even when focused to a wavelength-scale cross-section[6]. Bessel beams have been used to build optical traps that extend for millimetres along the optical axis and also to push particles precisely over very large distances[7]. Although the finite aperture of the lens system imposes some limitations on the range of the beam[6], the proposal of Chen *et al.* could lead to the practical realization of a Bessel tractor beam acting on particles at relatively far (macroscopic) distances from the light source.

Because the optical pulling force is strongly related to the existence of interference effects between multipole radiation fields, it is appropriate to investigate the different types and sizes of particle that could be pulled by such a tractor beam. Chen *et al.* show that the optical pulling force should be possible for polystyrene spheres with relatively low refractive index. For a given laser wavelength, the required interference condition is only obtained for particular ranges of sphere radii, and, vice versa, for a given sphere radius, the pulling force is only possible in specific spectral regimes. It would not be possible to pull arbitrary particles with the proposed tractor beam. On the other hand, the dependency of this technique on particle size provides intriguing possibilities for particle sorting.

Chen *et al.* also explore the response of particles that have both electric and magnetic responses (that is, non-zero magnetic permeability, μ, and electric permittivity, ε). In 1983, Kerker *et al.*[8] theoretically established the remarkable scattering effects of simple dipole scatterers by concerning the suppression or minimization of backwards scattering for cases when $\varepsilon = \mu$. As expected, the optical pulling force considered by Chen *et al.* is optimized around this condition for such particles[1]. However, researchers have yet to propose a particle that might present these interesting properties in the visible or infrared regions.

Researchers recently demonstrated that submicrometre semiconductor spheres are capable of exhibiting dipolar magnetic and electric responses[9]. In these particles, interference between the electric and magnetic dipolar fields can lead to anisotropic angular distributions of scattered intensity, including zero backwards and almost-zero forwards scattering intensities at specific wavelengths[10]. Related interference phenomena between electric dipolar and quadrupolar resonances in plasmonic nanoparticles lead to strong forwards scattering at specific wavelengths[11].

In addition, these particles could be attached to almost transparent objects such as biological macromolecules. The optical pulling force could then be used to attract even transparent objects by using multipolar particles as pulling probes. The results of Chen *et al.* represent a clear breakthrough in our current understanding of optical forces, and the concept of the optical pulling force will have a significant impact in the multidisciplinary field of optical manipulation. ❐

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NONLINEAR OPTICS

Exploiting disparity

Although two-photon absorption in a semiconductor is typically a very weak effect, the rate of absorption increases dramatically when the two photons have very dissimilar wavelengths, enabling applications such as ultrafast optical sampling and room-temperature mid-infrared detection.

Thomas E. Murphy

Two-photon absorption (TPA) is a nonlinear process that occurs in semiconductors when two photons are absorbed simultaneously, generating a single electron–hole pair in the process. When this process takes place in a photodiode, the resulting photocurrent can be directly measured for use in practical applications. However, TPA is typically regarded as a weak nonlinear effect.

Now, reporting in *Nature Photonics*, Dmitry Fishman, Claudiu Cirloganu and colleagues from the University of Central Florida in the USA demonstrate that the rate of TPA can be increased by 2–3 orders of magnitude when the two interacting photons involved in the process have very disparate wavelengths[1]. The researchers exploit this surprising effect to achieve the time-gated detection of mid-infrared radiation at 5.6 μm, with a sensitivity comparable to that of a cooled HgCdTe detector.

In conventional linear photodetection, photons with energy above the bandgap are absorbed to produce a photocurrent that is proportional to the optical power incident on the detector. In contrast, TPA can be observed even when the photon energy is below the bandgap, and results in a photocurrent that grows quadratically with the incident power. Unlike linear absorption, the rate of TPA can be enhanced by concentrating the photons in space (through focusing) or time (by using short pulses).

Two-photon absorption is widely used in ultrafast pulse measurements as a convenient substitute for sum-frequency generation, followed by photodetection of the mixing product[2]. Indeed, TPA provides several significant advantages over sum-frequency generation: it does not require phase matching, operates over a very broad optical bandwidth, exhibits less polarization dependence and the mixed product of the two optical signals can be measured directly as a photocurrent, without the need for external detection. In the telecommunications band, TPA can be observed using inexpensive silicon photodiodes even with continuous-wave (CW) illumination at milliwatt levels, and has been used for optical clock recovery, optical sampling and performance monitoring[3,4].

Most applications of TPA focus on the degenerate or nearly degenerate case, in which the two interacting photons have equal or commensurate wavelengths. In contrast, the work by Fishman *et al.* exploits the non-degenerate case, where the two absorbed photons have different wavelengths such that although the sum of their photon energies exceeds the bandgap, neither photon alone has sufficient energy to overcome the bandgap. When the two interacting optical beams coincide in time and overlap in space, the photocurrent generated by non-degenerate TPA is proportional to the product of their respective optical powers. In this way, the presence of a strong optical 'gating' pulse at one wavelength can enable the linear detection of photons at the other wavelength.

