

to the electric field of the incident beam transients at the sampling moment.

The EO sampling technique usually requires optical probe pulses to be synchronized to the THz radiation being detected. Usually, femtosecond pulses from a Ti:sapphire laser are split into two: one is used to generate THz transients by exciting PC switches or nonlinear materials, the other is used as the probe pulse. The synchronization maintains the temporal relation between THz transients and the probe pulses. By scanning the time delay between the THz transient and probe pulse, an EO signal reproduces the waveform of the incident radiation. The quality and bandwidth of the measurement depends on the duration of the sampling pulses, and the ability to achieve phase-matching in the EO crystal at the wavelength in question.

In the past, the EO sampling technique has not been considered to be applicable to measurements of a non-synchronized source as it was understood that EO signals will be averaged to zero. Contrary to this intuition, Gaal's experiment has shown that it is possible to retrieve meaningful information from such non-synchronized measurements.

The approach taken by Gaal *et al.* is to sample the electric field of the signal beam (one line of a CO<sub>2</sub> laser) with a probe beam (femtosecond pulses from a Ti:sapphire laser) inside a ZnTe crystal and store the sequential data (Fig. 1). This raw data, as expected, does not show any meaningful structure as shown by Fig. 2b in ref. 3. However, the autocorrelation of the sampled signal does

show structures that can be interpreted as the temporal coherence of the signal radiation (Fig. 3 of ref. 3). The autocorrelation is compared to an interferogram reconstructed from the sampled data. Further by Fourier transformation, the autocorrelation signal can be converted into the spectrum reproducing the signal radiation, thus giving its emission spectrum.

The highest detectable frequency (shortest wavelength) is determined by the inverse of the temporal width of the sampling probe beam and the phase-matching condition in the ZnTe crystal. Although Gaal *et al.* have performed all of their initial experiments at a wavelength of 10  $\mu\text{m}$ , it is their opinion that 12-fs probe pulses should be able to make measurements down to wavelengths as short as 7.5  $\mu\text{m}$ . Using a 12-fs probe pulse and a ZnTe crystal has already enabled synchronized radiation at up to 40 THz to be detected<sup>5</sup>. The spectral resolution of a measurement is determined by the signal accumulation time and is equivalent to the length of the sampled sequential data. The timing accuracy of the sampling, important for determining the absolute temporal length of the sampled data is determined by the repetition rate of the probe beam. Thus the demonstrated technique is advantageous in that the repetition rate of a mode-locked laser can be tuned to any precision that may be required for most practical uses, as proved by the optical comb technique<sup>6</sup>.

As it should make it possible to detect any radiation source in the THz range, this

non-synchronized EO sampling technique is potentially very useful, but to fully exploit its potential it could benefit from improved sensitivity. The signal quality of the EO sampling technique depends on two parameters: the amount of phase retardation given by the EO crystal; and the optical noise, which determines the smallest phase retardation that can be detected. Future approaches for improving sensitivity could include ensuring that the optical noise at the detectors is at the shot noise level and using an EO crystal with a stronger nonlinearity or a thicker sample if phase-matching can be retained. The sampling rate also limits the spectral range of the measurement according to the Nyquist theorem. In competing methods, such as Fourier transform infrared measurements, the scanning speed of the interferometer can be lowered in accordance with the response time of the detector to obtain the desired frequency range. In the proposed method, however, the spectral range can be broadened simply by increasing the sampling rate. Investigations into alternative or optimized mixing materials will help open the door to practical applications.

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## MOLECULAR SENSORS

# Cavities lead the way

A group at Caltech have used an optical microcavity to perform label-free detection of single molecules for the first time. The work represents a milestone in the application of optical cavities in sensing, and could lead to the realization of ultrasensitive lab-on-a-chip systems.

