Anapole Mediated Giant Photothermal Nonlinearity for Super-Localization Nanoscopy

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Featured with a plethora of electric and magnetic Mie resonances, high index dielectric nanostructures offer an unparalleled platform to concentrate light-matter interactions at the nanoscale for exploring exotic physical phenomena and underpinning tremendous emerging applications. By integrating extraordinary features of far-field scattering control and near-field concentration from radiationless anapole states, here, we demonstrate a giant photothermal nonlinearity in single subwavelength-sized silicon nanodisks for super-localization nanoscopy. The nanoscale energy concentration and consequent near-field enhancements mediated by the anapole mode yield a reversible nonlinear scattering cross-section modulation up to 200%, unveiling a record-high nonlinear index change up to 0.5 at mild incident light intensities of MW/cm². The observed photothermal nonlinearity showcases four orders of magnitude enhancement compared with that of unstructured bulk silicon, as well as nearly one order of magnitude higher than that through the radiative electric dipolar mode. The anapole-driven nonlinear scattering behaviors of such silicon nanodisks can empower distinctive point spread functions in confocal reflectance imaging, enabling super-resolution nanoscopy with a striking localization accuracy approaching 40 nm. Our findings shed new light on active silicon photonics leveraging all the benefits from optical anapoles and open up an unprecedented perspective in non-invasive super-localization nanoscopy.
All-dielectric, high refractive index nanostructures offer marvelous ability to efficiently confine and manipulate light at the nanoscale based on their enticing potentials to control both optically induced electric and magnetic Mie resonances\textsuperscript{1-4}. During recent years, the interplays of a wealth of Mie-type resonant modes have unveiled many novel physical phenomena such as unidirectional scattering\textsuperscript{5-8}, magnetic Fano resonances\textsuperscript{9}, bound states in the continuum\textsuperscript{10,11} and nonradiating optical anapoles\textsuperscript{12,13}. Among these intriguing observations originating from multimodal interference in dielectric nanostructures, optical anapole holds extraordinary features characterized by vanishing far-field scattering accompanied with highly nontrivial near-field distributions. The anapole state is strongly related to the excitation of a toroidal dipole (TD), which is out-of-phase with the coexisting electric dipole (ED), resulting in the destructive interference in the far field\textsuperscript{14}. In the meantime, the optical induced displacement currents inside the nanostructures produce tightly confined near-fields to resonantly enhance the local density of photonic states.

The discovery of the general existence of optical anapoles in dielectric nanostructures immediately spurred intensive investigations on diverse applications. Engineered anapole states have been used to tailor light scattering in the far field for inducing optical transparency\textsuperscript{15,16} or rendering novel pure magnetic dipole source\textsuperscript{17}. Functional anapole metamaterials and metasurfaces featuring high quality factors have been shown great potentials in optical modulation and sensing\textsuperscript{18,19}. More importantly, energy concentration in the subwavelength volume associated with anapole modes facilitates tremendous near-field effects. As such, onrushing developments in boosted light–matter interactions utilizing anapole-mediated hotspots including nonlinear harmonic generation\textsuperscript{20-22}, nanoscale lasing\textsuperscript{23}, broadband absorption\textsuperscript{24}, strong coupling with plasmon\textsuperscript{25} or molecular excitons\textsuperscript{26,27} and enhanced Raman spectroscopy\textsuperscript{28,29} have been witnessed. With rapid developments featured, the full potential amalgamating all the benefits in both far-field and near-field from optical anapoles to further advance nanophotonic devices remains tantalizing perspective.
In this report, we discover a giant photothermal nonlinearity mediated by anapole states within a single silicon (Si) resonator and demonstrate its nonlinear scattering for super-localization nanoscopy. Leveraging their maximized electromagnetic near-fields, anapole modes mediate a boosted photothermal nonlinearity by four orders of magnitude higher than that of bulk Si, or nearly one-order-of-magnitude outperforming the radiative ED-driven enhancement for similar sized Si nanostructures. A record-high photothermal refractive index change $\Delta n$ up to 0.5 upon a mild laser radiance intensity $I$ of $MW/cm^2$ can be achieved through optically pumping a Si nanodisk at the wavelength close to the anapole mode. The giant photothermal nonlinearity thus offers an active mechanism for dynamic tuning of far-field radiation from multipolar modes. The normalized scattering cross-sections can be reversibly regulated from -0.9 to 1.1, corresponding to 200% self-modulation spanning from scattering suppression to enhancement, due to the progressive transition of dominant modes from bright state to low-radiating dark state and further moving towards the bright state. Consequently, distinctive point spread functions (PSFs) of single Si nanodisks in confocal reflection imaging are revealed at various scattering states. Employing such exotic optical mode imaging, super-localization of silicon nanodisks with a striking accuracy approaching 40 nm has been achieved. Compared to existing techniques, anapole-mediated photothermal nonlinearity offers extraordinary features like non-invasive all-optical, fast and large modulation range. Our findings shed new light on active photonics harnessing dielectric nanostructures for on-demand tunability and open up an unprecedented perspective in non-invasive super-resolution nanoscopy.

