Directional Modulation of Exciton Emission Using Single Dielectric Nanospheres

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Optimized functionalities. For instance, the low quantum efficiency and poor emission directionality of monolayer transition-metal dichalcogenides (TMDs) have limited their practical applications in integrated photonics[4] and flexible optoelectronic systems[5] due to unwanted signal degradation. Tailoring the incident and radiation fields is thus particularly meaningful. However, challenges exist in both efficiency and complexity. When optical components are coupled at the nanoscale, not only do material losses cause decreased efficiency, but also very fine alignment between the elements is required. Therefore, a single subwavelength modulator working at visible wavelengths is highly desired for achieving higher coupling efficiency and miniaturized device size.

The last decades have witnessed the rapid development of optical nanoantennas as a promising solution to manipulating optical fields at the nanoscale, [6] leading to drastically enhanced light emission,[6b,7] photodetection,[8] and optical sensing.[9] Both plasmonic resonances based on noble metals[10] and Mie resonances from high-index dielectric materials[11] have been explored to facilitate strong light–matter interactions in the near field. Uniquely, dielectric nanoantennas allow simultaneous directional excitation and exciton emission at the subwavelength scale with high efficiency. Low-loss dielectric nanoantennas hold particular promise for this purpose, owing to their strong Mie resonances. Herein, a highly miniaturized platform is explored for the control of emission based on individual subwavelength Si nanospheres (SiNSs) to modulate the directional excitation and exciton emission of 2D transition metal dichalcogenides (2D TMDs). A modified Mie theory for dipole–sphere hybrid systems is derived to instruct the optimal design for desirable modulation performance. Controllable forward-to-backward intensity ratios are experimentally validated in 532 nm laser excitation and 635 nm exciton emission from a monolayer WS2. Versatile light emission control is achieved for different emitters and excitation wavelengths, benefiting from the facile size control and isotropic shape of SiNSs. Simultaneous modulation of excitation and emission via a single SiNS at visible wavelengths significantly improves the efficiency and directionality of TMD exciton emission and leads to the potential of multifunctional integrated photonics. Overall, the work opens promising opportunities for nanophotonics and polaritonic systems, enabling efficient manipulation, enhancement, and reconfigurability of light–matter interactions.

Recent advances in 2D semiconductors,[1] quantum dots,[2] and color centers[3] have showcased several opportunities for next-generation integrated photonic devices, such as nanoscale light sources. Effective control over the emission properties is of great importance for these novel emitters in order to realize optimized functionalities. For instance, Jie Fang, Dr. K. Yao, Prof. Y. Zheng
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excitation of magnetic and electric resonances,[11h,12] whereas generating magnetic responses at optical frequencies may be challenging in plasmonic structures.[11h,13] Dielectric nanoantennas can therefore implement interesting optical field manipulations within extremely simple geometries,[14] e.g., a single nanosphere, as compared to complex shapes and arrays typically required in plasmonic designs.[15] In addition, metals fundamentally suffer higher material losses than dielectrics, especially in the visible region,[16] which may further decrease the already low efficiency of nanoemitters. Consequently, the use of dielectric resonators appears to be an ideal solution to developing efficient subwavelength emission modulators.

The mutual interference of size-dependent magnetic and electric modes in dielectric nanoantennas can be used to efficiently modulate the far-field radiation pattern.[11h,17] For example, directional scattering of plane waves has been readily achieved at microwave,[18] THz,[19] and optical frequencies,[20] where directional modulation of both excitation and emission can provide more degrees of freedom to control the overall emission process. Cihan et al. have demonstrated directional modulation of monolayer MoS$_2$ emission with silicon (Si) nanowires.[21] But the challenge on the device size and the inconsistency in modulation depth along different device orientations (i.e., the radial and axial orientations of nanowires) remain, limiting the future device integration. From this perspective, dielectric nanospheres represent a compelling platform. On the one hand, thanks to their subwavelength nature in all dimensions, they can strongly enhance the emission of the coupled emitters along all sample orientations; on the other hand, they also provide smaller footprints as well as effective trapping of the excitation light, which both exerts the advantage of compactness and remedies the low efficiency. Finally, given their mature industrial base, Si-based nanoantennas[22] can ensure better compatibility with the existing complementary metal–oxide–semiconductor and emerging integrated photonic platforms.[23]

