Enhanced light-matter interaction in atomically thin MoS₂ coupled with 1D photonic crystal nanocavity

TAO LIU,[1,5] HAODONG QIU,[1,5] TINGTING YIN,[2] CHUNGCHE HUANG,[3] GUOZHEN LIANG,[1] BO QIANG,[1,2,3] YOUDE SHEN,[2] HOUKUN LIANG,[4] YING ZHANG,[4] HONG WANG,[1] ZEXIANG SHEN,[2] DANIEL W. HEWAK,[3] AND QI JIE WANG[1,2,*]

[1]*Centre for OptoElectronics and Biophotonics, School of Electrical & Electronic Engineering & The Photonics Institute, Nanyang Technological University, 639798, Singapore*
[2]*CDPT, Centre for Disruptive Photonic Technologies, School of Physical and Mathematical Sciences, The Photonics Institute, Nanyang Technological University, 639798, Singapore*
[3]*Optoelectronics Research Centre, University of Southampton, Southampton SO17 1BJ, UK*
[4]*SIMTech, 2 Fusionopolis Way, 138634, Singapore*
[5]*Equal contribution*
[*]*qjwang@ntu.edu.sg*

Abstract: Engineering the surrounding electromagnetic environment of light emitters by photonic engineering, e.g. photonic crystal cavity, can dramatically enhance its spontaneous emission rate through the Purcell effect. Here we report an enhanced spontaneous emission rate of monolayer molybdenum disulfide (MoS₂) by coupling it to a 1D silicon nitride photonic crystal. A four times stronger photoluminescence (PL) intensity of MoS₂ in a 1D photonic crystal cavity than un-coupled emission is observed. Considering the relative ease of fabrication and the natural integration with a silicon-based system, the high Purcell factor renders this device as a highly promising platform for applications such as visible solid-state cavity quantum electrodynamics (QED).

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1. Introduction

Monolayer transition metal dichalcogenides (TMDs), emerged as a new class of two-dimensional (2D) materials [1], have attracted a lot of attention due to remarkable properties in recent years. Monolayer Molybdenum disulfide (MoS$_2$), a typical example of TMDs, is a direct band gap semiconductor in the visible range which overcomes the gapless problem of graphene, thus making it promising in future optoelectronics. The energy band gaps of monolayer MoS$_2$ are located at two non-equivalent K and K' valleys of the Brillouin zone. Due to inversion symmetry broken, giant spin-orbit coupling splits the highest valence bands into two bands with spin up and spin down states. On one hand, inversion symmetry is broken making MoS$_2$ exhibit rich valley-contrasting physics. On the other hand, valley and spin degrees are strongly coupled owe to time-reversal symmetry suppressing significantly valley and spin relaxations. Consequently, we can control and manipulate valley pseudospin using circularly polarized light. This could open up opportunities for developing valley-based optoelectronic applications based on optical valley control in monolayer MoS$_2$ [2]. Additionally, optical response is dominated by tightly bound excitons in MoS$_2$ monolayer. In contrast to traditional inorganic semiconductors, monolayer MoS$_2$ show large exciton binding energy with hundreds of meV due to strong spatial confinement and reduced screening effect [3, 4], which provides the opportunity to achieve excitonic devices operating at room temperature. However, use of MoS$_2$ in optoelectronics applications requires overcoming photoluminescence (PL) low quantum yield. Therefore, novel approaches to manipulate light emission and enhance light-matter interaction are essential.

