Wavelength Tunable Microdisk Cavity Light Source with a Chemically Enhanced MoS$_2$ Emitter

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ABSTRACT: In this work, we report an integrated narrowband light source based on thin MoS$_2$ emissive material coupled to the high quality factor whispering gallery modes of a microdisk cavity with a spatial notch that enables easy out-coupling of emission while it yields high spatial coherence and a Gaussian intensity distribution. The active light emitting material consists of chemically enhanced bilayer MoS$_2$ flakes with a thin atomic layer deposited SiO$_2$ protective coating that yields 20-times brighter chemically enhanced photoluminescence compared to as-exfoliated monolayers on the microdisk. Quality factors $\approx$ 1000 are observed as well as a high degree of spatial coherence. We also experimentally achieve effective index tuning of cavity coupled emission over a full free spectral range. The thermal response of this system is also studied. This work provides new insights for nanophotonic light sources with atomically thin active media.

KEYWORDS: 2D Semiconductor, MoS$_2$ photoluminescence enhancement, microdisk optical cavity, narrowband light source

Transition to a direct electronic bandgap in emerging transition metal dichalcogenide (TMDC) materials at the monolayer limit has enabled novel optoelectronic devices such as photodetectors, solar cells, and light-emitting diodes, which take advantage of neutral and charged excitons in these 2D semiconductors.$^{1-4}$ Arguably, one of the most prominent applications of bulk direct bandgap semiconductors is integrated photonic light sources based on optical waveguides and resonators.$^{5-7}$ However, such integrated photonic devices based on 2D semiconductors that may potentially form the discrete components of future optoelectronic systems are yet to be explored. Here, we demonstrate a wavelength tunable integrated narrowband light source enabled by chemically enhancing MoS$_2$ emission. A subwavelength notch coupler integrated into a microdisk cavity with high quality-factor (Q) whispering gallery modes (WGMs) enables easy extraction of MoS$_2$ emission while yielding high spatial and temporal coherence. We envision that this work will pave the way for nanophotonic light sources with atomically thin active media for next generation optoelectronic sensors and systems.

Among other emerging 2D materials, naturally occurring molybdenum disulfide (MoS$_2$) as direct bandgap semiconductor has gained a great deal of attention for novel discrete optoelectronic devices.$^{1-4}$ While a suspended monolayer MoS$_2$ flake shows photoluminescence (PL) efficiencies 100 times higher compared to a suspended bilayer flake,$^8$ its overall quantum yield (QY) is still below 1%, and excitonic emission is spectrally broad at room temperature. In contrast, when transferred onto substrates for device integration, QY for MoS$_2$ monolayers is even lower and only 3.5 times higher than that of a bilayer. Therefore, enhancement of its emission efficiency is necessary to realize the potential of atomically thin MoS$_2$ as a nanoscale active material in a light source. Modification of MoS$_2$ PL emission has been reported through addition of plasmonic nanoparticles,$^{9,10}$ chemical dopants,$^{11}$ and photonic crystal cavities.$^{12,13}$

Recent developments in nano-optics have made possible the manipulation of light at subwavelength scales.$^{14-17}$ For light sources such as lasers in particular, micrometer scale dielectric optical microcavities that support high-Q WGM resonances have been extensively studied as potential candidates for providing optical feedback.$^{14,18}$ These resonances arise as the light is confined via total internal reflection to a circular cavity. However, for efficient out-of-plane extraction of light, the strong confinement of the cavity modes necessitates sophisticated evanescent coupling schemes that require extremely precise placement of optical fibers or fabrication of waveguides next to the cavity.$^{19}$ This is a significant obstacle for the practical implementation of these devices as narrow band light-sources integrated with emerging nanomaterials with efficient yet simple output coupling schemes.

For efficient integrated photonic light sources with MoS$_2$ active region, one must address (1) high QY on substrates and (2) efficient and simple input–output coupling to the devices with an atomically thin optical active region. To this end, in this work, we demonstrate the coupling of excitonic PL from...
chemically enhanced MoS₂ flakes to the high-Q WGMs of a SiO₂ microdisk optical resonator with an engineered broadband notch input—output coupler for efficient optical excitation and light extraction. This unique combination enables a tunable narrowband light source with a high degree of temporal coherence due to the high-Q WGM resonances and spatial coherence by localizing the out-of-plane emission near the subwavelength notch coupler. The light emitting material in this case is chemically enhanced bilayer MoS₂ that is embedded in the SiO₂ by the micromechanical exfoliation technique.²⁰ The SiO₂ cavity was designed to have WGM resonances coinciding with the peak PL near 664 nm. A subwavelength (~λ/2) notch was introduced at the rim of the disk to significantly increase both in- and out-coupling efficiencies and simultaneously localize emission. This notch enables the structure to function effectively as an optically pumped, spectrally narrowband light source with an active material that is only a few atoms thick.