In the literature, the controllable emission of a dipole coupled with a dielectric sphere has been demonstrated at microwaves.[24] However, it is still challenging to practically realize such a subwavelength platform in the visible region using silicon nanospheres. To this extent, a universal analytical model for 3D systems[22b,23] can provide physical insights into how the dipole-excited Mie resonances interfere and describe the evolution of the radiation patterns under various conditions. Sphere size control and good sphericity are also important, but can hardly be realized,[20a,27] especially for smaller spheres that supports lower-order modes at visible wavelengths.[11b,d,h,28]

In this work, we resolve these challenges by proposing a highly miniaturized emission control platform based on single subwavelength nanospheres. A rigorous multipoles model is first derived based on Mie theory,[22] to describe the far-field radiation pattern of the nanosphere-modulated dipole emission. Based on reciprocity theorem, this model instructs the possibility and optimal conditions of directional modulation of both excitation and emission processes via single nanospheres (Figure 1a,b). Then, using spherical dielectric nanoresonators, we experimentally demonstrate versatile directional modulation of 532 nm laser excitation and 635 nm exciton emission from a monolayer WS$_2$ with controllable forward-to-backward intensity (F/B) ratios in 125 samples, showing statistical agreement with our theoretical model and numerical simulations. The employed nanoantennas are single hydrogenated amorphous Si nanospheres (a-SiNS:Hs) with facile size control (200–500 nm)[29] and excellent sphericity (see Figure S1 in the Supporting Information). The a-SiNS:Hs support low-loss multipolar resonances down to 450 nm wavelength, spanning the whole visible range.[30] Unless otherwise noted, we will

Figure 1. Directional control on both excitation and emission processes of a dipole via single SiNS resonators. a, b) Analytical calculation of the far-field radiation patterns of a 532 nm (wavelength) dipole coupled to a 390 nm (diameter) SiNS (a) and a 635 nm (wavelength) dipole coupled to a 250 nm (diameter) SiNS (b). Both (a) and (b) are normalized. 0° is defined as forward (F), while 180° is defined as backward (B). Considering the reciprocity theorem (see Figure S5 in the Supporting Information), the extracted forward-to-backward (F/B) ratio of the emission process (schematic in (b)) can also be used to describe the forward and backward excitation of the near field at the position of the dipole (schematic in (a)). c) Numerical simulation of the F/B ratio mapping as a function of wavelength and SiNS diameter. A glass substrate is considered as compared to the free-standing SiNS in the analytical model. The corresponding cases in (a) and (b) are denoted by the green and red rhombi, respectively. Influence of the glass substrate on the F/B ratio mapping as a function of wavelength and SiNS diameter. A glass substrate is considered as compared to the free-standing SiNS in the analytical model. The corresponding cases in (a) and (b) are denoted by the green and red rhombi, respectively. Influence of the glass substrate on the F/B ratio mapping as a function of wavelength and SiNS diameter. A glass substrate is considered as compared to the free-standing SiNS in the analytical model. The corresponding cases in (a) and (b) are denoted by the green and red rhombi, respectively.
refer a-SiNS:HS as SiNSs for the ease of reading. Based on this platform, we tailor with large flexibility the emission properties. Various emitters and excitation wavelengths are used to demonstrate highly directional forward emission with maximized forward excitation efficiency, matching well with the predicted performance. Under optimized directional excitation, highly directional emission with a total enhancement up to fivefold is observed, which significantly enhances the efficiency and directionality of the emission by 2D TMDs or other nanoemitters. Integration of TMDs to resonant nanostructures has the advantage of controllable and accurate assembly and low-cost fabrication.\cite{1a,b,23} Our results manifest the efficient and versatile modulation of exciton emission at visible wavelengths via a single subwavelength nanosphere and thus promote the device miniaturization in all dimensions, opening promising opportunities for nanophotonics,\cite{3b} valleytronics\cite{31} and polaritonic systems.\cite{32}

