Far infrared synchrotron near-field nano-imaging and -spectroscopy

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I. SUPPLEMENTARY INFORMATION

Here we provide additional technical information about the ultrabroadband s-SNOM experimental setups and Ge:Cu detector.

SINS: IR synchrotron radiation is focused onto the apex of a sharp metallic tip of an atomic force microscope (AFM). In order to discriminate the weak near-field interaction from the strong diffraction-limited far-field background, the AFM typically operates in intermittent contact mode, and the detector signal is demodulated at harmonics of the tip tapping frequency Ω (typically 75 - 300 kHz) using a high speed digital lock-in detector (HF2-LI, Zurich Instruments). The complex near-field scattering signal is obtained as $S_n(\omega) = |S_n|e^{i\Phi_n}$, where $|S_n|$ and Φ_n are the SINS amplitude and phase, respectively, demodulated at the nth tip harmonic. Owing to the nonlinear distance dependence of the near-field interaction between the tip and sample, progressively higher order demodulation results in increasing suppression of far-field background, isolating the near-field scattering signal (see Fig. 2 in main text).

Back-scattered light from the tip-sample interface is combined interferometrically with a reference beam in an asymmetric Michelson configuration, analogous to dispersive Fourier-transform spectroscopy, and focused onto a cryogenically-cooled IR detector, giving the the complete nano-resolved ultrabroadband near-field response $|S_n(\omega)|e^{i\Phi_n(\omega)}$. Conventional mid-IR s-SNOM spectroscopy configurations typically operate using a KBr beamsplitter and HeCdTe (MCT) photoconductive or photovoltaic detector with speed > 5 MHz. To realize far-infrared SINS, we utilize either a Si or KRS5 beamsplitter, in conjunction with a novel home-built Ge:Cu photoconductor, described below. Significant modifications were required to minimize sources of thermal and electronic noise, while allowing for detection speeds > 2 MHz, necessary for higher-order demodulation at AFM tip frequencies.

Ultrabroadband SINS is available in two experimental configurations at ALS: Beamline 5.4, employing a specially modified AFM (Innova, Bruker) coupled to commercial FTIR Spectrometer (Nicolet 6700, Thermo-Scientific), and Beamline 2.4 using a commercial nanoscope (Neaspec GmbH).

Extending into the far-infrared: Besides the scarcity of sufficiently intense light sources, access to the FIR region is also hindered by the lack of suitable detectors having the necessary sensitivity and speed of response (on the order of 1 MHz) as required for s-SNOM measurements. A particular challenge for this spectral range is the intrinsic noise from fluctuations in the 300K thermal radiation background. When other noise sources (electronic noise in the detector itself and the read-out electronics) can be kept smaller, the detector is said to reach background limited infrared photodetection or BLIP. Mid-IR detectors based on small bandgap semiconductor, such as $Hg_{1-x}Cd_xTe$ (MCT) having an alloy composition x that places the long wavelength cutoff at < 16 μm, can fulfill this condition, and operate at speeds up to 10s of MHz. But alloy compositions for longer wavelengths are challenged by material limitations. Liquid helium cooled bolometers that sense the heat of absorbed infrared can achieve BLIP, but have a response speed on the order of 1 kHz in their standard IR spectroscopy configuration and are therefore unable to detect at typical tip-modulation frequencies. Extrinsic impurity photoconductors, based on doped Si or Ge, offer fast intrinsic response times (< 100 ns) and have small bandgaps for detection in the far-infrared (FIR). These detectors can achieve BLIP, but are typically configured with a moderately large optical acceptance for conventional spectroscopy. For example, an infrared detector designed for general spectroscopy has a $\sim 14^\circ$ field-of-view (corresponding to f/4) and an effective area of 20 mm^2 (5 mm diam.) or more to view a macroscopic sample. In the case of s-SNOM, the required detector area is at the diffraction-limit, or about 10λ for f/4, which is less than 0.5 mm at a wavelength of $\lambda = 30\mu$m. Thus the background noise could be 10x smaller, offering the potential for improved signal-to-noise and an opportunity to reach much longer wavelengths than MCT.

Ge:Cu: The detector was developed from a commercial Ge:Cu photoconductor system from Infrared Laboratories (IR Labs, Tucson AZ) originally intended for general infrared spectroscopy over the 5 μm to 30 μm wavelength range. The photoconductive element is mounted inside a small liquid helium cryostat that keeps it, plus an f/4 Winston cone and entrance aperture, at T = 4.2K. The external vacuum window is KRS-5. The original ~ 6 mm diameter aperture was decreased to 1 mm, reducing the background noise by a factor of 6. The other modifications were to the read-out electronics, consisting of replacing the amplifier components with ones appropriate to achieving a 1 MHz bandwidth (Fig. S1). The original detector wiring was left as-is.

We note that additional improvements in detector performance may be possible, depending on the limiting noise sources. If the thermal background is the limit, then an aperture 3x smaller could be installed and not restrict the ability to view a diffraction-limited source. If

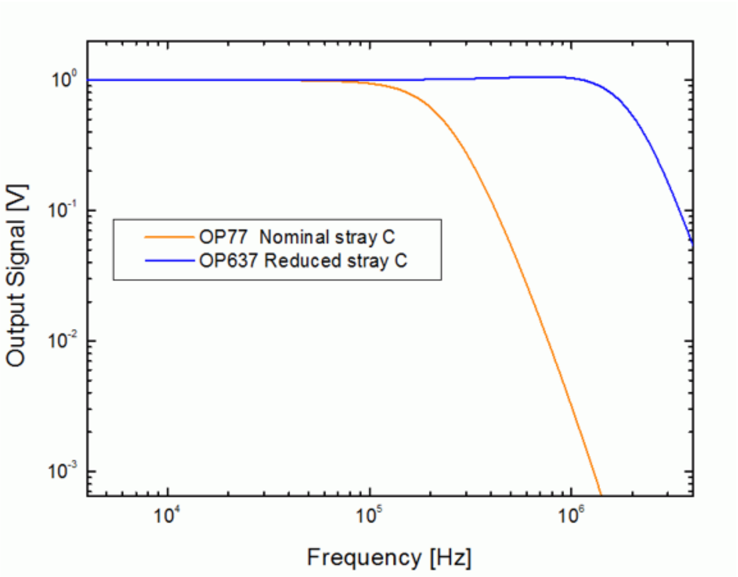

FIG. S1: Detector bandwidth comparison between original and modified Ge:Cu pre-amplifier, with additional reduction of stray capacitances, increasing sensitivity and speed of operation in excess of 2 MHz.

the noise limit is from the read-out electronics (typically from Johnson noise), then cooling the critical resistor elements can reduce noise. We have not had the opportunity to perform a detailed noise study to see what improvements would provide the most impact.