

limits inherent to the scheme, the method demonstrated by Lavoie *et al.* has the potential to be compatible with the optical components typically used in telecommunications applications.

Work remains to be done, however, before this method can be used in applications. For an average laser pulse power of 300 mW, the observed upconversion efficiency was only 0.06% — that is, out of 1,667 single photons sent into the nonlinear crystal, only one resulted in the emission of a frequency-shifted and bandwidth-compressed single photon. In addition, the compressed bandwidth of 43 GHz is still significantly larger than the megahertz-to-gigahertz bandwidths of typical quantum memory implementations⁴. And although the compression was shown to preserve temporal correlations, the researchers did not demonstrate that entanglement was preserved throughout the process. More work is needed to address these

challenges, but the solutions certainly seem to be within reach and the future looks promising. For example, higher upconversion efficiencies should be possible by increasing the power of the strong laser pulses and using periodically poled crystals⁶ for the upconversion process. Bandwidth compression to 1 GHz should ultimately be achievable by this method and polarization-insensitive upconversion⁷ should enable entanglement preservation.

In addition to quantum-information applications, arbitrary waveform manipulation of quantum states of light has many other possible uses. One example noted by Lavoie *et al.* is its potential to enable ultrafast timing measurements by using slow detectors to transform arrival time differences into frequency differences. Assuming the experimental parameters described by Lavoie *et al.*, this approach would allow the discrimination of time bins separated by as little as 0.6 ps. Through continued refinement of these

techniques, the work of Lavoie *et al.* may ultimately make made-to-order single-photon waveforms commonplace, and consequently help realize next-generation technologies in computing and communication. □

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GRAPHENE PLASMONICS

Damping of plasmons in graphene

The damping of surface plasmons hinders the realization of nanophotonic devices. Researchers have now uncovered some of the mid-infrared damping mechanisms for plasmons in graphene, which offer a number of unique and interesting properties.

Hrvoje Buljan, Marinko Jablan and Marin Soljačić

The development of nanophotonic devices requires the ability to confine and control light at scales much smaller than the wavelength of light. One, and perhaps the only, viable strategy for attaining this goal is to use surface plasmons — collective excitations of electrons and light at the interface between a conductor and a dielectric¹. Tight confinement can be attained at light frequencies close to the resonant frequency of the conductor because the plasmon wavelength is generally much smaller than the wavelength of light in air. A shorter wavelength implies a smaller diffraction limit in the plane of propagation, and thus rapid exponential decay (scaled by the plasmon wavelength) of the electromagnetic field away from the interface. However, there is a trade-off: the strong subwavelength confinement of light is generally accompanied by stronger interaction of the light with the lossy conductor, resulting in shorter propagation lengths (greater damping) of plasmonic excitations. This is a major obstacle to the

development of nanophotonic devices. With the isolation of graphene — a single sheet of carbon atoms organized in a honeycomb lattice with extremely interesting electrical and optical properties — on a dielectric substrate², graphene plasmons became a hot research topic in nanophotonics because of their ability to confine light strongly, the possibility of control using the gate voltage, and potentially smaller losses than previous systems³.

Writing in *Nature Photonics*, Yan and co-workers from IBM (USA) and Instituto de Ciencia de Materiales de Madrid (Spain) have determined some of the damping pathways for mid-infrared plasmons in graphene nanostructures⁴. Their experiment is depicted in Fig. 1. Graphene ribbons 50–250 nm wide were placed on a dielectric substrate and then illuminated from above by mid-infrared radiation with wavelengths in the range 4–15 μm . The transmission of radiation through this system was measured, which provided information on the excitations

in the system at a given frequency and wavenumber. The key result is the ability to disentangle various mechanisms that induce plasmon damping. The plasmon linewidth was found to increase greatly with decreasing ribbon width, indicating a dominant contribution from edge scattering effects. Furthermore, a substantial increase in the plasmon linewidth was observed at excitation energies higher than the energy of graphene's intrinsic optical phonons (a type of lattice vibration) at 1,580 cm^{-1} (0.2 eV, or a wavelength of $\sim 6 \mu\text{m}$ for photons in air); this indicates a damping channel through which a plasmon excites an optical phonon and an electron–hole pair.

The desirable strong subwavelength confinement of light is associated with the difficulty to excite plasmons experimentally. The plasmon wavelength can be much smaller than the wavelength of free-space photons. Consequently, these free-space photons have insufficient momentum to excite plasmons directly. To overcome this

momentum mismatch, Yan and co-workers use graphene sheets of finite width, called graphene ribbons. When the polarization of the incident radiation is perpendicular to the ribbons, the electric field induces oscillations of the charge across the ribbons, yielding a reduced transmission T_{\perp} . These charge oscillations are in fact plasmon oscillations whose wavelength is approximately half the nanoribbon width (Fig. 1). The transmission T_{\perp} decreases as the frequency of the incident wave approaches the plasmon frequency. Plasmons cannot be excited when the polarization of the incident wave is parallel to the nanoribbons because of a momentum mismatch, but the transmission T_{\parallel} serves as a reference point for obtaining the so-called extinction spectrum $1 - T_{\perp}/T_{\parallel}$, which contains information on the excitations in this system. To obtain the plasmon dispersion relation, the researchers produced nanoribbons with widths down to 50 nm. Measurements of the extinction spectra in samples with different nanoribbon widths provide the plasmon dispersion $\omega_{\text{pl}}(q_{\text{pl}})$ and the lifetime (Fig. 1). Experiments with micrometre-sized ribbons have generated plasmons in graphene at terahertz frequencies^{5,6}. The plasmons supported by nanoribbons have infrared frequencies used in telecommunications, thus providing strong motivation for their study^{3,4}. However, the physics becomes more complex because graphene plasmons in the infrared region interact with optical phonons.