In the 1980s, researchers developed a simple quantum-mechanical theory that has been used to successfully predict the observed scaling relationship between the bandgap, photon energy and TPA rate over a wide range of experimental conditions[5]. In the degenerate case, the TPA coefficient increases from zero at the half-bandgap ($\hbar\omega = E_{gap}/2$) to a maximum value at 2/3 of the bandgap. This theory was later generalized to include the case of non-degenerate TPA[6], for which the nonlinear photocurrent is predicted to scale in proportion to

$$F_2^{\,symm}(x_1, x_2) = \frac{2(x_1 + x_2 - 1)^{3/2}(x_1 + x_2)}{(4x_1x_2)^4} \qquad (1)$$

where $x_1 = \hbar\omega_1/E_{gap}$ is the photon energy normalized to the bandgap, and x_2 is similarly defined for the second wavelength. Many had assumed that the nonlinear absorption rate depends primarily on the photon energy sum $x_1 + x_2$, and not on the individual factors x_1 and x_2. However, equation (1) exhibits a remarkable singularity when either x_1 or x_2 becomes small. Figure 1 shows the enhancement factor $F_2^{\,symm}(x_1, x_2)$ over the region where TPA is observable. What many had considered to be the optimal condition ($x_1 = x_2 = 2/3$) is actually a saddle-point: along the degeneracy line ($x_1 = x_2$), the function $F_2^{\,symm}$ reaches a maximum, whereas in the orthogonal direction, it reaches a minimum.

Although the theoretical scaling relationship for non-degenerate TPA was predicted several years ago, the dramatic enhancement in nonlinear absorption that occurs for highly non-degenerate photons is not widely appreciated. The work of Fishman *et al.* is the first experimentally verification and exploitation of this enhancement.

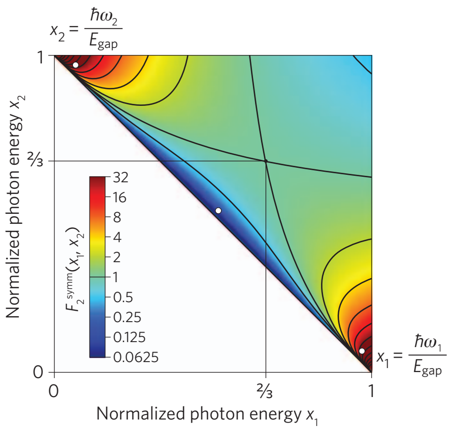

Figure 1 | Theoretically predicted enhancement in the rate of non-degenerate TPA, $F_2^{\,symm}(x_1, x_2)$, as a function of the normalized photon energies x_1 and x_2. The triangular region contained in the plot spans the range of conditions for which TPA can occur ($x_1 + x_2 > 1$) and where linear absorption is prohibited ($x_1 < 1$, $x_2 < 1$). Adjacent contour lines are separated by a factor of two, and the contour heights are given relative to the saddle-point at $x_1 = x_2 = 2/3$. The white circles mark the experimental conditions reported by Fishman *et al.*[1] for a GaN detector, illustrating the dramatic enhancement in nonlinear absorption that can occur when the two interacting photons have very disparate wavelengths.

The white circles shown in Fig. 1 indicate three of the experimental conditions considered by Fishman *et al.* using a GaN photodiode[1]. The two circles in the corners correspond to non-degenerate TPA at wavelengths of 5.6 μm and 390 nm, and the point near the centre corresponds to degenerate TPA at a wavelength of 790 nm. All three cases have approximately the same cumulative photon energy, yet the extremely non-degenerate cases are predicted and experimentally shown to produce a much stronger nonlinear photocurrent than the degenerate case. The significance of this result is not simply that a wide-bandgap GaN photodiode can be used to detect mid-infrared photons, but that the efficiency of this process is 100–1,000 times higher than that of degenerate TPA[1]. This dramatic enhancement is what allowed Fishman *et al.* to achieve mid-infrared detection sensitivities comparable to cryogenically cooled HgCdTe detectors.

The converse case, in which the researchers used mid-infrared pulses to perform gated detection of 390 nm light[1], also deserves equal mention. Although conventional photodiodes are capable of detecting 390 nm radiation, the use of a long-wavelength gating pulse with a wide-bandgap photodiode allows the signal of interest to be sampled at a temporal resolution that is limited primarily by the sampling pulsewidth. In this case, the strong sampling pulses do not generate a background signal of their own through degenerate TPA, which could allow for background-free optical sampling with unprecedented sensitivity.

The work of Fishman *et al.* represents more than simply an important experimental verification of previously derived theoretical scaling laws: it also confirms that the singularity present in equation (1) is a readily observable effect that can be exploited for a variety of important applications, including mid-infrared detection.

At a fundamental level, it remains to be seen whether the third-order nonlinearity retains its ultrafast response time in the highly non-degenerate regime, where the nonlinear properties are expected to be far more dispersive. The polarization dependence and anisotropy of TPA in this regime also deserve further experimental investigation. Another compelling question posed by this work is whether the real part of the nonlinear susceptibility exhibits a similar enhancement under non-degenerate conditions, which could enable cross-phase modulation and optical switching with unprecedented sensitivity.

From an engineering point of view, the GaN photodiode used in this work was clearly not intended for nonlinear optics. By optimizing the structure of the detector, or perhaps by using waveguide-based detection, it may be possible to achieve mid-infrared detection using CW or quasi-CW gating signals. Another potential application is for mid-infrared imaging, which could employ either an array of parallel detectors or a spatially scanned gating signal.

At both a fundamental and practical level, the use of highly disparate wavelengths to achieve dramatic enhancements to nonlinear absorption is an intriguing effect that holds promise for a host of future applications. ❒

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