Unveiling the Optical Emission Channels of Monolayer Semiconductors Coupled to Silicon Nanoantennas

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ABSTRACT: Monolayers (MLs) of transition metal dichalcogenides (TMDs) such as WSe$_2$ and MoSe$_2$ can be placed by dry stamping directly on broadband dielectric resonators, which have the ability to enhance the spontaneous emission rate and brightness of solid-state emitters at room temperature. We show strongly enhanced emission and directivity modifications in room-temperature photoluminescence mapping experiments. By varying TMD material (WSe$_2$ vs MoSe$_2$) transferred on silicon nanoresonators with various designs (planarized vs nonplanarized), we experimentally separate the different physical mechanisms that govern the global light emission enhancement. For WSe$_2$ and MoSe$_2$, we address the effects of Mie resonances and strain in the monolayer. For WSe$_2$, an important additional contribution comes from out-of-plane exciton dipoles. This paves the way for more targeted designs of TMD-Si nanoresonator structures for room-temperature applications.

KEYWORDS: dielectric nanoantennas, Mie-tronics, transition metal dichalcogenides, 2D materials, strain, excitons

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ntegrated efficient quantum light sources need to be developed for different applications, including quantum computing and transmitted or stored information for cryptography. Key technologies are plasmonic or dielectric nanostructures (NS) coupled to various quantum emitters position and orientation with respect to the nanoantenna are important to optimize the emission rate and the emitted light directivity and waveguiding. Therefore, monolayers of semiconducting transition metal dichalcogenides (TMDs), such as WSe$_2$ and also heterobilayers, are very promising as an active medium. Their atomic-monolayer thickness makes them uniformly sensitive to the near field around a nanoantenna, and they host several kinds of emitting dipoles at different wavelengths, such as the bright exciton (X$_b$, dipole in the monolayer plane), the so-called dark exciton (X$_d$, dipole out-of-plane), and other identified complexes, such as charged excitons, localized excitons, and biexcitons. TMDs coupled to plasmonic NS have shown strongly enhanced emission and tailored directivity. However, plasmonics is limited by high losses in metals, heating, and incompatibility with complementary-metal-oxide-semiconductor (CMOS) fabrication processes. High refractive index dielectric NS, such as silicon-NS or GaP-NS, can also provide reduced mode volumes, but in contrast to plasmonics, they show low nonradiative losses and are compatible with large-scale semiconductor fabrication processes. The emission properties of excitons in TMDs placed on NS have been investigated for several combinations of NS geometry and TMD materials. Despite these promising first results, the physical mechanisms that govern changes in the TMD emission as they are placed in proximity to the nanoresonators need to be understood. In addition to the targeted modification of the local electric field due to the resonator, several other mechanisms, such as strong local strain induced by the resonator and changing optical dipole orientation with respect to the detection, contribute to brightening and change in directivity of the emission.

In this work, we experimentally separate the three main mechanisms that govern the photoluminescence emission properties of the coupled dielectric NS-TMD monolayer system at room temperature, namely, (i) interactions with broad-band Mie resonances, (ii) strain in the TMD layer, and (iii) orientation of the exciton dipole with respect to the detection. First, we address the impact of the dipole
orientation by comparing on identical nonplanarized resonator samples (see dimer resonators sketched in Figure 1a) the emission of WSe$_2$ (in-plane and out-of-plane dipoles) with MoSe$_2$ (only in-plane emission). For WSe$_2$, we show a brightening of the global emission by 1 order of magnitude on the NS. This is accompanied by a change in overall spectral shape due to a variation of the relative contributions to the emission from the in-plane and out-of-plane dipoles, which emit at slightly different energies. In the PL mapping, we find a strong dependence of the spectral shape on the detection spot position on the NS (edge/center/suspended region). Second, we address the impact of strain as we compare TMD monolayer emission from samples on Si nanostructures (height about 100 nm) with samples on the same Si nanostructures surrounded by silica (planarized samples), where the TMD is strain-free, compare Figure 1c,e. We measure in nonplanarized samples a clear redshift of the PL energy (correlated to the TMD phonon shift measured in Raman mapping), showing the contribution to enhanced emission of the tensile strain-induced band gap lowering. Strained MoSe$_2$ behaves differently compared to WSe$_2$: although, we see a PL energy redshift due to strain, the spectra consist only of a main, single peak corresponding to the in-plane dipole in MoSe$_2$ (bright exciton). On planarized samples, we observe WSe$_2$ and MoSe$_2$ emission with less PL intensity enhancement (50%) as compared to the strained case, but with a clear impact of NS geometry on the PL emission pattern and dependence on the excitation laser polarization. Our experiments show an enhanced brightness for all NS-TMD configurations as compared to the TMD alone, so the Si-NS is beneficial in all cases.

Samples and Optical Spectroscopy Techniques. In this section we describe the two different kinds of nanoresonators used, which show broad resonances covering the wavelength region that corresponds to the MoSe$_2$ and WSe$_2$ monolayer emission and excitation.

Si-NS are obtained by a top-down approach in a Silicon on Insulator (SOI) wafer, where a hydrogen silsesquioxane (HSQ) resist is patterned by electron beam lithography followed by reactive ion etching. The thickness of the single crystal overlayer which determines the Si-NS height is $H = \text{90 nm}$. After etching and annealing, a SiO$_2$ layer (from the HSQ
Figure 2. WSe$_2$ monolayer on nonplanarized nanoresonators. (a) Normalized WSe$_2$ PL color maps of 12 dimer resonators (from $G = 300$ nm, $L = 300$ nm to $G = 50$ nm, $L = 200$ nm), for $x$-polarized incident light (along the antennas). (b) Normalized PL color map at the vicinity of resonator $G = 300$ nm, $L = 300$ nm for $x$-polarized incident light (corresponding to white circle in (a)). Dimer position and shape are marked by the black rectangles on the map. (c–e) Typical PL spectra taken along the white line on (b). Here we discern three contributions, namely, the bright exciton $X_0$ (blue curves and data points), the trion (green), and the dark exciton $X_D$ (red). (f) Top: Normalized total PL amplitude (black line) and individual Lorentzian fitting component amplitude (red, green, and blue) measured across the white line in (b); Bottom: energy of the individual Lorentzian peaks vs position. The dimer position is shown by the blue square, dashed gray lines mark the positions where the PL spectra shown in (c)–(e) have been taken. (g) is the same as (a) but for $y$-polarized incident light (perpendicular to the antennas). (h) Normalized PL color map at the vicinity of resonator $G = 300$ nm, $L = 300$ nm for $y$-polarized incident light. Dimer position marked by white rectangles. (i–k) Typical PL spectra taken along the white line in (h). Experimental data appear in a black continuous line. Blue, green, and red dashed lines are the constituting Lorentzian peak used to fit the PL; the corresponding global fit is shown as black dashed lines. (l) is the same as (f) but for $y$-polarized incident light.
deposition) of about 20 nm is left on top of the Si-NS. We will focus on dimers consisting of two nanorods of rectangular cross section having dimensions of $L \times W \times H$ and separated by a gap $G$ (Figure 1c). Values of $L$ and $G$ are varying across the resonator array from 50 to 300 nm, for constant $W = 150$ nm and $H = 90$ nm. An array of fabricated structures can be seen in Figure 1b. Before the TMD monolayer transfer, we perform dark-field scattering experiments to probe the optical Mie resonance bandwidth of the different dimers. A typical dark-field spectrum is shown in Figure 1h, where the measured resonances extend up to the relevant exciton transitions in the TMDs. As it can already be seen in Figure 1g by the different colors, the characteristics of the Mie resonances can be varied by changing $L$, $W$ and $G$.

