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Direct amplitude-phase near-field observation of higher-order anapole states

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Abstract

Anapole states associated with the resonant suppression of electric-dipole scattering exhibit minimized extinction and maximized storage of electromagnetic energy inside a particle. Using numerical simulations, optical extinction spectroscopy and amplitude-phase near-field mapping of silicon dielectric disks, we demonstrate high-order anapole states in the near-infrared wavelength range (900-1700 nm). We develop the procedure for unambiguously identifying anapole states by monitoring the normal component of
the electric near-field and experimentally detect the first two anapole states as verified by far-field extinction spectroscopy and confirmed with the numerical simulations. We demonstrate that higher-order anapole states possess stronger energy concentration and narrower resonances, a remarkable feature that is advantageous for their applications in metasurfaces and nanophotonics components, such as non-linear higher-harmonic generators and nanoscale lasers.

Keywords

SNOM, near-field microscopy, anapole, multipole decomposition, all-dielectric nanoparticles

High-refractive index dielectric particles have recently attracted considerable attention of nanophotonics community due to their exciting property of supporting both electric and magnetic resonances while exhibiting low absorption.\textsuperscript{1-8} Nowadays a variety of simulation tools is available to accurately find electromagnetic responses of arbitrarily shaped particles to any excitation field. However, the knowledge of full electromagnetic field distribution and its derivatives does not always ensure the transparent explanation and intuitive understanding of the physical phenomena involved. One way to proceed would be to further analyze numerically found electromagnetic field distributions by using a multipole decomposition (see Methods). With this approach, a complete (often very complicated) optical response will be decomposed into well-known responses of different multipoles. One can decompose either scattered electromagnetic fields outside the particle (known as the decomposition in spherical multipoles\textsuperscript{9}), or the electric field inside the particle (known as the decomposition in Cartesian multipoles\textsuperscript{10}). Recently it was shown that the spherical multipole decomposition can also be carried out using only fields inside the particle.\textsuperscript{11}

The spherical multipole family contains classical multipoles: electric/magnetic dipoles, quadrupoles, octupoles, etc., as described in the Mie theory.\textsuperscript{12} Due to the orthogonality of their fields, the total scattering (power or cross section) is simply a sum of contributions from each individual multipole moment. On the contrary, the Cartesian multipole family
includes other additional terms, which have particular field distributions inside the particle but the same far-field radiation patterns as the above mentioned spherical multipoles. For example, both Cartesian toroidal dipole (third-order multipole) and Cartesian electric dipole moments radiate in the same manner - that of the spherical electric dipole.\textsuperscript{11-18} When calculating the total scattering, one should therefore take into account the phase of the toroidal dipole moment relative to that of the Cartesian electric dipole moment (see Methods). This implies that the toroidal moment can be considered as the electric dipole-like moment. In general, Cartesian electric and toroidal dipole moments represent simply the first two terms of the spherical electric dipole being expanded in series within the long-wavelength approximation.\textsuperscript{11} Hereafter, to avoid confusion, we refer to spherical multipoles as full multipoles (for example, a full electric dipole).

The full electric dipole (FED) moment compared to other multipole moments provides usually a dominant contribution to the optical response of a particle. Therefore, when the FED moment is reduced, it leads to a significant decrease in the total scattering. Under certain conditions, the FED moment can even vanish completely, an effect which was observed for dielectric spheres in vacuum with the corresponding scattering mode being termed anapole.\textsuperscript{19} This term was borrowed from elementary particle physics, where it meant a non-radiating source.\textsuperscript{20} However, the ideal non-radiating anapole cannot be excited by a propagating optical wave due to the reciprocity theorem. To our knowledge, a pure anapole was excited only by a standing wave with a specific field profile.\textsuperscript{21} After conducting multipole analysis of dielectric spheres in vacuum, it was found that, at the anapole condition (i.e., when the FED is at zero), the Cartesian electric and toroidal dipoles completely cancel each other.\textsuperscript{19} In the same work, the definition of the anapole mode was relaxed with the sufficient requirement being that the Cartesian electric and toroidal dipole moments interfere destructively, i.e., when the FED experiences a local minimum. Hereafter, we adopt the latter (relaxed) definition without explicitly mentioning toroidal dipole moment, since the FED scattering, in general case, should include contributions of Cartesian electric, toroidal,
and other electric dipole-like moments. To avoid confusion with previous works, we refer to this condition as the anapole state.

