Engineering Purcell factor anisotropy for dark and bright excitons in two dimensional semiconductors

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Abstract

Tightly bound dark excitons in atomically thin semiconductors can be used for various optoelectronic applications including light storage and quantum communication. Their optical accessibility is however limited due to their out-of-plane transition dipole moment. We thus propose to strengthen the coupling of dark excitons in two dimensional materials with out-of-plane resonant modes of a cavity at room temperature, by engineering the anisotropy in the Purcell factor. A silica micro-disk characterised by high confinement of light in small modal volume, high $Q$-factor and free spectral range is used to couple to the excitons in monolayer transition metal dichalcogenides (TMDCs). We show numerically that the tapering of sidewalls of the micro-disk is an extremely versatile route for achieving the selective coupling of whispering gallery modes to light emitted from out-of-plane dipoles to the detriment of that from in-plane ones for four representative monolayer TMDCs.

Keywords: microresonators, 2D semiconductors, dark excitons, Purcell factor

(Some figures may appear in colour only in the online journal)

1. Introduction

Two dimensional (2D) semiconductors are of great interest for photonic and optoelectronic applications due to their unique optical and electronic properties [1, 2]. Within this class, the 2D transition metal dichalcogenide (TMDC) materials exhibit direct bandgaps over a wide range from visible to near-infrared with strong excitonic emission [3]. The strong confinement of the charges in 2D TMDCs results in high binding energy ($E_b$) of the excitons which is about 1–2 orders of magnitude greater than that in quasi-2D systems such as GaN, GaAs quantum wells [4]. