Coupling Interlayer Excitons to Whispering Gallery Modes in van der Waals Heterostructures

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ABSTRACT: Van der Waals heterostructures assembled from two-dimensional materials offer a promising platform to engineer structures with desired optoelectronic characteristics. Here we use waveguide-coupled disk resonators made of hexagonal boron nitride (h-BN) to demonstrate cavity-coupled emission from interlayer excitons of a heterobilayer of two monolayer transition metal dichalcogenides. We sandwich a MoSe$_2$−WSe$_2$ heterobilayer between two slabs of h-BN and directly pattern the resulting stack into waveguide-coupled disk resonators. This enables us to position the active materials into regions of highest optical field intensity, thereby maximizing the mode overlap and the coupling strength. Since the interlayer exciton emission energy is lower than the optical band gaps of the individual monolayers and since the interlayer transition itself has a weak oscillator strength, the circulating light is only weakly reabsorbed, which results in an unaffected quality factor. Our devices are fully waveguide-coupled and represent a promising platform for on-chip van der Waals photonics.

KEYWORDS: interlayer excitons, transition metal dichalcogenides, h-BN photonics, whispering gallery mode resonator

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an der Waals (vdW) heterostructures made of a vertical assembly of two-dimensional (2D) materials like transition metal dichalcogenides (TMDCs) and graphene have opened up new possibilities for optoelectronic devices. Since adjacent 2D materials are held together by weak van der Waals forces, they can be stacked without need of lattice matching and can be placed on almost any substrate. In their monolayer form, TMDCs exhibit a direct bandgap, which makes them excellent candidates for optoelectronic devices as well as for integration with optical cavities for enhanced light−matter interactions.

By combining two dissimilar TMDC monolayers into a heterobilayer (HBL), interlayer excitons with spatially separated electrons and holes can be formed. Their charge carrier dynamics, electrically tunable exciton resonances, and the possibility of creating p−n junctions are providing intriguing possibilities for materials engineering. Recently, interlayer excitons were coupled to photonic crystal cavities, as well as grating cavities, that led to the demonstration of surface emitting, optically pumped lasers at room temperature.

However, the cointegration of light emitting devices (LEDs) and lasers made of vdW heterostructures with on-chip photonic structures, such as waveguides and resonators, requires in-plane light guiding. Coupling via evanescent fields has been achieved, for example, by placing vdW materials on top of silicon-based waveguides and ring resonators, but the coupling strength obtainable in this manner is modest because the highest field intensity is found inside the light confining dielectric medium. Optimal mode overlap can be achieved by integrating the active layer into dielectric waveguides or resonators. For optically excited systems, this has been shown by sandwiching a monolayer WS$_2$ into a microdisk made of Si$_3$N$_4$/hydrogen silsesquioxane (HSQ) or hexagonal boron nitride (h-BN). However, in both of these structures, the disk was freestanding and is not easily compatible with on-chip photonics.

Here we present vdW heterostructures patterned into waveguide-coupled disk resonators, as illustrated in Figure 1a. The stacking allows us to place TMDCs into regions of high field intensity. Our approach combines two important attributes: (1) an enhanced optical mode overlap and (2) waveguide coupling for on-chip integration. These are enabling factors for the development of on-chip optoelectronic devices, such as LEDs, modulators, and photodetectors. For
the dielectric material, we chose h-BN, which has already been used to fabricate photonic structures like resonators or photonic crystals21−23 and has been proposed as a confinement layer in integrated photonics.24 In addition to its favorable photonic attributes, h-BN also provides a means of encapsulation; that is, by placing TMDCs in between h-BN, we prevent them from degradation.25

As shown in Figure 1a, we integrate a HBL into an h-BN disk resonator, which can be excited by laser light propagating along a nearby waveguide. Photoluminescence (PL) associated with the interlayer exciton is coupled out by the adjacent waveguide. The unique feature of our approach is that both the active material (TMDCs) and the dielectric (h-BN) are assembled into a single vDW heterostructure and subsequently patterned into the final photonic circuit (disk resonator and waveguide). This differs from previous works,1,2,20 where TMDCs were transferred on top of prefabricated photonic elements. As illustrated in Figure 1b, the interlayer exciton emission energy is lower than the optical band gaps of the individual monolayers, which leads to weak reabsorption of the interlayer emission.16,26 As a result, the quality (Q)-factor of the coupled mode is largely unaffected by reabsorption.27

