

Application Note #204

Spatiospectral Nanoimaging of Surface Phonon Plasmons

With their high spatial confinement, surface plasmon polaritons (SPPs) and surface phonon polaritons (SPhPs) in 2D materials can open up new opportunities to enhance light-matter interaction, and enable the development of super lenses, subwavelength metamaterials, and other novel photonic devices. In-situ characterization of these polaritonic excitations across different applications requires a versatile optical imaging and spectroscopy tool with nanometer spatial resolution and wide spectral coverage. Through a noninvasive near-field light-matter interaction, s-SNOM provides a unique way to selectively excite and locally detect electronic and vibrational resonances in real space.

The Anasys nanoIR3-s Broadband system provides a unique capability for nanoscale imaging and spectroscopy over the whole mid-infrared spectral range (2.5 μm - 15 μm / 4000 - 670 cm^{-1}) by coupling with a super-bright broadband light source based on a femtosecond OPO/DFG laser. While featuring high laser power and wide spectral range, this laser source can also switch its linewidth for imaging (narrow-linewidth mode and a spectral resolution of 4 cm^{-1}) and spectroscopy (broad-linewidth mode >200 cm^{-1}).

The power of this technique can be demonstrated by studying the SPhPs dispersion of hexagonal boron nitride (hBN). hBN has a hyperbolic optical permittivity near its two Reststrahlen bands in the mid-infrared region. Both its in-plane and out-of-plane optical permittivity has been plotted, with two resonances at 760 cm^{-1} for out-of-plane (ϵ_z) and 1370 cm^{-1} for in-plane (ϵ_t), respectively (see Figure 1).

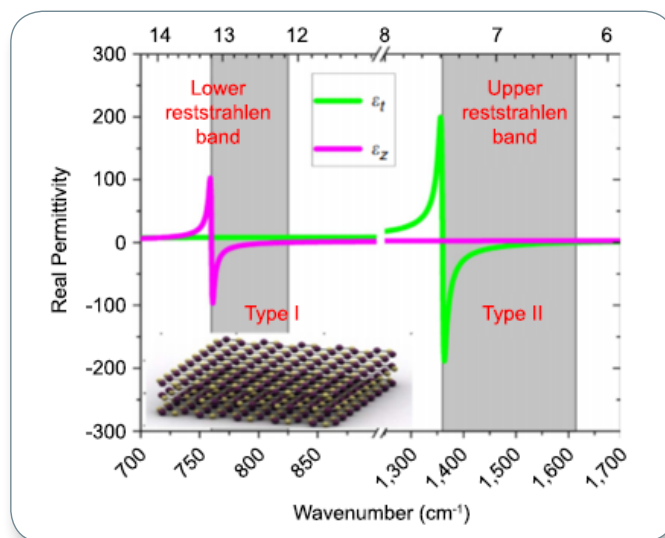


Figure 1. Optical permittivity of hBN in the mid-infrared.¹

Figure 2 shows the experiment schematics of a nanoIR3-s Broadband system imaging a sample of hBN nanoflake. The IR light is tightly focused on the AFM probe and launches hBN SPhPs at the tip apex. The excited SPhPs propagate out along the surface and reflects back from sample edges, forming a standing wave pattern. Figure 3 shows an example of nano-FTIR spectrum collected at different locations on the hBN sample.

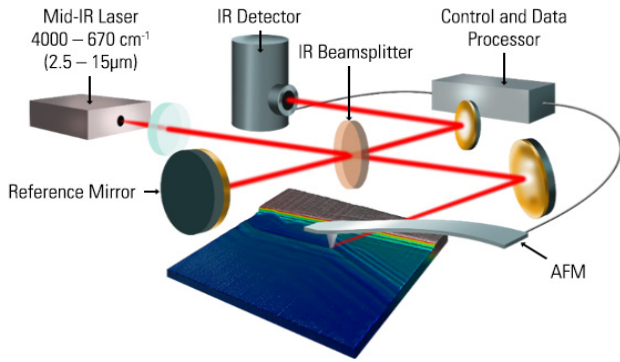


Figure 2. Schematics of a nanoIR3-s Broadband system consisting of a super broadband mid-IR laser source and a compact nano-FTIR microscope.

When operating in narrow-linewidth mode for imaging, the nanoIR3-s Broadband system can acquire high-resolution 2D nanoimaging of SPhPs at targeted wavelengths. When operating in broad-linewidth mode for spectroscopy, a full SPhPs spectrum over 670–4000 cm^{-1} can be collected at a targeted sample location with nanometer resolution. Collecting spectrum at an array of locations gives spatio-spectral nanoimaging of the full SPhP dispersion in a single measurement.