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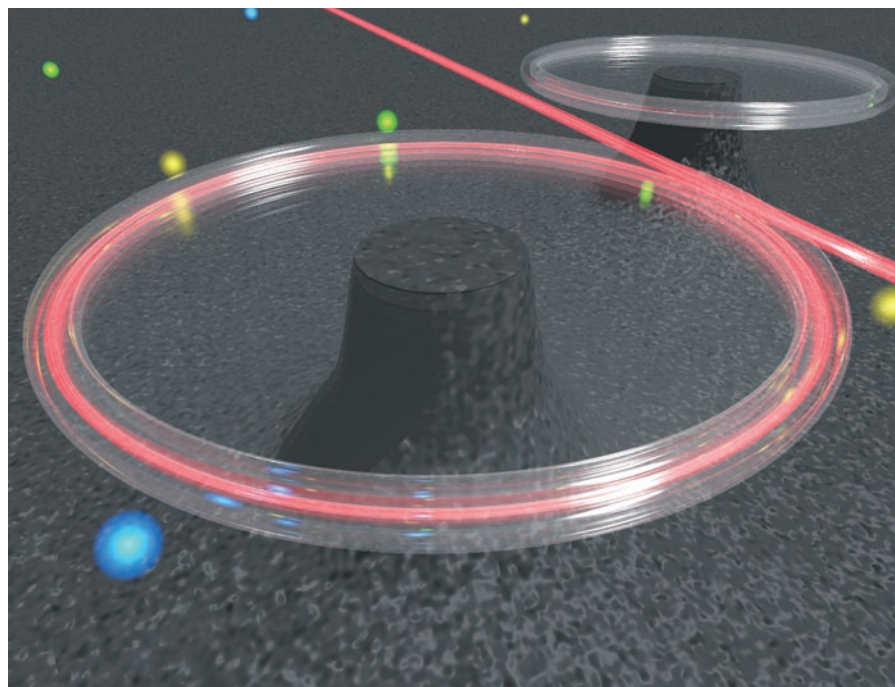
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**A** number of breakthroughs in the field of biochemical detection, made possible by advances in nanotechnology<sup>1,2</sup>, have greatly improved our understanding of the basic properties of matter at the single-molecule level.

Optical techniques, including Raman and fluorescence spectroscopy, are particularly suited to single-molecule detection because they are versatile, non-invasive and specific (that is, they can ignore uninteresting molecules). However, both of these techniques require molecules to be chemically labelled, either by metal nanoparticles or light-emitting nanocrystals<sup>2</sup>, to enhance their interaction with light. Groundbreaking work by Andrea Armani and co-workers

at Caltech, published in *Science*<sup>3</sup>, overcomes this labelling difficulty by taking advantage of photon recycling through the use of ultrahigh-quality-factor (-Q) optical cavities<sup>4</sup>.

The ideal biochemical sensor would be compact, portable, extremely sensitive (in other words, able to pick out individual molecules of interest) and would involve minimal specimen preparation and modification. A sensor platform with these properties would find immediate use



**Figure 1** A schematic of the microtoroidal resonator used by Armani *et al.*, which is immersed in a solution containing molecules to be studied. The coloured spheres represent the molecules under investigation, the red circle represents the light trapped in the ultrahigh- $Q$  microtoroidal cavity, and the straight red line represents the tapered optical fibre evanescently coupled to the cavity. The interaction of the cavity mode with molecules bound to the surface of the resonator offers a versatile mechanism by which single molecules can be detected. Image courtesy of Andrea Armani.

in important areas, such as biomedicine, pollution monitoring and national security. Single-molecule detectors could, for example, help to detect diseases early by monitoring ultrasmall quantities of body fluids.

In contrast to other optical single-molecule sensing techniques that require small (roughly 10 nm to 100 nm in size) ‘hot spots’ of light to irradiate the target, the approach taken by Armani and colleagues takes advantage of the long lifetime of photons trapped in an ultrahigh- $Q$  cavity. The  $Q$  of a cavity is a measure of its ability to store light, and is related to the ratio of energy stored in the cavity to the energy exchanged with the surroundings within one optical cycle. Therefore, each photon trapped in the ultrahigh- $Q$  cavity can have many chances to interact with a molecule under study. This photon-recycling mechanism is essential for the realization of the very-sensitive molecular probe reported by Armani and colleagues.

