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Broadband mid-infrared plasmon-polaritons in metallic-dielectric interfaces

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Abstract

We report real-space near-field images of mid-infrared (IR) surface plasmon-polariton (SPP) waves in the insulating/metal/insulating (IMI) heterostructure: hexagonal boron nitride/gold/silicon dioxide (hBN/Au/SiO₂). The SPPs are observed in the 750 – 1500 cm⁻¹ (~13.3 – ~6.7 μm) range, feature micrometer-sized wavelengths and propagation lengths \( L_{SPP} \) exceeding 20 μm at room temperature. Comparatively, real-space mapping of SPP waves in the mid-IR has been shown only in graphene, but with nanometer sized-wavelength and \( L_{SPP} \sim 10 \) μm at cryogenic temperatures. Interestingly, we show interference between different polariton types in the IMI since the lower momenta SPPs in the metal surface interfere with higher momenta hyperbolic phonon polaritons (HPhP) in the hBN top layer creating SPP-HPhP overlapped waves. In agreement with theory, we quantify momentum and damping governing the SPP waves. Tunability is discussed upon changing the IMI heterostructure. Our theory predicts SPP group velocities reaching 20 % of the light velocity in vacuum and 0.2 – 0.4 ps lifetimes. We further demonstrate that the SPP waves interact with SiO₂ and hBN phonons in the strong coupling regime. As a general effect of the metal/dielectric interface, the mid-IR SPP waves can be compelling for fast metal-based plasmonics, whilst their ability to strongly couple to phonons can be further explored for enhanced sensing in the mid-IR.
Surface plasmon-polaritons (SPP)\textsuperscript{1–3}, quasi-particles originated from the coupling of photons to collective oscillations of free charge carries, are intrinsically two-dimensional (2D) modes of the interface between a material with metallic character and a dielectric insulator. Such ultimate 2D confinement originates from the fact that SPPs are non-evanescent in the interface plane, but evanescent along the normal direction. Being photon-charge coupled modes, they feature a wave-particle behavior endowing them optical properties from the photon part, permitting guiding and focusing for instance, whilst the charge-inherited character can provide for desirable electronical control. In the ultraviolet-visible to near-infrared (IR) range, such compelling virtues of SPPs have been exploited in waveguiding\textsuperscript{3}, plasmonic circuits\textsuperscript{4,5}, sensing\textsuperscript{2}, lasing\textsuperscript{6–8}, light harvesting\textsuperscript{2} and for quantum information processing\textsuperscript{9}. In the technologically attractive mid-infrared (IR) range, SPPs can access and retrieve information from interface states associated to molecular vibrations, electronic transitions of energy around 50 – 300 meV and room temperature thermodynamical reactions\textsuperscript{10}. But, in analogy to free space waves or propagating optical modes in fibers, the use of SPPs as information carriers is critically dependent on their propagative properties in media supporting mid-IR plasmonic waves.

Despite semiconducting\textsuperscript{11} and metallic\textsuperscript{12} micro- and nanostructures can bear stationary and localized SPPs enabling functionalities as antennas\textsuperscript{13–18} and resonators\textsuperscript{10,14,19–21} in the mid-IR, these media are reported to have high ohmic losses\textsuperscript{11,12,22,23} that hamper the efficiency of propagative modes. Only graphene\textsuperscript{24–27}, to date, was shown to support tunable and high momenta (10\textsuperscript{5} cm\textsuperscript{-1}) mid-IR SPP waves of propagation length reaching 10 \(\mu\)m at cryogenic temperatures\textsuperscript{28}. As a promising alternative, we present here the first real-space, nanoscale resolved imaging/spectroscopy of micrometer-sized wavelength SPP waves in insulator/metal/insulator (IMI) heterostructures with propagation lengths exceeding 20 \(\mu\)m at room temperature in the 750 – 1450 cm\textsuperscript{-1} mid-IR range. By scattering-type scanning near-field optical microscopy (s-SNOM) and synchrotron infrared nanospectroscopy (SINS), which are optical nanoscopies\textsuperscript{29} able to fully characterize polariton waves\textsuperscript{30,31}, we measure the plasmonic waves in two IMIs: air/Au/SiO\textsubscript{2} and hexagonal boron nitride (hBN)/Au/SiO\textsubscript{2}. An antenna-like structure and a groove in the metal layer\textsuperscript{32} are the main couplers of free space light into the observed plasmon waves. The s-SNOM images and SINS spectral linescan provide for a complete data set permitting us to quantify the complex momentum \(q_{\text{SPP}}\) \((q_{\text{SPP}} = q_{\text{SP}} + i\gamma_{\text{SP}}, q_{\text{SP}}\) is propagative momentum part and \(\gamma_{\text{SP}}\), the damping\) for each excitation frequency (\(\omega\)), thus, composing the experimental frequency-momentum dispersion relation, \(\omega = q_{\text{SP}}\cdot\) governing the SPP waves. Overall, \(q_{\text{SP}}\) scales from \(\sim 0.8 \times 10^4\) cm\textsuperscript{-1} at \(\omega = 750\) cm\textsuperscript{-1} to \(\sim 1.6 \times 10^4\) cm\textsuperscript{-1} at \(\omega = 1450\) cm\textsuperscript{-1}. By nanoimaging in resonance with the hBN upper Reststhralen (RS) band, defined in between the transversal optical phonon (\(\omega_{\text{TO}}^{\text{hBN}}\)) at 1365 cm\textsuperscript{-1} and the longitudinal optical phonon (\(\omega_{\text{LO}}^{\text{hBN}}\)) at 1610 cm\textsuperscript{-1} where hBN is a type II hyperbolic medium supporting hyperbolic
phonon-polaritons (HPhP), we observe SPP-HPhP overlapped waves consisting in high momenta HPhPs ($q_{\text{HPhP}} \sim 10^5 \text{ cm}^{-1}$) modulating lower momenta SPPs. In general, type II hyperbolic materials occur when the real in-plane components of the electrical permittivity tensor are negative, $\text{Re}[\varepsilon_{xx}], \text{Re}[\varepsilon_{yy}] < 0$, while the real out-of-plane one is positive, $\text{Re}[\varepsilon_{zz}] > 0$.