One effective approach to modify light emission and enhance PL quantum yield is to couple the monolayer MoS$_2$ into photonic crystal (PC) nanocavity [5] where spontaneous emission (SE) rate can be enhanced through Purcell effects, and emission direction can be modified. PL enhancement in TMDs members MoS$_2$ [6] and WSe$_2$ [7] monolayers coupled to two dimensional (2D) photonic crystal (PC) cavities has been demonstrated. The 2D photonic crystal is made of material systems of gallium phosphides (GPs) with relatively high refractive index in the visible range. For material systems operating in the visible, an alternative but probably more promising compared to GPs is the material of silicon nitride due to its ease of fabrication and compatibility with advanced silicon nanofabrication processes. The moderately low refractive index of about 2 of silicon nitride is an impediment to high quality-factor 2D PC nanocavity designs in visible range. However, an ultrahigh quality-factor PC nanocavity can be achieved when silicon nitride is fabricated into one dimensional (1D) nanobeam [8]. In this paper, we demonstrate a strong enhancement of light-matter interaction in monolayer MoS$_2$ coupled with 1D photonic crystal nanocavity made of silicon nitrides with low refractive index in visible range. A 4 times stronger PL intensity of MoS$_2$ in photonic crystal cavity than un-coupled emission is observed. Due to nature of ultrahigh Q factor and significant mode confinement of this 1D PC, this work pays the way to observing the strong light-matter interaction (i.e. exciton-polariton) and studying spin-valley based cavity QED phenomena by further optimizing the nanofabrication processes. Considering the relative ease of fabrication and the natural integration with silicon-based system, the high Purcell factor renders this device as a highly promising platform for applications of visible solid-state polaritonic devices such as bosonic laser.

2. Device fabrication and characterization

The enhancement of SE rate in optical cavity, compared with that in free space, is characterized by Purcell factor which is proportional to ratio of Q/V_m with quality factor Q and mode volume V_m [5]. Thus, the SE intensity increases as raised quality factor and reduced mode volume of optical cavity. Here, our experiment employs one dimensional (1D) PC nanocavity to integrate MoS_2, which shows a much higher Q factor and smaller mode volume than corresponding 2D PC used in low refractive index materials [9]. The 1D nanocavity is based on a suspended silicon nitride (Si_3N_4) PC structure of circular holes with lattice constant $a = 260$ nm and hole radius $r = 70$ nm. The nanobeam width and thickness are 310 nm and 210 nm, respectively. The cavity defect mode is defined by a linearly increased hole radius from 54 nm to 70 nm with a step increment of 4 nm, and lattice constant from 208 nm to 260 nm with a step increment of 13 nm at the nanobeam center. Figure 1(a) shows the simulated cavity mode profile with calculated quality factor $Q = 1.3 \times 10^5$ and mode volume $V_m \simeq 0.55 \left(\lambda/n \right)^3$ where λ is the peak wavelength of the mode and n is refractive index of nanobeam. The cavity is fabricated by a combination of electron beam lithography and dry/wet etching steps in low-pressure chemical vapor deposition (LPCVD) grown Si_3N_4 film on silicon substrate. The scanning electron micrograph of fabricated structure before MoS_2 transfer is shown in Fig. 1(c). The monolayer MoS_2 is grown on c-plane sapphire substrates by atmospheric pressure chemical vapor deposition (APCVD) [10]. Figure 2 shows the PL and Raman spectra of APCVD grown MoS_2 thin films. The peak separation between A_{1g} and E_{2g}^1 is 19.02 cm^{-1}. This value of peak seperation between two Raman peaks (E_{2g}^1 and A_{1g}) can be used to identify the number of MoS_2 layers, which is single layer for our grown MoS_2 [11]. The narrow full width at half maximum of E_{2g}^1 peak with 3.94 cm^{-1} implies MoS_2 thin films grown on c-plane sapphire are close to single crystal structures (See Fig. 2(c)). The monolayer MoS_2 on sapphire substrates is transferred onto PC nanocavity based on standard 2D materials transfer method [12] (See Fig. 1(a)).
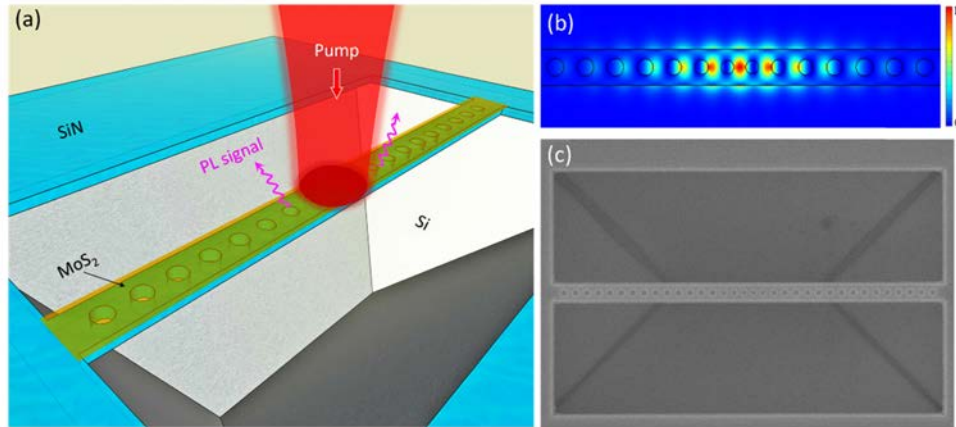

