

Magnetic and Electric Hotspots with Silicon Nanodimers

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ABSTRACT

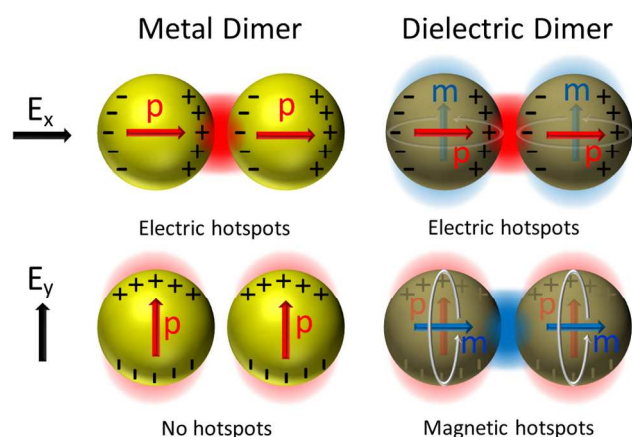
The study of the resonant behavior of silicon nanostructures provides a new route for achieving efficient control of both electric and magnetic components of light. We demonstrate experimentally and numerically that enhancement of localized electric and magnetic fields can be achieved in a silicon nanodimer. For the first time, we experimentally observe hotspots of the magnetic field, at visible wavelengths for light polarized across the nanodimer's primary axis, using near-field scanning optical microscopy.

KEYWORDS. Nanoantenna, Silicon nanoparticle, Optically-induced magnetic resonances

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5 Nanostructured dielectric materials with a high value of the refractive index have recently risen
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7 to prominence in the nanophotonics toolkit for control of light in the near-field [1-21]. They
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9 offer a novel way to directly engineer a magnetic field response at optical frequencies in addition
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11 to the electric field response in plasmonic structures. The basic building block – *a single*
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13 *dielectric spherical nanoparticle* – was shown to exhibit both electric and magnetic dipole
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15 resonances [1-10]. The magnetic resonance, recently experimentally demonstrated at visible
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17 frequencies in silicon (Si) [7-9] and gallium arsenide [10] nanoparticles, originates from circular
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19 displacement currents driven by an incident electric field, inducing a magnetic dipole moment
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21 perpendicular to the incident electric field. Similar to the case of plasmonic nanoparticles with
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23 electric dipoles, the induced magnetic dipole can be used to create more complex all-dielectric
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25 structures such as nanoantennas, metasurfaces and metamaterials [11-16]. Compared to metallic
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27 nanostructures employed in plasmonics, the near-fields in dielectric nanostructures immediately
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29 adjacent to the particles are less intense but can have larger far-field scattering cross sections
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31 [17]. Dielectric nanoparticles can reduce quenching and provide higher quantum efficiency of
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33 localized emitters than plasmonic structures which can be important for engineering emission at
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35 the subwavelength scale [17]. In addition to a single nanoparticle, *a nanodimer structure* should
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37 offer not only further tunability and engineering capabilities of the enhanced electric and
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39 magnetic fields but it should also reveal the physics of hotspots and near-field distributions
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41 known to be very important for nanoparticle oligomers with Fano-type resonances and strong
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43 inter-particle interactions [18, 19]. Dimer structures made of high-refractive-index materials have
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45 recently been studied through numerical calculations [17, 20-22] and also in experiments at
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47 microwave frequencies [23].
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This letter reports the first experimental demonstration of both electric and magnetic hotspots of an isolated silicon nanodimer in the visible spectral range. Experimental data sets are obtained by using near-field scanning optical microscopy (NSOM), and they are compared with finite-difference time-domain (FDTD) simulations and multipole decomposition.

For the case of the high-index dielectric nanodimer when magnetic dipoles couple with each other, enhanced near-fields are expected, in analogy with the case of electric dipoles. The response of a nanodimer can be studied for two basic configurations: light polarized parallel to the dimer axis (X-polarized light) and light polarized perpendicular to the dimer axis (Y-polarized light), as illustrated in Figure 1. The physics of a plasmonic (metallic) dimer is well known. It produces a coupled resonance and subsequent electric field hotspot for incident light polarized along its primary axis but does not exhibit strong coupling for light orthogonal to its axis [17]. The dielectric nanodimer should exhibit coupled resonances for incident light polarized both along and orthogonal to its primary axis. As illustrated in Figure 1, for X-polarized light, the dielectric dimer exhibits coupling of the induced electric dipoles while for Y-polarized light there is coupling between the magnetic dipoles.



