

decay into fundamental ones, which then shift deeper and deeper into the infrared with further propagation. For ideal conditions, this continuous shift will only be terminated by the onset of absorption as dictated by the fibre's core material. Meeting the optimum conditions of soliton fission typically requires a substantial adjustment of the zero-dispersion wavelength of the core material by means of suitably designed waveguide dispersion. In the case of AsSe, a shift of 1–2 μm was necessary to match the fibre to the employed pump wavelengths in the mid-infrared. This requirement puts tight constraints on the core diameter of the fibre, which then inadvertently results in a second problem — the appearance of higher spatial modes and the breakdown of the single-mode guidance of the fibre. Therefore, it seems quite remarkable that all these constraints could be managed to enable a supercontinuum with a bandwidth greater than three octaves.

The demonstrated supercontinua substantially enhance the capabilities of spectroscopy in the mid-infrared, a spectral region that has also been dubbed the molecular fingerprint range. This wavelength range is rich with characteristic vibrational spectra of molecules and thus enables the unambiguous identification of various species using transmission spectroscopy (Fig. 1). The greatly increased luminous intensity of laser-generated supercontinua, when compared with other broadband sources, immediately translates

into a welcome gain in sensitivity and wavelength selectivity.

Although the immediate benefits for conventional spectroscopy are clear, it may also be possible to use these sources for frequency comb spectroscopy. In particular, it seems appealing to extend the use of mid-infrared continua to dual-comb or Vernier spectroscopy⁷. This method requires the use of more complex laser oscillators with repetition rates on the order of megahertz, but as a benefit it promises virtually unlimited spectral resolution for spectroscopy. Such advanced methods may require further development and refinement of chalcogenide fibres. For example, microstructuring the fibre geometry may lead to greater optical confinement and thus lower the pulse energy requirements of the pump laser, similar to what has been done with photonic crystal fibres⁸. In these structures, much tighter guiding is achieved by surrounding the core with air-filled channels.

In conclusion, it is expected that the availability of multi-octave mid-infrared supercontinua will open a new avenue for spectroscopy in this regime. Higher selectivity and an enhanced sensitivity may help enable the unambiguous detection of even the faintest molecular traces. It is conceivable that the resulting technology may even outperform the nose of a dog, which still sets the gold standard for a range of applications. Highly sensitive mid-infrared spectroscopy may be used

in a wide range of applications, including the remote detection of explosives and the detection of certain diseases from the analysis of a patient's breath. It may also be employed for standoff-detection in environmental studies. Other applications will certainly be in the area of fundamental scientific studies, and, if pumped by a simple oscillator, mid-infrared supercontinua could be interesting for seeding parametric conversion schemes. Finally, it needs to be mentioned that the achieved limit of 13.3 μm does not constitute the absolute limit for further extension of supercontinua into the infrared. Using tellurite-based chalcogenide glasses, it may be possible to push the cut-off beyond 20 μm — so, there is still much work to be done. \square

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SPECTROSCOPY

Nanoscale terahertz spectroscopy

The advent of terahertz spectroscopy schemes that offer single-photon sensitivity, femtosecond time resolution and nanometre spatial resolution is creating new opportunities for investigating ultrafast charge dynamics in semiconductor structures.

Hidemi Shigekawa, Shoji Yoshida and Osamu Takeuchi

The desire to make semiconductor devices smaller and faster is a strong driving force behind developments in nanoscale science and technology. Marked progress in device design and fabrication in the past decade means that we are now in need of a new set of terahertz (THz) spectroscopy tools that can analyse semiconductor structures and devices on unprecedented spatial and temporal scales. Semiconductor devices have now become as small as

10 nm. At this scale, fluctuations in the spatial distribution of dopants — introduced to control the electronic and optoelectronic properties of devices — and in the nanostructured interface between dissimilar materials can directly affect device operation.

In addition, there is a need to be able to probe transient charge carrier dynamics that change on the femtosecond timescale. Traditional techniques that rely on temporally-averaged or integrated

data are often too slow to provide this level of resolution. Now, as they report in *Nature Photonics*, Max Eisele and colleagues¹ have developed a microscopy technique to simultaneously meet these demanding spatial and temporal requirements.

The recent development of quantum optics has provided techniques for observing ultrafast phenomena that occur on femtosecond to attosecond timescales. For example, snapshots of

ultrafast dynamics can be captured using an ultrashort pulse laser and the pump–probe method^{2,3}, allowing time-resolved analysis to be performed. However, the spatial resolution is generally limited by the wavelength, which dictates the size of the light spot. On the other hand, scanning tunnelling microscopy (STM), which was invented in 1981, has enabled the analysis of local electronic structures with atomic resolution. Many valuable results have been obtained using STM and the related technique of scanning probe microscopy (SPM). However, the temporal resolution is limited, in general, to millisecond to microsecond timescales.

Since the invention of STM, combining it with the techniques of quantum optics has been a sought after goal. To achieve this objective, several microscopy techniques have been realized^{4–8}. Near-field optical microscopy (NSOM) using an aperture has achieved a temporal resolution below 20 fs (ref. 4). The modulation of STM bias-voltage using THz pulses has enabled differential-conductance imaging with simultaneous sub-picosecond time resolution and nanometre spatial resolution⁶. The ultrafast spin dynamics of carriers confined in a quantum well have also been probed by STM with circularly polarized light⁸. However, greater understanding of the carrier dynamics of nanoscale materials requires the development of time-resolved spectroscopy schemes based on new concepts. In particular, the development of time-resolved THz spectroscopy with nanometre spatial resolution is highly desirable⁹.

Eisele *et al.*¹ have combined sub-cycle, field-resolved detection of THz waves (with a temporal resolution below 10 fs) with scattering-type NSOM (s-NSOM)^{10,11}. The result is ultrabroadband time-resolved THz spectroscopy (in the 20–50 THz frequency range) on the sub-nanoparticle scale (less than 10 nm). For the first time, the scheme reveals information about the carrier dynamics in a single InAs nanowire using three-dimensional probing of the time-dependent dielectric function.

A schematic of the experimental set-up used by Eisele *et al.*¹ is shown in Fig. 1. Atomic force microscopy (AFM) was used to obtain a high spatial resolution and a high signal-to-noise ratio. To obtain high temporal resolution, a 10 fs pulse was used to control the gate of the detector. The sample surface below the AFM tip was excited by a pump pulse and its effect on the dielectric function of the sample was measured through the change in the shape of the scattered electric field of the subsequent probe pulse.

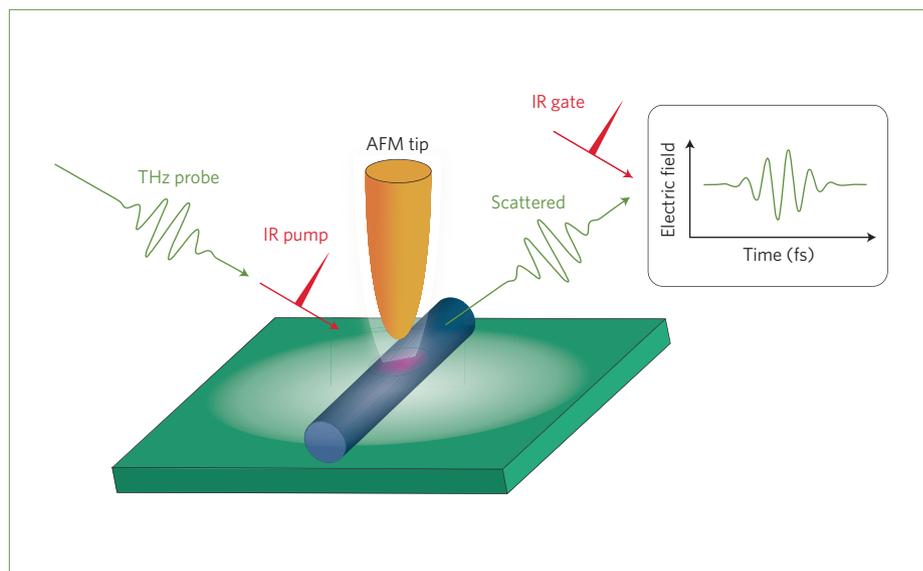


Figure 1 | State-of-the-art implementation of transient terahertz (THz) time-domain spectroscopy with a single-photon-level signal-to-noise ratio, which allows the THz response from a nanoscale object — an InAs nanowire (blue) on a diamond substrate (green) — to be measured. The field enhancement effect of a sharp atomic force microscopy (AFM) tip confines the probed volume (red) in the sample to around 10 nm. Using electro-optic sampling, the scattered electric field of the probe pulse was directly traced with the 10 fs infrared (IR) gate pulse.

A PtIr-coated AFM tip strongly enhances the p-polarized component of the electric field of excitation, which enables local probing of the signal below the AFM tip in a space much smaller than the applied light spot. To detect a weak signal, the AFM was operated in the tapping mode to enable the use of highly sensitive lock-in detection. The AFM cantilever was oscillated at a fixed frequency and the separation between the AFM tip apex and the sample was sinusoidally varied between 0 and, for example, 40 nm. The third harmonic of the tapping frequency was measured in the signal to reduce the large nonlocal background.

To perform electro-optic sampling with high temporal resolution, the scattered electric field of the probe pulse was directly traced with the 10 fs gate pulse. An instantaneous THz electric field and the waveform were recorded with sub-cycle time resolution by scanning the delay between the THz pulse and the pump pulse while keeping the delay of the pump gate fixed. In this way, information about the absolute amplitude and phase was obtained, thus providing the evolution of the dielectric function of the sample. From the analysis of the plasma frequency, included in the dielectric function, the local carrier density following photoexcitation was directly tracked.

Eisele *et al.*¹ applied this microscopy technique to an InAs nanowire. High-sensitivity detection made it possible to operate with a signal corresponding to only 50 photons per THz pulse. Therefore, the detection of electric field transients from a part of a single nanowire was carried out with a noise floor of less than one coherent photon per pulse by optimizing electro-optic sampling.

Two components of the carrier density decay — an ultrafast decay (less than 50 fs) and decay with a constant of 2 ps — were observed. Interestingly, the decay was dependent on the AFM tapping amplitude, suggesting the possibility of controlling the probing depth in the sample when the tip approaches it. From the measurement of the tapping amplitude dependence of the signal, the fast component, which dominates for small tapping amplitudes, was attributed to the formation of a depletion layer. In contrast, the slow component, dominant for large tapping amplitudes, was attributed to carrier trapping in defect states. A more accurate depth profile can be obtained with a deeper understanding of the physical origin of the tapping amplitude dependence. Such a three-dimensional analysis is expected to become a powerful technique for investigating nanoscale dynamics. Varying the light polarizations of both pump and probe pulses, thus changing the condition

of tip-induced electric field enhancement, may provide further information on local carrier dynamics. Development of the system so that it can operate in ultra-high vacuum may make it suitable for use with a wider variety of samples.