Fig. 1. (a) Schematic diagram shows the 1D photonic crystal integrated with monolayer MoS_2. (b) Simulated electric field intensity $\left| E \right|^2$ for the cavity optical mode. The resonance wavelength is 667.9 nm, and simulated Q is about 1.3×10^5 without considering the waveguide losses and mode volume is $V_m \simeq 0.55 \left(\lambda/n \right)^3$. (c) Scanning electron micrograph of a fabricated cavity structure before MoS_2 transfer. The substrate is silicon, and nanobeam of silicon nitride is suspended above it.
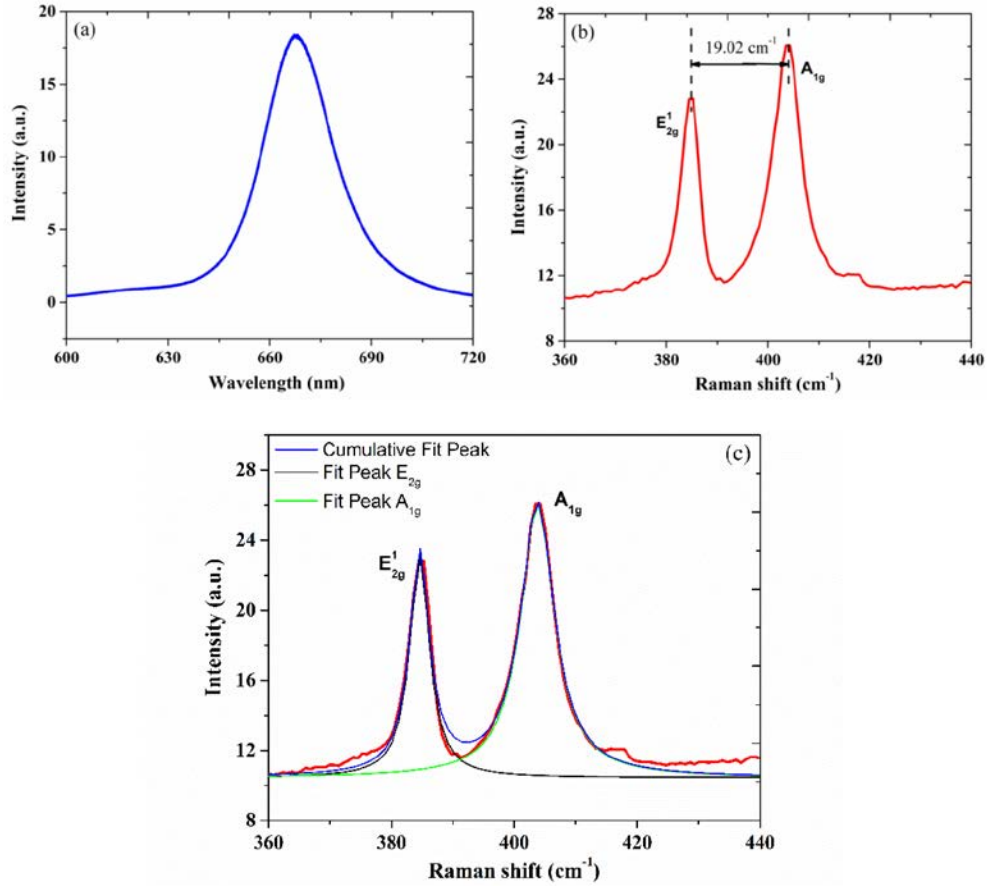
Fig. 2. (a) PL and (b) Raman spectra of atmospheric pressure chemical vapor deposition (APCVD) grown MoS_2 thin films on *c*-plane sapphire substrate. The peak separation between A_{1g} and E_{2g}^1 is 19.02 cm^{-1}. This value of peak separation between two Raman peaks (E_{2g}^1 and A_{1g}) can be used to identify the number of MoS_2 layers, which shows a single layer for our grown MoS_2. The narrow full width at half maximum of E_{2g}^1 peak implies MoS_2 thin films grown on *c*-plane sapphire are close to single crystal structures. (c) Lorentzian fit of Raman peaks. The full width at half maximum of peak E_{2g}^1 is 3.94 cm^{-1}.