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3 **Figure 1.** Comparison of metallic (plasmonic) and dielectric nanodimers. Plasmonic dimers
4 (left) only have electric dipoles which couple under a single polarization while dielectric dimers
5 (right) have both electric dipoles and magnetic dipoles that couple under the orthogonal polarizations.
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11 In this study, a nanodimer consisting of two silicon (Si) nanocylinders of diameter 140 nm and
12 height 150 nm placed on a quartz substrate is considered. Experimentally, gaps of 30 nm, 60 nm
13 and 120 nm are tested for varying degrees of coupling between the two particles. The system is
14 studied at visible wavelengths for both X- and Y-polarizations of incident light (using the
15 notation in Figure 1).
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23 Electromagnetic simulations are performed using the Finite Difference Time Domain method
24 (Lumerical FDTD). A silicon dimer is placed on a quartz substrate and illuminated using a
25 linearly polarized plane wave from the far-field, beneath the sample. Material parameters are
26 taken from ellipsometric measurements of amorphous silicon films used in the experiments.
27 Field monitors are used to observe the fields through and around the dimer. The key simulation
28 results for a dimer with a 30nm gap, are illustrated in Figure 2. For X-polarized light, the dimer
29 shows a single scattering peak at a wavelength of 600nm (see Figure 2a). For Y-polarized light,
30 the dimer shows two scattering peaks at 560nm and 650nm (Figure 2d). This is consistent with
31 earlier theoretical studies of far-field scattering spectra of a silicon nanodimer system [17].
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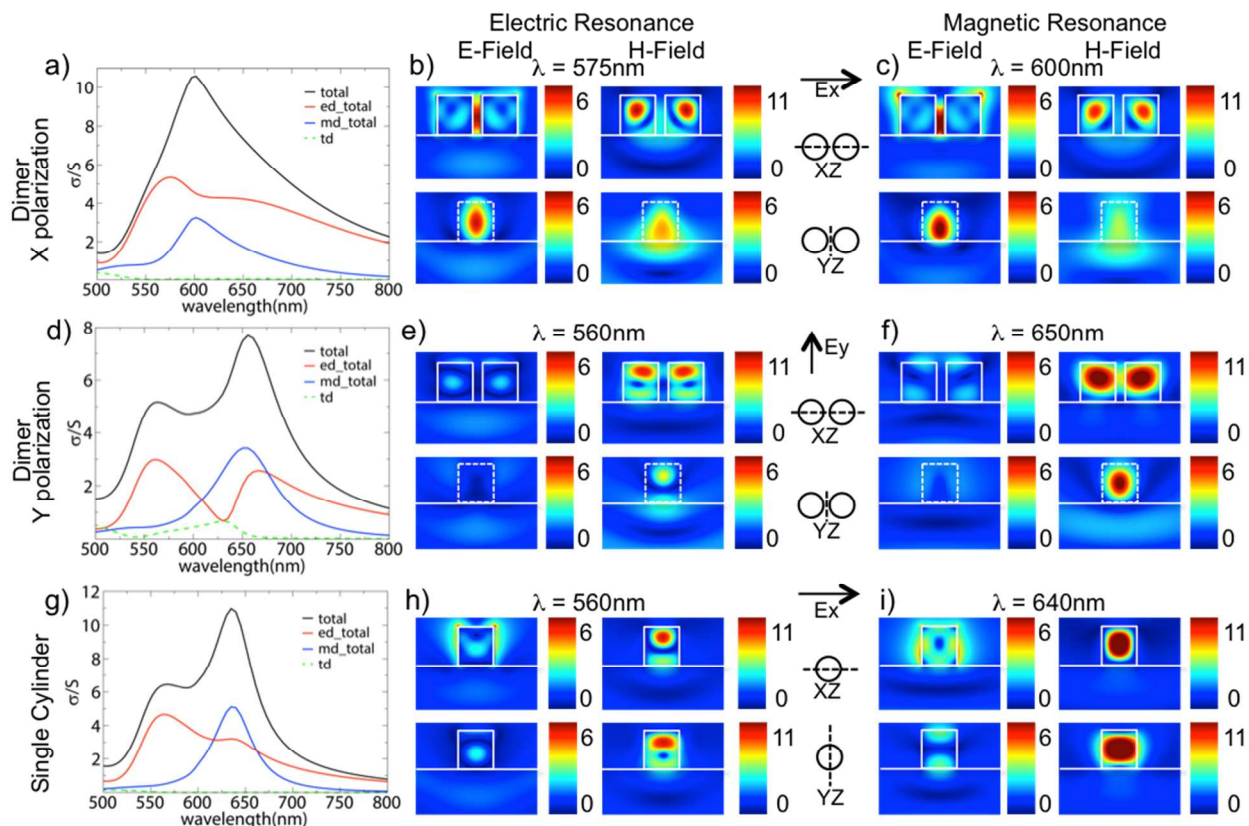