Results

The principle is schematically illustrated in Fig. 1. A Si nanodisk illuminated by a continuous wave (CW) laser beam converts incident light into heat, which raises the disk temperature substantially. Photothermal mechanism responsible for the refractive index variation, manifested by the continuous red-shifting of Mie resonances under light excitation, allows for tunable optical responses of the silicon
nanodisk. With rationally designed dimensions of the Si nanodisk, the associated anapole mode can be actively engineered and tuned in the vicinity of the excitation wavelength. Hence, the illumination radiance required for the large refractive index modulation of the Si nanodisk can be efficiently reduced via anapole-assisted absorption enhancement. As the laser intensity increases, excitation of initial scattering bright mode experiences shifting towards anapole mode (will be discussed in detail below), which induces saturation of scattering (SS)\textsuperscript{33} and significant reduction of the scattering intensity (Fig. 1b and c). Further shifting the excitation away from non-radiating anapole to ED mode exhibits a sharp increase of scattering intensity, which we denote as reverse saturation scattering (RSS). We will show in the following paragraphs that anapole-driven nonlinear dependence of scattering on excitation herein can be actively controlled in a reversible manner without the need to physically alter the dimensions of the nanostructure or change the environment.

The anapole state is an engineered superposition of toroidal and electric multipoles presenting in well-designed dielectric structures. Pioneering work has uncovered the disk geometry with its extreme structural simplicity providing efficient platforms to support the anapole mode in the visible region\textsuperscript{14}. To generate anapole mode in the spectral position close to 532 nm (excitation wavelength) in the measurements, Si nanodisks with diameter D of 200 nm and height h of 50 nm were used in the present system. Well-dispersed silicon nanodisks on a glass substrate were fabricated by colloidal-mask lithography\textsuperscript{34,35} with step-by-step fabrication sketched in Fig.2a (see Methods). Scanning electron microscopy (SEM) images (Fig. 2b) show high-quality Si nanodisks with an average spacing of micrometers to avoid the coupling effect. A 30° tilted-view of a single nanodisk is also presented. Scattering images of individual Si nanodisks under the 532 nm CW laser illumination are measured with a reflectance confocal laser scanning microscope (Fig.2c, see Methods).

As we have shown in Fig.1b, the scattering intensity displays a strong nonlinear response with increasing excitation radiances. Such nonlinearity can be analyzed by examining the PSFs of scattering signals at the focus of the microscope\textsuperscript{33,36-39}. Fig.2d presents how PSFs of scattering from isolated Si
nanodisks evolve with the increase of excitation intensities. When the laser intensity is low, the PSF fits well to a Gaussian function with full width at half maximum (FWHM) of 230 nm. The shape of the PSF changes dramatically with saturation of scattering. At deep saturation, a doughnut-like PSF appears with its intensity profile manifesting a deep valley (Fig. 2d (d-3)), corresponding to the radiationless nature of anapole. Further increases of excitation intensities, a sharp peak emerges from the center, indicating the onset of reverse saturation (shown in Fig. 2d (d-4)). Strong RSS which is indicated by the central peak dominates the PSF as the excitation continues to increase (Fig. 2d (d-5)).

The evolution of PSF profiles can be ascribed to an intensity-dependent nonlinear scattering behavior of Si nanodisks, which can be quantified by taking the ratio of scattering over incident intensity derived from the curve in Fig. 1b. As shown in Fig. 2e, the normalized scattering cross-section stays constant at the initial linear regime, then decreases to 0.1 for the largest SS, and again drastically rises to 1.1 for RSS, corresponding to 200% modulation spanning from scattering suppression to enhancement. The nonlinear scattering modulation is fully repeatable and reversible (Fig. 2f). Switching the laser intensity from a high power density level (which is adequate to induce RSS) to a lower one, full recovery of scattering intensity, as well as corresponding PSFs measured from the same nanodisk have been examined. This ensures the scattering behavior of a single silicon nanodisk can be actively and reversibly engineered.

