1.3.15 Near-field nanoscopy

Near-field imaging with terahertz (THz) waves is emerging as a powerful technique for fundamental research in photonics and across physical and life sciences. Spatial resolution beyond the diffraction limit can be achieved either by collecting THz waves from an object through a small aperture placed in the near-field or via scattering in a near-field optical microscopy set up. However, both approaches suffer from severe limitations in the far infrared. Here we report on near-field THz nanoscopy in both detectorless and ultrafast architectures and discuss intriguing applications in plasmonics and ultrafast polaritonics.

Light transmission through a sub-wavelength size aperture is fundamentally limited by the wave nature of light. To overcome the above limit, we conceived a novel architecture that exploits the inherently strong evanescent THz field arising within the aperture to mitigate the problem of vanishing transmission. The sub-wavelength aperture is originally coupled to asymmetric electrodes, which activate the thermo-electric THz detection mechanism in a transistor channel made of flakes of black-phosphorus or InAs nanowires (Fig. 1a). The proposed novel THz near-field probes enable room-temperature sub-wavelength resolution coherent imaging with a 3.4 THz quantum cascade laser, paving the way to compact and versatile THz imaging systems and promising to bridge the gap in spatial resolution from the nanoscale to the diffraction limit (Fig. 1b).

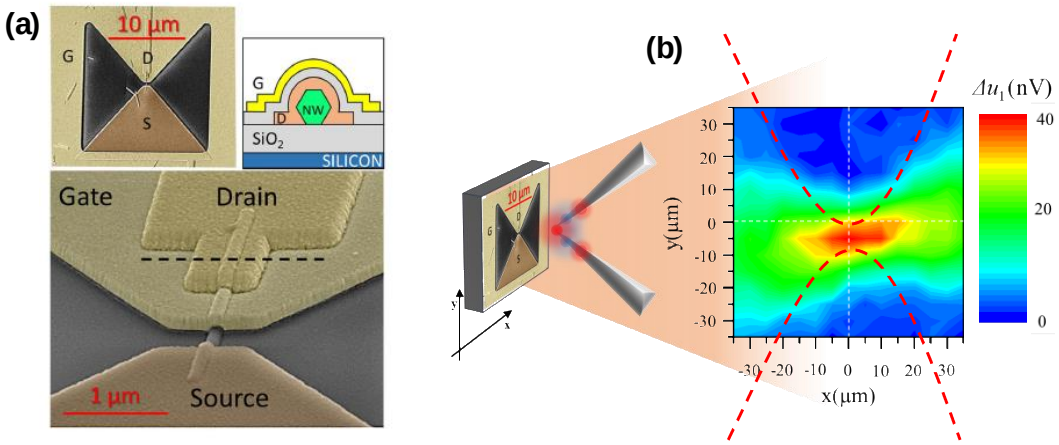

Figure 1. (a) Upper left and lower left: Scanning electron microscope (SEM) images of the near-field probe with an embedded FET-based THz nanodetector (view angles of 0° and 70°). A top gate contact (G) defines the aperture; the aperture size is 18 µm × 18 µm; the InAs nanowire detector is at the aperture center; and the source (S) and drain (D) contacts are isolated from the gate with a layer of SiO_2. (upper right) Schematic diagram of the cross-sectional view of the device. **(b)** Left: schematics of two needles employed for focusing the THz beam to a sub-wavelength spot. The needles are placed in front of the NW nanodetector probe. Right: spatial distribution of the near-field probe photovoltage; the red dotted lines mark the simulated confinement provided by the drain and gate contact.

At THz frequencies, scattering-type scanning near-field optical microscopy (s-SNOM), that is the alternative near field configuration, mostly relies on cryogenic and bulky detectors, which represents a major constraint for its practical application. Amplitude- and phase–resolved near-field imaging is particularly appealing to enable access to the spatial variation of complex-valued dielectric responses of THz frequency resonant 2D materials, heterostructures and low dimensional systems.

We devised the first THz s-SNOM system that provides both amplitude and phase contrast, and achieves nanoscale (60-70nm) in-plane spatial resolution. It features a quantum cascade laser that simultaneously emits THz frequency light and senses the backscattered optical field through a voltage modulation induced inherently through the self-mixing technique (Fig. 2a). We demonstrated its performance by probing a phonon-polariton-resonant CsBr crystal, doped black phosphorus flakes, ink jet printed graphene and propagation of THz acoustic plasmons in graphene (Fig. 2b).