3. Results and discussions

A micro-PL confocal microscope is used to characterize the fabricated PC nanocavity at room temperature. The devices are pumped with a 532 nm continuous-wave excitation laser through an objective lens with focused beam spot diameter of about 450 nm. The microscope is equipped with a three-axis piezoelectric stage allowing for spatial resolved PL measurements through samples scanning. Figure 3(a) shows the spatially resolved PL intensity map of fabricated PC nanocavity before MoS_2 transfer. The brightest area corresponds to region of PC nanocavity *i.e.* tapering part. The corresponding device area is illustrated in Fig. 3(b). We plot the PL spectra measured on PC and cavity regions in Fig. 3(c), indicated by the circles in Fig. 3(b). Only PL spectra in the cavity region is observed. The PL signal vanishes when measurement is taken on the PC region away from the cavity center. By fitting the resonance PL spectra, measured with a high-resolution grating (inset of Fig. 3(c)), to a Lorentzian profile, we experimentally observe a quality factor Q = 2050,

which is two order of magnitude smaller than that theoretically calculated owing to imperfections and roughness introduced in the fabrication process.
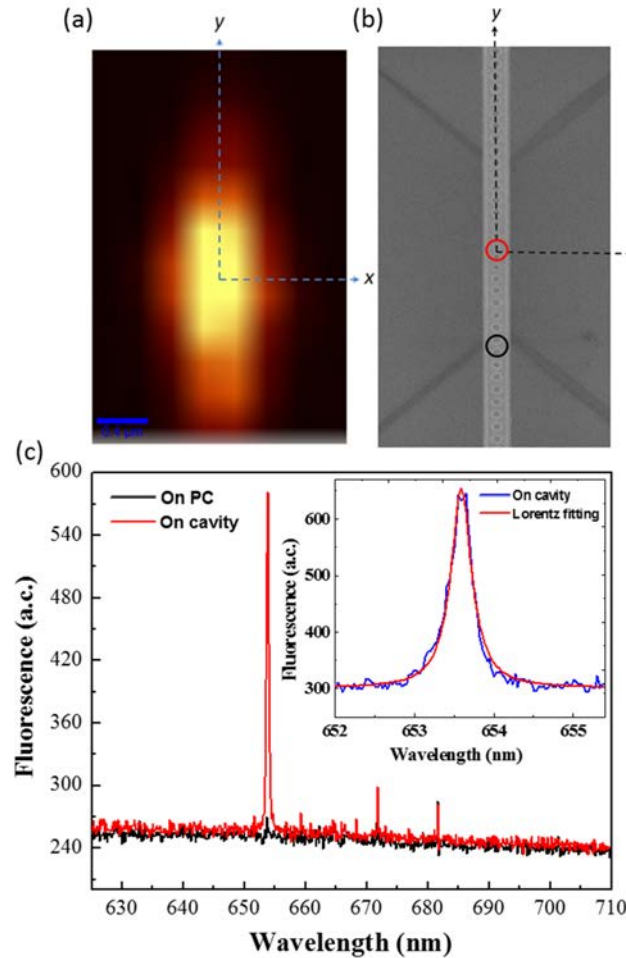

Fig. 3. (a) Micro-PL spatial intensity mapping of the fabricated nanocavity before MoS_2 transfer. The brightest area corresponds to area of cavity part of PC nanocavity. (b) The corresponding SEM image of the PC area with defined axes. (c) PL spectra with 600/mm grating, are measured on PC and cavity regions, as indicated by the circles with corresponding colors in (b). The inset: Cavity resonance measured with a high-resolution 1800/mm grating and Lorentzian fit. A fit to a Lorentzian gives Q = 2050.