To explain the mid-IR SPP modes, we model the IMIs as a multilayer system to calculate the theoretical $-q_{\text{SPP}}$. The theoretical dispersion is given by the solution of the Maxwell’s equation, applying the boundary conditions in which are obtained by the continuity of the fields at the interfaces (see suppl. mat.). These calculations intrinsically account the coupling of the plasmonic modes at each I/M interface yielding a resultant parameter-free $\omega - q_{\text{SPP}}$ in excellent quantitative agreement with the experimental observations. Moreover, the theoretical and the experimental $\omega - q_{\text{SPP}}$ reveal spectral gaps, near the SiO$_2$ and in-plane hBN phonon absorption peaks, where the amplitudes of the SPP waves are considerably attenuated. We attribute such gaps to anti-crossing (AC) regions owing to the interaction of SPPs and phonons of the surrounding media in a strong coupling regime as evaluated from the couple-oscillator modelling. Additionally, the theoretical $\omega - q_{\text{SPP}}$ enables us to predict that the SPP waves travel with an average group velocity of $0.2 \ c$ ($c$ is the light velocity in vacuum) and lifetimes between 0.2 to 0.4 ps.
Figure 1. s-SNOM imaging of SPP and HPhP waves. (a) s-SNOM scheme probing a SPP-HPhP wave propagating from the groove edge (air gap) in an antenna-hBN/Au/ SiO$_2$ heterostructure on the Si substrate. (b) Topography of a similar heterostructure (material thicknesses shown in a) investigated in (c-m). c) s-SNOM third harmonic amplitude ($S_3$) image, with excitation at $\omega = 1470$ cm$^{-1}$. The selected region of the hBN crystal (gray square) is displayed in (d). (e) Experimental profiles (○) extracted from different regions in (d) with the corresponding model-fits (red curves) and $q_{\text{SPP}}$ and $q_{\text{HPhP}}$ values. (f) $S_3$ image at $\omega = 1470$ cm$^{-1}$ of an Au/SiO$_2$ region located on the right of (b) with the corresponding experimental profile, model-fit and $q_{\text{SPP}}$ value (g). Comparative images and analyses, for excitation at $\omega = 1200$ cm$^{-1}$, are depicted in (h-m). Scale bars: 5 $\mu$m.
The sketch in Fig. 1a illustrates a HPhP-SPP wave in a cross-section of an hBN/Au/SiO$_2$ heterostructure, with an Au disk-like antenna atop, probed by s-SNOM. The s-SNOM microscope is an atomic force microscope (AFM) equipped with a suitable optical apparatus for the measurement of optical near-field. Basically, this nanoscopy uses a metalized AFM tip as a high-spatial resolution and high-momenta optical probe to detect optical phenomena belonging to the near-field regime, including SPP and HPhP waves (see methods for details). Fig. 1b presents the AFM topography of such heterostructure composed by a hBN crystal lying on a Au film surface deposited by electron beam onto SiO$_2$/Si surface. Part of the hBN flake is suspended onto a 2.5 µm-width groove made by lithography in the Au film. The Au disk-like antenna resides atop the hBN crystal (see methods for more details on sample construction). In Fig. 1c, the s-SNOM third harmonic amplitude ($S_3$) image at $\omega = 1470 \text{ cm}^{-1}$ on-resonant with in-plane HPhPs of the upper RS band of hBN, exhibits distinguishable near field patterns on hBN/Au/SiO$_2$ and Au/SiO$_2$ stemming from polariton fringes of different spatial periods, namely, SPPs and HPhPs. As seen in Fig. 1d depicting a zoomed area on the hBN/Au/SiO$_2$, we observe the interference between SPP and HPhP waves launched both by the disk-like antenna and by the groove edge. To better comprehend the polaritonic interferences, in Fig. 1e we plot profiles extracted from different sample regions. The profile $P_1'$ (Fig. 1e) permits a good visualization of a SPP-HPhP superposed wave: the SPP is the larger wavelength ($\lambda_{\text{SPP}}$) component and HPhPs, the smaller wavelength ($\lambda_{\text{HPhP}}$) one as will be quantitatively confirmed in the following analysis. One can see that the groove edge is the primary launcher of the SPP-HPhP waves since their wavefronts are characteristically parallel to it. To extract quantitative information from those waves, we model (eq. 1) the resultant polaritonic field ($E$),