While the optical properties of nanoantennas can be evaluated numerically, it is of both theoretical and practical interest to perform an analytical study to gain physical insights into the role of each resonance in the total directional modulation. For the sake of convenience, we omit the presence of a substrate in this analysis and consider a SiNS with diameter 2a coupled with a tangential electric dipole emitter positioned at the distance d = 1 nm. The localized dipole models the exciton emission from the monolayer TMD. Because of the Mie resonances supported by the SiNS, this configuration is able to redistribute both the excitation power on the dipole\cite{22a} and the emitted power from the dipole\cite{11b,12b,d} under different conditions, resulting in tunable directional excitation and emission. The directional modulations of excitation and emission processes are reciprocal of each other (see Figure 1a,b and Section SIV in the Supporting Information).

The scattering problem of a sphere excited by a coupled electric dipole has been analyzed in the literature but mainly in the near-field region to study the modification of the emitter’s decay rates.\cite{33} In order to quantify the directionality, referred to as F/B ratio in the present work, we revisit this problem and perform rigorous analysis in the far-field region. Following a similar procedure as in standard Lorenz–Mie theory, we expand the incident field, i.e., the exciton emission, and the scattered field, into a series of spherical functions.\cite{34} Compared with the standard Mie theory under plane wave illumination, the local excitation by an electric dipole results in very different coefficients of the series. The 3D nature of the present system also introduces much more significant complexity in the algebra than in the case of nanowires.\cite{23} For a tangential electric dipole located at (a + d, 0, 0) in the spherical coordinates, the entire field can be derived from two Debye potentials u and v, which correspond to the transverse electric and transverse magnetic fields with respect to the radial direction, respectively. The expression of the potentials reads

\[
\frac{1}{r} \sin \theta \cos \phi \sum_{n=1}^{\infty} \frac{(2n+1)}{n(n+1)} \left[ a_n \mathcal{C}_n(k_0r) - \phi_n(k_0r) \right] \mathcal{C}_n(kr) P_n(\cos \theta) \tag{1}
\]

and

\[
\frac{1}{r} \sin \theta \sin \phi \sum_{n=1}^{\infty} \frac{(2n+1)}{n(n+1)} \left[ b_n \mathcal{S}_n(k_0r) - \phi_n(k_0r) \right] \mathcal{S}_n(kr) P_n(\cos \theta) \tag{2}
\]

for the region \( r > r_c = a + d \). Here, \( a_n \) and \( b_n \) are the Mie coefficients, known as functions of the size parameter \( k \cdot a \); \( k_0 \) and \( k \) are the wave vectors in free space and in SiNS, respectively; \( \phi_n \) and \( \mathcal{C}_n \) are the Riccati–Bessel functions related to the spherical Bessel function and spherical Hankel function of the first kind, respectively; \( P_n \) is the Legendre polynomials and the prime denotes a derivative. Details of the derivation can be found in Section SIII in the Supporting Information. The dependence of the Mie coefficients on \( k \cdot a \) suggests that the size parameter plays an important role in the directional modulation, showing the potential for versatile designs in both excitation and emission processes via SiNS sizes.

Utilizing the developed model, we first examine the radiation patterns for different parameters by evaluating the outgoing Poynting vector in the far-field region. The dielectric function of Si is confirmed by fitting the scattering spectra as discussed in Section S1 in the Supporting Information. In Figure 1a,b, two examples are presented for SiNSs of different radii and at different emission wavelengths. A highly forward directed modulation can be found at 532 nm wavelength with a 390 nm SiNS (Figure 1a), while more backward components can also be achieved at 635 nm wavelength with a 250 nm SiNS (Figure 1b). Importantly, reciprocity (Section SIV, Supporting Information) ensures that the near field intensity at the position of the emitter excited by a 532 nm plane wave will be similarly modulated by a 390 nm SiNS, with the same F/B ratio. 532 and 635 nm wavelengths are chosen for illustration according to the excitation and emission studied experimentally in the next section.