The experiments of Yan *et al.* have revealed the roles of the two types of phonons that participate in this interaction. Phonons of the first type are generated when graphene is on a polar dielectric substrate such as SiO₂; these are surface polar phonons at 806 cm⁻¹ and 1,168 cm⁻¹ that interact strongly with plasmons. In the long-wavelength limit, the dispersion of graphene plasmons is given by $\omega_{\text{pl}} \propto \sqrt{q_{\text{pl}}}$. This was observed in the terahertz range^{5,6}; it also applies to infrared frequencies⁴, with at least one exception: when the plasmon frequency is close to the surface phonon frequency, they form a coupled (hybrid) plasmon–phonon mode^{4,7} that yields a characteristic dispersion (see Fig. 3 in ref. 4). The second type covers intrinsic optical phonons at 1,580 cm⁻¹ in graphene, which contribute to plasmon losses at frequencies above 1,580 cm⁻¹.

It is essential to gain an understanding of plasmon losses in graphene before plasmons can be used in nanophotonics³. There are several possible damping pathways for plasmons in graphene. The dominant decay mechanism is Landau damping, in which a plasmon emits an electron–hole pair. This damping can be eliminated by using doping

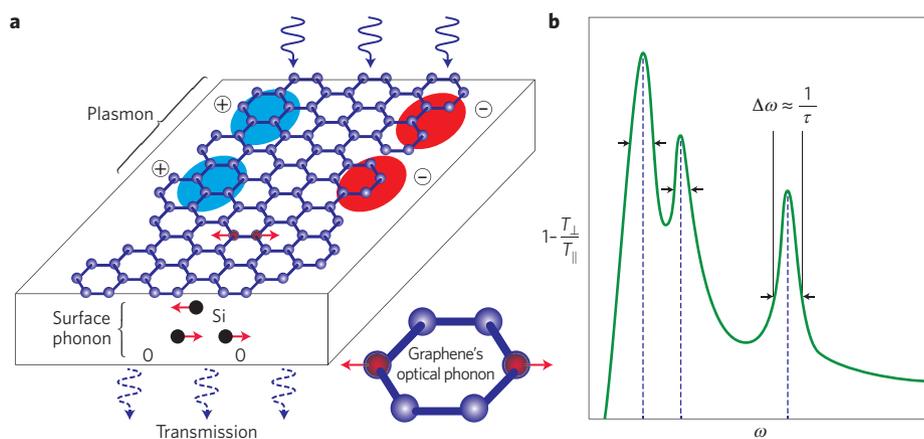


Figure 1 | Sketch of the experiment by Yan and co-workers⁴. **a**, Illustration of plasmons, phonons and experimental set-up. **b**, The characteristic response (extinction spectrum) used to measure the frequency and lifetime of plasmonic excitations. The peaks correspond to coupled plasmon surface phonon modes, and their widths (damping rates) correspond to edge scattering and interaction with graphene's optical phonons.

to increase the carrier concentration, as the Pauli principle then prohibits such transitions in a large region of the (q, ω) space. In contrast to bulk three-dimensional metallic structures, two-dimensional graphene can be doped by applying an electrostatic gate voltage. This leads to exciting and intriguing possibilities for potential applications as gating can be used to change dispersion or even turn plasmons on and off.

Other possible damping pathways involve scattering from various types of impurities or defects, interactions of electrons with phonons and mutual electron–electron interactions. The effect of electron scattering from impurities can be partially extracted from d.c. measurements, which have indicated very large mobilities (that is, very long electron lifetimes). Because improving the quality of the samples should reduce such effects, scattering from impurities is not expected to prevent the realization of low plasmon losses. Analysis of the influence of graphene's optical phonons on plasmon losses revealed that when the plasmon frequency exceeds the optical phonon frequency of 1,580 cm⁻¹, there will be non-negligible losses through simultaneous emission of an optical phonon and an electron–hole pair³. The experiment of Yan *et al.* indeed demonstrated that the plasmon linewidth strongly increases at frequencies above 1,580 cm⁻¹. Potential applications requiring lower losses should therefore preferably use frequencies below optical phonon frequencies.