$$E = A_{\text{HPhP}}e^{-i(q_{\text{HPhP}}-i\gamma_{\text{HPhP}})x-i\theta} + A_{\text{SPP}}e^{-i(q_{\text{SPP}}-i\gamma_{\text{SPP}})x} + C$$

(Eq. 1),

as the summation over damped plane waves attributed to the HPhP and SPP components and a complex non-propagative background, $C$, assigned to the background of the surrounding media. The damped plane wave is defined by an amplitude $A_\alpha$, a momentum $q_\alpha$ and a damping $\gamma_\alpha$, with $\alpha = \text{SPP or HPhP}$. For generalization, we include the relative phase difference $\theta$ between the waves. These parameters are determined by fitting the eq. 1 to the experimental profiles. It is noteworthy that our model includes only edge- and antenna-launched waves. There is no experimental evidence of tip-launched waves which would introduce half-wavelength contributions from reflected polariton waves as occurring at crystal edges$^{33-35,37-40}$. 


The model-fit to the profile $P_1'$ (Fig. 1e), considering the sum of a HPhP wave and a SPP wave in eq. 1, yields $q_{\text{SPP}} = 1.12 \times 10^4 \text{cm}^{-1}$ and $q_{\text{HPhP}} = 18.8 \times 10^4 \text{cm}^{-1}$. As the wavelength $\lambda = \frac{2\pi}{q}$, we verify that $\lambda_{\text{SPP}} > \lambda_{\text{HPhP}}$ in $P_1'$ as stated above. The model-fit to the profile $P_2'$, extracted from the hBN onto the groove (Fig. 1d), determines $q_{\text{HPhP}} = 6.14 \times 10^4 \text{cm}^{-1}$ and $q_{\text{SPP}} = 1.57 \times 10^4 \text{cm}^{-1}$. Interestingly, the SPP component in $P_2'$, which is extracted from hBN/air without metal at the bottom interface, stems from the plasmonic waves travelling near the groove in the metal/dielectric interface. In this case the SPP optical field reaches the hBN/air region and interfere with the HPhP component propagating in the hBN crystal, albeit with a smaller amplitude than that of the HPhP wave as confirmed from the amplitude ratio $A_{\text{SPP}}/A_{\text{HPhP}} \approx 0.34$. For comparison, the components in $P_1'$ have equivalent amplitude, $A_{\text{SPP}}/A_{\text{HPhP}} \approx 1.1$, as the waves propagate in the media immediately underneath the tip. In agreement with the literature $^{33-35,37-41}$, our analysis finds a larger value of $q_{\text{HPhP}}$ on Au from the $P_1'$ fit than that on air from the $P_2'$ fit. Moreover, using the model described in ref. $^{34}$ for a 60 nm thick hBN, the theoretically predicted values of $q_{\text{HPhP}}$ are $19.6 \times 10^4 \text{cm}^{-1}$ on Au and $7 \times 10^4 \text{cm}^{-1}$ on air, which are reasonably close to the model-determined ones from the $P_1'$ and $P_2'$ experimental profiles, respectively: $18.8 \times 10^4 \text{cm}^{-1}$ on Au and $6.14 \times 10^4 \text{cm}^{-1}$ on air. With a single wave in eq. 1, the model-fit to the profile $P_3'$, acquired on hBN/Au from the antenna in a direction parallel to the groove edge (Fig. 1d), gives $q_{\text{SPP}} = 1.62 \times 10^4 \text{cm}^{-1}$. We support the choices of the number of components used in the model-fits on Fourier Transform numerical analyses of each profile (see sup. mat.). In comparison with $P_1'$, the model-fits to $P_2'$ and $P_3'$ generate more accurate values of $q_{\text{SPP}}$ since their profile lengths accommodate more oscillations of the SPP waves. Fig. 1f depicts the $S_3$ image at $\omega = 1470 \text{cm}^{-1}$ of edge-launched SPP waves in the Au/SiO$_2$ (Fig. 1f inset). The model-fit to the profile $P_4'$ (Fig. 1g) yields $q_{\text{SPP}} = 1.35 \times 10^4 \text{cm}^{-1}$.