In the first investigations, it was shown that, for high-refractive index discs with high diameter-to-thickness aspect ratios, the anapole state results in a decrease in the total scattering cross-section and, at the same time, increase in the total electric field at the disk top center.\textsuperscript{19} Later on, it was also demonstrated that the anapole state features a substantial boost of the electric energy inside the disc.\textsuperscript{22,23} Very recently, the existence of high-order anapole states at shorter wavelengths was revealed with high-order states featuring the same properties as the fundamental anapole state.\textsuperscript{24} It was also found that higher-order anapole states possess stronger energy concentration and narrower resonances, resulting in a stronger third-harmonic generation. However, the multipole analysis was not implemented, making the study of higher-order anapole states incomplete.\textsuperscript{24} Interestingly, upon close examination of the previous studies one can also find the presence of the second-order anapole state revealed by minima in the total scattering and maxima in total energy spectra.\textsuperscript{19,22} Finally, it should be mentioned that anapole states were also investigated with a completely different Fano-Feshbach analysis of internal modes.\textsuperscript{25}

Since the electric dipole radiation is suppressed in the anapole state, it is reasonable to assume that there should be a signature of the anapole state in the near-field distribution. Here we demonstrate, for the first time to our knowledge, amplitude-phase resolved near-field detection of the anapole states. It is revealed that the normal component of the electric near-field on the top of a disc experiences a local minimum at the anapole states. This is a very surprising feature (confirmed also by numerical simulations) of the anapole states, when taking into account that the total electric energy (and, consequently, the average total electric field magnitude) inside the disc is enhanced at the anapole states. There is, however, no contradiction to the main laws of electromagnetics, since these states feature strong spatial variations of the tangential field components inside the disc. It should be noted that the experimental near-field investigation of anapole states by mapping the intensity of
fiber collected radiation was already reported.\textsuperscript{19} However, this near-field technique is known to have severe resolution limitations and a complicated response with interfering \textit{in-plane} electric and magnetic field contributions,\textsuperscript{26,27} a circumstance that makes a direct identification of anapole states very problematic. The near-field detection method presented in this work probes the \textit{out-of-plane} (normal) component of the electric near-field with very high spatial resolutions,\textsuperscript{28–30} providing a straightforward tool for identification and observation of anapole states. In addition, this method is valid for different illumination wavelengths and different orders of anapole states.

The paper is composed as following. First, we conduct simulations and multipole analysis of isolated Si discs in free space and show the existence of different-order anapole states. They are defined by the drop in the FED scattering and are accompanied by a peak in the electromagnetic energy concentration inside the particle and a drop in both the total scattering cross-section and the amplitude of the normal electric near-field component, measured on top of the disc and averaged over the disc area. We also verify that higher-order anapole states feature higher energy accumulation and narrower resonances, compared to the first (fundamental) anapole state. We make a far-field experimental detection of anapole states by measuring the transmission and correspondent extinction spectra for a variety of disc diameters. Then we proceed to investigation of the Si discs in a scattering-type scanning near-field optical microscope (SNOM). Using a simple symmetry-based procedure, we extract the normal component of the near-field $E_z$. The drop in $|E_z|$, averaged over the disc area, is then used for the experimental near-field identification of anapole states. Finally, we summarize our findings and show that the anapole states can be detected by the drop in near-field $|E_z|$ and/or in the far-field extinction, or by the peak in the electric energy accumulated inside the particle.
Results and discussion