Flakes from TMD bulk crystals are obtained by multiple folding in adhesive tape. The adhesive tape with the flakes of various thicknesses is pressed against a transparent polymer stamp, here PDMS, to transfer the flakes. Monolayers on the PDMS stamp are identified by optical contrast (confirmed by AFM). As a last step, the desired WSe$_2$ or MoSe$_2$ monolayer is placed above the targeted nanoresonator array and then transferred on the patterned substrate, covering 30 Si-NS with the same flake in Figure 1b. An atomic force microscope (AFM) profile in Figure 1d shows that the WSe$_2$ ML is acting as a nearly conformal coating, following the Si-NS morphology. These samples are referred to as nonplanarized and will be used to investigate the impact of strain and dipole orientation on the PL emission (Figure 1c).

Figure 3. MoSe$_2$ monolayer on nonplanarized nanoresonators. (a) Normalized MoSe$_2$ PL color maps of 12 dimer resonators (same nanoresonators as in Figure 2a), for x-polarized on nonplanarized nanoresonators. (b) Normalized PL color map at the vicinity of nanoresonator $G = 300$ nm, $L = 300$ nm for x-polarized (corresponding to white circle in (a)). Dimer positions and shape is marked by the black rectangles on the map. (c) Normalized PL integrated amplitude (black line) and energy of the PL maximum is (blue line) versus position, cross section taken along the white dashed line on (b). The dimer position is marked by the blue squares. (d) Typical PL spectra taken at three different positions marked by matching color dashed vertical lines in (b). The spectra are normalized, the normalization factor is indicated.

Figure 4. Strain-free WSe$_2$ monolayer on planarized nanoresonators. (a) Normalized PL images of WSe$_2$ monolayer deposited on $G = 300$ nm, $L = 300$ nm planarized nanoresonator for x-polarized laser excitation. Dimer position shown as black rectangles. (b) Typical PL spectra taken along the white line on panel (a). The PL spectra are fitted using the same three Lorentzian peaks, as detailed in Figure 2. (c) Top: Normalized total PL amplitude (black line) and individual Lorentzian fitting component amplitude (red, green, blue) measured across the white line on (a). The dimer position is marked by the blue squares. Bottom: Energy of the individual Lorentzian peaks versus position along the white line on panel (a). The positions where the PL spectra have been taken appear as dashed line, with matching color. (d), (e), and (f) are the same as (a), (b), and (c), but for y-polarized incident light.
In order to eliminate strain effects and focus solely on the impact of the Si antenna on the TMD emission, another set of samples was fabricated. We followed the same fabrication procedure as for the nonplanarized Si-NS and added a planarization step involving HSQ deposition and subsequent annealing and chemical etching. This leads to Si nano-antennas buried in silica and we refer to this as planarized samples, see Figure 1e. The distance between the top Si-NS and the silica smooth surface (see AFM profile Figure 1f) is about 30 nm over 1 μm. Although the surface is not totally flat, the height variations are smooth enough to avoid strain in the TMD, as confirmed in PL and Raman mappings (see below). The active region is here protected by a 10 nm thick hexagonal BN layer (high crystal quality, atomically flat) on top of the TMD ML.

Room-temperature photoluminescence and Raman experiments are performed using either a Horiba JY Labram HR or a Horiba JY XPlora MV2000 setup. Hyper-spectral maps are obtained by raster scanning the sample under a laser spot (532 nm excitation wavelength, power of tens of nW) tightly focused through a microscope objective (0.9 numerical aperture, NA). The spectra are obtained in backscattering using the same objective. The high NA allows us to collect mainly emission from in-plane but also to a small extent from out-of-plane optical dipoles. Although the diffraction limited spot has a diameter comparable to the dimer, the recorded PL emission map of the sample reveals fine details in lateral resolution due to the high signal-to-noise ratio. The laser polarization is kept constant and the so-called x (linear polarization along the dimer axis) and y polarization (linear polarization perpendicular the dimer axis) are obtained by rotating the sample by 90°. Here we define as the dimer axis a straight line across the dimer gap that connects the centers of the two Si rectangles.