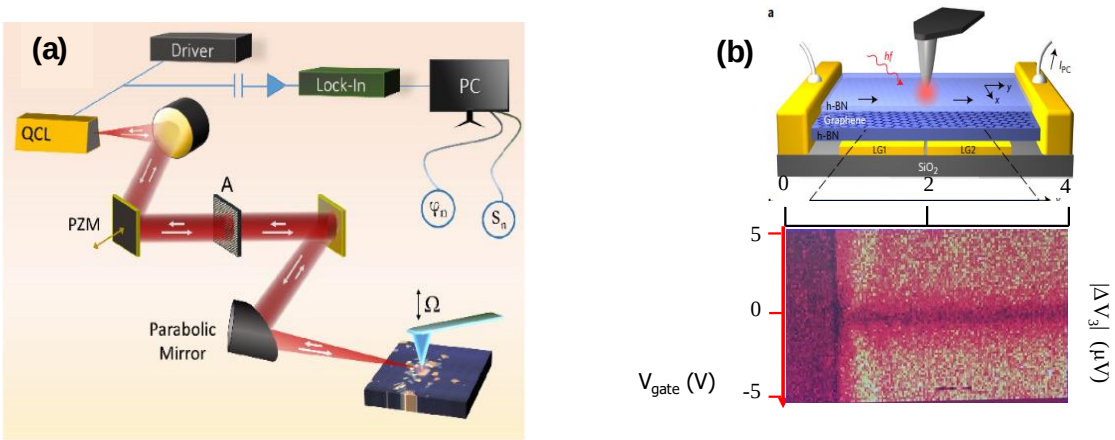

Figure 2. (a) Self-detection scattering type near field optical microscope with nanometer resolution at terahertz frequencies: schematic diagram showing the experimental arrangements; (**b)** Propagating THz acoustic plasmons in graphene. Upper: Schematics of the employed device; bottom: third harmonic self-mixing near field signal collected as a function of the gate voltage (V_{gate}).

Furthermore, we devised lightning-fast switch for electron waves in the far-infrared based on black phosphorus (BP) heterostructures, in which interface polaritons can be activated by photo-induced interband excitation within 50 fs, with switching times on the femtosecond scale (Fig. 3), many orders of magnitude faster than the fastest existing transistors.

The possibility of hybridizing collective electronic motion with mid-infrared light to form surface polaritons has made van der Waals layered materials a versatile platform for extreme light confinement and tailored nanophotonics.

We design a SiO_2/black phosphorus/SiO_2 heterostructure in which the surface phonon modes of the SiO_2 layers hybridize with surface plasmon modes in black phosphorus that can be activated by photo-induced interband excitation. Within the reststrahlen band of SiO_2, the hybrid interface polariton assumes surface-phonon-like properties, with a well-defined frequency and momentum and excellent coherence. During the lifetime of the photogenerated electron–hole plasma, coherent hybrid polariton waves can be launched by a broadband mid-infrared pulse, coupled to the tip of a scattering type scanning near-field optical microscopy set-up. The scattered radiation allows us to trace the new hybrid mode in time, energy and space.

The excellent switching contrast and switching speed, the coherence properties and the constant wavelength of this transient mode make it a promising candidate for ultrafast nanophotonic devices.

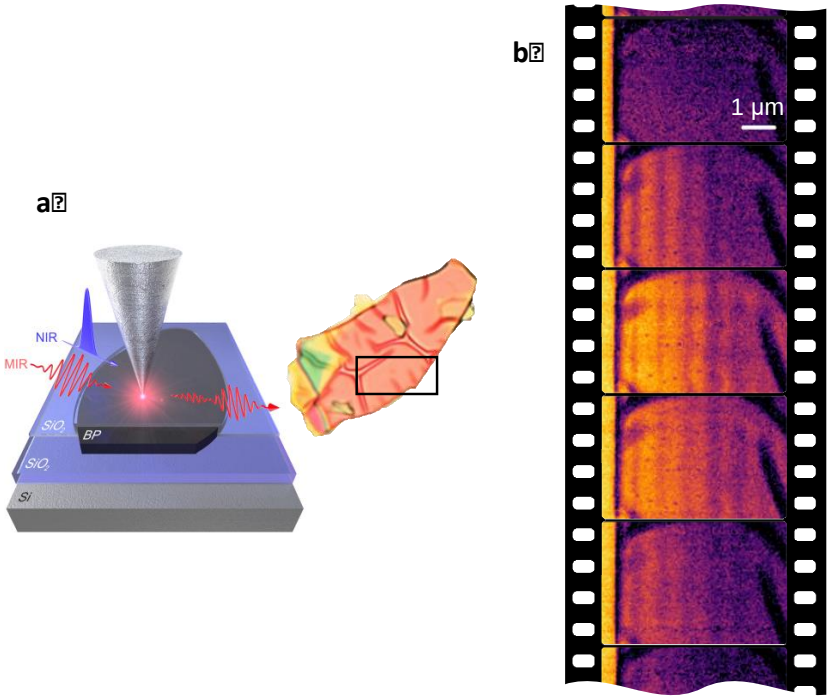

Figure 3. (a) Schematic of set-up. An ultrafast near-infrared pump pulse is focused onto a heterostructure of SiO_2 and BP on a SiO_2-capped Si wafer. The sample is probed in the near field with a mid-infrared pulse coupled to a sharp metallic tip. Right side: optical image of a single BP flake capped with SiO_2; **(b)** Scattered near-field intensity images of the SiO_2/BP/SiO_2 heterostructure, plotted for different delay times between the pump and probe pulses.