To account for the substrate effect and make the model better suited to the experimental demonstration, we also conduct full-wave simulations with the presence of a semi-infinite substrate of glass using CST Studio Suite. Figure 1c shows that the ratio of incident/emitted power in the forward direction to the one in the backward direction (F/B ratio), as a function of wavelength and the size of SiNS. The mapping by the analytical approach shows basically the same tendency in Figure S6 in the Supporting Information. Since the emission property is determined by both dipole excitation and its decay channels, the modulation is twofold: on the one hand, the nanoemitter needs to be coupled with an SiNS of optimal size to realize the most efficient excitation at a given wavelength and from a certain direction; on the other hand, the SiNS should be tailored to also enhance radiation in the preferred direction at the emission wavelength. Here, Figure 1c and Figure S6 in the Supporting Information can serve as a graphical guide to determine the dimension of SiNS, excitation wavelength, and incident direction for implementing the desired emission properties.

To experimentally demonstrate controllable F/B ratios, a chemical vapor deposition (CVD)-grown monolayer WS_2 flake is selected as the emitter (see Section SIII in the Supporting Information), and SiNSs are drop-cast on the top, as shown in Figure 2a. We modify our microscope system with laser excitation from both top and bottom (Figure 2b) to implement forward and backward modulated excitation/emission when the sample faces either up or down. In order to study the directional modulation of the excitation and emission individually, we have to separate these two processes efficiently.

\[
I_{F/B}^{\text{Em}} = f_{F/B}^{\text{Em}} \left( I_0^{\text{Em}}, R_0 \right) \equiv A_{F/B}^{\text{Em}} I_0^{\text{Em}} \tag{3}
\]
Figure 2. Schematic diagram and experimental demonstration of controllable directional modulations of excitation and emission separately. a) Schematic of the monolayer WS$_2$ emitter modulated by a single SiNS on it. F: forward; B: backward. b) Sketch of the experimental setups measuring the forward modulated WS$_2$ emission (sample facing down) under forward excitation (right) and backward excitation (left), respectively. Backward modulated emission can be measured similarly with sample facing up. The blue triangles refer to CVD-grown monolayer WS$_2$ flakes. c,d) Measured (black dots) and simulated (green/red curves) F/B ratios as a function of SiNS diameter on both 532 nm excitation (c) and 635 nm WS$_2$ emission (d). The ranges of y axes are selected for better display. See the whole curves for simulated F/B ratios in Figure S11 in the Supporting Information.

where $P_0$ is the incident power density, $I_0^{\text{Ex(Em)}}$ is the excitation (Ex/emission (Em)) light intensity without modulation, and $I_{F(B)}^{\text{Ex(Em)}}$ is the forward (backward) modulated intensity via SiNS. A constant $\alpha_{F(B)}^{\text{Ex(Em)}}$ can be used to represent the modulation function $I_{F(B)}^{\text{Ex(Em)}}$ in the low power (linear) regime ($P_0 \leq 70 \, \mu W \, \mu m^{-2}$). Experimental proofs and detailed discussions can be found in Section SVII in the Supporting Information.

The comparison of the forward and backward modulated emission is carried out when the modulation of excitation is along a fixed direction, and vice versa for studying the directionality of excitation. For example, as shown in Figure 2b, with our sample facing down we collect the forward modulated emission signals from the bottom and determine the F/B ratio for excitation along a fixed direction, and vice versa for studying the directionality of emission. More details on the measurements can be found in Section SVIII in the Supporting Information.

Finally, the results from 125 samples are summarized in Figure 2c,d, showing a good agreement with our theoretical predictions statistically. The fluctuation of the data can be further avoided by introducing effective diameters as illustrated in Figure S17 in the Supporting Information. They reveal that controllable directional modulation can be simultaneously achieved in both excitation and emission. The combined effect can thus be designed to tailor the overall exciton emission properties, given that the size of SiNSs can be well controlled across several hundreds of nanometers. It is worth noting that the directionality can become much larger if the part of monolayer WS$_2$ not covered by the SiNS is etched$^{[33]}$ or a smaller numerical aperture is used for signal collection. Exciton diffusion may further exacerbate the unwanted signals from the surrounding WS$_2$. That is why we see an obvious difference in the F/B ratio between experiments and simulations when it is larger than one. As for the relatively similar F/B ratio when it is smaller than one and other discussions, please see the detailed explanations in Section SVIII in the Supporting Information.