**WSe₂ Monolayer on Nonplanarized N ano-resonators.**

In Figure 2a we plot a normalized, spectrally integrated PL map of nonplanarized nanoresonators covered by one large WSe₂ monolayer. The nanoresonator positions can be seen directly on the map, PL away from the resonators is very weak (uniform dark blue area). To allow for a direct comparison of the results in Figures 2, 3, and 4, we show spectra from the Si dimer nanoresonator with the same dimensions in all cases of gap G = 300 nm and length L = 300 nm (white circle in Figures 2a,g and 3a). This dimer has broad optical Mie resonances at the emission as well as the laser excitation energy (see Figure 1h). Strain will clearly play an important role for nonplanarized nanoresonators and similar structures are used to generate localized excitons (0D) at low temperature for quantum optics. Note that we avoid exciton localization in our room temperature measurements as the thermal energy of excitons is larger than the localization energy.

In our experiment in Figure 2a, the excitation laser is polarized along the main axis of the dimer (x), Figure 2g shows identical measurements, but carried out with laser polarization perpendicular to the dimer (y) (white arrows indicating the electric field in the figures). We first comment on x-polarized excitation. The WSe₂ PL intensity is substantially enhanced by 1 order of magnitude on top of the nanoresonators. In addition to intensity changes, we clearly observe that the spectra recorded on top and next to the nanoresonator shown in Figure 2b are very different, compare panels Figure 2c–e. The PL emission is fitted using three peaks, in agreement with the different transitions identified in the literature for WSe₂ ML even at room temperature: the free exciton at about 1.66 eV (745 nm; dashed blue line), the charged excitons (trions; dashed green line) at about 1.64 eV (756 nm), and the dark (out-of-plane) exciton near 1.61 eV (770 nm; dashed red line). Please note that we have identified the respective dipole orientation of the dark and bright excitons previously by imaging the Fourier plane of the microscope objective in experiments on ML WSe₂. The relative weight of the different peaks strongly varies as a function of the position along the map, especially around and above the Si-NS (see Figure 2f, top panel).

The impact of strain on the optical transitions is revealed in Figure 2f, bottom panel, where we plot the emission energy of the three PL components as a function of position. A shift of the PL emission to lower energy for all three transitions is a clear confirmation for tensile strain in the layer. The same conclusion can be independently drawn from the Raman maps, presented in the supplement. The enhancement observed for emission on the nanoresonators benefits therefore from the combined effect of strain (local lowering of the band gap that results in funneling of excitons) and optical resonances, which we aim to separate in the next section. Interestingly, as one can see in Figure 2f, top panel, the emission linked to the dark exciton (red line), which has an optical dipole out-of-the monolayer plane, shows strongest enhancement on the sample edges. Here we suggest that, as the monolayer is folded with respect to the microscope objective axis (by 30° to 40°, as given by AFM profile), more light emitted by the out-of-plane dipole is detected. These arguments are developed in more detail in the discussion section. In addition, enhanced emission from excitons with out-of-plane dipoles might be due to lowering of the crystal symmetry. The impact of strain for WSe₂ MLs on GaP dimers is discussed in refs 13 and 22.

In Figure 2g we plot the same intensity map as in Figure 2a, but the excitation laser polarization is now perpendicular to the dimer axis. We observe clear differences: the global enhancement on the NS is now a factor of 3, as compared to roughly 8 for the other laser polarization, and the spatial region around the NS position that shows enhancement is smaller. The maximum enhancement is localized in the gap between the two rectangular rods constituting the dimer, as can be seen in Figure 2h. In the dimer gap the monolayer is suspended and, therefore, presumably less strained than on the dimer edges. As a result, the PL emission energy does not change as a function of position as we scan across the gap, see Figure 2l, bottom panel.