In comparison with the just discussed HPhP resonant case (Fig. 1c-e), we present in Fig. 1h-j the $S_3$ image of the same antenna-hBN/Au/SiO$_2$ heterostructure illuminated at $\omega = 1200 \text{cm}^{-1}$, which is off-resonant with HP$_2$s. As highlighted in Fig. 1i,j, solely SP$^3$ waves are unveiled at $\omega = 1200 \text{cm}^{-1}$, requiring the use of one propagative term as confirmed by Fourier Transform analysis (sup. mat.), to fit the profiles $P_1$ to $P_3$ (Fig. 1j) with eq.1. As discussed above, the fits $P_2$ and $P_3$, which have more oscillations, yield more accurate values of $q_{\text{SPP}}$. In Fig. 1l, we observe edge-launched SP$^3$ waves in the Au/SiO$_2$, for $\omega = 1200 \text{cm}^{-1}$, with $q_{\text{SPP}} = 1.14 \times 10^4 \text{cm}^{-1}$ determined from the model-fit to $P_4$ (Fig. 1m).
Table 1. Values of $q_{SPP}$ and $\gamma_{SPP}$ in $10^4 \text{cm}^{-1}$ for the different excitations and sample regions. For hBN/Au/SiO$_2$, it is shown results from the $P_3'$ model-fit at $\omega = 1470 \text{ cm}^{-1}$ and the $P_3$ model-fit at $\omega = 1200 \text{ cm}^{-1}$.

| IMI   | $\omega$ (cm$^{-1}$) | hBN/Au/SiO$_2$ | Au/SiO$_2$ |
|-------|-----------------------|----------------|------------|
|       |                       | ($q_{SPP} \mid \gamma_{SPP}$) | ($q_{SPP} \mid \gamma_{SPP}$) |
|       | 1200                  | 1.2 | 0.12 | 1.14 | 0.16 |
|       | 1470                  | 1.62 | 0.16 | 1.35 | 0.16 |

In Table 1 we compare the model-determined values of $q_{SPP}$ and $\gamma_{SPP}$ for hBN/Au/SiO$_2$ and Au/SiO$_2$ at the two excitation frequencies, for the two frequencies $q_{SPP}$ is larger on hBN than on air whilst $\gamma_{SPP}$ presents small variations upon changing the material. To further confirm these analyses, we inspect the SPP waves in a broader range by a spectral linescan from SINS nanoscopy (Fig. 2). Concisely, SINS employs the highly brilliant and broadband synchrotron IR radiation as the excitation source for a s-SNOM microscope. Thereby, SINS produces the broadband near-field spectrum with the high spatial resolution of s-SNOM via interferometry with an asymmetric Michelson interferometer. The spectral linescan measurement consists of a distance $\times \omega$ map built by plotting SINS spectra acquired along a defined direction$^{30}$. 
Figure 2 – Hybrid waves in hBN/Au/SiO₂ over the 750 – 1450 cm⁻¹ broad range. a) Scheme of SINS nanoscopy. The IR broadband synchrotron beam enters an asymmetric Michelson interferometer formed by a beamsplitter (BS) and a scanning mirror (SM). The interferometry is realized between the back-scattered light from tip-sample (hBN/Au/SiO₂) and the light from SM using a mercury-cadmium-telluride (MCT) detector. b) Normalized second harmonic amplitude ($S_2$) of the SINS spectral linescan on the heterostructure in (a) from the groove edge in the Au film. c) Profiles extracted from (b) along the x-axis with the respective model-fits. Experimental (d) and theoretical (e) $\omega - q_{SPP}$ dispersion. Inset in (d): experimental $\omega - q_{HPhP}$ with the corresponding theoretical prediction (white dashed curve). Inset in (e): imaginary parts ($Im[\varepsilon]$) of the electrical permittivities of the hBN in-plane component ($\varepsilon_{XX}^{hBN}$) and of the SiO₂ ($\varepsilon_{SiO₂}$).