Figure 1, panels a and b show SEM images of a microfabricated notched microdisk light source with integrated MoS₂ layer. The orange shaded area in Figure 1, panel a and the white dotted outline in Figure 1, panel b show the device active region covered with the MoS₂ flake that was transferred from a bulk natural molybdenite crystal. These microdisks are fabricated from thermally grown SiO₂ released from the bulk natural molybdenite crystal. These microdisks are able to support high-Q-factor WGMs. These resonances can be characterized by the azimuthal mode number $m$, which determines the number of field maxima or minima around the microdisk circumference, and the radial mode number $n$ corresponds to the number of radial nodes in the field.²¹⁻²³ For operation at a fixed wavelength, the Q-factor for a WGM resonance increases with increasing radius.²¹⁻²³ However, in the case of exfoliated 2D materials, because of the small areas available with transferred flakes, the microdisk cannot be excessively large to allow for considerable spatial overlap with the WGMs. Because of the proximity of the support pedestal to the edge of the microdisk cavity, only the modes with $n = 1$ achieve high-Q. A disk of radius 3.8 μm was chosen, as it provides reasonably high-Q within the size limitation of the exfoliated MoS₂ flake. The microdisk height is constrained by the use of the 285 nm thermal oxide required for locating, through thin-film interference, low optical contrast MoS₂ flakes on Si substrates.²⁴

Figure 1, panel c shows the spatial distribution of the electric field intensity for the transverse-electric (TE), $n = 1$, $m = 46$ WGM of a perfectly circular microdisk. The mode shown is the PL from MoS₂ flake near the edges of the disk, which is expected to couple well to the WGMs due to the good spatial overlap with the mode. However, the WGM coupled light will not scatter efficiently out of the cavity in the out-of-plane direction. By incorporating a 200 nm × 300 nm semicircular notch, the light can be efficiently coupled in and out of the microdisk. Because of the subwavelength size of the notch, the Q is only minimally perturbed, which indicates that in-plane losses are dominated by leakage due to the small size of the cavity.²⁵ The modification to the WGMs from the notch can be seen in Figure 1, panel d. Since the PL emission of MoS₂ originates entirely from excitonic transitions polarized in the plane of the material, we only consider the TE modes of the microdisk resonator.²⁶

We calculated the emitted optical power from the notch by modeling the MoS₂ PL with a Lorentzian (Figure 1e, red curve) dipole source. This is shown in Figure 1, panel e along with the spectral response of the notched microdisk cavity. We observe that the system supports multiple $TE_{m=1,n}$ modes (Figure 1e, black curve). As a result, the far-field emission spectrum from the notch becomes almost entirely dominated by these WGMs whose relative intensities are modulated by the assumed Lorentzian spectrum.

To determine the cavity coupled absorption efficiency with the notch coupler, we assumed a Gaussian beam focused on the notch with a spectrum matching the experimental excitation wavelength of 532 nm. The coupler first scatters light efficiently into the plane of the silica microdisk to excite cavity modes. The optical power in these resonant modes circulating in the microdisk is then absorbed by the MoS₂ layer. The interaction of the cavity modes with the active layer is strong due to high cavity Q and good spatial overlap of the MoS₂ flake with the
performed PL and Raman microscopies on a 2L flake. We define "E-2L" as an exfoliated bilayer MoS₂ flake that has undergone the process of ALD etching, chemical enhancement, and encapsulation by SiO₂. Similarly, we denote as-exfoliated monolayer MoS₂ as "1L." Figure 2, panel a shows the separation of the E₁₂g and A₁₁g Raman active vibrational modes for 1L, 2L, and E-2L MoS₂ samples. For 2L and 1L MoS₂, the Raman peak separation values are around 22.3 and 19.5 cm⁻¹, respectively, which match closely to other reported values for the separation at these layer thicknesses. After exposure to the ALD process, the peak separation of an E-2L flake gradually decreases from 22.3 to 20.3 cm⁻¹ (Figure 2a). This implies that as more SiO₂ is deposited through the ALD process, more and more of the top monolayer of material is etched. The disappearance of initially 1L flakes further indicates the etching process.

