Direct observation of ultraslow hyperbolic polariton propagation with negative phase velocity

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Polaritons with hyperbolic dispersion are key to many emerging photonic technologies, including sub-diffraction imaging, sensing and spontaneous emission engineering[1–8]. Fundamental to their effective application are the polaritons' lifetimes, as well as their phase and group velocities[7,9]. Here, we combine time-domain interferometry[10] and scattering-type near-field microscopy[11] to visualize the propagation of hyperbolic polaritons in space and time, allowing for the first direct measurement of all these quantities. Particularly, we study infrared phonon polaritons in a thin hexagonal boron nitride[8,12,13] waveguide, exhibiting hyperbolic dispersion and deep subwavelength-scale field confinement. Our results reveal - in a natural material - negative phase velocity paired with remarkably slow group velocity of 0.002c, and lifetimes in the picosecond range. While these findings show the polariton's potential for mediating strong light-matter interactions and negative refraction, our imaging technique paves the way to explicit nanoimaging of polariton propagation characteristics in other 2D materials, metamaterials, and waveguides.

Materials with hyperbolic dispersion[1–8] are characterized by an anisotropic permittivity (or permeability) tensor, $\boldsymbol{\varepsilon}$, where one of the principle components (axial component, indexed z in the following) is opposite in sign to the other principle components (transverse components, indexed x and y). Electromagnetic waves in hyperbolic materials (called hyperbolic polaritons, HPs) thus propagate with wavevector $\boldsymbol{k}$ described by:

$$k_z^2/\epsilon_x + \left(k_x^2 + k_y^2\right)/\epsilon_z = (\omega/c)^2 \qquad (1)$$

where $\varepsilon_x = \varepsilon_y$. The solutions of Eq. 1 are open hyperboloids of either type I (Re $\varepsilon_z < 0$, Re $\varepsilon_x > 0$) or type II (Re $\varepsilon_z > 0$, Re $\varepsilon_x < 0$). Owing to the possibility of large k-values, HPs offer many exciting applications, such as hyperlensing[3] and extreme light confinement[8,12,14].

Intriguingly, hyperbolic dispersion leads to exotic behavior in the alignment of the velocity and energy-flow vectors; the Poynting vector $\boldsymbol{S}$ is orthogonal to the phase velocity $\boldsymbol{v}_p$ ($\boldsymbol{v}_p$ parallel to $\boldsymbol{k}$), and both $\boldsymbol{S}$ and the group velocity $\boldsymbol{v}_g = \partial\omega/\partial\boldsymbol{k}$ have a fixed angle relative to the z-axis ($\boldsymbol{v}_g$ parallel to $\boldsymbol{S}$). As a result, $\boldsymbol{v}_p$ and $\boldsymbol{v}_g$ can have opposite signs with respect to individual axes (i.e. the x or z directions), leading to a rich variety of optical phenomena including negative refraction and backward waves[15]. To fully understand, exploit and verify the large potential of propagating HPs, research and development will require tools for comprehensive experimental characterization of their various properties. Vitally important among these are the signs of the group and phase velocities relative to the energy flow (decisive, for example, in distinguishing between slow and superluminal light[16,17]), as well as dispersion and lifetimes. The latter determines the performance of HP applications, which can be difficult to predict theoretically as they often depend on device fabrication quality.

The values and signs of a light pulse's $\boldsymbol{v}_p$ and $\boldsymbol{v}_g$ can be measured by time-domain or spectral interferometry[10,16,18,19]. This technique has been combined with aperture-type scanning near-field optical microscopy (a-SNOM) to study surface plasmon polaritons and light in photonic crystals[15,20,21]. The use of apertures, however, limits the spatial resolution and restricts applications to the visible and near-infrared spectral

ranges. In contrast, scattering-type scanning near-field optical microscopy (s-SNOM), where light scattered at the tip of an atomic force microscope is recorded, achieves 10 nm spatial resolution at visible, mid-infrared and terahertz frequencies[11,22]. This spectral freedom – combined with its unprecedented spatial resolution – has allowed s-SNOM to image, for example, graphene plasmons at mid-infrared frequencies[23,24]. Method-intrinsic interference phenomena, however, generally make image analysis challenging, and direct measurements of pulse group velocities and lifetimes by s-SNOM have been elusive so far.