For hBN, $\omega_{TO}^{hBN} = 1365$ cm⁻¹. For SiO₂, $\omega_{TO}^{L} = 805$ cm⁻¹ and $\omega_{TO}^{U} = 1100$ cm⁻¹.

In Fig. 2b, we present the SINS spectral linescan of the hBN/Au/SiO₂ heterostructure illustrated in Fig. 2a. The linescan direction is normal to the groove, the same direction as $P_1'$ in Figure 1d. The linescan data in the hBN upper RS band (1365 - 1610 cm⁻¹) clearly show SPP-HPhP waves, thus, experimentally corroborating our observations from the s-SNOM on-resonant image (Figure 1c-e). Such overlapped waves are model-fitted using eq. 1 with two propagating components similarly to the one employed for the correlate waves in Fig. 1d,e. As an example, we exhibit in Fig. 2c the
profile data of the SPP-HPHP wave at $\omega = 1415$ cm$^{-1}$ and its corresponding model-fit. In the remaining spectral range from 750 to $\sim 1365$ cm$^{-1}$, however, the waves are model-fitted with a single component in eq. 1 due to having only the SPP component. It is worth commenting that in the hBN lower RS band (749 – 816 cm$^{-1}$), out-of-plane HPhPs emerge but with much shorter amplitude than the SPP waves. Thus, the contribution from HPhPs in the fits of the corresponding spectral interval is neglected. Hence, the full data analysis yields the $\omega - q_{SPP}$ throughout the whole 750 – 1450 cm$^{-1}$ probed range. Such experimental dispersion is shown in Fig. 2d from the real part of the Fourier Transform of a damped plane wave, Re$[F(\omega, q)]$ (see supp. mat. for details) using the values of $q_{SPP}$ and $\gamma_{SPP}$ obtained from the fitting procedure. This method allows for presenting simultaneously such key parameters governing the polariton waves. In the inset of Fig. 2d, the same procedure is used to construct the experimental HPhPs dispersion, $\omega - q_{HPhP}$, matching the theoretical prediction (white dashed curve) from the model in ref. 34. To understand the dispersion of the SPP waves, we calculate the dispersion of the IMI heterostructure considering it a multilayer system. In absence of external sources, the problem is reduced to solving the Maxwell’s equations in the metallic layer accounting for the boundary conditions of at all interfaces. We remark that this classical formalism automatically incorporates the coupling of SPP modes of the upper M/I and lower I/M interfaces. For the air/hBN/Au/SiO$_2$ heterostructures studied in Fig1a-e and Fig. 2, the modelling considers a 4-layer system whose theoretical dispersion (see supp. mat. for details) is found from the numerical solution of the determinant

$$\begin{vmatrix}
R_1 e^{-i k_0^z (a+b)} & R_4 e^{i k_0^z (a+b)} & 0 & 0 \\
0 & e^{-i k_0^z a} & 0 & R_4 + R_3 \\
0 & e^{i k_0^z a} & -e^{-i k_0^z a} & -R_3 e^{-i k_0^z a} \\
R_2 e^{-i k_0^z a} & -R_2 e^{i k_0^z a} & -R_3 e^{-i k_0^z a} & R_3 e^{i k_0^z a} \\
& & & \\
\end{vmatrix} = 0 \quad (Eq. 2),$$

with the subindexes’ correspondences $j = 1 \rightarrow$ air, $2 \rightarrow$ hBN, $3 \rightarrow$ Au and $4 \rightarrow$ SiO$_2$. ($j =$ air, hBN, Au and SiO$_2$) assuming a heterostructure identical to the one measured in Fig. 2a. In the case of isotropic materials (air, Au and SiO$_2$), $R_j = \frac{i k_0^z}{\varepsilon_j}$ is defined from the z-axis momentum $k_0^z = \sqrt{\varepsilon_j k_0^2 - q_{SPP}^2}$ and the electrical permittivity $\varepsilon_j$. Owing to the hBN out-of-plane anisotropy$^{42}$, $R_2 = \frac{i k_0^z}{\varepsilon_{hBN}^{xx}}$ and $k_0^z = \sqrt{\varepsilon_{hBN}^{xx} k_0^2 - \frac{\varepsilon_{hBN}^{xx}}{\varepsilon_{hBN}^{zz}} q_{SPP}^2}$ where $\varepsilon_{hBN}^{xx} (= \varepsilon_{hBN}^{yy})$ and $\varepsilon_{hBN}^{zz}$ are the in- and out of plane components of the hBN electrical permittivity tensor. $k_0$ is the momentum of light in free space. The parameters $b$ and $a$ are the hBN and Au thicknesses, respectively. Hereupon, we plot the modeled $\omega - q_{SPP}$ via $Re[F(\omega, q)]$ in Fig. 2e using the values of $q_{SPP}$ and $\gamma_{SPP}$ calculated from eq. 2 as inputs. The theoretical dispersion quantitatively matches the experimental one presented in Fig. 2d. This correspondence validates our interpretation on
the observed mid-IR SPP waves of the IMI heterostructure as the plasmon waves resulting from the modes at each I/M interface.