Our measurements also show, in contrast to laser excitation parallel to the dimer axis, the global shape of the PL spectra is not affected in the perpendicular case (Figure 2i,j,k). The relative amplitude of the different PL components remains constant when scanning over an antenna (Figure 2l top panel). We suggest in the discussion section below, that this striking dependence on laser excitation polarization (compare Figure 2a,b with g,h) is linked to an optical antenna effect.

**MoSe₂ Monolayer on Nonplanarized N ano-resonators.**

For WSe₂, we observe on several Si-NS dimers a strong PL intensity enhancement above each element of the dimer, showing amplification as high as 8 as compared to the bare monolayer. We now investigate emission from a MoSe₂ monolayer deposited on an identical set of resonators, see Figure 3a. We choose this material for comparison as the emission wavelength is close to WSe₂ and can be covered by the broadband resonances of the Si-NS, as shown in Figure 1.
An additional advantage for this material is the comparatively simple emission spectrum as the dark, \( z \)-polarized states (optical dipole out-of-plane) are in energy above the bright states and therefore have only very small impact on PL emission here, see spectra in Figure 3d and the scheme in Figure 5a.

In Figure 3a we plot the PL emission map above the resonators and we see strong PL emission enhancement (up to \( 14 \times \)) on several resonators covered by the ML flake. PL emission next to the resonators is weak (uniform dark area in the map). The most striking observations are First, a strong enhancement of the PL emission intensity above the Si-NS, see Figure 3b. Second, in contrast to our WSe\textsubscript{2} results, the PL emission for MoSe\textsubscript{2} does not change shape, as can be seen in Figure 3d. The PL band could be fitted using a single peak attributed to the neutral bright exciton. Thus, the red shift of the PL energy shown in Figure 3c is deduced from the maximum of the measured PL peak. As in the case of WSe\textsubscript{2}, this is a signature of tensile strain but resulting in a smaller energy shift, in qualitative agreement with previous studies on strain tuning.\textsuperscript{33} Although due to the manual stamping method, the coupling efficiency of the TMD monolayer might not be uniform across the nanoresonator array, we observe for the investigated MoSe\textsubscript{2} monolayer very similar enhancement for several nanoresonators analyzed, as can be seen in the map in Figure 3a. Interestingly, for dimers with narrower gaps, two emission lobes appear, which might be due to an antenna effect that redirects the light at the dimer edge toward our microscope objective. The simple PL shape makes MoSe\textsubscript{2} a suitable material for analyzing more complex effects related to Mie resonances, that might be obscured by the multiple transitions in WSe\textsubscript{2}. Please note that MoSe\textsubscript{2} monolayers have, in general, comparatively weaker emission than WSe\textsubscript{2} monolayers at room temperature\textsuperscript{33–37} and, hence, a wider margin for increasing emission efficiency.

Concerning the dependence on laser excitation polarization for MoSe\textsubscript{2}, we observe a stronger emission for excitation along the dimer axis than perpendicular to it. The difference is about

![Figure 5.](https://dx.doi.org/10.1021/acsphotonics.0c01175)

Figure 5. Numerical simulations of electric field amplitudes and optical dipole emission. (a) Top: schematics of the energy levels in MoSe\textsubscript{2} (left) and WSe\textsubscript{2} (right). Bottom: orientation of the in-plane neutral exciton \( X_{\text{o}} \) (red arrow) and the out-of-plane dark exciton \( X_{\text{D}} \) (blue arrow) in the \( x-z \) plane with respect to the TMD layer. The gray shaded areas represent the emission lobes of the dipoles. (b) Near-field intensity in the TMD layer of the nonplanarized system for two polarizations parallel and perpendicular to the dimer axis computed in the \( x-y \) plane for an incident plane wave at the excitation wavelength \( \lambda = 532 \text{ nm} \). (b) Geometry of the TMD on a nonplanarized dimer in the \( x-z \) plane, direction of \( X_{\text{o}} \) (red) and \( X_{\text{D}} \) (dark blue) are indicated by arrows. (d) Emission patterns of a dipole emitting at 750 nm and placed at different positions and heights (\( z \)) with respect to the nonplanarized dimer shown in (b). The relative positions of the Si-NS and the dipoles are displayed in the \( x-y \) plane. The dotted circle corresponds to the 0.9 NA of the microscope objective; we, hence, detect emission from inside this circle. The dipole can be assimilated to an in-plane (red arrow) or out-of-plane exciton (dark blue arrow), depending on its orientation and position. The surrounding refractive index is \( n = 1 \). (e) Geometry of the TMD on a planarized dimer in the \( x-z \) plane. (f) Emission patterns of a dipole emitting at 750 nm and placed at different positions and \( z = 100 \text{ nm} \) with respect to the planarized dimer shown in (e). The surrounding refractive index is \( n = 1.45 \) (HSQ refractive index).
15%, see supplement, much less pronounced than for the WSe$_2$ case, which suggests that the contributions from out-of-plane dipole emission plays an important role for the strong polarization difference observed in WSe$_2$. Note that we analyze for both materials the same nanoresonators with the same optical resonances (as shown in Figure 1h), so major differences in our comparative studies between MoSe$_2$ and WSe$_2$ emission have their origin in the different band structure, sketched in Figure 5a.

**Strain-Free WSe$_2$ Monolayer on Planarized Nanoresonators.** In the case of the planarized samples, neither strain-induced Raman shifts of the TMD phonon can be detected (see supplement) nor a systematic redshift of the PL energy, as shown in our measurements in Figure 4c.f. The AFM profile in Figure 1f shows that the maximum roughness above a Si-NS is limited to about 30 nm extended over 1 μm. For this strain-free WSe$_2$ monolayer on planarized resonators we make the following observations: We record an increase of the PL of 50% due to the presence of Si-NS, this is less than the factor of about 8 achieved on the same nanoresonator but nonplanarized, compare Figure 2a and Figure 4a. Interestingly, the overall emission spectrum for the planarized sample of WSe$_2$ does not change shape, but only the amplitude is modified by the antenna, see Figure 4b,e. The bright exciton contribution in the PL band is the strongest peak according to our fits and the weight of the different contributions (dark and charged excitons) are constant when we scan across the Si-NS. The PL maps in Figure 4 are dependent on the laser polarization for WSe$_2$, compare panels Figure 4a,c with Figure 4d,f. For γ-polarized excitation, we see a maximum in PL signal over the dimer gap, see Figure 4d,f. Our measurements show that the emission of WSe$_2$ is governed by an optical antenna effect in the planarized samples, whereas strain effects play a key role in the nonplanar samples, as pointed out in earlier work on strain WSe$_2$ on GaP dimers. The measured PL enhancement is up to 50% of the integrated intensity, which can be further improved. Our measurements on planarized MoSe$_2$ samples gave very similar results.

In future experiments, the Si-NS shape and dimensions can be optimized in order to reach higher electric field amplitudes. It is important to take into account that, after HSQ planarization and hBN deposition, the dielectric contrast between Si-NS and the surrounding is lower than for nonplanarized resonators. This lowers light confinement and needs to be compensated by optimizing the antenna design.

**Numerical Simulations and Discussion.** Our measurements of the PL emission of ML WSe$_2$ and MoSe$_2$ coupled to nonplanarized and planarized nanoresonators revealed for all cases an enhancement of the PL emission as compared to the bare monolayer. In the discussion below we suggest different mechanisms at the origin of this enhancement, taking into account the polarization-dependent near field at the excitation energy and the interaction of dipole in-plane and out-of-the-ML-plane with the Mie resonances of the dimer structures at the emission energy.

The Si-NS is optically resonant at the excitation wavelength. We first simulate the spatial distribution of the electric near-field intensity in the plane of the TMD monolayer (see Figure 5c) with the finite-difference time-domain (FDTD) method using an open-source software package, see results in Figure 5b. For laser excitation parallel to the dimer axis, our simulation shows a stronger near-field enhancement than in the case where the polarization is perpendicular to the dimer axis. The near-field enhancement is localized above and around the Si blocks in the parallel polarization case. This can be seen, in particular, along the edges (γ-oriented) perpendicular to the polarization. In contrast, there is almost no enhancement for the perpendicular case (even lower field above each block of the Si dimer). This behavior, relevant at the laser excitation wavelength, is in qualitative agreement with the PL maps of the chosen example described previously, compare Figure 2a and g. This indicates that optical Mie resonances contribute to PL enhancement. Nevertheless Mie resonances alone cannot explain the differences in the enhancement and the polarization dependence observed in the four configurations (ML WSe$_2$ and ML MoSe$_2$ on nonplanarized and planarized Si-NS, respectively); otherwise, the observations for MoSe$_2$ and WSe$_2$ for the polarization dependence would have been identical.

To go further, we study the antenna effect induced by Si-NS on the emission of the two kinds of exciton dipoles (the bright exciton X$_b$ and the dark exciton X$_D$). In a first approximation, we consider a single oscillating dipole with orthogonal orientation depending on the nature of the exciton (in-plane X$_b$ and out-of-plane X$_D$, see Figure 5a). Considering the nonplanarized and planarized geometries (Figure 5c,e), we compute the intensity of the electric field collected in backscattering geometry through the microscope objective NA for several key locations of the emitting dipoles. The electric field radiated by the dipolar source in the presence of a Si-NS is obtained by the Green Dyadic Method using the pyGDM toolkit for electrodynamic simulations. In Figure 5d–f is shown the emission patterns of different dipoles emitting at 750 nm (approximately the average of the bright and dark exciton wavelengths). Depending on the position around the Si-NS, the dipole represents either a dark exciton (out-of-plane dipole, black arrow) or a neutral exciton (in-plane, red arrow).

In the upper left part of Figure 5d, we represent a dipole marked by a black arrow that is located at one Si-NS edge, at mid-height (z = 55 nm) and oriented along the x-axis of the dimer. Now we need to take into account that our monolayer changes orientation: as represented in Figure 5c, the TMD ML is nearly conformal to the Si-NS, that is, following its shape. That means, at the outer dimer edges, a dipole oriented along the x-axis of the dimer is oriented out of the folded monolayer plane. Hence, it can be considered as a dark exciton X$_D$ with a dipole out of the TMD ML plane. For this dipole, our calculations show that the intensity measured through the microscope objective NA (represented by the dotted circle) is very high. In contrast, the collected intensity originating from the dipole in the TMD plane at the same position (represented by a red arrow in Figure 5c) is negligible. Indeed, the emission pattern (not shown) is oriented along the x-axis due to the combined effects of the dipole orientation parallel to the microscope axis and the Si nanoantenna. These simulations give an indication why the emission signal detected at the X$_D$ energy is particularly strong at the dimer edges.

In contrast, if the dipole is positioned at midheight of the long side of the nanoblock (upper right part of Figure 5d), the collected intensity drops. We believe that this antenna effect plays an important role in the increase in the measured signal. It adds to the polarization-dependent enhancement presented in Figure 5b, which leads to the creation of more excitons (including X$_D$ ones) in the regions of high electric field. Thus, the combination of these two fundamental mechanisms leads to the boost in the PL enhancement observed for WSe$_2$ on
nonplanar Si-NS, only for a parallel polarization of the laser with respect to the Si dimer axis.

In the gap of the Si dimer (lower part of Figure 3d), the integrated intensity corresponding to Xo emission is approximately the same as the one due to Xe. Both are contributing, thus, leading again to the signal increase. Compared to the emission pattern of the dipole in a TMD monolayer alone hosting Xo (WSe2; see Figure S1a), it is clear that the relative contributions of Xo and Xe are very different as a function of the emitting dipole position in the case of the nonplanar sample.