The anapole mode supported by the Si nanodisk is clarified both by simulation and experimental measurements (supplementary Fig. S1). The anapole state is featured by a significant dip in the total far-field scattering spectrum, accompanied by unique near-field distribution as we show in Figs. 3a and b. Notably, the associated maximum in the near-field energy directly contributes to the absorption peak at the anapole wavelength. Leveraging anapole-induced absorption enhancement, the temperature within the Si disk rises substantially. To corroborate the local temperature rise of Si nanodisks under 532 nm laser illumination by different intensities, we perform Raman spectroscopy measurements. By taking the intensity ratio of anti-Stokes to Stokes of the Raman spectra,
temperature increment within the Si nanodisks under various incident intensities can be extracted (Fig.3b, see Methods). Experimental results from Raman thermometry reveal that the Si nanodisks experience a huge temperature rise more than 900°C relative to room temperature (RT) during the nonlinear scattering processes (Fig. 3c and supplementary Fig. S2). Such elevated temperatures will lead to essential tuning of complex refractive index in both real and imaginary parts, particularly in the thermal sensitive visible wavelength region\textsuperscript{40} (also see Supplementary Fig. S3). In the temperature range from RT to 950°C, the change of the refractive index in real part is $\Delta n = 0.5$ at a moderate laser intensity of $1.25 \times 10^6 \text{W/cm}^2$. This equivalently gives the effective nonlinear refractive index as $n_2,\text{@532nm} = \Delta n/I = 0.4 \text{cm}^2/\text{MW}$. Compared with the photothermal induced index change $\Delta n \sim 3 \times 10^{-5}$ in bulk Si under the same laser excitation intensity\textsuperscript{41}, anapole-enhanced photothermal nonlinearity manifests a striking improvement by four orders of magnitude. Such substantial temperature rise of Si nanostructures induces large modulations of refractive indices, also known as thermo-optic effect, holding fundamental importance in nonlinear Si photonics. We will show in the following that, unlike the absorption ascribed to all multipole modes of the nanodisk keep increasing with temperature (Fig. 3d), the scattering can be desirably manipulated in response to optical heating of anapole mode, thus yielding exotic nonlinear optical scattering.

The two-dimensional simulation map of scattering cross-section $C_{sc}$ is plotted in Fig.3e to examine the spectral response and the tuning range of Mie resonances with temperature increments. The corresponding photothermal-induced Mie resonance shifts thus are under simple estimation to be $\Delta \lambda \approx \lambda \Delta n/n \approx 60 \text{nm}$. Such spectral shifts are indeed observed in Fig.3e. The pronounced scattering maxima and minima (marked by the dashed lines for eye guidance) undergo continuous redshifts with elevated temperatures. Approximate $\Delta \lambda \approx 50 \text{nm}$ of anapole mode shifting is obtained, in good agreement with the above estimation. The achieved resonance tuning $\Delta \lambda /\lambda \sim 10\%$ represents a remarkable improvement compared with previous studies using liquid crystals or thermo-optic effect\textsuperscript{42-45}. We also remark here that free-carrier contributions are ruled out by the fact that they lead to a negative $\Delta n$, which causes the blue shift of the resonance\textsuperscript{46}. 
Increasing the temperature from room temperature (RT) to around 500°C by laser illumination, allows for suppressing the scattering cross-section from $2.3 \times 10^{-14}$ m$^2$ to $0.6 \times 10^{-14}$ m$^2$ (Fig. 3e-g), corresponding to 74% modulation. This agrees qualitatively with the experimental observation of 90% suppression of normalized cross-section from linear to SS in Fig. 2e. The large modulation depth is attributed to the fact that the excitation laser delicately operates in the vicinity of photothermal-tuning anapole positions. The remarkable slope in far-field scattering spectrum near anapole mode enables pronounced changes of scattering cross-sections under a small spectral tuning (Fig. 3f). The bottom panels in Fig. 3f further depict the near-field distributions excited at the wavelength of 532 nm, providing a clear progress that the illumination laser initially excites the lower-energy-side of anapole mode, and then gradually approaches resonant with the anapole mode and finally excites the ED mode again.

To this end, we are able to establish the relationship between nonlinear scattering $I_{sca}$ and the incoming light intensity $I_{exc}$ for single Si nanodisks. The intensity-dependent scattering can be derived as $I_{sca} = C_{sca} \cdot I_{exc}$ and is depicted in Fig. 3h, which shows a considerable similar trend with the experimental results shown in Fig. 1b. It is worth noting that, in the experiments, we actually recorded the backward scattering with PMT. Hence, more simulations are performed for total scattering and the forward scattering as a function of temperature variations (supplementary Fig. S4). Similar results are observed which excludes the scattering modulation originating from the redistribution of forward and backward radiation.

To illustrate the crucial importance of anapole-driven nonlinearity, calculations are also performed for another two representative sizes of Si nanodisks (Fig. S5). For a smaller-sized nanodisk (D=170nm), the overall photothermal tuning occurs near its ED mode. We show that ED-mediated process presents much weaker photothermal nonlinearity by the fact of temperature increasing less than 200°C at the laser intensity of $1.25 \times 10^6$ W/cm$^2$. The corresponding nonlinear refractive index $n_2 = 0.08 cm^2/MW$, which is five times lower than anapole-assisted process. In addition, its
scattering cross sections undergo slight decreasing when increasing the excitation intensity, resulting in negligible SS. On the contrary, for large-diameter nanodisk, its original anapole mode coincides with 532 nm laser at RT. Elevated temperatures induce the anapole red-shifting away from the exciting wavelength, leading to a monotonical increase of scattering cross sections. Thus, a directly sharp RSS is achieved. Similarly, such saturation scattering induced by photothermal nonlinearity has been reported by optical heating the magnetic quadrupole.

