and b), most strikingly the emergence of hyperbolic, volume-confined modes. Additionally, surface phonon polariton modes are split into symmetric and anti-symmetric thin-film modes, resulting in strong compression of the dispersion. Adding the SiC substrate in c) leads to additional modifications of the dispersion. Most notably, the strong SiC surface phonon polariton shown in d) results in a mode repulsion/compression of the high-frequency (~840 cm\(^{-1}\)) XH polariton. Above the XH Reststrahlen region (wavenumbers > 860 cm\(^{-1}\)) where all components of the XH dielectric function are positive, the XH acts as an effective (but dispersive) index-shifter for the SiC polariton. Notably, the dispersion of the hyperbolic modes is only mildly affected by the SiC substrate, apart from the type-II hyperbolic band (740-770 cm\(^{-1}\)) which only emerges with the SiC substrate.
Figure S12 XH slab-thickness dependent polariton dispersion. Large momentum modes \((k/k_0 > 1.5)\) are analyzed for a) 200 nm, b) 400 nm, c) 600 nm, and d) 800 nm thick slabs of the XH on a SiC substrate. The typical mode progression of the hyperbolic modes\(^6,8\) proves their volume-confined nature. We did not detect any of the weak type-II hyperbolic modes (740-770 cm\(^{-1}\)) beyond \(m=1\) mode number\(^8\). Notably, also the elliptical surface modes show a pronounced amplitude behavior and dispersion modification with the XH slab thickness.
Figure S13 Optical field profiles for localized excitation of hyperbolic and elliptical phonon polaritons in a 585 nm thick XH on SiC. The wavenumber as well as the in-plane and out-of-plane dielectric function values are given in each subplot, marking the expected nature of the mode, i.e. hyperbolic or elliptical. The calculations were performed as described in the Methods section of the main text. All plots are scaled to the field maximum of the polariton modes inside the XH.
**Figure S14** Theoretical XH-thickness dependent reflectance of the XH/SiC structure, calculated with the transfer matrix approach as in Fig. 4 of the main text, but without the prism. The angle of incidence was set to 60°.
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