Here, we describe a technique that combines (i) s-SNOM, (ii) time-domain interferometry and (iii) near-field data processing to map in space and time the dispersion, lifetimes and both the value and sign of the phase and group velocities of deep subwavelength-scale polariton pulses. We demonstrate our technique's potential by mapping phonon polaritons propagating in a thin hexagonal Boron Nitride (h-BN) slab. Owing to its layered crystal structure, h-BN is a natural material in which phonon polaritons exhibiting hyperbolic dispersion exist. Type I HPs are found in the spectral region between 760 – 825 cm^{-1} (HPI) and type II HPs between 1370 – 1610 cm^{-1} (HPII). The existence of h-BN HPs has only recently been verified[8,12,13,25–27]. Strong field confinement, ultrashort wavelengths compared to the illumination wavelength, and extremely low losses have been found, making HPs in boron nitride promising candidates for enhanced molecular spectroscopy, tailored thermal emission[28], flatland optics[29] and super-resolution imaging[3,26,27].

We employ s-SNOM (i) to image h-BN polaritons launched at the edge of a semi-infinite gold film deposited on top of a h-BN layer, in contrast to previous work where the s-SNOM probe was used for both launching and imaging of the HPs[12,13]. We thus first describe the basic details of our technique (Fig. 1, see also Methods) and its advantages. The gold film serves as a broadband antenna for converting the incident p-polarized field E_{in} into strongly confined near fields at the gold edge, providing the necessary momentum for launching HPs in the h-BN slab. The numerical simulation in Fig. 1b (monochromatic illumination) shows the edge-launched HP rays reflecting from the top and bottom surfaces of the h-BN slab, forming a guided mode that can be described as a superposition of eigenmodes Mn with n = 0,1,2… (Fig. 1c), each described by wavevector $K_x = k_x + i\gamma$. Although the

high-order modes have a higher Q factor[8], they travel a shorter distance, essentially due to their dramatically shorter wavelength (see Fig. 3). As a result, only the fundamental mode HP-M0 remains to propagate over several micrometres. The HP-M0 fields, $E_{\text{HP-M0}}$, interfere with E_{in}, yielding interference fringes above the h-BN slab that are parallel to the gold edge. This pattern is mapped by recording the field scattered by the metal tip of our s-SNOM, E_{sca}, while the sample is scanned (Fig. 1d). The image also shows weaker and more tightly spaced fringes parallel to the edge of the h-BN slab. They result from the interference of radially propagating HPs that are launched by the tip and reflected at the h-BN edge[12,25] (Supplementary Sect. A). In the following, we image and study HPs launched at the gold edge, as they offer the dual advantages of stronger signals and plane-wave propagation. Note that the decay of plane wave HPs directly reveals the imaginary part, γ, of the wavevector, while the tip-launched HPs decay faster (clearly seen by the fringes in Fig. 1d) due to their radial propagation (Supplementary Sect. A).

To study the polariton propagation in space and time, we apply time-domain interferometry (ii) as illustrated in Fig. 2a. HP pulses were launched at the metal edge by mid-infrared laser pulses of about 100 fs duration which were chosen to cover either the HPI or the HPII spectral regions (see Methods). The tip-scattered light E_{sca} is collected and superimposed at the detector with reference pulses E_{ref}, i.e. the cross-correlation of E_{sca} and E_{ref} is detected. We repeatedly scanned perpendicularly across the gold edge for increasing time delay τ between the tip-scattered field and the reference field, yielding a space-time near-field map with a characteristic fringe pattern. The background-subtracted space-time near-field map $I'_{\text{det}}(x,\tau)$ (Supplementary Sect. B) recorded in the HPII spectral region is shown in Fig. 2c. Close to $\tau = 0$ ps, we see fringes (marked E_{dir}) between $x = 0$ and 14 μm, caused by the interference of the direct tip-scattering of the incident field, E_{dir}, with the reference beam, E_{ref} (illustrated in Fig. 2b, upper panel). We also observe fringes (marked E_{HP}) moving away from the gold edge with increasing τ, which we assign to the tip-scattered propagating HP field, E_{HP} (illustrated in Fig. 2b, lower panel).

