Experimental demonstration of tunable directional scattering of visible light from all-dielectric asymmetric dimers

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**ABSTRACT** Due to the presence of strong magnetic resonances, high refractive index dielectric nanoantennnas have shown the ability to expand the methods available for controlling electromagnetic waves in the subwavelength region. In this work, we experimentally demonstrate that an asymmetric dielectric dimer made of silicon can lead to highly directional scattering depending on the excitation wavelength, due to the interference of the excited magnetic resonances. A back focal plane imaging system combined with a prism coupling technique enables us to explore the scattering profile parallel to the substrate. The directivity of scattering along the substrate is high enough to produce selective guiding of light along the substrate. These results showing tunable control of directional scattering will encourage the realization of novel optical applications, such as optical nanocircuitry.

TOC Graphic



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Optical nanoantennas have been intensely studied due to their ability to manipulate visible light at the nanometer scale. A variety of applications, such as enhanced spectroscopic techniques, optical nanocircuitry, or solar cells, are expected to be boosted by optical nanoantennas1–4. Plasmonic nanoparticles made of noble metals could be a unit cell for highly efficient optical nanoantennas since they are capable of confining light on scales well below the diffraction limit5,6 and also of controlling the direction of light scattering depending on their shape and configuration7–12. Obtaining high directionality of scattered light from nanoantennas is desirable for the effective operation of many practical optical devices envisioned for optical nanocircuitry. Metallic nanoantennas, however, generally suffer from substantial absorption and energy losses in the optical and near infra-red (NIR) regimes because of both inter- and intra-band transitions13. Overcoming this issue has been a long-term challenge in designing optimal optical nanoantennas14.

Recently, optical nanoantennas made of high refractive index dielectric materials, such as silicon, gallium arsenide, gallium phosphide, and germanium, have been of high interest as an alternative to metallic nanoantennas15–26. Dielectric nanoantennas show near field enhancement and high scattering intensity due to the capability of exciting Mie resonances in these structures15,16. In contrast to metals, dielectric materials have low losses throughout the visible and NIR regimes since they have negligible conductivity. Dielectrics therefore have small to no imaginary components of their refractive indices owing to the absence of free electrons. Furthermore, not only electric but also magnetic resonances can be excited, even with simple nanoparticles such as nanospheres and nanodisks, made of such dielectric materials15–18. Accessing both electric and magnetic responses of dielectric nanoantennas offers more flexible ways to control the light propagation direction. For instance, the interference between the electric and magnetic dipolar resonances has reportedly shown unidirectional forward or backward scattering using simple configurations, such as a single nanoparticle or dimer, of high-index dielectric materials27–34.

The presence of magnetic resonances enables dielectric nanoantennas to exhibit another intriguing property, bidirectional scattering showing a dependence on the excitation wavelength. Whereas plasmonic nanoantennas require complex configurations, such as bimetallic structures to attain such functionality35,36, dielectric nanoantennas can offer easier ways to achieve bidirectionality. A V-shaped nanoantenna made of silicon has previously shown that scattered light can be routed depending on the excitation wavelength37. However, V-shaped antennas are only suitable to steer the scattering of light propagating towards the substrate at normal incidence. Tunable control of directional scattering along the substrate would be preferable for many applications, including optical nanocircuitry. Recently, we reported theoretical work showing that simple asymmetric dimers of silicon nanoparticles can route the direction of scattered light in the left or right directions depending on the wavelength of excitation38. We also showed that the interference between two magnetic dipole moments excited in each particle is the mechanism responsible for guiding the scattered light with the incidence polarization oriented along the dimer axis. One of the advantages of the asymmetric dimer system over other configurations is that they can achieve the desired directional tuning of scattered light travelling along a substrate; when illuminated by an incident wave propagating along the substrate, a dimer of dielectric nanodisks on the substrate can produce bidirectional scattering in the plane parallel to the substrate, depending on the excitation wavelength. Another theoretical study reported that asymmetric dimers of dielectric nanoparticles can exhibit the tuning of light scattering direction by varying illumination pumping intensity due to the excitation of electron-hole plasma.39