Moreover, experiment and theory also consistently reveal gaps in the dispersion relation, i.e., regions without SPP modes, near the hBN in-plane and SiO$_2$ phonons. These lattice vibrations correspond to peaks in the imaginary part of the electrical permittivities of these materials (Fig. 2e inset) centered at the $\omega_{TO}$s of each material. In Fig. 2c, we remark that inside such gaps the SPP modes present more attenuated amplitudes (1365 cm$^{-1}$ profile) and even uncharacterized wave shapes ($\omega = 1077$ and 818 cm$^{-1}$ profiles) resulting in overdamped model-fits. We assign these spectral gaps to anti-crossing regions (AC) that rise owing to the coupling of pure SPPs to lattice phonons of the dielectric layers. To evaluate these effect, we use a classical model of two coupled classic harmonic oscillators$^{37,43}$ described by a pair of coupled equations of motion:

$$\begin{cases}
\ddot{x}_{SP}(t) + \Gamma_{SP}\dot{x}_{SP}(t) + \omega_{SP}^2 x_{SP}(t) - \Omega \omega \bar{x}_{PP}(t) = F_{SP}(t) \\
\ddot{x}_{PP}(t) + \Gamma_{PP}\dot{x}_{PP}(t) + \omega_{PP}^2 x_{PP}(t) - \Omega \omega \bar{x}_{SP}(t) = F_{PP}(t)
\end{cases} \tag{3}$$

where ($\omega_{SP}$, $\omega_{PP}$), ($\Gamma_{SP}$, $\Gamma_{PP}$) and ($x_{SP}$, $x_{PP}$) are the resonant frequencies, dampings and displacements of the coupled SPP and phonon-polaritons (PP) modes, respectively. ($F_{SP}$, $F_{PP}$) are the effective driving forces, proportional to the excitation electric field, acting on the two modes. By definition, $\bar{\omega} = (\omega_{SP} + \omega_{PP})/2$. The solution of eq. 3, setting the approximation $F_{SP} = F_{PP} \sim 0$, leads to the upper $\omega^+$ and lower $\omega^-$ eigen-frequencies (eq. 4) of the coupled modes. From that, the coupling strength $\Omega$ is determined by the smallest difference between $\omega^+$ and $\omega^-$. Whether the strict condition $\eta = \frac{2\Omega}{\Gamma_{SP} - \Gamma_{PP}} > 1$ is fulfilled$^{37}$, the system is classified in the strong coupling regime. Following the method adopted in ref. $^{37}$, we determine those coupling parameters by fitting the extinction coefficient $C_{ext}(q_{SP}, \omega)$ in eq. 5 to the experimental iso-momentum spectra extracted from vertical profiles near the AC regions as shown in Fig. 3d-f. By applying this analysis to different iso-momentum spectra in the AC regions, we compute the corresponding $\omega^+$ and $\omega^-$ dispersion branches, which are displayed as red circles in Fig. 3a-c, overlapping the experimental $\omega - q_{SP}$s. As expected, $\omega_{SP}$ and $\omega_{PP}$ branches, which would take place in absence of coupling, show less agreement with the data. From our calculations, we find that the interaction between SPP waves to both SiO$_2$ and in-plane hBN phonons happens in the strong coupling regime.
\[ \omega^\pm = \bar{\omega} \pm \frac{1}{2} \text{Re} \left[ \sqrt{\Omega^2 + \left[ \omega_{SP} - \omega_{PP} + i \left( \frac{\Gamma_{SP}^2}{2} - \frac{\Gamma_{PP}^2}{2} \right) \right]^2} \right] \] (4)

\[ C_{\text{ext}}(q_{SP}, \omega) \propto \langle F_{PP} \cdot \dot{x}_{PP} + F_{SP} \cdot \dot{x}_{SP} \rangle \] (5)