To verify these fringes as polaritons and analyze their properties, we first perform a 2D-Fourier Transform (FT) to obtain a data set in the wavevector-frequency (k_x, ω) domain (Fig. 2d and Supplementary Sect. C). We immediately recognize a polariton-

like dispersion, which can be assigned to the fundamental mode M0 (see Fig. 3a). Analogously, we mapped and analyzed the HPI spectral region (Supplementary Sect. B), and display its (k_x, ω) map in Fig. 2f where the fundamental HPI mode is also marked. Intriguingly, we observe positive k_x values for the HPII modes, but negative k_x values for the HPI modes (positive x-direction points away from the gold edge and thus in the same direction as the Poynting vector $\boldsymbol{S}$). Further, we find a positive dispersion (which we define as $\partial\omega/\partial|k_x| > 0$) for the HPII-M0 mode, and a negative dispersion ($\partial\omega/\partial|k_x| < 0$) for the HPI-M0 mode. Both the sign and the slope of $\omega(k_x)$ provide direct and unambiguous experimental evidence that the HPII mode propagates with both positive phase and group velocity, while the HPI mode propagates with negative phase but positive group velocity.

We explain our observations by illustrating the propagation of the HP rays (blue and red arrows of Fig. 2e,g representing $\boldsymbol{S}$, see also Fig. 1b) in the h-BN slab, which is described by the solutions of Eq. 1 (blue and red isofrequency curves). For type II HPs, the wavevector $\boldsymbol{k}$ (black arrows in Fig. 2e) is perpendicular to $\boldsymbol{S}$ (blue arrows) and exhibits a positive component $k_x > 0$. With increasing frequency ω, the Poynting vector $\boldsymbol{S}$ rotates towards the z-axis, resulting in a shorter periodicity of the HP rays, as indicated by the grey arrows. Consequently, k_x of the HPII-Mn modes increases with frequency ω, yielding positive dispersion. Type I HPs (Fig. 2g), in contrast, exhibit a negative wavevector component $k_x < 0$, while $\boldsymbol{S}$ still points away from the gold edge (because of causality). The Poynting vector $\boldsymbol{S}$ rotates away from the z-axis for increasing ω, resulting in larger periodicity of the HP rays. Thus, k_x of the HPI-Mn modes decreases, yielding a negative dispersion.

Negative dispersion of h-BN HPs can also be inferred by far-field spectroscopy of h-BN resonators[8] and nanoimaging experiments[26,27]. However, its interpretation requires knowledge of the sign of k_x relative to the Poynting vector. Indeed, without considering the sign of k_x, one could even interpret the negative dispersion of the HPI modes as a negative group velocity $\boldsymbol{v}_g$. Negative $\boldsymbol{v}_g$, however, would imply a backward propagating HP pulse (i.e. opposite to the Poynting vector $\boldsymbol{S}$), and subsequently superluminal propagation[7,16,30]. While such behavior can indeed be found in metamaterials[16], it can be excluded for HPs propagating along an h-BN slab, owing to our direct experimental measurement of the sign of group and phase velocities. We

note that Ref. 8 predicts negative values of $\boldsymbol{v}_g$: the sign of $\boldsymbol{v}_g$ in this case is referenced to the coordinate system of the sample, while in our work the signs are referenced to the direction of $\boldsymbol{S}$. As can be seen in Fig. 1 of Ref. 8, however, $\boldsymbol{v}_g$ and $\boldsymbol{S}$ are always parallel and thus $\boldsymbol{v}_g$ is positive with respect to $\mathbf{S}$, which is consistent with our experimental results.

Figures 2d and f also show faint lines next to the fundamental modes HPII-M0 and HPI-M0 (marked with white arrows). They could indicate higher-order HP modes or backreflection of tip-launched modes at the gold edge. In the HPII region (Fig. 2d), the large mismatch between the faint lines and the expected dispersion of higher order HPII modes (see dashed lines in Fig. 3a) indicates that the latter scenario is observed. For that reason, we consider in our following analysis only the HPI-M0 and HPII-M0 modes.

