Status of THz-to-Visible Nanospectroscopy Development

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Abstract. Quite unexpectedly, THz and infrared spectroscopy has now a real chance to solve problems in the nanosciences. This rests on a new microscope technique that overcomes the Abbe diffraction limit, by using the near field of a metal antenna in close proximity to a scanned sample surface. Here I briefly summarize present activities in the microwave, mid-infrared and visible spectral ranges. It seems straightforward and highly desirable to fill the existing gap between about 20 GHz and 20 THz, and attain spatial resolution of 10 nm and below also in this important part of the electromagnetic spectrum.

Key words: infrared microscopy, infrared spectroscopy, microscopy, near-field microscopy

It has been known for decades if not a whole century that electromagnetic waves can be concentrated to narrow spaces much smaller than the wavelength. An example is any two-wire cable used in electronics. Metal wire antennas have routinely been operated to concentrate the power of free-space or guided waves to tiny spaces, as for example to the μm -sized transistor elements of microelectronics. Interestingly, such antenna focusing has already enabled harmonic generation within the THz range, from 1 GHz up to 60 THz, as early as the 1970s [1–3].

That such antenna-confined electromagnetic fields can achieve microscopy with sub-wavelength spatial resolution was first discovered in the microwave region [4–9]. By coupling 20 cm-wavelength microwaves to the tip of an STM we were able to demonstrate microwave microscopy with 200 nm resolution [10], a breakthrough because this is better than that of a classical optical microscope. Further developments have introduced microwave resonators to enhance the sensitivity [11–13], yet according to recent reviews [14, 15] no higher spatial resolution seems to have been reported. Very recently, the first antenna-focused mapping that operates with broadband 0.1–2.5 THz radiation was reported, with as yet modest resolution of about 18 μm [16].

Much better resolution of < 30 nm is now firmly established in the 30 THz region offered by the CO_2 laser (wavelength range 9.2–11.2 μm). Here the infrared imaging channel is superimposed on the basic scanning probe setup which is either a STM [17] or an AFM [18–29]. The typical experimental setup employed is sketched in Figure 1. The infrared laser is focused with a high numerical aper-

Figure 1. Schematic view of the AFM-based infrared near-field microscope that has operated with 9.2–11.2 μm radiation, attaining < 30 nm spatial resolution.

ture mirror objective to a focal spot with 10–20 μm diameter. This illuminates a metallized tip located at the end of a slightly oscillating AFM cantilever. A sample is approached and raster-scanned in close vicinity of the tip. The AFM feedback loop (not shown) assures a constant average distance between tip and sample (AFM tapping mode). Simultaneous with generating the topographical map of the sample surface, the ‘scattering’-type scanning near-field infrared microscope (s-SNIM) also records an image of the scattered infrared power. This infrared image can contain considerable false contrast because an overwhelming amount of the signal can come from uninteresting ‘background’ scattering that originates from the tip shaft and cantilever. Our development has shown that demodulating the detector signal at a higher harmonic of the tapping frequency Ω can completely suppress the background signal [28, 30], allowing to map genuine near-field contrasts that can directly be related to the local refractive index of the sample as extensively studied also in the visible [31, 32]. Using interferometric signal detection we have shown that indeed amplitude and phase of the near field can be mapped simultaneously, and that this allows to map the *complex* local dielectric function [31] as well as cope with interesting resonant phenomena such as plasmons [33] and phonons [29].

Figure 2 illustrates the status of microwave-to-visible near-field microscopic resolution attained so far in our laboratory. The arrows give the achieved sub-wavelength factors of the experiments cited. We have reason to expect, backed by the pioneering work of the Wickramasinghe group which found 1 nm resolution in the visible [34–36] as well as by theoretical analyses [37–39] that a spatial resolution of about 1 nm may be attainable at any frequency from the visible through the infrared and THz ranges down to microwave frequencies. This is indeed a

Figure 2. Status of the achieved near-field microscope spatial resolution vs wavelength of illuminating visible, infrared and microwave radiation, as obtained in the author's laboratory.

tremendous outlook, as it predicts the tool of nanospectroscopy not dreamt of a decade ago.

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