Figure 1. (a) Illustration of a waveguide-coupled disk resonator with an integrated heterobilayer (HBL). The HBL is sandwiched between two h-BN flakes. The emission from the interlayer exciton is coupled to whispering gallery modes (WGMs) of the disk resonator and then outcoupled through the same waveguide used for excitation. (b) Free-space photoluminescence (PL) spectrum of the HBL, showing strong emission from the interlayer exciton (ILX) and quenched intralayer exciton emission. (c) Cavity-coupled photoluminescence spectrum featuring distinct peaks separated by the free-spectral range of the WGMs of the disk resonator. The resonator is excited at _λ_ = 633 nm from one end of the waveguide, and the photoluminescence is detected at the other end. The intralayer exciton emission is strongly suppressed because of reabsorption. The scale bar is 5 μm.

Flakes of h-BN and monolayers of WSe₂ and MoSe₂ were prepared by mechanical exfoliation and assembled into vDW heterostructures using a polymer-based stacking technique.27,28 The crystallographic axes of the two monolayers were rotationally aligned. Atomic force microscopy was utilized to identify h-BN flakes of the desired thicknesses. Parts b and c (magnified) of Figure 2 show optical microscope images of such an h-BN–MoSe₂–WSe₂–h-BN heterostructure on a glass substrate. As illustrated in Figure 2a, waveguides and disk resonators were fabricated from the vDW stack using electron-beam lithography and reactive ion etching with aluminum as a hard mask. Figure 2d shows the final device. The disk resonator has a total thickness of ~310 nm and a design radius of 4.25 μm.

Figure 2. (a) Schematic of the fabrication procedure, showing cross sections of the van der Waals (vDW) heterostructure after stacking and after subsequent patterning into a waveguide-coupled disk resonator (not to scale). (b, c) Optical microscope images of the h-BN–MoSe₂–WSe₂–h-BN heterostructure after stacking. The heterostructure has a total thickness of ~310 nm (bottom h-BN, ~150 nm; top h-BN, ~158 nm). (d) Electron-beam lithography and reactive ion etching were utilized to pattern the vDW heterostructure into a waveguide-coupled disk resonator with a design radius of 4.25 μm.
procedure can be found in the methods section in the Supporting Information.

In order to determine the emission energies, we performed PL measurements of the TMDC monolayers and of the HBL region using a 633 nm continuous wave (cw) He–Ne laser. The PL spectra of the individual monolayers reveal direct intralayer exciton emission peaks at 755 nm (1.64 eV) and 793 nm (1.56 eV) for WSe₂ and MoSe₂, respectively, and are in agreement with previous works\textsuperscript{14,17,29,30} (details in the Supporting Information). In Figure 1b, we show the PL spectrum of the HBL, with a pronounced peak at around 923 nm (1.34 eV). This feature is identified as the interlayer exciton emission and can be explained by the type-II band alignment of MoSe₂ and WSe₂ in the overlap region.\textsuperscript{13,19} In this process, the absorption of a photon in a monolayer is followed by a charge transfer to the energetically favorable band of the neighboring monolayer, leading to a spatial separation of electron and hole. As a result, a strong decrease of the intralayer exciton emissions can be observed. Furthermore, for two rotationally aligned monolayers, the interlayer exciton exhibits a direct band gap character.\textsuperscript{31} The strong interlayer peak in combination with the quenching of intralayer exciton emission in Figure 1b consequently veriﬁes the good electrical coupling and the near perfect rotational alignment of the two layers.

To investigate the properties of h-BN waveguide-coupled disk resonators, we fabricated reference structures without integrated TMDCs. Figure 3a shows a typical transmission spectrum of a fabricated device. It was obtained by focusing a broadband supercontinuum laser on one waveguide facet and detecting the transmitted light at the other end of the waveguide. The recorded spectrum features sharp dips at wavelengths that satisfy the resonance conditions of the disk resonator. The envelope of the spectrum is deﬁned by the excitation spectrum and the transmission characteristics of the experiment. The Q-factors of the resonances reach values of ~1000. The spectral positions of the resonance peaks, the Q-factors, and the free-spectral range depend sensitively on the h-BN thickness and the disk radius (details in the Supporting Information). Using ﬁnite element simulations as a guide, we deﬁne the disk radius for a given stack thickness in order to optimize the resonances for the interlayer emission spectrum (details on simulations in the Supporting Information). As shown in Figure 3c, the computed electric field distribution is highest near the rim of the disks and near the vertical center. Therefore, to optimize the coupling strength to the interlayer exciton, the HBL should be sandwiched into the h-BN resonator.