For a quantitative analysis of the polariton propagation, we isolate the HPI-M0 and HPII-M0 modes by spatial filtering (iii) of the data in Figs. 2d,f and correct for the illumination angle (Supplementary Sect. D). The results are shown in Figs. 3a,d, where we also plot the calculated dispersion. We find an excellent agreement between experiment and theory for the fundamental modes M0. By displaying the data in the space-frequency domain (x,ω) we can see the evolution of the polariton fields with increasing distance x from the edge. In both bands we find an exponential decay of the amplitude signal, while the phase linearly increases (decreases) in the HPII (HPI) region (Fig. 3b,e). When representative frequencies are plotted in the complex plane, we observe an anticlockwise rotating spiral in the HPII region (blue line), while it is clockwise in the HPI region (red line), corroborating the positive and negative phase velocities in these regions respectively. We also find that HPII-M0 (HPI-M0) modes exhibit the largest propagation length close to TO (LO) frequencies, which essentially stems from the mode wavelengths being longest at these frequencies. Theory confirms this observation, which can be appreciated by comparing the calculated (white lines) and experimental (blue and red lines respectively) propagation lengths (defined as $L = 1/\gamma$) in Figs. 3b and e, reaching up to 6 μm (1.5 μm) in the upper (lower) HP band (for further details see Supplementary Sect. E).

Finally, by displaying the filtered data from Figs. 3a,d in the space-time domain (Figs. 4b,e) we can visualize the HP pulse propagation and directly measure the lifetimes and group velocities. In contrast with Fig. 2c, we now clearly observe the fringes moving away from the metal edge with increasing time delay τ. They exhibit falling (rising) slopes (see inset, Fig. 4b,e), signifying a positive (negative) fringe velocity v_f (Fig. 4c,f), again confirming the positive (negative) phase velocity v_p of the HPII-M0 (HPI-M0) mode. At distances between x_1 and x_2 we measure the *positive* envelope velocities $v_{e,\text{HPII-M0}} = \Delta x/\Delta\tau = +0.027c$ and $v_{e,\text{HPI-M0}} = +0.002c$ (Figs. 4a,d). These numbers match the calculated group velocities ($v_g = \partial\omega/\partial k_x$) of the HPII-M0 and HPI-M0 modes at frequencies of ω = 1440 cm^{-1} and 802 cm^{-1}, respectively, as obtained from the theoretical dispersion curves in Figs. 3a,d. These frequencies lie rather centrally within their respective Reststrahlen bands, as the envelope velocity reveals the average group velocity[10,18–20] of the broadband HP pulse. Our measurements thus corroborate experimentally the slow propagation of HPs – strikingly being two orders of magnitude smaller than plasmons on gold nanowires[18,19] and comparable to photonic crystal waveguides[21] – which may open exciting new possibilities for strong light-matter coupling in the mid-infrared spectral range, for example in sensing applications.

The space-time maps in Fig. 4 also reveal the impressive lifetimes (field amplitude decay) of the fundamental HP pulses to be in the picosecond range. By fitting the envelope maximum as a function of time delay τ to an exponential function, we obtain the 1/e decay times $t_{\text{HPII-M0}} \approx 0.8$ ps and $t_{\text{HPI-M0}} \approx 1.8$ ps. These experimental values are in good agreement with the calculated HP-M0 lifetimes within an accuracy of 20% (Supplementary Sect. E,F). Notably, these lifetimes are about one order of magnitude larger than the lifetimes of gold plasmon polaritons at visible frequencies. Our measurements also confirm that the pulses live longer for type I HPs[31]. Without the lossy SiO_2 substrate, we expect lifetimes of around 5 ps (Supplementary Sect. E).