Figure 4 shows near-field images collected in narrow-linewidth mode on a hBN nanoflake. By raster scanning the sample at a specific wavelength, AFM height (Figure 4a)

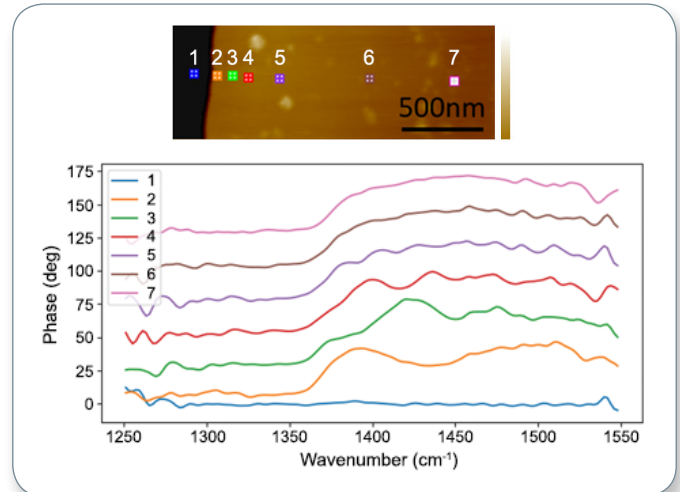


Figure 3. Near-field optical spectrum on a thin hBN nanoflake shows a systematic spectral shift varying with distances to the edge.

and a near-field image at the selected wavelength are collected simultaneously. Figure 4b–w shows IR near-field images at different laser wavelengths at a tuning step of 10 cm^{-1} . Across the range, a standing-wave pattern along the sample edge in each image can be seen. One can also observe the standing-wave pattern systematically varying with laser frequency and distance from the sample edge. The polariton wavelength λ_p can be obtained simply by doubling the fringe period.