Anyhow, this work mainly focuses on controllable directionality, and the current results in Figure 2c,d clearly demonstrate the phenomenon. From this perspective, we extract the system-dependent relation (Equation (S15), Supporting Information) between simulated and measured F/B values (see y axes in Figure 2c,d) and apply it to the prediction of performance in more versatile emission designs in the following.

The F/B ratio can be up to 2.5 and down to 0.5 owing to the mutual interference of different resonances supported by SiNS. The mode contributions at different wavelengths can be extracted from our modified Mie theory for dipole excitation, yielding the scattering efficiency

$$Q_{\text{dipole}} = \frac{3}{4 \pi k_0^2 r_s^2} \sum_{n=1}^{N} \left[|a_n|^2 |\zeta_n' (k_0 r_s)|^2 + |b_n|^2 |\zeta_n (k_0 r_s)|^2 \right]$$

Compared to the standard model for plane-wave excitation, $a_n$ and $b_n$ here are modulated by the spherical functions $\zeta_n$ and their derivatives, respectively. This difference stems from the rich near-field components of the dipole emission. Two examples are presented in Figure 3a.c. For fixed wavelengths, the multipolar superposition at different SiNS sizes is also drawn in the shaded background in Figure S11 in the Supporting Information for both 532 nm excitation and 635 nm emission. Figure 3b illustrates the forward-enhanced 532 nm excitation enabled by a 390 nm SiNS, as analytically studied in Figure 1a. As another typical scenario of interest highlighted in Figure 1b, backward-enhanced 635 nm emission enabled by a 250 nm
SiNS is shown in Figure 3d. All these results are normalized based on the isolated monolayer WS$_2$ emission without modulation (black curves in Figure 3b,d). For a given SiNS modulator, we can straightforwardly understand the directional modulation as the difference between the solid (forward) and dashed (backward) colored curves. However, in Figure 3b,d, each emission spectrum represents the total modulation of the emission property, instead of the separate modulation of excitation and emission discussed so far. For instance, the forward-enhanced 532 nm excitation via a 250 nm SiNS (Figure 3d) makes the total emission always larger than the pure WS$_2$ emission no matter whether it emits in the forward or backward direction. Similarly, we can find that the green curves are both larger than the black curve in Figure 3b, due to the backward-enhanced modulation of emission. The schematics of the two-step modulation according to Figure 3b,d can be found in Figure S13 in the Supporting Information.

In order to study the directional modulation exclusively on the excitation (emission) process, the modulation of emission (excitation) must be fixed along a specific direction, either forward or backward. This provides us two choices in experiments, and we can choose the fixed direction that gives us a higher signal-to-noise ratio in the measurements (e.g., stronger peaks in green and red in Figure 3b,d). The mechanism behind such a choice can be attributed to the different magnitudes and phases of the multipoles in a SiNS when excited by a 532 nm plane wave or a 635 nm dipole emission. The field quantities at arbitrary positions modulated by these multipolar resonances can be easily extracted from our analytical model and straightforwardly displayed in phasor diagrams, as shown in Figure 4 and detailed in Section SXII in the Supporting Information. Then, based on the universality of our model and the control on the SiNS size, the phasor diagrams can further suggest the optimal parameters for emission control at any excitation wavelength.

