NONLINEAR OPTICS

Graphene–silicon fusion

A single sheet of graphene dramatically changes the nonlinear response of a silicon photonic crystal, enabling ultralow-power optical bistability, self-induced regenerative oscillation and coherent four-wave mixing.

Kinam Kim, Seong-Ho Cho, and Chang-Won Lee

The realization of an ultrafast, all-optical switching device has long been a goal for scientists in the field of optical information technology. In 2010, researchers successfully demonstrated an optical switch based on the nonlinear optical response of a high-Q photonic crystal structure, which was made possible by the maturation of silicon fabrication technology[1]. Unfortunately, the low optical nonlinear coefficients of silicon and group III–V materials caused the final device to exhibit extremely narrow operating margins. The need for a wide operating margin has made it very difficult to realize a micro- or nanocavity capable of exhibiting nonlinear responses.

Reporting in *Nature Photonics*, Tingyi Gu and co-workers from Columbia University in the USA and the Institute of Microelectronics in Singapore have demonstrated that placing a sheet of graphene on top of the silicon photonic crystal structure can significantly enhance its nonlinear response[2].

The zero-bandgap linear electronic band structure of graphene — known as the Dirac-cone — has been studied for its potential use throughout a wide range of optoelectronic applications[3]. Graphene's linear dispersion provides a universal light absorption of around 2.3% from infrared to visible wavelengths. Therefore, unlike other semiconductor-based photonic devices, graphene can in principle be used to achieve ultrabroadband operation.

Gu *et al.* exploited an intriguing but lesser-known property of graphene: its optical nonlinearity[2]. In a previous study, Hendry *et al.* reported the use of graphene flakes in four-wave mixing[4]. They found that the nonlinear susceptibility $\chi^{(3)}$ of the graphene flakes was 10^{-7} esu, which is two orders of magnitude higher than that of a 4-nm-thick gold film. This value is 10^5 times higher than the values reported for silicon[5] — no other material has shown a nonlinear coefficient comparable to that of graphene. The peculiar electronic band structure of graphene enables all interband transitions to be resonant[6]. Low-carrier concentration and easy band filling provide very low-power saturated absorption, leading to mode-locking operation and hence to ultrafast pulsed laser operation[7,8].

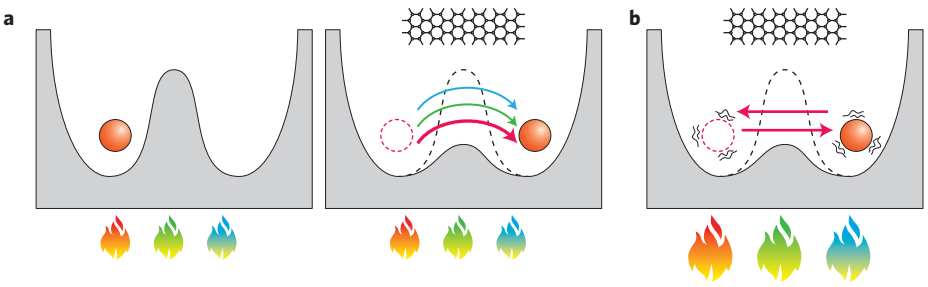

Figure 1 | Double-well model of excitation-dependent nonlinear optical phenomena in a graphene–silicon hybrid system. **a**, A graphene-on-silicon photonic crystal cavity reduces the potential barrier to facilitate optical bistability during heating. The colours of each flame represent the detuned wavelengths of the input laser. **b**, Increased heating power induces regenerative oscillation between the two potential wells.

The researchers chose to exploit the best features of both approaches: the large Q-factor of a silicon photonic crystal and the large nonlinearity of graphene. For a nonlinear response to be effective, the electromagnetic field inside the cavity must be captured and maintained using a photonic crystal with a very high Q-factor. However, in this approach, the fabrication tolerance is extremely narrow. Graphene, in contrast, does not require an ultrahigh Q-factor to induce a suitable nonlinearity in the cavity. Gu *et al.* adopted a silicon photonic crystal cavity scheme designed by Akahane *et al.*, the result of which showed an intrinsic Q-factor of 23,000 and a loaded Q-factor of 7,500 (ref. 9).

In order to convert the $\chi^{(3)}$ susceptibility of the hybrid structure into a straightforward nonlinear Kerr coefficient (n_2), the team employed a four-wave mixing set-up and a vectorial model that relied on finite-difference time-domain calculations. They obtained a value of $n_2 = 7.7 \times 10^{-17}$ m^2 W^{-1}, which is 17 times larger than that of silicon with the same structure and is similar to that of chalcogenide glass on a silicon photonic structure (7.0×10^{-17} m^2 W^{-1}; ref. 10). The hybrid graphene–silicon structure also exhibited an estimated threshold cavity energy of hundreds of attojoules.

This enhanced nonlinearity allowed Gu *et al.* to demonstrate two interesting behavioural aspects of the graphene–silicon hybrid structure: steady-state optical bistability and regenerative oscillation, both of which can be understood by considering a ball oscillating in a double potential well (Fig. 1). In this analogy, the ball is being heated by an external source (the laser). Without graphene, the potential barrier is too high for the ball to jump into the next well. The role of graphene is to lower the potential barrier and thus allow the ball to jump over the wall (Fig. 1a). The researchers observed input–output optical bistability in both steady-state and pulsed-mode operation, even at modest Q-factors. Owing to fast carrier relaxation followed by slow thermal relaxation, only the positive detuning mode showed bistability during the quasi-steady-state sweeping of the input power.