We begin with numerical simulations of an isolated 80-nm-thick Si disc with 800 nm diameter in free space, illuminated normally with an $x$-polarized plane wave (Figure 1a). First we compute the total scattering of the disc for the near-infrared wavelength range of 750-2000 nm (see Methods), which features three dips (Figure 1b). In order to understand the scattering spectra, we apply the spherical multipole decomposition (by using the calculated E-field inside the disc, see Methods) and find the accompanying dips in the contribution of the full electric dipole (FED) scattering, which we use to define the anapole states. The decomposition into Cartesian multipoles (see Methods) verifies that the first anapole state (at $\lambda \approx 1425$ nm) is indeed caused by destructive interference of the toroidal and electric dipole moments (Supporting Information, Figure S1). However, the Cartesian multipole decomposition fails to describe accurately higher-order anapole states, appearing at shorter wavelength (at $\lambda \approx 985$ and 795 nm, correspondingly), since it intrinsically assumes a small parameter of $2\pi r/\lambda$. Thus, higher-order anapole states can be analyzed with the spherical multipole decomposition, and the dip in FED scattering is not simply a result of destructive interference between Cartesian electric and toroidal dipole moments.

The similarity between total and FED scattering shows that the far-field scattering is dominated by the electric dipole moment (Figure 1b). Thus, it is natural to assume that the near-field is also dominated by the same electric dipole. We found that it is indeed so for the normal $E_z$ component of the near field. When its amplitude is calculated at a certain altitude above the disc and averaged over the disc area, then it also features three dips, corresponding to anapole states (Figure 1c). This near-field method of anapole identification is robust to the altitude, where $E_z$ is calculated. Later we use the altitude of 50 nm, since it equals to the oscillation amplitude of a SNOM probe in the experiment. Finally, we compute the electric and magnetic energy inside the disc ($\varepsilon_0 \varepsilon \iiint |E|^2 \, dV$ and $\mu_0 \mu \iiint |H|^2 \, dV$), which now features three pronounced peaks at the anapole states (Figure 1d). This correlation can be intuitively understood by the increase of the quality factor of the resonator, causing higher
Figure 1: Anapole states. (a) Illustration of the anapole state excitation in a Si disc upon normal $x$-polarized plane-wave illumination. Overlay: $|\mathbf{E}|$-field distribution inside the disc at the first anapole state, with arrows representing in-plane electric field direction. At anapole state the electric dipole moment is reduced, causing reduction in both near- and far-field scattering. (b) Full electric dipole (FED, green) and total scattering cross-section (blue) for an 80-nm-thick silicon disc with a diameter of 800 nm. (c) Normalized amplitude $|E_z|$, averaged over the disc area and calculated at disc surface (violet) and at the altitude of 20 nm (green), 50 nm (red), and 100 nm above the disc surface (black). (d) Total electric ($\varepsilon_0 \int \int |\mathbf{E}|^2 \, dV$, solid black) and magnetic energy ($\mu_0 \mu \int \int |\mathbf{H}|^2 \, dV$, dashed blue), accumulated inside the 1100 nm disc. 1 a.u. corresponds to $2 \cdot 10^{-26}$ J at the irradiance of 1 W/m$^2$. Inserts show $|\mathbf{E}|$-field distributions inside the disc, normalized to the amplitude of incident field and calculated at $\lambda = 794$, 985, 1116, and 1425 nm (scale bar is 500 nm). The white double arrow shows the polarization of the incident wave.
energy accumulation inside the resonator resulted from suppressed scattering. It should be noted that higher-order anapole states possess narrower resonances, indicating their higher quality factor compared to the first (fundamental) anapole state. Similar simulations and multipole decomposition were also done for a larger Si disc of 1100 nm in diameter (Supporting Information, Figures S1-S2). As expected, with an increase of the size all resonances are red shifted. We also verified that, with absence of losses, higher anapole states possess stronger energy concentration, compared to the first anapole state (Supporting Information, Figure S2). Additionally we simulated the same 800 nm Si disc on a glass substrate (Supporting Information, Figure S2), as used in our experiments. In this case we didn’t apply the multipole decomposition, since it is rather complicated for particles in uneven environment. Nevertheless, obtained scattering and electric field spectra are rather similar to spectra of the disc in air, featuring same signs of anapole states.