Our simple model is already sufficient to understand the underlying physics leading to the strong PL enhancement when the TMD hosts out-of-plane excitons. For instance, since this cumulative effect mainly occurs along the y-oriented edges (small side of a nanoblock) for a parallel polarization, it helps explain the extension of the PL patterns along the x-axis well beyond the dimer limits in the maps of Figure 2. For the same polarization configuration, it is also in good agreement with the PL band shape modification related to the strong enhancement of the fitted (red) peak associated with Xo at the y-oriented (outer) edges of the Si-NS in the spectra of Figure 2.

On the other hand, for the planarized sample, the dark exciton contribution is always very minor, and the PL band is dominated by the bright exciton contribution, compare Figure 4e and Figure 2e. The radiation pattern of Xo is weakly redirected by the antenna in the angular window defined by the microscope objective NA, neither when it is positioned along the edges nor in the gap region (Figure 5f). As a consequence, no enhancement associated with the Xo is expected. It is in good agreement with the experimental observation where this dark exciton is intrinsically eliminated (MoSe2 samples) or does experience no enhancement (WSe2 on planarized samples). In both cases, the polarization-dependent enhancement, resulting from a pure optical effect associated with the Mie resonances in these Si-NS, is about 50%, far from the roughly 10X enhancement in PL from WSe2 on nonplanar resonators.

We unveil here three intertwined mechanisms that contribute to the PL enhancement. The Mie-assisted enhancement of the dark exciton contribution plays, therefore, an important role in the global PL response of the WSe2—resonator coupled systems. Interestingly, its contribution seems to be of the same order of magnitude as the strain one. The amplitude of the latter can be estimated with MoSe2 on nonplanar samples. Indeed, about 15% variation of the PL is observed as the polarization is rotated, indicating a very strong contribution to the global factor PL enhancement (14X) originates from the strain generated by the Si nanoblocks in this sample.

Finally, as expected for nonoptimized resonators, the pure optical effect associated with Mie resonances observed in planarized samples in Figure 4 (no strain, no dark exciton) is relatively weak compared to the other two mechanisms. It should easily be further improved by optimizing the geometry of the nanoantennas.

**CONCLUSION**

we have shown strong PL enhancement for WSe2 and MoSe2 on nonplanarized nanoantennas due to the combined effect of amplifying contributions from out-of-plane dipole emission, strain and pure electric field amplification. On planarized nanoantennas we show that the TMD monolayer is strain free and the PL emission is only enhanced by the Mie resonances of the Si dimer. In addition to the presence and absence of strain in the active medium for nonplanarized and planarized samples, respectively, also the dielectric contrast plays a role. For nonplanarized resonators, the dielectric contrast is high, whereas it is lower for planarized samples, which demands further work on planarization techniques such as the use of low refractive index dielectric around the silicon nanostructures and a very small thickness to position the TMD as close as possible to the Si nanoantenna. Finally, the optimization of the nanoantenna design will be essential to control strain, resonances at both the excitation and emission wavelength, and directivity of the emitted light. WSe2 is a very promising material as the in-plane and out-of-plane exciton emission can be controlled independently to increase the global emission yield. MoSe2 shows also strong PL emission enhancement and a simple emission spectrum for targeted interaction with Mie resonances.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsphotonics.0c01175.

Raman mappings of WSe2, monolayers transferred on nonplanarized (under strain) and planarized (strain-free) nanoantennas are shown in Figures S1 and S3, respectively. Figure S2 presents the photoluminescence mapping of a MoSe2 monolayer on nonplanarized nanoantenna as a function of the excitation laser polarization (PDF)

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REFERENCES
(15) Kuznetsov, A. I.; Miroshnichenko, A. E.; Brongersma, M. L.; Kivshar, Y. S.; Luk’yanchuk, B. Optically resonant dielectric nanostructures. Science 2016, 354, 844–847.
(18) Lee, Y.-C.; Tseng, Y.-C.; Chen, H.-L. Single type of nanocavity structure enhances light outcouplings from various two-dimensional materials by over 100-fold. ACS Photonics 2017, 4, 93–105.

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