Here, we experimentally demonstrate that tunable optical bidirectional scattering can be obtained along the substrate using an asymmetric high-index dielectric dimer, specifically silicon in this case. Simple fabrication methods using electron beam lithography combined with lift-off and etching techniques were used to produce silicon nanodisk dimers on a transparent silica substrate. Far field radiation patterns of the scattered light were collected using a back focal plane (BFP) measurement technique combined with a prism coupling setup. The prism coupling setup with grazing incidence illumination generates an evanescent wave, which propagates along the substrate but decays exponentially in the air, and excites the nanoantenna. The results reported here demonstrate a proof of concept for the tuning of the scattering direction along the substrate with a simple dimer configuration and pave the way for realizing practical applications such as nanocircuitries or selective spectroscopy techniques.

Silicon nanoantennas were fabricated by electron beam lithography followed by reactive ion etching. First, silicon layer was deposited onto a silica substrate via plasma-enhanced chemical vapor deposition, followed by an etching procedure to achieve the thickness of 170 nm. The optical properties of the deposited silicon layer are shown in Figure S1 of the supporting information. The real part of the refractive index of silicon was larger than 3 over the visible-NIR range (400-800 nm), which is sufficiently high to generate the required electric and magnetic resonances of the silicon nanodisks on silica substrate40. Positive tone resist, PMMA (poly(methyl methacrylate)), was spin coated onto the substrate and baked at 180 °C for 5 minutes. The substrate was exposed with an electron beam followed by a development procedure in a MIBK (methyl isobutyl ketone) : IPA (isopropanol) = 1 : 3 solvent. The nanostructured PMMA was covered with a 40 nm Cr layer deposited by thermal evaporation, and then removed by a lift-off process in acetone. The remaining Cr mask pattern was transferred to the silicon layer via reactive ion etching. Subsequent Cr removal with commercial etchant produced the final sample. Figure 1a shows scanning electron microscopy (SEM) images of the fabricated asymmetric silicon dimer. The SEM observations confirmed that the side walls of the structures were vertical relative to the substrate and the gap distance between the pillars of the dimer was uniform from the bottom to the top of the disks. Isolated single nanodisks were also fabricated for comparison (see Figure S2 in the supporting information).

First, the backward scattering spectra of the fabricated samples was measured. An optical dark field microscope (Nikon, Ti-U) with a spectrometer was used to collect the back-scattered field from the silicon nanoantennas. The antennas were fabricated with a pitch of 5 um between antennas, which was sufficient to insure that scattering was collected from a single antenna at a time Incident light was polarized along the dimer axis using a linear polarizer along the illumination path. A dark field objective (Nikon LU Plan ELWD 100x NA0.80) was used both to provide the incident illumination and to collect the scattered light. The normalization of the measured signal intensity was carried out using a commercially available flat pellet of barium sulfate fine powder which can generate reflection close to the Lambertian pattern. Numerical simulations were conducted with finite difference time domain software (Lumerical) to calculate the scattering spectra of silicon nanodisks on a silica substrate. A dark field source was used for the numerical simulations to reproduce the experimental settings. The procedure the numerical analysis has previously been described elsewhere 34. The experimental and simulated scattering spectra are shown in Figure 1b. Theoretical and experimental results show good agreement, verifying that the experimentally fabricated samples have both electric and magnetic resonances as theoretically predicted. The experimental spectrum was shifted to shorter wavelengths by around 30 nm. This is likely due to small imperfections in the fabricated samples and the presence of a native oxide layer covering the surface of the nanoantennas. Such a native oxide layer would act to reduce the effective volume of the nanoantennnas17.



**Figure 1.** (a) SEM image of the fabricated asymmetric dimer of silicon nanodisks. The scale bar is equal to 100 nm. *H* = 170 nm, *D*1 = 180 nm, *D*2 = 250 nm and *d* = 40 nm correspond to height, diameters of the small and large nanodisks, and the gap distance, respectively. (inset) SEM image from tilted angle. (b) Dark field scattering spectra of the dimer obtained from experiment and simulation.