The insets in Figure 1d show the total electric field inside the disc at different illumination wavelengths (more detailed field profiles can be found in Supporting Information, Figure S3). At the first anapole state (also shown in Figure 1a) the electric field distribution features three hot spots with alternating direction of the in-plane electric field, forming two opposite vortices, which generate a magnetic quadrupole.¹⁹ Thus the outer part of the disc has the opposite direction of the in-plane electric field relative to the central part, which results in a reduced electric dipole moment, while electric field amplitude is high inside the disc. At higher-order anapole states the number of E-field hot spots increases,²⁴ similarly to the standing-wave patterns formed in brick antennas at high-order modes.³¹

To confirm the far-field response discussed above, we performed a series of experiments to observe and detect the excitation of anapole states. Si discs of 80 nm thickness and diameters varied from 470 to 970 nm were fabricated on a fused silica substrate (see Methods). These discs, separated by 50 μm to reproduce the isolated condition, were used for far-field transmission measurements (see Methods). The measured transmittance T was used to plot extinction 1 – T, which should be proportional to the total scattering, since absorption is
negligible for wavelengths above 900 nm (see Supporting Information, Figure S4). Points in Figure 2 show the position of the dip in simulated scattering spectra (squares and circles correspond to the first and second anapole state, respectively), which demonstrates a good agreement between experimental and numerical results.

![Experimental extinction spectra and SEM images](image)

Figure 2: Experimental far-field detection of anapole states. Experimental extinction spectra of isolated silicon disks with a diameter ranging from 600 to 880 nm. The baseline for each spectrum has an offset of 1 a.u. Points show the position of the dip in simulated scattering spectra (squares and circles correspond to the first and second anapole state, respectively). Right: SEM images of corresponding disks (scale bar is 500 nm).

Then another set of discs with the same diameters, but arranged in a four-by-four array with a period of 2 μm was investigated with the scattering-type amplitude- and phase-resolved scanning near-field microscope (SNOM) at wavelengths of 900-1640 nm (see Methods). It should be noted that SNOM transfer function is rather complicated, on the one hand because SNOM probe is not significantly small compared to a studied object, and on the other hand because of the sophisticated detection process, when the scattered signal is first modulated due to the tip’s oscillations and then demodulated to suppress constant background. However, in previous studies there was a good enough agreement between SNOM maps and simulated normal component of the near-field $E_z$, recorded at the altitude of 50 nm above the object without the tip. $^{29,32,33}$ The in-plane near-field components $E_x$ and $E_y$ are taken into account only in special cases. $^{30}$ It is already clear from the SNOM amplitude distribution (Figure 3b,c) that a disc with a diameter of 800 nm corresponds to the mini-
maximum average near-field amplitude (it is encircled with the white dashed line, the incident wavelength is $\lambda = 1000$ nm). However, there is a clear difference between SNOM maps for the $x$- and $y$-polarized incident beams. This discrepancy can be explained by anisotropy of the tip, which results in different sensitivity to $E_x$ and $E_y$ near-field components. However, assuming a linear contribution of the components to the detected near-field, and taking into account the different symmetry of each near-field component along $x$- and $y$-axes, it is possible to decompose the recorded near-field $E$ into $E_x$, $E_y$, and $E_z$ (see Methods). This processing method, applied to different SNOM maps for both polarizations of the incident beam, resulted in nearly the same $E_z$ distribution (Figure 3d).

Figure 3: Near-field characterization. (a) SEM and (b, c) pseudocolor SNOM maps of near-field amplitude for silicon disks under normal and linearly polarized illumination (depicted with double arrow). Disk diameters are shown in a. Scale bar, 1 $\mu$m. The disk with the lowest near-field amplitude is encircled with the white dashed line in (b), (c). (d) Processing of near-field maps for 800 nm disk by using symmetry in order to get a clean near-field component $E_z$.