The new, label-free, single-molecule detector is based on a silica microtoroidal cavity<sup>4</sup> whose surface is specially functionalized so as to bond with target molecules. In particular, the cavity surface was coated with antibodies to capture

interleukin-2 antigen from serum. The detection of small quantities of interleukin-2 is important because it is an indicator that the immune system is impaired, and its concentration in serum changes in patients with childhood leukemia, for example. The cavity used by the Caltech group has a diameter of approximately 80  $\mu\text{m}$  and a  $Q$  value greater than  $10^8$ . Such a large  $Q$  gives rise to a long photon lifetime during which a molecule can be sampled by light in the cavity over a million times. By immersing the microcavity in a carrier fluid containing molecules of interest, a molecule can become attached to the cavity surface and induce a shift in the cavity resonance, owing to the direct effect of the molecule’s polarizability on the optical path length of the system. This shift can be easily picked up by monitoring the transmission of light through a tapered optical fibre that is evanescently coupled to the cavity (Fig. 1).

Similar sensing approaches have already been investigated<sup>5</sup> but single-molecule resolution has not been achieved until now. This is because the polarizability of an individual molecule unfortunately offers too small a perturbation to the comparatively large optical path length of a cavity (about 400  $\mu\text{m}$ ). To overcome

this limitation, Armani and co-workers rely on a thermo-optic effect<sup>3</sup> that helps to induce much larger shifts of the cavity resonance. A molecule attached to the surface of the cavity is slightly warmed up by absorbing a fraction of the large optical energy that propagates within the cavity. For example, approximately 100 kW of optical power circulates in the cavity when only 1 mW input power is injected into the fibre. According to Armani *et al.* the temperature change in the immediate vicinity of the molecule is on the order of a few degrees Celsius. This is not large enough to introduce observable changes in the molecule’s biological activity or to damage the molecule, but it is enough to warm up the microcavity and change the refractive index of the constituent silica. This indirect effect can produce a marked change in the optical path length of light in the cavity and the subsequent shift in the resonant frequency of the cavity (on the order of 25 fm) can be detected on a molecule by molecule basis.

The strength of the thermo-optic effect harnessed by the Caltech group is proportional to the amount of optical energy that propagates in the cavity. This, in turn, is related to the cavity  $Q$ . It is interesting to note that the sensing scheme used by Armani and co-workers — which is based on measuring only the power of the transmitted optical signal and not its spectrum — exploits the ultrahigh  $Q$  of the microtoroidal cavity twice. First, the large  $Q$  enables the cavity to be heated by the hot molecules. Second, it gives rise to narrow resonances, and thus smaller frequency shifts of these resonances can be easily resolved. The sensitivity of this cavity-based detector is proportional to  $Q^2$ , and is the key to achieving single-molecule detection in this system.

Several exciting possibilities could be explored to improve the sensitivity and specificity of the sensor, and, in particular, to take advantage of the thermo-optic effect at play. One approach would be to match the wavelength of the incident light used and the cavity resonance with the molecule’s absorption peaks, thereby opening up the possibility of performing single-molecule absorption spectroscopy. Another avenue would be to increase the energy stored in the cavity to obtain nonlinear Raman scattering from the detected molecule. Combining absorption or Raman spectroscopy (or both) in the same device would allow for not only the detection of a single molecule, but also for its complete identification.

It would also be interesting to take advantage of other cavity geometries and sizes<sup>6</sup>, as size does play a role in the

thermo-optic effect. As Armani and co-workers explain, the strength of the thermo-optic effect is actually proportional to  $Q/V$ , where  $V$  is the optical mode volume<sup>6</sup> of the cavity and is comparable to its physical volume. Planar photonic-crystal cavities<sup>7,8</sup> with  $Q$  values of around  $10^6$  and optical mode volumes well below a cubic wavelength have emerged as an attractive geometry for applications that require large  $Q/V$ . Therefore, the thermo-optic effect can be substantial in these cavities and single-molecule sensitivity could be possible. Owing to their small size and two-dimensional nature, photonic-crystal cavities could also be integrated into large

networks capable of sensitive, selective, and parallel interrogation of different molecules. Gain could even be introduced into the photonic-crystal cavity to further reduce the linewidth of its resonances, and improve the sensing scheme<sup>8</sup>.