Leveraging this appealing photothermal nonlinear scattering, we demonstrate the potential for super-resolved localization of dielectric nanostructures. As we already show in Fig. 2d, the nonlinear behaviors of scattering dramatically change the PSFs under a confocal laser scanning microscope. In particular, RSS is featured by a sharp peak that emerges from the center of PSF. Such small spike in the central PSF provides the opportunity to super-localize the position of the nanodisk. As shown in Fig.4a, we extract the narrow FWHM by taking the difference between two scattering images obtained at RSS and SS stages. The outer contour of the RSS image can be subtracted over the SS image with r to be the subtractive factor, leaving a clear and sub-diffraction spot in the image. The exhibited 41-nm FWHM displays a striking localization capability which is remarkably smaller than the size of the nanodisk itself.

Notably, the scattering image of a nanodisk at high irradiances is drastically different from the confocal imaging in homogeneous media where the giant photothermal nonlinearity leads to the failure of the spatial invariance. To gain better understanding of the super-localization imaging, we begin with the raster scanning scattering image of a nanodisk. The scattering image of a nanodisk located at a given position (r) actually depicts the scattering-rate distribution, which can be given as:

$$S(I, r) = c|p(I) \cdot E(I, r)|^2$$

where $p(I)$ denotes the unit vector of excited multipolar mode which is intensity dependent, $E(I, r)$ represents the electric field at the position of the nanodisk at certain impinging intensity, and $c$ is a constant (supplementary Fig. S6). The bright and dark regions in the scattering image at high powers clearly indicate the different scattering state by different multipole
modes. In specific, the scattering pattern of a spike in the center of a doughnut shape in RSS manifests the gradual transition from radiationless anapole mode to ED when scanning close to the peak of the focal spot. Nevertheless, the differential image with 41-nm FWHM showcases a superior accuracy for localizing the center of both the nanodisk and supported multipole modes remarkably beyond its physical size.

As a proof-of-principle demonstration in super-localization nanoscopy, we fabricated periodic Si nanodisk arrays with diameter of 200 nm, height of 50 nm and pitch size of 300 nm (supplementary Fig. S7 for AFM characterization). Combination of SEM and optical images has been applied in order to unambiguously correlate the nonlinear scattering images and size/morphology for each individual nanodisk. The confocal image at low excitation intensities for such nanodisk array is blurry without surprise. When gradually increasing the laser power, highly saturated scattering emerges with the signature of PSFs being dark spots on top of bright background. At higher power, appearance of small bright spot in the center indicates the onset of RSS. The RSS then quickly dominates the PSF as the excitation continues to increase (see supplementary Fig. S8, and video for more results). By utilizing the same differential excitation technique, super-localization images shown in Fig.4c are achieved, and they are all in perfect correspondence with the SEM image in Fig.4b.

The resolution of images acquired at different stages is plotted in Fig. 4d. The best resolution corresponds to the very beginning of RSS whilst the signal to noise ratio is barely satisfactory. Strong RSS can induce better contrast while sacrificing a bit of resolution. Interestingly, PSF of SS generates a negative contrast. Taking the similar analogue of definition of resolution in ground state depletion (GSD) microscopy\textsuperscript{3}, FWHM of the valley at the measured PSF can be regarded as the experimental resolution. The FWHM of the valley exhibits 150 nm, which is indeed remarkably below the diffraction limit. As SS corresponds to anapole mode excitation, this mode imaging method can be utilized to directly visualize the nonradiating optical state. Noteworthily, during the whole photothermal
Discussion

In summary, we have demonstrated a novel super-localization nanoscopy based on discovered giant photothermal nonlinearity by harnessing all benefits of anapole states sustaining in a single silicon nanodisk. Taking advantage of the maximized electromagnetic near-fields, the absorption was resonantly enhanced at the anapole’s excitation wavelength, leading to extraordinary temperature rises along with record-high refractive index changes under mild optical irradiances. Utilizing low-radiating nature of anapole modes, far-field scattering was dynamically controlled by photothermal tuning the anapole spectral positions, allowing for active scattering engineering with external light stimuli. By fully exploiting the near- and far-field advantages, anapole-driven photothermal nonlinear scattering was experimentally measured. The dramatically changed PSF in the confocal reflectance images enabled by the nonlinear scattering behaviors at various scattering states has allowed the development of super-localization nanoscopy. As a proof-of-principle demonstration, super-localization imaging of Si nanodisks with an outstanding location precision approaching 40 nm corresponding to $\lambda/13$ has been achieved. Considering the compatibility with the existing semiconductor fabrication infrastructure, our work not only unlocks the full potential of Si photonics for active control, but also facilitates non-invasive high-resolution imaging which is highly desirable in silicon photonics components.
Methods

Preparation of silicon nanodisks: Polystyrene (PS) spheres were firstly spin-coated on to the layer film consisting with Si onto the glass. The size of the PS mask was reduced by the RIE process using oxygen gas. And then such PS spheres serve as the mask for the subsequent fluorine-based inductively coupled plasma reactive ion etching (ICP-RIE) using CHF3 gas. Finally, the PS mask was removed with sonication in acetone. The sizes of the resulting silicon disks can be precisely tuned by changing the size of the PS mask with accurate control of the etching time. When fabricating large array of periodic Si nanodisks, self-organized PS spheres assembling in a hexagonally close-packing manner were prepared as the monolayer mask.