Ultrabroadband electrooptic sampling of few-cycle field transients can even reveal novel dynamics that occur faster than a single oscillation cycle of light. However, conventional terahertz spectroscopy is intrinsically restricted to ensemble measurements by the diffraction limit. As a result, it measures dielectric functions averaged over the size, structure, orientation and density of nanoparticles, nano-crystals or nano-domains. We extended ultra-broadband time-resolved THz spectroscopy to the sub-nanoparticle scale (10 nm) by combining subcycle, field-resolved detection (10 fs) with scattering-type near-field scanning optical microscopy (s-NSOM). We trace the time-dependent dielectric function at the surface of a single photoexcited InAs nanowire in all three spatial dimensions and reveal the ultrafast (<50 fs) formation of a local carrier depletion layer.

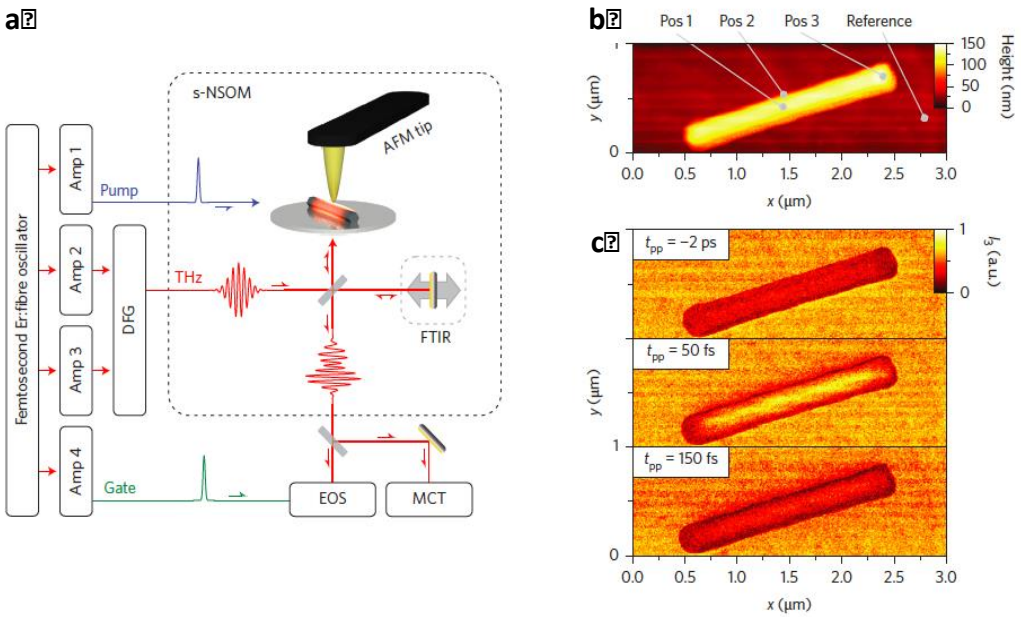

Figure 4. (a) Schematic of the experimental set-up. A femtosecond Er: fiber oscillator seeds four separate Er:fiber amplifiers, which are used to produce the near-infrared (1.56 μm) pump pulses (Amp 1), near-infrared (1.3 μm) electro-optic gate pulses (Amp 4) and phase-stable multi-terahertz probe pulses (Amp 2 and Amp 3) via difference frequency generation (DFG). The THz transients are focused onto the atomic force microscope (AFM) tip of an s-NSOM and the scattered electric near field is detected by electro-optic sampling (EOS) with sub-cycle temporal resolution. Alternatively, a mercury cadmium telluride (MCT) photodiode records the time integrated scattered intensity and enables Fourier transform infrared (FTIR) spectroscopy (>60 fs time resolution). **(b)** Topography of the InAs nanowire (on diamond) studied in the experiments, measured by AFM. (**c)** Ultrafast terahertz nano-movie frames of nanowire photoexcitation. Free carriers are photoinjected into the InAs nanowire by near-infrared pump pulses and time-resolved near-field terahertz intensity images are measured as a function of pump–probe delay time tpp. The pump fluence is 1.0 mJ cm^{-2} and the tapping amplitude is 130 nm.

References

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