Measurements of the scattering patterns of the fabricated asymmetric dielectric dimer were then performed using the BFP imaging setup combined with prism coupling. The position of pixels in the BFP images corresponds to the light propagation angle12,35,37, making it a useful tool to characterize the directionality of the electromagnetic field of light scattered from nanoantennas. BFP techniques reported to date for measuring the scattering patterns from nanoantennas were, in general, designed to monitor the scattered field propagating towards the substrate with the incident illumination normal to the substrate10,12,35,37. That setup has not yet been used to investigate the scattering light in a plane along the substrate, which is the main objective in this study due to its importance for applications such as optical nanocircuitry or efficient light guiding in a medium on the substrate. Here, we employed a prism coupling setup combined with the BFP imaging technique. This system provides an evanescent wave to excite the nanoantenna and allows us to explore the distribution of the scattering field for light travelling along the substrate. A schematic of the experimental setup is shown in Figure 2. A Yb:KGW PHAROS laser system was used as the pump for an ORPHEUS collinear optical parametric amplifier with a LYRA wavelength extension option (Light Conversion Ltd, pulse duration 200 fs, repetition rate 100 kHz). The incident laser was directed towards the nanoantenna from through the silica substrate which was combined with the silica prism. The grazing incident light, which was set at an angle of 80 °, resulted in total reflection at the interface between the silica substrate and air, generating an evanescent wave to excite the nanoantenna. Light scattered from the nanoantennas was collected by an objective (Nikon CFI Plan Fluor 100x NA0.90). BFP images of the objective which corresponds to the scattering patterns were recorded with a CCD camera after being magnified by a set of two lenses (*f* = 75 mm and 175 mm) so that full BFP images can be collected in the sensor of the camera. There have been reported some measurement techniques to improve image quality of BFP from a single nanoparticle41,42. In this study, we used a pinhole to restrict the sample area and to collect signal from a single nanostructure. The enlarged view in Figure 2 also shows the scattering pattern calculated at  = 480 nm, ** = 0-180 ° and ** = 7 ° (details will be described later). The scattered light has a certain lobe width extending from the surface of the substrate to at least 45 °, which is detectable by the objective of NA0.90.



**Figure 2**. Schematic diagram of the optical setup for measuring the scattering along the substrate using a BFP imaging technique combined with a prism coupling. Evanescent field, which is generated by the total internal reflection at the interface of the substrate and air, propagates along the surface and decays exponentially into the air. The enlarged view on the right hand side includes the scattering pattern calculated at  = 480 nm, ** = 0-180 ° and ** = 7 °.

Figure 3 compares the calculated and measured scattering patterns for incident illumination at λ = 480 and 590 nm. Here, we define scattering towards the smaller particle side as the positive direction and scattering towards the larger particle side as the negative direction, as shown in Figure 3a. The scattering patterns obtained experimentally agree well with the calculated patterns. At λ = 480 nm, the scattered field was steered to the positive direction. On the other hand, at λ = 590nm the scattering direction is clearly seen to be towards the negative direction. From the results obtained in the numerical simulation, the steering angle was estimated as +9 ° at λ = 480 nm and -47 ° at λ = 590 nm. These angles would selectively guide light/signals with different wavelengths to two different angular regions38. For comparison, the experimental and simulated scattering patterns for single disks are also shown in Figure S3 in the supporting information. Even though the shape of the simulated and measured scattering patterns did not fully agree, due to the small signal collected from the isolated cylinders and the limitation in the sensitivity of the CCD camera, the single disks did not show any asymmetric side scattering in either the positive or negative directions, but only along the incident axis (0 °).



**Figure 3**. Scattering patterns from an asymmetric dimer of silicon nanodisks. (a) Schematic image describing how the scattering angle was defined. Scattering patterns were monitored for incident illumination at  = 480 nm (b,d) and  = 590 nm (c,e) in the simulation and experiment. Note that experimental results include some speckles, which is an undesirable artefact of light being scattered from the optical measurement setup or some defects/roughness on the sample’s surface.