Then we performed such decomposition for near-field maps of all disc sizes, recorded at
five different illumination wavelengths: 900, 1000, 1425, 1500, and 1640 nm (Figure 4 and Supporting Information, Figures S7-S8). It was found that there is a clear drop in $|E_z|$ for a certain disc size, which in turn depends on the wavelength. Simulations confirm the drop in the average near-field $|E_z|$, calculated 50 nm above the disc surface (the position of the dip is shown with vertical lines in Figure 4a,b). According to simulations, such drop appears both for first and second anapole states. Interestingly, that near-filed $|E_z|$ map always exhibits two-lobe pattern for all disc sizes and anapole orders, while the profiles of the in-plane components $|E_x|$ and $|E_y|$ feature more complicated patterns, with a number of lobes being proportional to the order of the anapole state (Supporting Information, Figure S3).

To sum it up, we compare different techniques, used for the identification of anapole states. First we plot simulated average near-field $|E_z|$ (Figure 5a), calculated 50 nm above the top disc surface, and the total scattering cross-section, normalized to the disc cross-section area (Figure 5b), for different wavelengths and disc diameters. Even though they represent either near- or far-field properties, they both look very similar, with dips corresponding to the anapole states. At such states there is a clearly pronounced peak in the total electric energy inside the disc, normalized to its volume (Figure 5c). Finally, Figure 5d shows positions of the extrema, found with the above mentioned techniques, and the experimental findings - dips in extinction spectra for each disc size and dips in average near-field $|E_z|$ for each selected wavelength. As can be seen, there is a good agreement between experimentally measured and simulated results. Also it was found that different techniques provide almost the same position of the anapole states with deviation within 5%. The origin of such deviation is the well-known mismatch between the peak positions in the near- and far-fields.\textsuperscript{34}
Figure 4: Near-field detection of anapole states. Experimental near-field $|E_z|$ distribution and its average value for silicon disks, measured at different illumination wavelength: (a) 900 (magenta down triangles), 1000 (orange stars), (b) 1425 (black squares), 1500 (red circles), and 1640 nm (blue up triangles). The thick transparent curves are for eye guidance. Disk sizes are labeled on top. The disk with the lowest near-field amplitude is encircled with the white dashed line. Vertical lines in graphs show the disk diameter with the lowest simulated $|E_z|$ and are related to the (b) first-order and (a) second-order anapole states.
Figure 5: Various observation methods of anapole states. (a-c) Simulated (a) average near-field $|E_z|$, (b) total scattering cross-section, normalized to the disc cross-section area, and (c) average electric energy density inside the disc for silicon discs with a diameter ranging from 500 to 1000 nm. (d) Resonance position, experimentally measured and calculated with different techniques. Error bars are evaluated from measurements of different samples (for far-field extinction) and from the discrete set of disc diameters (for near-field).
Outline

In summary, we have thoroughly investigated the anapole states of Si discs in free space and on top of a glass substrate. We defined an anapole state as a condition when the full electric dipole contribution to the scattering cross-section is resonantly suppressed. This suppression originates from destructive interference of electric-dipole-like moments corresponding to the Cartesian multipole decomposition, i.e., electric and toroidal dipoles for the first-order anapole state. We have found that the FED scattering at higher-order anapole states, appearing for larger disc sizes or at shorter wavelengths, cannot be described as simply destructive interference of only Cartesian electric and toroidal dipoles, since the high-order Cartesian electric-dipole-like moments cannot be neglected. We have shown that the anapole states in the discs can be identified by a drop in the total scattering cross-section and a local maximum in the total accumulated electromagnetic energy inside the particles. Additionally we have found that the anapole states also feature a reduction of the normal near-field electric component $|E_z|$. This property has been successfully used for experimental identification of the anapole states. All three identification techniques are found to provide similar results with deviations less than 5%. The near-field detection of anapole states can be particularly useful for sets of particles with subwavelength variations, for example in metasurfaces, when other far-field techniques might fail due to the far-field resolution limitations. We expect that the near-field anapole identification method used in our work is limited to flat and symmetric particles, because, for arbitrary shapes, the near-field distributions might become rather complicated, impeding the implementation of the developed near-field map processing. Finally, we have demonstrated that higher-order anapole states possess stronger energy concentration and narrower resonances, a remarkable feature that is advantageous for their applications in nanophotonics components, such as non-linear higher-harmonic generators and nanoscale lasers.
Methods