Next, we characterize an h-BN disk resonator with an integrated TMDC heterobilayer. We vary the location of the excitation in order to investigate the coupling of the interlayer emission to the WGMs of the disk. As shown in the sketch and the optical microscope image in Figure 4a and b, coupling to WGMs and interlayer excitons can be achieved by placing the excitation spot near the edge of the disk. We use the same objective for excitation and detection. The emission is detected either at the same position as the excitation spot or at the ends of the waveguide. The light coupled out at the end of the waveguide is partially refracted into the glass substrate and can be captured at high angles with the oil-immersion objective (NA = 1.4) that we use. This allows us to compare the spectra of free-space PL (detected at the excitation spot on the disk) and cavity-coupled PL (detected at the waveguide end). As shown in Figure 4c, the spectrum of the free-space PL is in agreement with the expected interlayer emission of the HBL (cf. Figure 1b). On the other hand, the cavity-coupled spectrum, detected at the waveguide end, exhibits distinct peaks that are separated by the free-spectral range of the WGMs of the disk resonator. We emphasize that, in contrast to several other recent studies,\textsuperscript{8,9,21} the waveguide-coupled spectrum features a reduced background because it is not superimposed to free-space PL.

As shown in the inset of Figure 5a, the two TMDCs forming the HBL do not overlap over the entire footprint of the disk. There are regions where only the MoSe₂ monolayer can be directly excited. Figure 5a shows the corresponding spectrum of the free-space and cavity-coupled PL. The main contribution of the cavity-coupled PL comes from the intralayer exciton emission of the MoSe₂ monolayer. Interestingly, however, the envelope function of the resonances does not coincide with the free-space PL measured at the excitation spot. Instead, the highest peak intensity of the cavity-coupled PL is positioned at wavelengths longer than the free-space PL emission peak. We attribute this observation mainly to reabsorption of the emitted PL in the cavity by the MoSe₂ monolayer itself, although other contributions have also been reported to be present\textsuperscript{32} (detailed discussion in the Supporting Information). This interpretation is supported by the absorption spectrum of MoSe₂ visualized by the extinction coefficient, i.e., the imaginary part of the refractive index (taken from ref 33), which we include in Figure 5a (dashed curve). Thus, the spectral overlap of the PL with the extinction coefficient causes significant reabsorption for wavelengths in this spectral range. As a result, the corresponding peak intensities of the cavity-coupled PL are reduced for these wavelengths. The peaks at longer wavelengths have an additional contribution from interlayer emission from the...
WSe$_2$ has been experimentally determined to be 2 orders of magnitude lower than that of the intralayer exciton. In MoSe$_2$, this provides further support for the reabsorption of monolayer emission discussed earlier. Because of the low oscillator strength of the interlayer transition, the reabsorption of interlayer emission is significantly weaker, which is evidenced by the high Q-factor at wavelengths near the interlayer transition. The decrease in Q-factor at longer wavelengths (>950 nm) can be explained by radiation losses, which increase at higher wavelengths (see the Supporting Information). Our Q-factors are currently limited by fabrication imperfections and the h-BN sidewall roughness arising from etching (see the scanning electron microscope image in the Supporting Information). Nevertheless, even with integrated TMDCs and a coupled waveguide, we achieve Q-factors that are slightly higher than those reported for h-BN ring resonators. Our Q-factors could be increased even further by removing the substrate underneath the outer regions of the disk where the WGMs are localized.

In summary, we have demonstrated the coupling of interlayer excitons from a TMDC HBL to WGMs of an h-BN disk resonator. We achieve high coupling strengths by sandwiching the HBL into the h-BN disk. The high oscillator strengths of interlayer transitions of the two monolayers lead to a high absorption efficiency. On the other hand, the low oscillator strength of the interlayer transition, the reabsorption of interlayer emission is significantly weaker, which is evidenced by the high Q-factor at wavelengths near the interlayer transition. The decrease in Q-factor at longer wavelengths (>950 nm) can be explained by radiation losses, which increase at higher wavelengths (see the Supporting Information). Our Q-factors are currently limited by fabrication imperfections and the h-BN sidewall roughness arising from etching (see the scanning electron microscope image in the Supporting Information). Nevertheless, even with integrated TMDCs and a coupled waveguide, we achieve Q-factors that are slightly higher than those reported for h-BN ring resonators. Our Q-factors could be increased even further by removing the substrate underneath the outer regions of the disk where the WGMs are localized.