The work by Armani and colleagues is an important step towards the realization of practical single-molecule, label-free biochemical sensors. It represents a milestone in the application of optical cavities to the field of biochemical sensing. Moreover, it is an excellent example of the advantages offered by optofluidic integration<sup>9,10</sup>, which combines cutting-edge optical devices with microfluidic delivery

schemes. It can be hoped that with this versatile detection scheme, new biological avenues can now be explored.

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## DISPLAYS

# Organic acquisition

The acquisition of one of the pioneers of polymer LED displays by a Japanese chemical giant looks set to give organic optoelectronics a welcome boost.

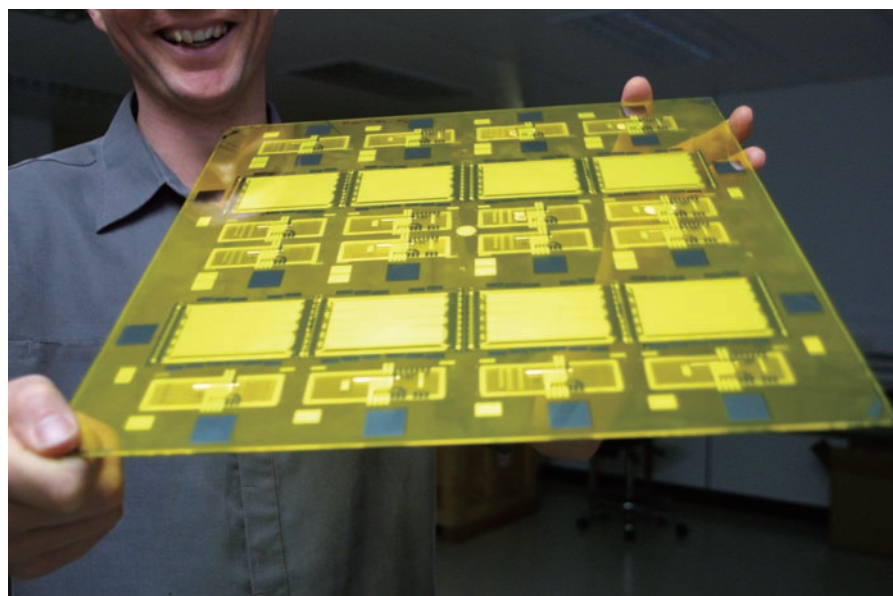
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**S**umitomo Chemical has announced the acquisition of UK-based Cambridge Display Technology (CDT) at the price of \$285 million. The investment by the Japanese chemical giant highlights the growing importance of organic materials to the optoelectronics market. Cambridge Display Technology is a spin-out company set up in 1992 to commercialize polymer-organic-LED (P-OLED) technology developed at Cambridge University. The company's main business is the licensing of their extensive intellectual property, including 60 granted patents with a further 70 pending, and fundamental and technical research aimed at enlarging this portfolio. Licensees already include Philips, DuPont, Seiko Epson and Osram. Over the past 15 years CDT has advanced the technology to the point that their polymer displays can now be found in a range of household products, including electric shavers and MP3 players.

The two big advantages of organic-based displays over competing technologies are their ease of production and low power consumption. Polymer OLEDs are made simply by spinning a thin film of



A welcome boost. The acquisition of CDT by Sumitomo Chemical could give P-OLED technology the investment it needs to increase its market share in the display sector.

a light-emitting polymer, the chemical structure of which determines the colour of the light emitted, onto a glass or plastic substrate that has transparent electrical contacts. A second contact can then be coated on top. The result is far less bulky

than cathode-ray-tube devices and does not require backlighting like LCDs. Polymer OLEDs make very thin displays, with an almost unlimited viewing angle, and can even be made on flexible substrates, opening the way to electronic paper.