Figure 2. Numerical results for a silicon dimer (a-f) and a single cylinder (g-i) excited by plane wave from the substrate side. Each cylinder has a diameter of 140nm, a height of 150 nm and a gap of 30nm; (a-c) X-Polarized light scattering spectrum with mode decomposition; ed: total electric dipole = Cartesian electric dipole + toroidal dipole, md: magnetic dipole and td: toroidal dipole, plus cross section E- and H- field maps for the electric dipole resonance at 575nm and magnetic dipole resonance at 600nm. (d-f) Same as (a-c) but for Y-Polarized light, the electric dipole resonance is plotted at 560nm and the magnetic dipole resonance at 650nm. (g-i) Same as (a-c) but for the case of a single cylinder, the electric dipole resonance is plotted at 560nm and the magnetic dipole resonance at 640nm. For all maps, both electric and magnetic field amplitudes are normalized to incident plane wave. The size of each map is 450nm x 350nm. White lines indicate the substrate and particle positions.

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6 To gain a deeper insight into the different modes of the system, the multipole decomposition
7 approach is employed [24, 25]. Due to the presence of the substrate, it is performed by
8 integration of the polarization currents induced within the particles [24]. The multipoles are
9 defined in a Cartesian basis with coordinates' center coinciding with the dimer's center of mass.
10 In addition to conventional electric and magnetic multipole moments this representation includes
11 toroidal moments, the magnetic dipole mean-square radius [26, 27] and fully takes into account
12 substrate interactions while computing equivalent multipoles. For simplicity, when computing
13 the cross-sections associated with each multipole, they are considered to radiate into air (thus, the
14 total cross section is slightly higher than the sum over contributions).

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27 The multipole decomposition for X-polarized light reveals that the single peak in the scattering
28 spectrum is due to an overlap of the dipolar electric and magnetic resonances (Figure 2a). For Y-
29 polarized light, the total scattering cross section has two peaks (Figure 2d) which closely
30 resembles that of a single silicon cylinder (Figure 2g). The multipole decomposition reveals a
31 feature that has not been reported in previous theoretical studies [17], namely, a pronounced dip
32 in the total electric dipole contribution. The dip stems from the interaction between the
33 Cartesian electric dipole moment and the toroidal dipole moment which, due to their similar
34 radiated fields, are able to destructively interfere [26, 27]. The toroidal dipole moment is
35 generated by the magnetic dipoles induced in the disks that point in the same direction but are
36 not fully parallel due to the electric-magnetic dipole interaction. As these features originate from
37 the electric to magnetic dipole interaction, they are a unique to systems with a magnetic dipole
38 and thus not present in metallic dimers (see Supplementary Figure S1 for more details).
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3 The simulated electric and magnetic near-field profiles at resonant wavelengths are also shown
4 in Figure 2. For the dimer with X-polarized light there is spectral overlap of both electric and
5 magnetic field resonances. For the electric field, a hotspot is clearly observed between the two
6 particles (Figures 2b, 2c). Enhancements are observed for the magnetic field as well, but these
7 are mostly confined inside the individual silicon cylinders (Figures 2b, 2c). For the case of Y-
8 polarized light, the 560nm resonance (Figure 2e) is dominated by the individual electric dipoles
9 aligned towards Y-direction while the 650nm (Figure 2f) resonance is dominated by the coupled
10 magnetic dipoles.
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22 It is clearly observed that there is an enhanced and accessible (outside of the particles)
23 magnetic field for both X- and Y-polarized light. In the gap between the particles, the magnetic
24 field has a higher enhancement value for Y-polarized light (over 6x) compared to X-polarized
25 light (~3x). The high enhancement for the Y-polarized case is due to coupling between two
26 aligned magnetic dipoles of the particles. The enhancement inside the gap is lower than that
27 inside the nanoparticles (>10) but still high enough to be accessible for near-field measurements
28 or radiation enhancement of localized emitters. Deeper theoretical analysis of the magnetic field
29 enhancement of the silicon dimer as a function of the gap shows that the magnetic field
30 enhancement in the gap can reach a value of 10 for a gap of 5nm (see Supplementary Figure S3).
31 This value is similar to the magnetic field enhancement inside of each silicon nanoparticle.
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46 The electric field shows strong localization in the gap for the case of X-polarized light (over
47 6) with smaller localization for the case of Y-polarized light (~4x). For X-polarized light, the
48 electric field enhancement is due to coupling of aligned electric dipole resonances. It can also be
49 affected by discontinuity of the normal component of electric field \mathbf{E} at high-dielectric contrast
50 interface similar to high-index dielectric slot waveguides [28]. For X-polarized light, the electric
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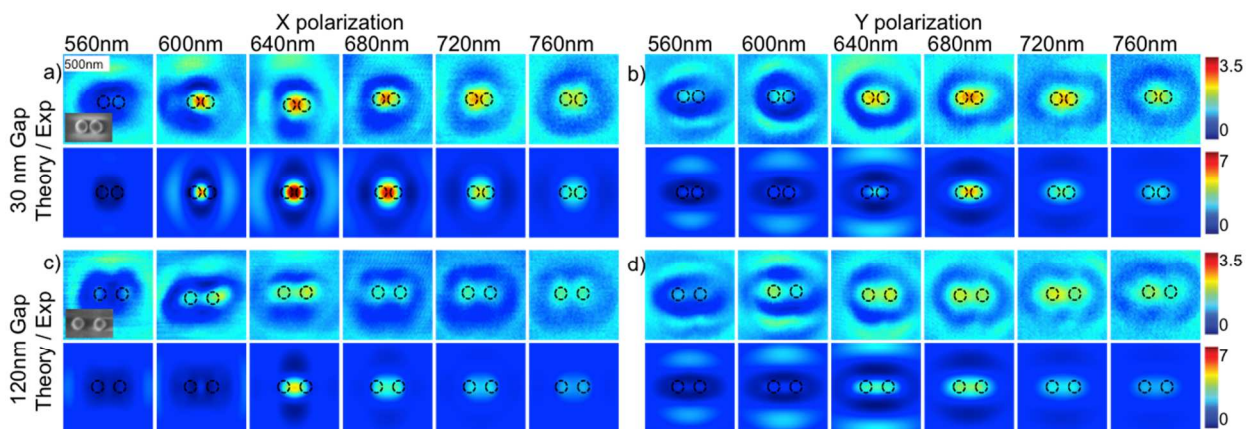
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3 field in the gap is stronger at the magnetic dipole resonance than at the electric dipole resonance.