Thermal calculations: The temperature growth inside the Si nanodisk is related to the absorbed power $Q = \sigma_{abs} I$ according to

$$\Delta T = \frac{\sigma_{abs} I}{4\pi R_{eq} \kappa \beta}$$

where $\kappa$ is the thermal conductivity of the surrounding medium. In the present case for Si nanodisks on the glass substrate and immersed in the oil environment, $\kappa$ was taken to be 0.38. $\beta$ is a dimensionless geometrical correction factor for a geometry with axial symmetry. For Si nanodisks with $D/h=4$, it is expressed as $\beta = \exp\{0.04 - 0.0124 \ln 4 + 0.0677 (\ln 4)^2 + 0.00457 (\ln 4)^3\}$. $R_{eq}$ is the corresponding equivalent radius, calculated as the radius of a sphere with the same volume as the nanodisk. The temperature rising from initial RT ($25^\circ$C) to the final temperature was divided into several intermediate steps and for each iteration, temperature-dependent optical absorption was firstly determined (Fig.3d) and substituted into the formula (1). The derived gradual temperature increase under photothermal effect shows a nice agreement with the results from Raman measurement. The linear trend shown in the red dashed line in Fig. 3c represents temperature rises linearly with incident light intensities, providing an underestimation of the actual temperature without taking into account photothermal refractive index change.
**Microscope system:** The nonlinear scattering measurements were performed based on Abberior 775 STED confocal microscope (Abberior Instruments GmbH, Göttingen). We coupled continuous-wave laser line (532nm) into the system for CW illumination\(^{37,39}\). The excitation beam was first spatially filtered and then focused onto the sample. Linear polarization excitation was obtained by imposing a half-wave plate on the laser beam. The backscattering signal was collected using the same objective lens, reflected by a beamsplitter and detected by a photomultiplier tube (PMT) after a confocal pinhole. The microscope images are obtained by synchronizing the PMT and the galvo mirror scanner.

**Multipole decomposition:** The Cartesian electric and magnetic dipole, quadrupole moments and the toroidal dipole moments of a nanodisk were calculated using the standard expansion formulas\(^{10}\):

- Electric dipole moment:
  \[
P_{\text{car}} = \frac{1}{i \omega} \int j d^3 \mathbf{r}
  \]
  \(^{(2)}\)

- Magnetic dipole moment:
  \[
  M_{\text{car}} = \frac{1}{2c} \int (\mathbf{r} \times j) d^3 \mathbf{r}
  \]
  \(^{(3)}\)

- Electric quadrupole moment:
  \[
  Q^{E}_{\alpha,\beta} = \frac{1}{i2\omega} \int [r_\alpha j_\beta + r_\beta j_\alpha - \frac{2}{3} \delta_{\alpha,\beta} (\mathbf{r} \cdot j)] d^3 \mathbf{r}
  \]
  \(^{(4)}\)

- Magnetic quadrupole moment:
  \[
  Q^{M}_{\alpha,\beta} = \frac{1}{3c} \int [(\mathbf{r} \times j)_\alpha r_\beta + (\mathbf{r} \times j)_\beta r_\alpha] d^3 \mathbf{r}
  \]
  \(^{(5)}\)

- Toroidal dipole moment:
  \[
  T_{\text{car}} = \frac{1}{10c} \int [(\mathbf{r} \cdot j) \mathbf{r} - 2r^2 j] d^3 \mathbf{r}
  \]
  \(^{(6)}\)

where \(j = i \omega \varepsilon_0 (\varepsilon_r - 1) E\) is the induced current in the structure, \(\mathbf{r}\) is the position vector with the origin at the center of the nanodisk and \(\alpha, \beta = x, y, z\).
**Raman spectroscopy:** Raman spectra was taken under a microspectroscopy system based on an inverted optical microscope (NTEGRA Spectra, NT-MDT). The experimental configurations were described in detail in a previous study. Briefly, Si nanodisks were excited using linearly polarized 532-nm lasers using an oil immersion objective (1.4 NA, 60×, Olympus). The resulting Raman signal with both stokes and anti-stokes lines was collected using the same objective, passed through a notch filter, and focused into the spectrometer with a cooled CCD (iDdus, Andor). Raman spectra were recorded over an acquisition time of 1 s.