Additionally, with increasing ALD cycles, the weak 2L MoS₂ PL transitions to a more luminescent monolayer-type emission after addition of SiO₂ (Figure 2b). As an internal calibration, the spectra are normalized to the MoS₂ Raman peak intensities. For a deposited SiO₂ thickness of ~20 nm, PL intensity dramatically increases ~70-fold, which corresponds to 20 times the brightness of the as-exfoliated monolayer MoS₂. We attribute this chemically enhanced PL to partial etching of the original bilayer MoS₂ yielding a mostly monolayer flake of higher quality than the as-transferred monolayer. In addition to the etching involved in the ALD process, the ozone-based chemistry used in the oxide deposition also might change the effective doping in the resulting monolayer, which further increases the luminescence. The thin SiO₂ layer also serves as an encapsulation for the resulting MoS₂ layer from further reactive etching processes needed for patterning of the microdisks that would detrimentally affect optical properties.

These E-2L MoS₂ flakes were then incorporated into SiO₂ microdisk optical resonators with the notch coupler. Figure 3, panel a, by probing the notch with a 532 nm laser, we see discrete emission peaks corresponding to the WGMs, which are modulated by the original MoS₂ PL intensity. Only certain wavelengths within the MoS₂ emission spectrum are coupled back into the microdisk cavity selected by the allowed cavity modes. It is these modes that are then scattered at the notch. These modes have Q factors as high as 900 (Figure 3b), which are in agreement with values in the literature for small optical microcavities in the visible. The free spectral range (FSR) of ~9.5 THz is also consistent across different samples (Supporting Information), which shows the sample uniformity.

It is also possible to tune the resonances by post processing. By slightly altering the dimensions of the microdisk, we can tune the cavity modes by more than the full FSR of the microcavity. To do this, we increased the disk height by sequentially adding SiO₂. Figure 3, panel c shows the tuning of the peaks as the SiO₂ thickness is increased. The thin SiO₂ layer also serves as an encapsulation for the resulting MoS₂ layer from further reactive etching processes needed for patterning of the microdisks that would detrimentally affect optical properties.

As shown in Figure 4, panel a, high-Q peaks with a line width of 2.44 meV can be achieved, which in turn means that the emitted light has a high degree of temporal coherence (coherence length of approximately 440 μm, in contrast to typical LED coherence lengths on the order of tens of micrometers). In addition, the notched microdisk geometry allows for a high degree of spatial coherence as well. Figure 4, panels c and d show FDTD simulations of the electric field intensity profile at the surface of the microdisk as well as 1 μm above the surface of the disk, where the only significant radiation into the far field comes from the locality of the notched area. The spatial coherence of the emitted light can be seen in the cross-sectional intensity plot of the electric field 1 μm above the surface of the disk in Figure 4, panel b. The intensity of the beam spot fits nicely to the Gaussian form with a width of 88 m (corresponding approximately to a divergence angle of 23.8°) and a coherence area of...
approximately 1.55 μm². This is larger than the observed beam spot size, which is approximately 0.805 μm² by treating it as an ellipse with major and minor axes of 1160 and 884 nm, respectively. Although we observe high spatial and temporal coherence, we do not observe lasing in this system, and any nonlinear pump intensity dependence is attributed to thermal effects.

In summary, we both experimentally and theoretically show that a 2D material, MoS₂, can be chemically enhanced and used to make a tunable light source with high spatial and temporal coherence when integrated with a notched microdisk optical cavity. Very high contrast emission in comparison to the MoS₂ PL can be seen at very low pump power densities. The success of recent efforts at synthesizing high-quality, large-scale MoS₂ by chemical vapor deposition (CVD) is extremely promising and will allow for device fabrication at a larger scale. Additionally, this platform can be utilized for other types of 2D TMDC materials, which gives us the ability to utilize and probe the optical properties of this emerging class of materials. This work will ultimately pave the way toward lasers with atomically thin gain media and has a great promise for the basis for lab-on-a-chip type sensing applications.

**ASSOCIATED CONTENT**

* Supporting Information
Methods, analysis of microdisk cavity design, and supplemental figures
This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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