Fabrication. The disks were fabricated in silicon deposited on silica wafers. The new silica wafers were first cleaned using a standard RCA clean, without the HF steps. Thereafter, 80 nm of amorphous Si was LPCVD grown. The thickness and refractive index of the grown Si were measured using a FilmTek 4000TM spectrometer. The structures were then defined using e-beam lithography. Since the standard resist used is a positive one, we used a combination of lift-off and etching to define them. Thus, after the spinning of the AR-P 6200.09 resist and the definition of the structures, a thin 50 nm layer of Al was deposited on the sample. Using a standard lift-off technique, we removed the rest of the resist and the Al layer deposited on top of it. This way, we obtained an Al hard mask for etching the Si. We used a BOSCH process to etch the 80 nm of silicon and define the structures. The last step involved removing of the Al such that the structures are now defined only in Si. A quick overview of the fabrication steps can be seen in Supporting Information, Figure S4. Designed disc diameters were 30 nm larger than those reported in this work (i.e., from 500 to 1000 nm). Then upon comparing the experimental and simulated anapole positions (Figure 5d), we found that the best agreement will be if we assume a 30 nm reduction in the fabricated disc diameter. This assumption is appropriate, since it is only a 3-6% reduction, and it is well within the accuracy of the used techniques for the mask fabrication and the following etching of the amorphous Si layer. Also it should be noted that the size of the fabricated discs cannot be measured more accurately neither with AFM nor with SEM (the latter is because of charging problems of non-conducting samples). Only this assumption was used as a fitting parameter, all other parameters (Si thickness and its dielectric permittivity) were taken from experimental measurements.

Numerical simulations. Scattering spectra and field distributions were calculated using a three-dimensional finite-difference time-domain (FDTD) method with a commercial software package (Lumerical). A simulation box of $3 \times 3 \times 3 \mu m^3$ was used with the perfectly matched layer conditions on every boundary and a mesh size of 5 nm over the volume of
the silicon disk. The silicon disk was excited by a normal-incident total-field/scattered-field plane wave source ranging from 750 to 2000 nm. The scattered power was then directly calculated by means of the transmission monitors surrounding the disk and the substrate. All calculations used the measured permittivity value of Si (Supporting Information, Figure S4), the refractive index of glass was set to 1.45.

**Multipole decomposition.** Multipole decomposition of scattering cross section was carried out in both Cartesian\textsuperscript{31,35} and spherical\textsuperscript{11} basis based on light-induced polarization $P(r) = \varepsilon_0 (\varepsilon_{\text{Si}} - 1) E(r)$, where $\varepsilon_0$ and $\varepsilon_{\text{Si}}$ are the vacuum permittivity and relative dielectric permittivity of silicon. $E(r)$ is the total electric field inside the disk, which can be obtained by the FDTD simulations. Thus, the Cartesian multipole moments can be defined as:\textsuperscript{16}

$$p_{\text{Car}} = \int P \, dr,$$
$$m_{\text{Car}} = -\frac{i}{2} \int [r \times P] \, dr,$$
$$\hat{Q}_{\text{Car}} = \int \left\{ 3 (r \otimes P + P \otimes r) - 2 [r \cdot P] \hat{U} \right\} \, dr,$$
$$\hat{M}_{\text{Car}} = \frac{\omega}{3!} \int \{ [r \times P] \otimes r + r \otimes [r \times P] \} \, dr,$$
$$T_{\text{Car}} = \frac{k^2}{10} \int \{ [r \cdot P] r - 2r^2 P \} \, dr,$$