We conclude that the extraordinary properties of h-BN HPs promise interesting potential for sub-diffraction imaging[26,27], sensing or slow light applications, while our imaging technique could be applied for studying exotic photonic properties in various 2D materials and metamaterials that have so far not been accessible experimentally. In combination with electro-optic detection and pump-probe spectroscopy[32,33] we

envision further exciting possibilities such as the precise mapping of length and shape of polariton pulses and the study of nonlinear properties.

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Author Contributions:

E.Y., M.S. and R.H. conceived the study. E.Y. and M.S. performed the experiments. E.Y., M.S. and R.H. analyzed the data and discussed the results. O.T. fabricated the sample. M.S., A.W. M.B.L. and A.N. carried out simulations. E.Y., M.S. and R.H. wrote the manuscript. F.C., L.E.H., F.K, and R.H. supervised the work and discussed the manuscript. All authors contributed to the scientific discussion and manuscript revisions.

Competing financial interests:

R.H. is co-founder of Neaspec GmbH, a company producing scattering-type scanning near-field optical microscope systems such as the one used in this study. All other authors declare no competing financial interests.

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Figure 1: Near-field imaging of metal-edge-launched HPs on a 135 nm thick h-BN layer on SiO_2 (a) Illustration of our experiment. The black, horizontal lines of the h-BN slab represent the orientation of the material's layers. (b) Numerical simulation of the field distribution (log scale) of type II HPs propagating in an h-BN layer (ω = 1563cm^{-1}). (c) Field intensity profiles of the M0,1,2 HP eigenmodes of the h-BN slab. (d) Left: Topography of a 55 nm thick gold film on a 135 nm thick h-BN slab on a SiO_2 substrate. Right: Infrared near-field image, revealing h-BN HPs launched at the gold edge, as well as h-BN HPs launched by the tip and reflected by the h-BN edge (see Supplementary Sect. A).

Figure 2: Time-domain interferometry of broadband HP pulses. (a) Experimental setup. (b) Tip-scattering of the incident field (top) and of the propagating HP field launched by the gold edge (bottom). (c) Background-subtracted space-time map of E_{sca} for the HPII spectral range. (d) 2D FT of (c), revealing the dispersion of the HPII fundamental mode (M0). Transverse (longitudinal) optical phonon frequencies are marked TO (LO). (e) Top: HPII rays (blue arrows) within the h-BN slab. Bottom: Isofrequency curve solutions for Eq. 1 at 1520 cm^{-1} (solid blue) and 1580 cm^{-1} (dashed blue). (f) 2D-FT of a space-time map for HPI range (shown in Fig. S1), revealing its fundamental mode (HPI-M0). (g) Isofrequency curve solutions for Eq. 1 at 777 cm^{-1} (solid red curve) and 800 cm^{-1} (dashed red). Note that in (g) the x-component of the wavevector, k_x, points towards the gold edge, corresponding to a negative phase velocity.

Figure 3: Dispersion and real-space maps of the isolated HP-M0 modes. (a,d) Filtered dispersion of HPII-M0 and HPI-M0 modes (Supplementary Sect. D). White solid lines show the calculated dispersion obtained by the transfer matrix method. White dashed lines show calculated higher order modes. (b,e) Amplitude and phase maps of the tip-scattered HP fields obtained by FT of (a,d). Blue and red solid lines mark the decay length obtained by fitting exponentially decaying plane waves $C\exp(iK_x x)$, where C is a complex valued constant and K_x is the complex-valued wavevector $K_x = k_x + i\gamma$. White solid lines mark the theoretical propagation length L (Supplementary Sect. E). (c,f) Amplitude and phase profiles (black dots) extracted

from Figs. 3b,e at ω = 1500 cm^{-1} and at ω = 782 cm^{-1}, displayed as a function of distance x (left panels) and in the complex plane (right panels). Blue and red solid lines show fitting with exponentially decaying plane waves.