To demonstrate our ability of versatile emission control, we start from the design of highly directional forward emission under maximum forward excitation efficiency by choosing suitable SiNSs for different emitters (monolayer WS$_2$ or MoS$_2$) and excitation wavelengths (446 or 532 nm). Figure 4a,b,d,e presents the design mechanism for a minimum backward component in both emission and excitation intensity distributions. Here, the maximum forward excitation is simply chosen for easier experimental demonstrations, while backward excitation can also be designed straightforwardly. Although we derive the complex-valued phasor vectors based on a common plane-wave-sphere model for excitation (Figure 4a,d) and a modified dipole–sphere model for emission (Figure 4b,e), respectively,

**Figure 3.** Interference between multipoles and simultaneous modulation of excitation and emission for total light emission control. a) Scattering efficiency and its multipolar contributions of a 390 nm SiNS based on the Mie theory modified for dipole excitation. The green and red vertical dashed lines denote the excitation and emission wavelengths, respectively. b) The backward emission from monolayer WS$_2$ under forward and backward excitation condition, modulated by a 390 nm SiNS. c) The same as (a), but with a 250 nm SiNS. d) The forward-excited emission from monolayer WS$_2$ collected in the forward and backward directions, modulated by a 250 nm SiNS. All the curves in (b) and (d) are normalized to pure WS$_2$ emission without any modulation. MD: magnetic dipole, ED: electric dipole, MQ: magnetic quadrupole, EQ: electric quadrupole, MO: magnetic octupole, and EO: electric octupole.
the reciprocity theorem still holds for every decomposed phasor (see Section SXI in the Supporting Information). Thanks to the multipolar resonances, the incident field (INC) can be almost cancelled out along the backward direction, as shown in the rational designs of 446 nm excited WS$_2$ via a 320 nm SiNS (Figure 4a) and 532 nm excited MoS$_2$ via a 385 nm SiNS (Figure 4d). Meanwhile, the forward direction shows significant enhancement in total as illustrated in Figure S14a,c in the Supporting Information. This is for the excitation process. Based on the same SiNS, good forward directionality is also achieved for emission process as shown in Figure 4b,e and Figure S13b,d in the Supporting Information. Comparing Figure 4a,d and Figure 4b,e, we notice that fewer higher-order modes are needed for emission phasor diagrams but still with enough accuracies. This is because of the longer wavelengths of the emission than that of the excitation. For a certain SiNS size, fewer resonances can be effectively excited by photons with lower energies. As another proof, changes in the radiation patterns can hardly be observed when additional higher-order modes are included in analysis, as shown in Figure S4 in the Supporting Information.

In summary, we have experimentally demonstrated effective control of exciton emission via a low-loss subwavelength SiNS at visible wavelengths through controllable directional modulation of both excitation and emission processes. The isotropic spherical shape allows consistent directional modulation along all sample orientations. Based on a modified Mie theory for dipole excitation, the control over the F/B ratio is attributed to the superposition of multipolar resonances supported by single SiNSs. Phasor diagrams are extracted from the analytical model and provide great insights into the size and wavelength
dependent multipolar contributions. Measurements performed on 125 SiNS resonators convincingly suggest a good performance of our highly miniaturized platform and highlight the two-step directional modulation of incident and radiation fields. Finally, thanks to the universality of our model and the facile SiNS size control, we achieved versatile emission property designs on various emitters. Two degrees of freedom of SiNS size and excitation wavelength in the design provide us opportunities for multifunctional nanophotonics. Moreover, with the rigorous consideration of the emitter at various position/orientation in derivations,\(^\text{[36]}\) our multipolar theory also allows for the analysis of the dipole oriented normal to the sphere’s surface, e.g., spin-forbidden dark exciton emission in TMDs.\(^\text{[37]}\)

Our work boosts the development of ultracompact and multiplexed integrated photonic devices at visible wavelengths by a silicon-based subwavelength nanoantenna. As the diffraction limit in our measurements significantly degrades the detected signals in directionality, we would expect enhanced performance when our platform is incorporated in silicon nanophotonics, e.g., waveguides.\(^\text{[25]}\) On-demand modulator assembly for the wanted functions might also be realized by a size-selective optical printing\(^\text{[18]}\) based on our SiNSs. The proposed antenna–emitter hybrid may give insights into the use of high-index dielectric nanoparticles as functional components in photonic circuits.

**Experimental Section**

The Experimental Section is available in the Supporting Information.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that support the findings of this study are available on request from the corresponding author.

**Keywords**

directional excitation and emission, directional modulation, Mie resonances, nanoemitters, silicon nanospheres, transition metal dichalcogenides

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