One alternative means of exciting plasmons — besides using nano- or microribbons or other structures such as disk arrays⁶ — is scattering-type scanning near-field optical microscopy^{7–9}. This

technique involves illuminating a metallic tip with infrared light. If the tip is sharp, it will 'emit' radiation over a large span in momentum space that can be used to excite plasmons locally. These plasmons will be reflected from the edge of the sample and couple, through the tip, with backscattered light; this light can then be measured, thus providing information about the plasmon amplitude below the tip.

Two groups have recently^{7–9} imaged plasmons by this method and investigated their properties (including their lifetimes) for different doping concentrations; experiments have shown that the lifetime is independent of the doping concentration⁹. A previous measurement of the absorption of normally incident light in graphene revealed large absorption, even in the region in which electron–hole excitations are forbidden¹⁰. However, this high absorption (which is related to plasmon losses) has yet to be quantitatively accounted by theory: theoretical calculations have predicted a lower absorption than that observed in ref. 10. One proposed way of resolving this discrepancy is to perform more sophisticated calculations that incorporate electron–electron interactions^{9,10}. Nevertheless, the reported performances of tunable-graphene-based plasmonic devices are much more promising than those of non-tunable metal-based structures^{4–9}.

The experiments of Yan *et al.* represent an important step towards unravelling the damping pathways of graphene plasmons in the infrared range. However, plasmon losses in the regime where Landau damping does not occur are not yet quantitatively understood. The complete resolution of this

issue is very important for nanophotonics and is thus attractive for further study. □

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VIEW FROM... 2013 PHOTONICS WEST

Solid cooling

Cooling materials using interaction with light has come a long way in the past 20 years. Researchers at the recent 2013 Photonics West showed that they can cool new types of materials and more can be expected in the future using new approaches.

David Pile

At the beginning of each year, San Francisco hosts one of the biggest and broadest optics conferences in the world. This year, SPIE's Photonics West conference took place on 2–7 February 2013 at the Moscone Center, which is conveniently located close to the Embarcadero, Mission District and numerous other popular destinations.

As the weight of the 415-page technical program indicates, the meeting was incredibly broad. Major themes were grouped under 'sub-conferences' named "Green Photonics", "BiOS", "LASE", "MOEMS-MEMS" and "OPTO". The laser cooling of solids and dense gases (also known as optical refrigeration), although not one of the 'biggest' topics at the meeting, provided some focused sessions that demonstrated the significant recent progress. Optical refrigeration is of interest because it can be realized using compact, cryogen-free systems and does not generate vibrations.

One of the early sessions addressed the problem of cooling optically important rare-earth-doped material systems. Seth Melgaard and colleagues from the USA (Air Force Research Laboratory and the University of New Mexico) and Italy (Università di Pisa) discussed several milestones. They have cooled a 5%-wt. Yb:YLF crystal to ~118 K — the minimum achievable temperature for their pump centred at 1,020.7 nm. Melgaard explained that they improved the cooling efficiency by investigating the effect of doping concentration using 1%, 5%, 7% and 10%-wt. Yb:YLF crystals. They noticed that the parasitic background absorption decreased with increasing



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Wim Leemans of Lawrence Berkeley National Laboratory giving a plenary talk on the acceleration of particles using laser-generated plasmas.

doping concentration. The ratio of the background absorption to the resonant absorption of the 10%-wt. Yb:YLF crystal was reduced giving the 10%-wt. Yb:YLF crystal a minimum achievable temperature of 93 K. So far, they have achieved a temperature of 114 K; Melgaard emphasizes that, although this is above the minimum achievable temperature, it is by far the coldest solid-state optical technology in the world. "We have identified a path towards achieving solid-state cooling to the liquid-nitrogen temperature of 77 K," Melgaard told *Nature Photonics*. "Because of the trend in parasitic background absorption, elemental analysis was performed and it

identified iron as the main contributor to parasitic heating. By reducing the iron concentration through purification of the starting materials, model predictions show that solid-state cooling to liquid-nitrogen temperature is within reach."

Also on the topic of cooling rare-earth-doped systems, Angel Garcia-Adeva (Universidad del País Vasco) and colleagues from Spain and France discussed work on using light to cool erbium-doped oxysulphide crystal powders. Garcia-Adeva explained to *Nature Photonics* that lanthanum oxysulphide, a uniaxial P3m wide-bandgap semiconductor material, is an excellent host lattice for trivalent rare-earth ions, as its maximum phonon energy of about 400 cm⁻¹ enhances efficient upconversion processes while strongly suppressing nonradiative multiphonon losses. "Our group is currently investigating Er³⁺-doped La₂O₂S crystal powders as a promising candidate for all-optical cooling. This material exhibits an efficient infrared-to-visible upconversion under excitation in the 800–870 nm band," Garcia-Adeva told *Nature Photonics*. "Indeed, we have obtained efficient upconversion-assisted local cooling when pumping in resonance with a two- or three-phonon annihilation process. Even though this investigation is still underway, our preliminary results suggest that efficient bulk optical cooling could soon be achieved in this system."

Galina Nemova from Polytechnique de Montréal (Canada) discussed a new theoretical scheme for the laser cooling of rare-earth-doped direct-bandgap semiconductors. Nemova explained that the cooling cycle in conventional laser cooling schemes is based on electron