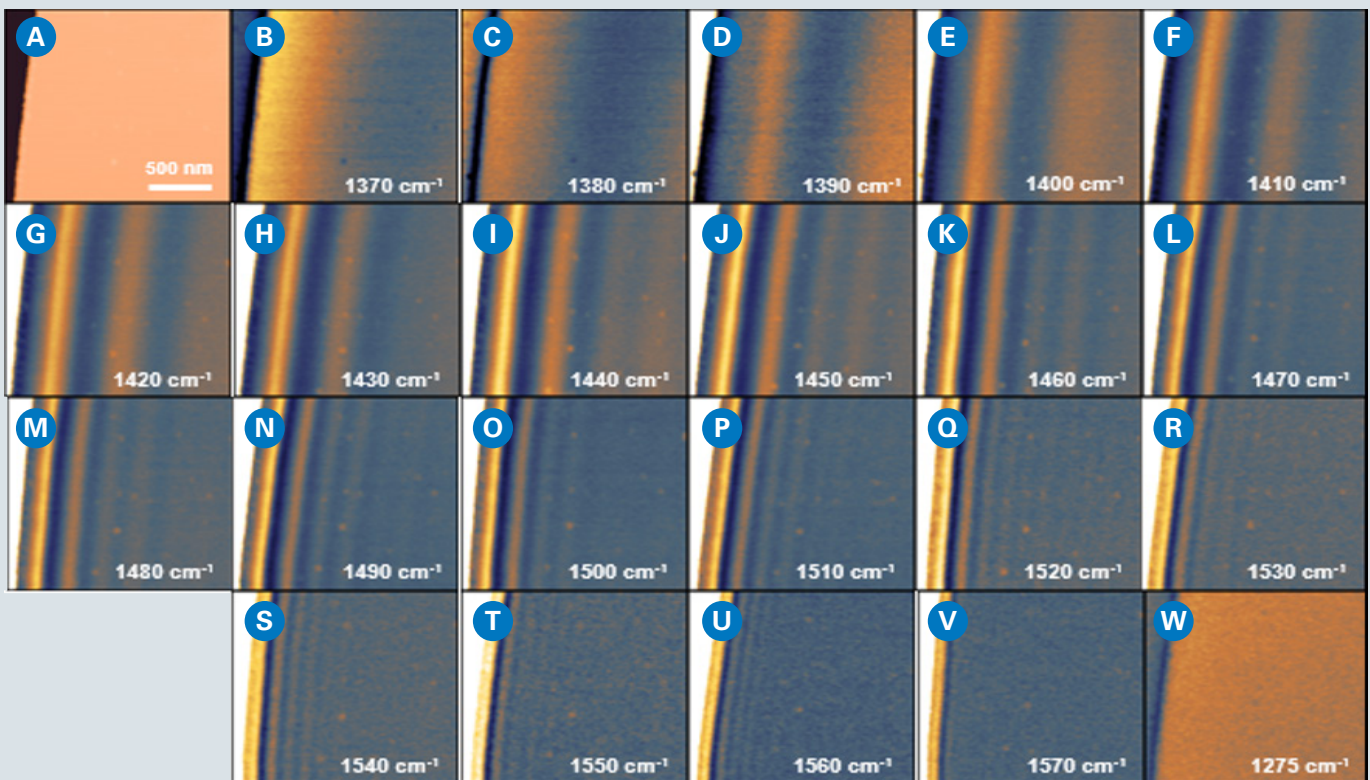


Figure 4. Near-field optical images on a hBN nanoflake at different wavelengths under narrow linewidth mode. Each image is $1.5 \mu\text{m} \times 1.5 \mu\text{m}$ with 10 nm pixel spacing: a) AFM height; b-w) near-field images at different wavenumbers, showing a systematic variation of surface phonon polariton (SPP) waves pattern.

Figure 5 shows spectroscopy under broad-linewidth mode. By acquiring spectra at an array of locations, spatio-spectral nanoimaging of the full SPhP dispersion can be plotted as a 3D datacube. Figure 5a shows the spatio-spectral nanoimaging by plotting a stack of spectra in a waterfall manner, with individual spectrum, like Figure 3. The line scan pixel spacing is 15 nm, and each spectrum of spectral resolution (3 cm^{-1}) has an acquisition time of 1 minute, giving a total measurement time of ~ 2.5 hours. The spatio-spectral maps in Figure 5, obtained in broad linewidth mode with $>200\text{ cm}^{-1}$ spectral linewidth, show the full s-SNOM amplitude and phase response of hBN, including the optical phonon at 1370 cm^{-1} and SPhP waves spanning the range from $1370\text{--}1550\text{ cm}^{-1}$.

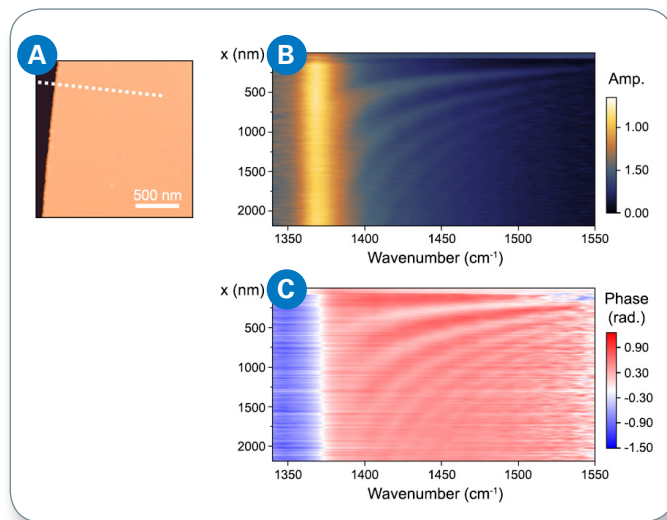


Figure 5. Spatio-spectral nanoimaging on a hBN nano flake: (A) The array of spectra is collected under broad-linewidth mode along the dotted white line; By plotting the stack of spectrum in a false color in a waterfall manner with position as vertical axis, a spatio-spectral nanoimaging map is created for amplitude (b) and phase (c), respectively. The spatio-spectral scan shows the complete SPP frequency response across the whole range, with high spectral resolution (3 cm^{-1}). Line scan pixel spacing is 15 nm, and each spectrum has an acquisition time of 1 minute, giving a total measurement time of ~ 2.5 hours.

Conclusion

In conclusion, combining imaging with broadband spectroscopy (and spatio-spectral imaging) the nanoIR3-s Broadband system provides a powerful tool for spatio-spectral nanoimaging of SPP and SPhPs in 2D materials with nanometer spatial resolution, wide spectral coverage, and high signal to noise.

Reference

1. Caldwell, J.D. et al., "Sub-diffraction, volume-confined polaritons in the natural hyperbolic material: hexagonal boron nitride," *Nat. Comms.* 5, 5221 (2014).