The microscopy technique demonstrated by Eisele and colleagues is expected to play an important role in advancing nanoscale science and technology. As is discussed above, a variety of time-resolved SPM techniques have already been realized^{4–8}. In addition, by combining a spin-polarized tip and pump–probe pulse using a bias voltage, the investigation of inelastic tunnelling has provided information on the spin dynamics in single atoms¹². Because the nanoscale properties of materials differ from those in

bulk structures due to various factors such as the morphology, temperature, band gap, dielectric function and electron-phonon coupling strength, the complementary use of these microscopy techniques is expected to be important for exploring the carrier and spin dynamics in nanoscale structures. In particular, combining microscopy techniques with local excitation using plasmonic superfocusing¹³ may be interesting in the future. The successive development of novel microscopy techniques based on new ideas will greatly aid the further development of nanoscale science and technology. □

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TRANSPARENT ELECTRODES

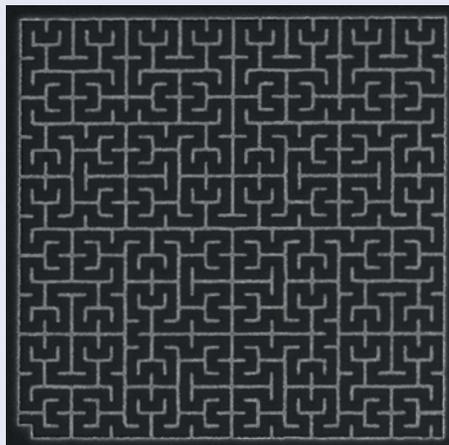
Fractal future

Fractals are beautiful patterns based on the principle of self-similarity. Well-known examples include the branching arrangements of trees, river networks or blood vessels. Now researchers have discovered that fractals can be put to good use in photonics by creating transparent ultrathin metallic electrodes with superior optoelectronic properties.

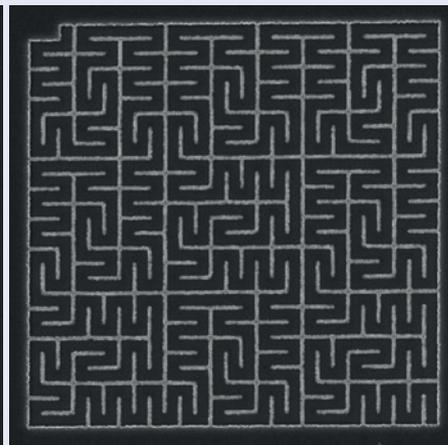
Transparent electrodes based on conducting oxides such as indium tin oxide or zinc oxide play a crucial role in devices like solar cells, LEDs and displays. However their performance is far from ideal; they are costly, brittle and not well suited to use with some organic materials. As a result, much research is devoted to finding alternatives. One option being explored is the use of ultrathin metal layers. Farzaneh Afshinmanesh and co-workers at the Geballe Laboratory for Advanced Materials in Stanford, USA as well as the Institute of Photonic Sciences and the Catalan Institution for Research and Advanced Studies in Barcelona, Spain have demonstrated that ultrathin aluminium electrodes formed with a fractal geometry have optoelectronic properties superior to their metal counterparts in a grating or grid geometry (*Nano Lett.* **14**, 5068–5074; 2014).

When it comes to transparent electrode design, a high optical transmission is a key figure of merit. The authors studied the performance of aluminium electrodes of various designs that were integrated into

Hilbert fractal



Peano fractal



a silicon photodetector. The investigated structures included linear gratings, grids and Hilbert and Peano fractal geometries. The wavelength- and polarization-resolved photocurrent responses of the detector revealed broadband responsivity in the 450–750 nm range without any polarization dependence apart from the case of the grating. Importantly, detectors featuring the Hilbert and Peano fractal shaped electrodes (pictured) benefited from significantly improved responsivity: they were about 40% and 34% better, respectively, compared with the grid, whereas the increase was 7% and 2%, respectively, compared with the grating.

Breaking down the fractal structures into regions where the metallic wires

are horizontally and vertically parallel helps elucidate the physics behind the observations. Horizontal gratings transmit primarily vertically polarized light, whereas the opposite is true for vertical gratings. As a result, reflection imaging reveals various bright and dark areas along the surface of the fractal, corresponding to areas of low and high transmission, respectively.

Following further research such fractal electrodes could prove useful in cameras and displays as well as solar cells. In the latter case, the properties of fractal networks may also be helpful for light trapping.

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