Then we characterize the devices after MoS_2 transfer using the same tools. Figure 4 shows PL spectra of devices measured on PC, cavity and substrate regions. Noted the PL of silicon nitride does not subtracted from that of MoS_2 due to quite weak signal from it compared with that from MoS_2. In the region of cavity part of PC, due to Purcell effect which is proportional to ratio of Q/V_m, the PL intensity is greatly enhanced by about 4 times stronger than the emission in substrate region. On the PC part of PC nanocavity, PL intensity is also enhanced compared with that on substrate. Because of the coupling between the periodic holes of PC nanocavity and single-layer MoS_2, in-plane emission is prohibited due to photonic bandgap, which overlaps with the emission band of the monolayer MoS_2. Therefore, the SE should be re-directed into near-vertical wave-vectors within the light cone [13]. This SE redistribution and the higher collection efficiency from the PC part enhance the collection of emission into the vertical direction via the suppression of emission into in-plane PPC modes. Hence, PL

intensity in the PC region is larger than that in the substrate region. In principle, a strong exciton-light interaction (i.e. exciton-polariton) should be achieved in 1D silicon nitride photonic crystal due to theoretical ultrahigh Q value and significant mode confinement. The major obstacle to achieve high Q in experiments is the roughness of 1D edge and mismatched dimensions of hole size and space caused by imperfect nanofabrication. By further optimizing nanofabrication processes, we could achieve a new quantum regime of exciton-polariton in monolayer TMD based on 1D PC.
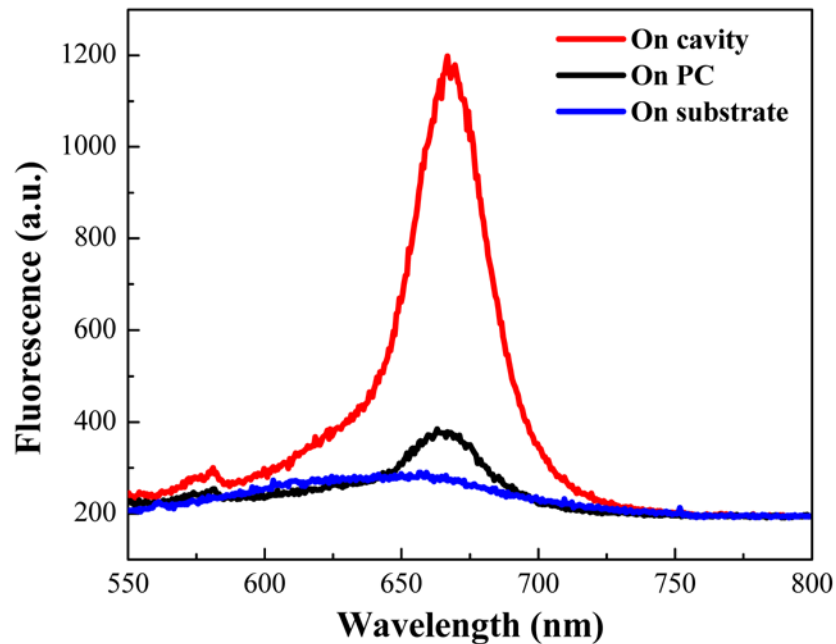

Fig. 4. PL spectra are measured on PC, cavity and substrate regions.

4. Summary

In conclusion, we have demonstrated the control of excitonic light emission from monolayer MoS_2 in an integrated photonic structure by coupling it to 1D silicon nitride photonic crystal. The high Purcell factor in the visible range can make this device highly promising platform for application in visible solid-state cavity electrodynamics (QED). A 4 times stronger PL intensity of MoS_2 in photonic crystal cavity than un-coupled emission is observed. Due to nature of ultrahigh Q factor and significant mode confinement of this 1D PC, the strong exciton-light matter interaction (i.e. exciton-polariton) could be observed in this device by further optimizing the nanofabrication processes.

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