**Figure 3. SPP-phonons strong coupling.** Color maps shown in a)-c) are the experimental \( \omega - q_{SP} \) cropped from Fig. 2d featuring the AC regions near SiO\(_2\) phonon resonances (a and b) and the in-plane optical phonon of hBN (c). In correspondence, d-f show the fits of eq. 4 to iso-momentum spectra (vertical dashed lines in a), for two different values of \( q_{SP} \). The fitting resulting dispersions for \( \omega^+ / \omega^- \) (white/yellow dots), \( \omega_{SP} \) (blue dots) and \( \omega_{PP} \) (red dots) are plotted for all analyzed cases.
In the mid-IR, SPP-phonon coupling has already been discussed but from the spectroscopic point of view by the use of far-field techniques lacking spatial resolution for imaging the plasmonic waves. In this context, it was recently reported the ultra-strong coupling between epsilon-near-zero (ENZ) SPP modes of coaxial Au nanocavities and SiO$_2$ phonons. It was also recently observed the strong coupling between propagating HPhP waves and molecular resonances for a hBN crystals lying onto a thin organic film. In the latter case, the strong coupling was characterized by ACs in the HPhP dispersion relation in the upper RS band wherein the HPhP waves presented increased damping for the frequencies coincident with the molecular phonons of the film. Such effects are precisely analogous to those we have describe for the SPP-phonon strong couplings. The notable difference is that the presented plasmon waves can couple to phonons in a broader range spanning over considerable portion of the molecular fingerprint.

![Figure 4](image)

**Figure 4** – Theoretical dispersions relations of the group velocity ($v_{g,SPP}$) and lifetime ($\tau$) of the SPP waves in a hBN/Au/SiO$_2$ heterostructure identical to that in Fig. 2.

Furthermore, in Fig. 4 we also examine the photonic properties of the SPP wave found in the group velocity, $v_{g,SPP}$ ($v_g = \frac{d\omega}{dq}$), and lifetime $\tau$, from the theoretical $\omega - q_{SPP}$ (Fig. 2d). Apart from the AC spectral regions, where $v_{g,SPP}$ tends to zero, one can see that the SPP waves reach up to $0.2c$ ($c$ is the light velocity in vacuum) which is one order of magnitude higher than that of graphene plasmons, and have $\tau$ spanning from 0.02 to 0.1 ps in the most part of the spectral range. The $v_{g,SPP}$ in the upper RS band of hBN assumes values about three orders of magnitude greater than that of HPhPs ($v_{g,HPhP}$). Thus, as $v_{g,SPP} \gg v_{g,HPhP}$, the SPP waves can also be used to further explore exotic phenomena as Cherenkov HPhP wakes reported from plasmon modes confined in silver nanowires residing on hBN and the $v_{g,HPhP}$ variation as a function of the distance between hBN and Au in hBN/SiO$_2$(wedge)/Au heterostructures.
Moreover, a broader perspective on the mid-IR SPP waves’ photonics is given from their quality factor $Q_{SPP} = \frac{q_{SPP}}{\gamma_{SPP}}$ spanning in the 4-13 interval in the 750-1450 cm$^{-1}$ range. These values of $Q_{SPP}$ are comparable to those reported for different plasmonic media studied at room temperature: $Q = 40$ for SPPs in graphene monolayers$^{24,25,47}$, $Q = 10$ for SPPs in graphene edges$^{48,49}$, $Q = 26$ for one-dimensional SPP in carbon nanotubes$^{50}$, and $Q = 3$ for Dirac plasmons in topological insulators$^{51,52}$.

In summary, we here report mid-IR SPP waves in IMI heterostructures with $L_{SPP}$ reaching 20 $\mu$m at room temperature. Using s-SNOM and SINS nanoscopies, we show that these plasmon waves exist throughout the broad mid-IR range from 750 to 1450 cm$^{-1}$. As confirmed from theoretical modelling in quantitative agreement with the experimental observations, the mid-IR SPP waves are the resultant waves from the interference between the plasmonic modes at each I/M interface. By analyzing hBN/Au/SiO$_2$ and air/Au/SiO$_2$, we demonstrated momenta tunability by changing the IMI heterostructure. Interestingly, the scan on hBN/Au/SiO$_2$ in resonance with the hBN upper RS band unveils the interference between SPP waves on the metal layer and HPhP waves in the hBN crystal, a SPP-HPhP superposed wave. In graphene-hBN$^{53,54}$, the momenta matching of graphene plasmons and hBN HPhPs was shown to form hybrid waves of one component. In the SPP-HPhP superposed wave, however, the interference happens between polaritons of different momenta scale, $q_{SPP} \sim 10^4$ cm$^{-1}$ and $q_{HPhP} \sim 10^5$ cm$^{-1}$, leading to a wave of two discernible components. This aspect can be useful to create spatial frequency modulation in 2D polariton waves as observed from the fact that the HPhP rises as a high spatial frequency modulation onto the SPP wave. We also display and quantitatively evaluate the strong coupling regime of the mid-IR SPP waves with SiO$_2$ and hBN phonons. Provided the spectral covering in the mid-IR, this ability of the mid-IR SPP waves to couple with phonons can be exploited for sensing in a considerably ample portion of the molecular fingerprint range. It is also relevant to comment that SPP waves can be employed as probes for exploring ENZ modes$^{55}$ that occur near the phononic resonances of polar crystals. Our predictions on the high group velocity and the lifetime of the mid-IR SPPs reveal their potential for use in high-speed plasmonic circuits. In the context of plasmonic devices, we envision that the free-space to SPP coupler, played by the groove herein, can be optimized to enhance the launch of SPP waves in previously defined spectral ranges. Additionally, different van der Waals materials$^{56-58}$ and substrates$^{59}$, supporting phonon-polaritons, can be combined in IMI heterostructures to determine the SPPs’ working bandwidth in between the phonon absorption bands. Such heterostructures can also serve as novel platforms for basic investigations. To conclude, mid-IR SPP waves are shown to be a general phenomenon of the I/M interface with attractive photonic properties that can be exploited in metal-based
2D circuits and devices designable in almost any desirable configuration by modern fabrication techniques, like photo- and electro-lithography, possessing a mature technology for manipulating metals at the 2D scale.