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5 This can be explained by enhanced circular displacement currents of the electric field, which
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7 generate the magnetic dipole resonance. At the electric dipole resonance (575 nm) the
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9 contribution from the magnetic dipole is low, therefore the observed enhancement is attributed to
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11 the electric dipole mode only. In comparison, at the magnetic dipole resonance (600 nm) a
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13 superposition of both ED and MD modes is observed, which leads to higher collective
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15 enhancement of electric field.
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20 To experimentally observe the discussed field enhancement, dimers of silicon nanoparticles
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22 were fabricated using a standard top-down nanofabrication approach. A 150nm thick amorphous
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24 silicon (Si) film is grown on a cleaned quartz substrate using inductively couple plasma chemical
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26 vapor deposition (Plasmalab System 380, Oxford Instruments). The dielectric function of the
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28 fabricated amorphous films is measured through the visible spectral range using ellipsometry.
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30 Electron beam lithography (Elionix, 100kV) is performed using Hydrogen silesquioxane (HSQ,
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32 Dow Corning, XR-1541-002) as the resist. Unexposed HSQ is removed with
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34 Tetramethylammonium hydroxide (TMAH, 25%). The sample is then etched using an
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36 inductively coupled plasma etcher (Plasmalab System 100, Oxford) to create Si nanodimers on
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38 the quartz substrate. The resultant structures are twin 150nm tall Si nanopillars with a diameter
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40 of 140nm. Gaps of 30nm, 60nm and 120nm are explored. The dimers are fabricated with a 5
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42 micron pitch to allow for measurements of single uncoupled dimers.
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48 The near-field profiles are studied experimentally with a Near-Field Scanning Optical
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50 Microscope (NSOM, from Nanonics Imaging Ltd). The sample is illuminated from the far-field
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52 using linearly polarized light, focused to a spot size of approximately 5 microns on the sample.
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54 The light originates from a supercontinuum source (SuperK Power, NKT Photonics) and specific
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wavelengths are selected using a variable bandpass filter (SuperK Varia, NKT Photonics) and then polarized with a Glan Thompson prism. Near-field maps are created by collecting light in a transmission configuration through a tapered fiber (coated with Chrome and Gold) with a 110nm aperture. The wavelength is adjusted throughout the visible with a bandwidth of 10nm. To avoid any contribution of polarization sensitivity of the tip, the sample was rotated 90° relative to the incident light polarization to measure the orthogonal polarization. Near-field maps are compiled for X- and Y-polarizations of incident light normalized to an averaged background signal >1 micron apart from the dimers.