In order to quantitatively analyze the directionality of the nanoantenna, we calculated the directivity, D, of the scattered light12,37. The directivity was estimated as the ratio of light intensity scattered to the two directions,

 (1)

where *S*(**,**) is the intensity of the scattered light at a given angle in the BFP image. **m = 57 °,

**p = 9 °, **n = -47 ° and ** = 7 ° were assigned for the calculation (see Figure 4a). Figure 4b shows a plot of the directivity as a function of the wavelength. The experimental results showed a reasonable agreement with the numerical ones obtained from simulation. The tuning of scattering direction from the positive to negative direction was clearly observed in the wavelength region from λ = 450 nm to λ = 650 nm. The range of the directivity reached -4.8 to +6.8 dB in the simulation and -3.1 to +3.1 in the experiment. The quantitative differences between the experiment and simulation may be caused by small errors in the fabrication and the undesirable speckles observed in the experimental BFP images. As shown in a recent theoretical study38, the scattering properties of an asymmetric dielectric dimer can be significantly affected by a variety of geometrical parameters of the constituent nanodisks. For example, the ellipticity of dielectric nanoparticles have been studied as a key factor to determine the scattering pattern in far field31,32,43,44. Also, the directivity defined by eq. (1) in this paper could be slightly shifted when strong noise is observed in the experimental BFP images. However, this does not change the conclusion that an asymmetric silicon dimer can steer light to selective directions depending on the wavelengths of excitation, as it can be seen in Figure 4b. To support the result of the scattering directivity, we plotted the peak scattering angle of *φ* in Figure S4 in the supporting information. The scattering angle was also clearly shifted from positive to negative by changing the excitation wavelength.



**Figure 4.** (a) A schematic image of the angular regions (blue and orange circles) used for the directivity calculation. (b) Directivity of the scattered field from the asymmetric silicon nanodimer calculated from the obtained scattering pattern in the simulation and experiment.

The scattered light measured in the experiment did not propagate exactly along the substrate since the objective used to collect the scattered light in this study had NA = 0.9, which corresponds to a collection angle of up to 64°. However, this NA value is enough as to observe the tail of scattering lobes from excited dipoles. This was confirmed via numerical simulation using an incident wave propagating along the substrate and the asymmetric dimer which has the same dimensions as in the experimental case. The calculated scattering patterns in the plane which is parallel to the substrate are shown in Figure S5 in the supporting information. Although the wavelengths and scattering angles used in Figure S5 were not the same as those used for the experimental demonstration due to the use of a different excitation method, the asymmetric nanodimer showed the capability of routing the scattering either to the left or to the right direction along the substrate. This result clearly suggests that the experimental demonstration in this study provides the proof of concept of controlling bidirectional scattering along the substrate using asymmetric dielectric antennas.

A monometallic Au dimer was reported to show side scattering, but not tuning of the scattering direction35. To highlight this, we conducted numerical FDTD simulations of the Au nanodimer under the same simulation condition as the Si nanodimer. As representative scattering patterns at  = 480 nm and 590 nm are shown in Figure S6 in the Supporting Information, the Au nanodimer showed a small steering of light and no tuning of the direction between negative and positive throughout the visible regime. That may be because the magnetic dipoles excited perpendicular to the dimer axis were mainly responsible to tune the scattering direction in the Si dimer case. However, in the case of the Au nanodisks, the asymmetric dimer cannot excite the magnetic resonances, resulting also in a very little steering of the scattered light.

In conclusion, we have experimentally demonstrated that an asymmetric dimer of silicon nanoparticles can tune the propagation direction of a scattered electromagnetic field to the left or right of the incident propagation axis depending on wavelength. The optical measurement setup using prism coupling and a BFP imaging technique enabled us to characterize the profile of the scattered field propagating along the substrate, demonstrating that the scattering direction can be routed by changing the excitation wavelength. This study shows that asymmetric high-index dielectric dimers could be a basic unit for spectroscopic techniques and nanocircuitry where tuning the light propagation direction would be beneficial to improve performance.

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*Supporting Information Available:* Additional information about refractive indices of the deposited silicon layer, the SEM image and scattering spectrum of single nanodisks, the scattering pattern of single nanodisks, and scattering patterns of asymmetric dimer in the plane which is parallel to the substrate are available free of charge via the Internet at http://pubs.acs.org.

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