As shown in Figure 5b, the Q-factors of the emission peaks are wavelength dependent, which is a result of the optical properties of the incorporated TMDC HBL. The highest values are ~570 and occur at wavelengths that correspond to the interlayer emission peak. In contrast, a considerably lower Q-factor is observed for wavelengths near the optical band gap of MoSe$_2$. This provides further support for the reabsorption of monolayer emission discussed earlier. Because of the low oscillator strength of the interlayer transition, the reabsorption of interlayer emission is significantly weaker, which is evidenced by the high Q-factor at wavelengths near the interlayer transition. The decrease in Q-factor at longer wavelengths (>950 nm) can be explained by radiation losses, which increase at higher wavelengths (see the Supporting Information). Our Q-factors are currently limited by fabrication imperfections and the h-BN sidewall roughness arising from etching (see the scanning electron microscope image in the Supporting Information). Nevertheless, even with integrated TMDCs and a coupled waveguide, we achieve Q-factors that are slightly higher than those reported for h-BN ring resonators. Our Q-factors could be increased even further by removing the substrate underneath the outer regions of the disk where the WGMs are localized.

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the Q-factor can be observed at the absorption wavelengths of the interlayer exciton transition of MoSe₂. No drop in Q-factor is observed for wavelengths of the interlayer exciton transition, indicating low reabsorption.

Figure 5. (a) Measured spectra at the waveguide end (cavity-coupled PL) and at the excitation spot (free-space PL), when exciting at the MoSe₂ monolayer region, as indicated in the inset (scale bar is 5 μm). The envelope function of the cavity-coupled PL does not coincide with the free-space PL, indicating strong reabsorption of interlayer emission by the MoSe₂ monolayer itself. The dashed curve indicates the MoSe₂ extinction spectrum (taken from ref 33). (b) Wavelength dependence of the Q-factor, determined by fitting Lorentzian functions to the measured resonances. Measurements were taken for different locations of excitation (HBL, monolayer, waveguide end) and detection at both waveguide ends. The means and the standard deviations are shown in the figure. A strong drop in Q-factor can be observed at the absorption wavelengths of the interlayer exciton transition of MoSe₂. No drop in Q-factor is observed for wavelengths of the interlayer exciton transition, indicating low reabsorption.

oscillator strength of the interlayer transition suppresses the reabsorption at the emission wavelength. The combination of these two effects gives rise to strong interlayer emission from our vdW disk resonators.

The presented platform would give rise to further quantitative analysis of the cavity quantum electrodynamics and experimental studies on interlayer excitons, their dynamics and emission properties, when placed at high field intensities. Considering the fact that patterning photonic structures out of h-BN and vdW heterostructures is still in its infancy, further improvements in performance with optimized fabrication techniques (e.g., optimization of the etching process) is expected. To proceed one step further toward on-chip optoelectronics, light sources need to be cointegrated with photonic structures. The recent progress in electrical control of interlayer excitons, as well as the realization of light emitting devices, demonstrates a wide range of possibilities for band structure engineering with HBLs. Therefore, combining our approach with electrically tunable interlayer excitons offers a promising platform for on-chip integrated photonics based on 2D materials.

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**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.0c02432.

Sample fabrication, methods of measurements, photoluminescence measurements, characterization of h-BN disk resonators, simulations of h-BN disk resonators, study of the vertical emitter position inside the h-BN disk resonator, transmission measurement of the waveguide-coupled disk resonator with integrated HBL, discussion of the effect of reabsorption inside the disk resonator, determination of Q-factor, as well as comparison of the resonance peak intensities for the waveguide-coupled disk resonator with integrated heterobilayer (PDF).

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**Author Contributions**

L.N. and R.K. conceived the project. R.K. and P.B. fabricated the devices and performed the experiments. A.J. and N.F. contributed to the experiments. S.N. and K.M. performed preliminary studies. T.T. and K.W. synthesized the h-BN crystals. R.K., P.B., N.F., A.J., and L.N. analyzed the data and cowrote the manuscript.

**Notes**

The authors declare no competing financial interest.

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