Increasing the input power causes the ball to bounce back and forth — a phenomenon known as regenerative oscillation (Fig. 1b). This oscillatory behaviour usually takes place under a high-intensity pulsed mode. In the graphene–silicon hybrid device, however, this regenerative oscillation appeared even

in a quasi-steady-state pumping condition with triangular input modulation due to competition between phonon (~10 ns) and carrier (~200 ps) relaxation. Gu *et al.* observed an oscillation period of 106 MHz, which will be useful for generating microwaves. This oscillation period differs from the theoretical coupled-mode theory value of 50 MHz due to insufficient information in the model, including a lack of knowledge about the time-varying *Q*-factor of the cavity.

The work of Gu *et al.* provides a very interesting outlook. The development of suitable fabrication techniques should allow graphene sheets to be placed between identical vertically stacked photonic cavities to achieve better mode-overlapping. Graphene sheets could also be used to reduce the threshold power of all-optical nonlinear switches and to achieve non-degenerate optical parametric generation. Graphene may be one of the simplest solid-state materials, but its wealth of fascinating behaviour continues to surprise us, year after year. ❐

Kinam Kim, Seong-Ho Cho and Chang-Won Lee are at Samsung Electronics, San 14, Nongseo-dong, Giheung-gu, Yongin-si, Gyeonggi-do 446-712, Korea.
e-mail: kn_kim@samsung.com

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MICROANALYSIS

Single-atom X-ray spectroscopy

Advances in electron optics and X-ray detection are opening up the periodic table to one of the ultimate goals of microanalysis — single-atom spectroscopy.

Michael Walls

The electron microscope is used both as a microanalytical tool and to produce high-resolution micrographs. Energy-dispersive X-ray spectroscopy (EDX; confusingly also known by a number of different acronyms, including EDS and EDXS) is one of the principal spectroscopic techniques used to identify and quantify chemical species in transmission electron microscopy. The technique involves measuring the energies of X-rays emitted by the targeted region of a sample under an incident electron beam. Peaks in the spectrum correspond to specific electronic transitions and thus yield chemical information about the sample. Writing in *Nature Photonics*, Suenaga *et al.* have now taken on what might be considered the ultimate challenge for a microspectroscopic technique such as EDX: detecting the signal from a single atom[1]. Similar experiments on silicon and platinum atoms were also recently reported by Lovejoy *et al.*[2]

In recent years, researchers have made significant improvements to transmission electron microscopy — particularly for devices operating in scanning mode. Among the many technological breakthroughs, perhaps the most important was the introduction of spherical aberration correctors, which enabled the generation of high-current sub-ångström-sized electron probes and thus opened up the path towards atomic-resolution spectroscopy[3].

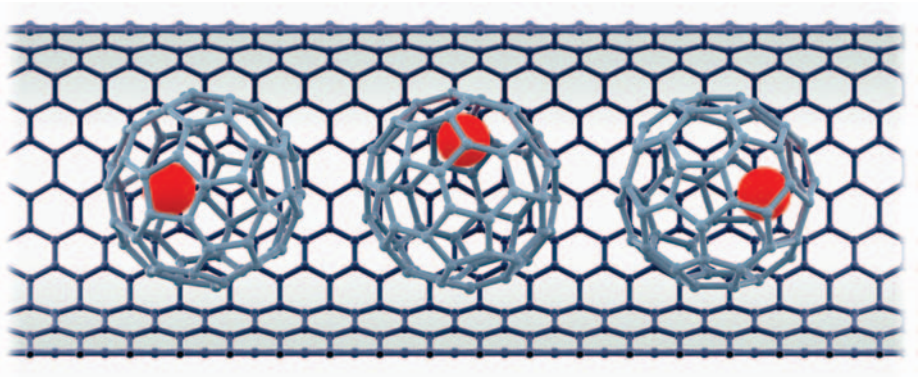

Figure 1 | The 'peapod' structure of Suenaga *et al.*: single erbium atoms trapped within fullerene cages.

Another spectroscopic technique typically employed in transmission electron microscopy is electron energy-loss spectroscopy (EELS), which involves measuring the energy loss of the fast electrons that have traversed the thin sample (usually no more than a few tens of nanometres). EELS has conventionally been considered as the more 'advanced' technique, particularly owing to its vastly superior energy resolution (<1 eV, compared with >100eV for EDX), but it is more difficult to apply and interpret. EELS was also the first technique used to map individual chemical species with atomic spatial resolution in crystals[4]. For many years, researchers assumed that EDX would never have the same spatial resolution as EELS, given the limitations that arise not only from the size of the probe that can be generated, but also from its interaction volume in a sample, which was thought to be much larger in EDX than in EELS. However, recent work has clearly overturned that assumption: the first atomic-resolution EDX maps were demonstrated around two years ago, showing (albeit faintly) individual columns of atoms in aligned crystals[5]. Since then, researchers have continued to improve the quality of the maps produced by EDX. This is due partly to great improvements in spectrometer sensitivities and speeds,