where $p_{\text{Car}}, m_{\text{Car}}, \hat{Q}_{\text{Car}}, \hat{M}_{\text{Car}}, T_{\text{Car}}$ are the Cartesian electric dipole moment, the Cartesian magnetic dipole moment, the Cartesian electric quadrupole moment, the Cartesian magnetic quadrupole moment, and the Cartesian toroidal dipole moment, respectively. $\omega$ is the frequency of incident light, and $\hat{U}$ is the $3 \times 3$ unit tensor. The scattering cross section of the disk in free space $\sigma_{\text{scat}}$ thereby can be calculated as:

$$\sigma_{\text{scat}} \approx \frac{k^4}{6\pi \varepsilon_0^2 |E_{\text{inc}}|^2} \left| p_{\text{Car}} + T_{\text{Car}} \right|^2 + \frac{k^4 \mu_0}{6\pi \varepsilon_0 |E_{\text{inc}}|^2} \left| m_{\text{Car}} \right|^2 + \frac{k^6}{720\pi \varepsilon_0 |E_{\text{inc}}|^2} \sum \left| \hat{Q}_{\text{Car}} \right|^2 + \frac{k^6 \mu_0}{80\pi \varepsilon_0 |E_{\text{inc}}|^2} \sum \left| \hat{M}_{\text{Car}} \right|^2 \, \, .$$
For spherical multipole moments we exploit the following equations:

\[
\begin{align*}
\mathbf{p}_{\text{sph}} &= & \int \mathbf{P} j_0(kr) dr + \frac{5}{16} \int \left\{ [\mathbf{r} \cdot \mathbf{P}] \mathbf{r} - \frac{1}{3} r^2 \mathbf{P} \right\} \frac{j_2(kr)}{(kr)^2} dr, \\
\mathbf{m}_{\text{sph}} &= & -\frac{i\omega}{2} \int [\mathbf{r} \times \mathbf{P}] \frac{j_1(kr)}{kr} dr, \\
\dot{Q}_{\text{sph}} &= & \int \left\{ 3 (\mathbf{r} \otimes \mathbf{P} + \mathbf{P} \otimes \mathbf{r}) - 2 [\mathbf{r} \cdot \mathbf{P}] \dot{\mathbf{U}} \right\} \frac{j_3(kr)}{kr} dr + 6k^2 \int \left\{ 5 \mathbf{r} \otimes \left[ \mathbf{r} \cdot \mathbf{P} \right] - (\mathbf{r} \otimes \mathbf{P} + \mathbf{P} \otimes \mathbf{r}) r^2 - r^2 [\mathbf{r} \cdot \mathbf{P}] \dot{\mathbf{U}} \right\} \frac{j_4(kr)}{(kr)^3} dr, \\
\dot{M}_{\text{sph}} &= & \frac{\omega}{3} \int \left\{ [\mathbf{r} \times \mathbf{P}] \otimes \mathbf{r} + \mathbf{r} \otimes [\mathbf{r} \times \mathbf{P}] \right\} \frac{j_5(kr)}{(kr)^4} dr,
\end{align*}
\]

where \( \mathbf{p}_{\text{sph}}, \mathbf{m}_{\text{sph}}, \dot{Q}_{\text{sph}}, \) and \( \dot{M}_{\text{sph}} \) are the spherical multipole moments expressed in the Cartesian coordinates, and \( j_n \) denotes \( n \)-order spherical Bessel function. The far-field scattering cross section \( \sigma_{\text{scat}} \) can also be subsequently treated in the same manner as for the Cartesian multipoles, with noticing the disappearance of the toroidal dipole in the spherical basis.\textsuperscript{14,19}

**Far-field spectroscopy.** Optical properties of the fabricated Si discs were studied using spatially resolved linear transmission spectroscopy. The spectroscopic transmission analysis was performed on a BX51 microscope (Olympus) equipped with a halogen light source and fiber-coupled grating NIR spectrometer NIRQuest (Ocean Optics) with the wavelength resolution of 7.2 nm. The light was collected in the transmission configuration using the MPAnFL (Olympus) objective with magnification \( \times 100 \) (NA = 0.9). By using a pinhole in the image plane, we collected spectra from an area with a diameter of \(~20 \mu \text{m}\). The extinction experimental data in Figure 2 represent \( 1 - T_{\text{str}}/T_{\text{ref}} \), where \( T_{\text{str}} \) is the transmission spectrum measured from a single disc and \( T_{\text{ref}} \) is the transmission spectrum recorded from the clean glass surface.