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**Author contributions**

X. L. conceived the idea and supervised the project. T. Z. and Y. C. performed the experiments. K. C. prepared the sample. T. Z. and Y. X. performed the multipolar expansion. T. Z., Y. C., S-W. C. and X. L. analyzed data and prepared the manuscript. All authors contributed to the discussion and manuscript writing.

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References

1. Jahani S, Jacob Z. All-dielectric metamaterials. *Nat Nanotechnol* **11**, 23-36 (2016).

2. Kuznetsov AI, Miroshnichenko AE, Brongersma ML, Kivshar YS, Luk’yanchuk B. Optically resonant dielectric nanostructures. *Science* **354**, aag2472 (2016).

3. Yang Z-J, Jiang R, Zhuo X, Xie Y-M, Wang J, Lin H-Q. Dielectric nanoresonators for light manipulation. *Physics Reports* **701**, 1-50 (2017).

4. Kruk S, Kivshar Y. Functional Meta-Optics and Nanophotonics Governed by Mie Resonances. *ACS Photonics* **4**, 2638-2649 (2017).

5. Fu YH, Kuznetsov AI, Miroshnichenko AE, Yu YF, Luk’yanchuk B. Directional visible light scattering by silicon nanoparticles. *Nat Commun* **4**, 1527 (2013).

6. Staude I, et al. Tailoring Directional Scattering through Magnetic and Electric Resonances in Subwavelength Silicon Nanodisks. *ACS Nano* **7**, 7824-7832 (2013).

7. Cihan AF, Curto AG, Raza S, Kik PG, Brongersma ML. Silicon Mie resonators for highly directional light emission from monolayer MoS2. *Nat Photonics* **12**, 284-291 (2018).

8. Shi T, et al. All-Dielectric Kissing-Dimer Metagratings for Asymmetric High Diffraction. *Adv Opt Mater* **7**, 1901389 (2019).

9. Miroshnichenko AE, Kivshar YS. Fano Resonances in All-Dielectric Oligomers. *Nano Lett* **12**, 6459-6463 (2012).

10. He Y, Guo G, Feng T, Xu Y, Miroshnichenko AE. Toroidal dipole bound states in the continuum. *Phys Rev B* **98**, 161112 (2018).

11. Liu Z, et al. High-Q Quasibound States in the Continuum for Nonlinear Metasurfaces. *Phys Rev Lett* **123**, 253901 (2019).

12. Baryshnikova KV, Smirnova DA, Luk’yanchuk BS, Kivshar YS. Optical Anapoles: Concepts and Applications. *Adv Opt Mater* **7**, 1801350 (2019).

13. Yang Y, Bozhevolnyi SI. Nonradiating anapole states in nanophotonics: from fundamentals to applications. *Nanotechnology* **30**, 204001 (2019).

14. Miroshnichenko AE, et al. Nonradiating anapole modes in dielectric nanoparticles. *Nat Commun* **6**, 8069 (2015).

15. Liu W, Zhang JF, Miroshnichenko AE. Toroidal dipole-induced transparency in core-shell nanoparticles. *Laser & Photon Rev* **9**, 564-570 (2015).

16. Ospanova AK, Labate G, Matekovits L, Basharin AA. Multipolar passive cloaking by nonradiating anapole excitation. *Sci Rep* **8**, 12514 (2018).