Figure 4: Simultaneous real-space mapping of fringe and envelope velocities, v_f and v_e, of (a-c) HPII-M0 and (d-f) HPI-M0 modes. The maps in the center (b,e) show the real part of the inverse FT of the filtered HP dispersion of Figs. 3a and d into the space-time domain, $I_{\mathrm{filt}}(x,\tau)$. To the left (a,d) and right (c,f) of the maps we show line profiles (green solid lines) for different distances x and time delays τ, respectively. The black curves represent the envelope of the fringe patterns. The insets in the upper right corners show zooms into the fringe patterns (exaggerated aspect ratio), clearly revealing their falling/rising natures, corresponding to positive/negative phase velocities. Note that the launching of HPs only within the Reststrahlen band frequencies acts like a spectral filter for the broadband incident pulses (see Methods 4). As a result, the time domain signal shows ringing, manifesting as small additional maxima at larger τ in (a) and (d).

Methods

1. Numerical Simulations

The near-field field distributions in Fig. 1b and the mode profiles in Fig. 1c were calculated by finite-difference time domain (FDTD) and finite-difference frequency domain (FDFD) simulations respectively, using a commercial software package (Lumerical Solutions). The theoretical dispersion curves in Fig. 3a,d and propagation lengths in Fig. 3b,e were found using the transfer matrix method. The values of the h-BN permittivity tensor $\boldsymbol{\varepsilon}$ were taken from Caldwell et al.[8] and the SiO_2 permittivity from Gunde[34].

2. Sample Preparation

The h-BN flake was prepared by mechanical exfoliation with blue Nitto tape and transferred onto a Si/SiO_2 (250nm) chip using polydimethylsiloxane[35]. The gold edge on top of the flake was fabricated as follows: first, high-resolution electron-beam lithography was performed on an electron-sensitive bilayer polymethyl methacrylate resist with 2 nm of gold on top; the thin gold layer was used to avoid charging of the sample during the electron-beam exposure. Second, the gold layer was removed by an etchant, and the resist was developed in methyl isobutyl ketone:isopropanol 1:3. Third, 5 nm of titanium (adhesion layer) and 50 nm of gold were deposited by electron-beam evaporation in an ultra-high vacuum chamber. Finally, the resist was removed by lift-off in acetone and the sample was rinsed with isopropanol.

3. Single Wavelength s-SNOM Imaging

Our commercially available s-SNOM (*Neaspec, Munich)* is based on an atomic force microscope (AFM). The vertically oscillating tip (Ω = 250 kHz, *NCSTAu, Nanosensors, Switzerland*) acts as a scattering near-field probe. For the images shown in Fig. 1d, p-polarised infrared light from a tuneable continuous-wave (CW) quantum cascade laser (ω = 1563cm^{-1}, *CW-PLS Laser, Daylight Solutions, USA*) was focused via a parabolic mirror onto both the tip and sample at an angle of 60 degrees to the surface normal. The tip-scattered light of field E_{sca} was recorded with a pseudo-heterodyne interferometer[36]. To suppress the background scattering from the tip shaft and sample, the detector signal is demodulated at a frequency 3Ω. With this

technique, we record both the amplitude and phase of E_{sca}. Fig. 1d shows the amplitude of E_{sca}.

4. Time-Domain Interferometry

For time-domain interferometry, we use the s-SNOM setup described above. We illuminate the sample and the oscillating AFM tip with broadband laser pulses of around 100 fs duration covering 1200 – 1700 cm^{-1} for the HPII spectral region and 700 – 1000 cm^{-1} for the HPI spectral region. The broadband laser pulses are generated by a difference frequency generator (DFG, lasnix.com), where two near-infrared, 100-fs pulse trains from a fiber-laser system (FemtoFiber pro IR and SCIR, toptica.com) are superimposed in a GaSe crystal[37]. The oscillating reference mirror of the pseudo-heterodyne interferometer is replaced by a piezo-controlled linearly moving mirror to precisely control the time delay τ between the tip-scattered field, E_{sca}, and the reference pulse, E_{ref} (Fig. 2a). We repeated a line scan across the sample's gold edge (see Fig. 1d) at increasing time delays τ to record the demodulated detector signal as a function of both position x and time delay τ (see Supplementary Sect. B), yielding the space-time map $I_{\mathrm{det}}(x,\tau)$ shown in Fig. 2. The detector signal was demodulated at frequency 3Ω to suppress the background scattering from the tip shaft and sample.

Methods References

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