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**Methods**

1. **Scattering-type Scanning Near-field Optical Microscopy /(s-SNOM)**

The s-SNOM (neaSNOM from neaspec, attocube systems AG) uses a metallic tip of an atomic force microscope (AFM) as an optical nanoprobe to measure the optical near-field. Hence, the s-SNOM nanoscope consists in an AFM embodied with a suitable optical arrangement. The AFM operates in semi-contact to the sample mode wherein the metallic tip is electronically-driven to vibrate in its natural mechanical frequency (Ω). By illuminating the tip-sample region with far-field light, it is induced the antenna effect to the tip as the incident electromagnetic field causes charge separation in its metallic coating. Thus, an optical near field rises at the tip surface with higher density at its apex. The optically polarized tip polarizes the surrounding material creating an effective tip-sample polarization. The tip antenna functioning reconverts, by scattering, such tip-sample optical near-field interaction into propagating far-field light that reaches the detector. This scattered light (S) is composed by the mentioned optical near field component and a higher intensity far field one. To separate such contributions, it is used the fact that the near-field light presents a non-linear
temporal dependence on the frequency $\Omega$, due to the strong light-matter interaction at short distances, the far field one has a linear dependence on $\Omega$. Such dependences can be mathematically expressed as $S = \sum_{n=0}^{\infty} S_n \cos(n\Omega t)$ where $n$ is the harmonic order. By lock-in electronics and pseudo-heterodyne (PS) interferometric amplification, the high harmonics, $n \geq 2$, that correspond to pure optical near-field are detected. In this work, such scheme was used to the acquisition of the polaritonic images in Figure 1. Quantum cascade lasers (QCL) were employed as the illumination sources. The detection of $S$ was achieved by a mercury-cadmium-telluride (MCT) detector.

2. **Synchrotron Infrared Nanospectroscopy (SINS)**

SINS underlies on the s-SNOM principles concerning the optical near-field excitation and the use of analogous detection scheme. Thereby, SINS comprises of using a s-SNOM microscope with the highly brilliant broadband IR radiation emitted by a synchrotron as the illumination source. Concisely, the IR beam induces the optical polarization to the tip-sample region and the antenna effect. The resulting scattered light ($S$), then, enters an asymmetric Michelson interferometer (Figure 2a). It is realized interferometry between $S$ and the beam in the reference arm. In analogy to the described for s-SNOM, a locking-based electronics provides for the detection of optical near-field interferograms given from the high harmonics of $S$ ($n \geq 2$). By Fourier Transform (FT), it is obtained the optical near field spectrum. The SINS experiments of this work were performed in the Infrared Nanospectroscopy Beamline of the Brazilian Synchrotron Light Laboratory (LNLS). It was used a MCT detector enabling measuring the optical near-field in the 650 to 3000 cm$^{-1}$ range.

3. **Sample construction**

The groove was created by standard photolithography onto SiO$_2$ (2 µm thick)/Si substrates followed by deposition of a 90 nm thick Au film by electron beam. The hBN flakes were then transferred onto the Au groove using PDMS assisted technique$^{60}$. Previously to the hBN transfer, the flakes were exfoliated by standard scotch tape method on the PDMS stamp and were selected based on optical contrast$^{61}$. After the hBN transfer, the Au disk-like antenna, with an average height of 150 nm and an elliptical basis measuring $\sim 2.4$ and $\sim 3.9$ µm for short and long axes respectively, was designed atop of the transferred hBN and at the edge of the groove by a new photolithography step, followed deposition of 90nm thick Au film by electron beam and lift-off.
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