17. Feng T, Xu Y, Zhang W, Miroshnichenko AE. Ideal Magnetic Dipole Scattering. *Phys Rev Lett* **118**, 173901 (2017).

18. Ospanova AK, Stenishchev IV, Basharin AA. Anapole Mode Sustaining Silicon Metamaterials in Visible Spectral Range. *Laser & Photon Rev* **12**, 1800005 (2018).
19. Wu PC, et al. Optical Anapole Metamaterial. ACS Nano 12, 1920-1927 (2018).
20. Grinblat G, Li Y, Nielsen MP, Oulton RF, Maier SA. Enhanced Third Harmonic Generation in Single Germanium Nanodisks Excited at the Anapole Mode. Nano Lett 16, 4635-4640 (2016).
21. Xu L, et al. Boosting third-harmonic generation by a mirror-enhanced anapole resonator. Light-Sci Appl 7, 44 (2018).
22. Grinblat G, Li Y, Nielsen MP, Oulton RF, Maier SA. Efficient Third Harmonic Generation and Nonlinear Subwavelength Imaging at a Higher-Order Anapole Mode in a Single Germanium Nanodisk. ACS Nano 11, 953-960 (2017).
23. Totero Gongora JS, Miroshnichenko AE, Kivshar YS, Fratalocchi A. Anapole nanolasers for mode-locking and ultrafast pulse generation. Nat Commun 8, 15535 (2017).
24. Wang R, Dal Negro L. Engineering non-radiative anapole modes for broadband absorption enhancement of light. Opt Express 24, 19048-19062 (2016).
25. Du K, et al. Strong Coupling between Dark Plasmon and Anapole Modes. J Phys Chem Lett 10, 4699-4705 (2019).
26. Liu S-D, Fan J-L, Wang W-J, Chen J-D, Chen Z-H. Resonance Coupling between Molecular Excitons and Nonradiating Anapole Modes in Silicon Nanodisk-J-Aggregate Heterostructures. ACS Photonics 5, 1628-1639 (2018).
27. Verre R, Baranov DG, Munkhbat B, Cuadra J, Kall M, Shegai T. Transition metal dichalcogenide nanodisks as high-index dielectric Mie nanoresonators. Nat Nanotechnol 14, 679–683 (2019).
28. Baranov DG, Verre R, Karpinski P, Käll M. Anapole-Enhanced Intrinsic Raman Scattering from Silicon Nanodisks. ACS Photonics 5, 2730-2736 (2018).
29. Yang Y, Zenin VA, Bozhevolnyi SI. Anapole-Assisted Strong Field Enhancement in Individual All-Dielectric Nanostructures. ACS Photonics 5, 1960-1966 (2018).
30. Zograf GP, et al. Resonant Nonplasmonic Nanoparticles for Efficient Temperature-Feedback Optical Heating. Nano Lett 17, 2945-2952 (2017).
31. Corte FGD, Montefusco ME, Moretti L, Rendina I, Cocorullo G. Temperature dependence analysis of the thermo-optic effect in silicon by single and double oscillator models. Journal of Applied Physics 88, 7115-7119 (2000).
32. Lewi T, Butakov Nikita A, Schuller Jon A. Thermal tuning capabilities of semiconductor metasurface resonators. Nanophotonics 8, 331–338 (2019).
33. Chu S-W, et al. Saturation and Reverse Saturation of Scattering in a Single Plasmonic Nanoparticle. ACS Photonics 1, 32-37 (2014).
34. Dao TD, Chen K, Nagao T. Dual-band in situ molecular spectroscopy using single-sized Al-disk perfect absorbers. Nanoscale 11, 9508-9517 (2019).
35. Dao TD, et al. Infrared Perfect Absorbers Fabricated by Colloidal Mask Etching of Al–Al2O3–Al Trilayers. ACS Photonics 2, 964-970 (2015).
36. Chu SW, et al. Measurement of a saturated emission of optical radiation from gold nanoparticles: application to an ultrahigh resolution microscope. Phys Rev Lett 112, 017402 (2014).
37. Xu J, et al. Plasmonic Nanoprobes for Multiplexed Fluorescence-Free Super-Resolution Imaging. Adv Opt Mater 6, 1800432 (2018).
38. Chen Y-T, et al. Study of Nonlinear Plasmonic Scattering in Metallic Nanoparticles. ACS Photonics 3, 1432-1439 (2016).
39. Ouyang X, et al. Invited Article: Saturation scattering competition for non-fluorescence single-wavelength super-resolution imaging. APL Photonics 3, 110801 (2018).
40. Chapter 3 - Thermo-Optic Coefficients. In: Handbook of Optical Constants of Solids (ed Palik ED). Academic Press (1997).
41. Horvath C, Bachman D, Indoe R, Van V. Photothermal nonlinearity and optical bistability in a graphene-silicon waveguide resonator. Opt Lett 38, 5036-5039 (2013).
42. Rahmani M, et al. Reversible Thermal Tuning of All-Dielectric Metasurfaces. Adv Funct Mater 27, 1700580 (2017).
43. Zangeneh Kamali K, et al. Reversible Image Contrast Manipulation with Thermally Tunable Dielectric Metasurfaces. Small 15, 1805142 (2019).
44. Sautter J, et al. Active Tuning of All-Dielectric Metasurfaces. ACS Nano 9, 4308-4315 (2015).
45. Shcherbakov MR, et al. Ultrafast All-Optical Switching with Magnetic Resonances in Nonlinear Dielectric Nanostructures. Nano Lett 15, 6985-6990 (2015).
46. Xiang J, Chen J, Dai Q, Tie S, Lan S, Miroshnichenko AE. Modifying Mie Resonances and Carrier Dynamics of Silicon Nanoparticles by Dense Electron-Hole Plasmas. Physical Review Applied 13, 014003 (2020).
47. Duh Yi-Shiou, et al. Giant photothermal nonlinearity in single silicon nanostructure: ultrasmall alloptical switch and super-resolution imaging, Preprint at http://arxiv.org/abs/2001.08421.
48. Zhao G, Kuang C, Ding Z, Liu X. Resolution enhancement of saturated fluorescence emission difference microscopy. Opt Express 24, 23596-23609 (2016).
49. Chen C, et al. Multi-photon near-infrared emission saturation nanoscopy using upconversion nanoparticles. Nat Commun 9, 3290 (2018).
50. Nawa Y, et al. Saturated excitation microscopy using differential excitation for efficient detection of nonlinear fluorescence signals. APL Photonics 3, 080805 (2018).