**Near-field microscopy.** We used AFM-based scattering-type SNOM from NeaSpec with standard platinum-coated Si tips (Arrow NCPt from NanoWorld). The AFM tip was tapping with frequency \( \Omega \sim 250 \text{ kHz} \) and amplitude of \(~50 \text{ nm} \). The sample was illuminated normally from below, using a parabolic mirror to focus the light on a structure. The illumination spot size at the sample surface was estimated to be \(~3 \mu \text{m} \). The scattered
light was collected using a second parabolic mirror (Supporting Information, Figure S5). A Mach-Zehnder interferometer with an oscillating mirror \((f \sim 300 \text{ Hz})\) in the reference arm is incorporated in our SNOM in order to resolve both amplitude and phase using a pseudoheterodyne detection scheme.\(^{36}\) In order to remove background, the detected signal was demodulated at the fourth harmonic of the tip’s tapping frequency. The sampling interval was equal to the pixel time = 20 ms.

**SNOM decomposition method.** From simulations it is found that different near-field components of the illuminated disc have different symmetry (see Supporting Information, Figure S6). For example, at \(x\)-polarization of the incident beam, the near-field component \(E_x\) is symmetrical for both axis, i.e., \(E_x(-x) = E_x(x)\) and \(E_x(-y) = E_x(y)\), while \(E_y\) is asymmetrical: \(E_y(-x) = -E_y(x)\) and \(E_y(-y) = -E_y(y)\). Finally, \(E_z(-x) = -E_z(x)\) and \(E_z(-y) = E_z(y)\). Therefore, if we assume linear superposition in the recorded field

\[
E(x, y) = C_x E_x(x, y) + C_y E_y(x, y) + C_z E_z(x, y)
\]

then the following operation will give

\[
E(x, y) + a_x E(-x, y) + a_y E(x, -y) + a_x a_y E(-x, -y)
= (1 + a_x)(1 + a_y)C_x E_x + (1 - a_x)(1 - a_y)C_y E_y + (1 - a_x)(1 + a_y)C_z E_z
\]

Thus, by choosing \(a_x = a_y = 1\), the operation will result in the \(E_x\) component, \(a_x = a_y = -1\) will give \(E_y\), and \(a_x = -a_y = -1\) will give \(E_z\) (see Supporting Information, Figure S6). After such decomposition, applied for both incident polarizations, it was found that our SNOM is sensitive mostly to the \(z\) component, moderately to the \(x\) component, and almost negligibly to the \(y\) component of the near-field (see Supporting Information, Figure S6). Thus the decomposition method provides a reliable near-field \(E_z\) for both incident polarizations and near-field \(E_x\) for \(x\)-polarized incident beam. At first glance the decomposed \(E_x\) doesn’t look similar to the simulated one, because our SNOM measures background-free near-field,
using tip’s oscillations and following demodulation of the scattered signal. If we remove the background in simulations and plot $E_x - E_t$ (bottom row in Supporting Information, Figure S6), where $E_t$ is a transmitted plane wave, then a resulting field distribution will be more similar to the measured near-field $E_x$, featuring a ring of high amplitude and almost constant phase. In order to get a better correspondence between simulations and experiment, one could make simulations with a presence of the tip, oscillate altitude of the tip, calculate scattering in a specific solid angle, and then demodulate the scattered signal at the used harmonic. However, such approach would lack intuitive understanding and easy treatment of experimental data. The accuracy of our SNOM decomposition method can be estimated by choosing $a_x = -a_y = 1$, since it should give zero for any linear superposition of the electric field components, therefore, when applied to recorded SNOM maps, its result can be claimed as the error.

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Supporting Information Available

Spherical and Cartesian multipole decomposition; refractive index of Si; fabrication procedure; SNOM setup; SNOM decomposition procedure; decomposed near-field $|E_z|$ data for all measurements.
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