Experimental near-field maps are illustrated in Figure 3 for wavelengths throughout the visible for both X- and Y-polarized light (For more in-depth mappings, see Supplementary Figures 4, 5 and 6). These are directly compared with simulations of electromagnetic fields collected by an NSOM tip. To account for the sensitivity of the NSOM probe in the simulations, a method based on the electromagnetic reciprocity theorem [29] is applied (this is similar to Refs. [30, 31]). According to this approach the signal collected by a near-field probe is determined by a specific convolution integral (mutual impedance) of the fields emerging from the aperture of the tip operating in excitation mode with the fields in the vicinity of the structure under study. The simulated near-field images correspond well to the experimental images.



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3 **Figure 3.** Experimental near-field maps of Si nanodimers compared with simulated near-field
4 maps, as a function of wavelength, dimer gap dimension and polarization. (a) and (b) show the
5 30nm gap dimer with X- and Y-polarized light respectively. (c) and (d) show 120nm gap dimer
6 with X- and Y-polarized light respectively. Each column represents a different wavelength,
7 labeled at the top. The area of each plot is $1 \times 1 \mu\text{m}$.
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16 Distinct resonance features and evolution as a function of wavelength are observed for the four
17 gap and polarization cases in figure 3. For the case of the 30nm gap and X-polarized light, the
18 dimer provides a near-field absorptive response at 560nm. This quickly turns into an
19 enhancement effect as the near-field grows to a maximum at 640nm with a circular feature
20 surrounded by near-field extinction. This feature evolves and decreases in intensity through the
21 red. At 760 nm, the map shows an elongated feature with two lobes.
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30 Looking at Y-polarized light for the 30nm gap, the mode starts out, at 560 nm, as a minor
31 elongated feature in the X-direction, surrounded by a null. As the wavelength shifts to the red,
32 an X-elongated feature grows in intensity at the center with a field suppression around it. It
33 reaches a maximum and then decreases, while maintaining its elongated shape as the wavelength
34 moves to red.
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42 For the case of the 120nm gap with X-polarized light, the NSOM features closely resemble
43 that of two separate particles. For 560nm, extinction in the form of two adjacent particles is
44 clearly observed. The near-field feature of each particle increases in intensity and then decreases
45 at longer wavelengths. The case of the 120 nm gap with Y-polarized light, is fairly similar to the
46 X-polarized case, but a slightly higher NSOM signal-enhancement value is measured along with
47 a stronger resonance through the red portion of the spectrum. This is attributed to moderate
48 coupling between the induced magnetic dipoles positioned end to end in the X-direction.
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3 Comparing the NSOM maps obtained for the dimers with 30 and 120 nm gap one can
4 immediately see that for the case of the smaller gap, the NSOM signal reaches larger
5 enhancement for both polarizations. This provides experimental evidence of field localization via
6 electromagnetic coupling of the two silicon nanocylinders.
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12 The collected NSOM signal consists of both electric and magnetic fields. As it can be seen
13 from simulated field components (see Supplementary Figures 3, 4 and 5) the experimentally
14 measured signal strongly resembles that of the transverse magnetic field, H_y for the X-
15 polarization case and H_x for the Y-polarization case, with more minor contributions from the
16 transverse electric fields.
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24 The electromagnetic response of isolated Silicon nanodimers of three different gaps (30nm,
25 60nm and 120nm) has been experimentally mapped in the near-field throughout the visible
26 portion of the spectrum. A strong spectral response is observed for each of the three gaps for
27 two orthogonal polarizations. The strongest enhancement (3.5) is observed for X-polarized light
28 with the 30nm gap; in this case, there is a coupled resonance response of the electric field and a
29 strong magnetic field response for dimer. The weakest enhancement (2.0) is observed for X-
30 polarized light and the 120nm gap; the electric field and magnetic field response resembles that
31 of two isolated nanoparticles.
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43 From the simulations, it is clear that the highest magnetic fields occur inside the silicon
44 nanoparticles. However, the simulations in conjunction with the near-field experiments clearly
45 show that enhanced magnetic fields are accessible outside of the silicon, both in the gap of the
46 dimer and in the plane above the dimer. These enhancements are available for both X- and Y-
47 polarizations with stronger magnetic fields for the Y-polarized case, where the magnetic dipoles
48 couple with each other.
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3 In conclusion, we have presented, for the first time to our knowledge, the experimental
4 evidence of near-field enhancement of the magnetic fields with silicon resonant nanodimers at
5 visible frequencies. The response of the system has been studied as a function of wavelength,
6 polarization and gap. Theory and numerical simulations predict that such a structure should have
7 a strong magnetic response; and our simulations confirm a very good correlation with the near-
8 field measurements. When the simulated near-fields are broken down into their components, it is
9 confirmed that the resonance measured in the near-field is strongly magnetic in nature, for both
10 X- and Y-polarizations. Additionally, we have conducted a complete analysis of the modes
11 excited inside the dielectric nanodimer system by means of the multipole decomposition. This
12 analysis has revealed excitation of a toroidal dipole moment which interferes with the Cartesian
13 electric dipole moment resulting in a dip in the total electric dipole radiation. This is the first
14 reporting of toroidal dipole excitation in dielectric nanodimer antennas. The dimer system is the
15 basic building block for many nanophotonics systems. The results demonstrated in this paper lay
16 the ground work for future engineering of the magnetic field response at optical frequencies with
17 high-index dielectric nanostructures for the ultimate control on the subwavelength scale.
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42 **Author Contributions**

43 RMB performed the near-field experiments and analysis, and wrote the manuscript with inputs
44 from all coauthors. DP performed simulation of near-field detected by the NSOM tip and
45 contributed to NSOM data acquisition and analysis. YFY performed nanofabrication of the
46 silicon dimers. DM performed FDTD simulation of far- and near-field properties of the silicon
47 dimers. RPD performed the multipole decomposition calculations and analysis. LG developed
48 and performed the Si etching for the sample. AKS contributed to NSOM data acquisition and
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3 analysis, and coordinated the simulation work. YSK supervised the work of ITMO team. BL
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5 supervised the work of DSI team. AIK proposed the initial idea, organized and coordinated the
6
7 whole work. All authors contributed to the manuscript preparation and reviewed the final version
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9 of the manuscript.
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24 25 **Notes**

26
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