51. Gu M. Point Spread Function Analysis. In: Advanced Optical Imaging Theory). Springer Berlin Heidelberg (2000).
52. Li X, Lan TH, Tien CH, Gu M. Three-dimensional orientation-unlimited polarization encryption by a single optically configured vectorial beam. Nat Commun 3, 998 (2012).
53. Rittweger E, Wildanger D, Hell SW. Far-field fluorescence nanoscopy of diamond color centers by ground state depletion. EPL (Europhysics Letters) 86, 14001 (2009).
54. Baffou G, Quidant R. Thermo-plasmonics: using metallic nanostructures as nano-sources of heat. Laser & Photon Rev 7, 171-187 (2013).
55. Baffou G, Quidant R, García de Abajo FJ. Nanoscale Control of Optical Heating in Complex Plasmonic Systems. ACS Nano 4, 709-716 (2010).
Fig. 1. Schematic illustration and experimental observation of anapole-mediated photothermal nonlinearity. (a) Illustration of strong optical heating that efficiently converts light into temperature rises within subwavelength volume of silicon nanodisks. (b) The nonlinear dependency of scattering on excitation intensity for 532 nm excitation of single Si nanodisk. When the excitation intensity is low, the scattering intensity is linearly proportional to excitation intensity. When the excitation intensity exceeds $8 \times 10^5$ W/cm$^2$, scattering deviates from linear trend into deep saturation. When the excitation intensity is higher than $1.25 \times 10^6$ W/cm$^2$, the scattering sharply increases, showing the reverse saturation of scattering. (c) Scheme of anapole-driven nonlinear scattering behavior.
Fig. 2. Experimental measurement of nonlinear scattering. (a) Schematic representation of the fabrication of isolated silicon nanodisks with desired sizes. (b) SEM images as well as a side-view, showing the resulting Si with diameter of 200 nm and height of 50 nm. (c) Optical setup of the reflected laser scanning confocal microscope. HWP: half-wave plate; BS: beam splitter; OL: objective lens; PMT: photomultiplier tube. (d) Measured PSF of 532 nm excitation beam under different laser intensities. The right panel gives the intensity lateral profile of a selected nanodisk (white dashed line). (e) The normalized scattering cross-section variation versus excitation intensity. (f) Reversible and repeatable nonlinear scattering. Full recovery of scattering intensity as well as corresponding PSF measured from the same nanodisk under multiple measurement are confirmed.
Fig. 3. Anapole-driven photothermal nonlinear scattering. (a) Simulations of optical scattering and absorption spectra of Si nanodisk. Grey arrow shown in the figure indicates the anapole signature by the scattering dip in the far-field, along with the absorption peak associated with the same anapole excitation position. (b) Multipolar decomposition of induced currents in Cartesian coordinates and the electric field distribution inside the nanodisk calculated at the total scattering dip wavelength position. The white arrow indicates the incident light linearly polarized in the y-direction. Scale bar, 100nm. (c) Temperature increases in silicon nanodisks as the intensity of 532 nm excitation beam. Dots represent Raman nanothermometry derived temperatures based on the ratio of anti-stokes and stokes signals. Solid lines denote thermal calculations based on iterative technique (see Method). The linear trend shown in red dashed line represents calculated temperature without taking the change of complex indices into account. (d) Simulation mapping of the absorption cross-section and (e) scattering cross-section of Si nanodisks as a result of increasing the temperature. The dashed lines in (d) indicate the series of absorption maximum. The red, gray and blue dashed lines in (e) denote the electric dipole
state, anapole state, and excitation wavelength, respectively. (f) Scattering cross-section at three representative temperatures (top panel) and the corresponding near-field distributions at the excitation wavelength of 532 nm (bottom panel). (g) Scattering cross-section corresponding to 532-nm excitation under elevated temperatures, showing that it undergoes firstly suppression and then increase during the photothermal tuning. (h) Calculated photothermal nonlinear scattering as a function of laser intensity.
Fig. 4. Super-localization nanoscopy based on the photothermal nonlinear scattering. (a) PSF of a single isolated Si nanodisk at different intensities. High resolution localization with fitted FWHM of 41 nm is obtained by differential images between RSS and SS with $r$ to be a subtractive factor. (b) PSFs of nonlinear scattering from periodic Si disk arrays evolve with increasing excitation intensities. Correlated SEM image is also presented. The conventional confocal at the low excitation intensity shows undistinguishable image due to the diffraction limit. SS image generates a negative contrast whilst in RSS a central spike emerges. The onset of RSS process is presented in image RSS-1. With the degree of reverse scattering increases, the central spike becomes more obvious (RSS-2). (c) Super-localization of nanodisk arrays by means of differential image between RSS-1/RSS-2 and SS. (d) Resolution scaling at PSF obtained at different stages of nonlinear scattering.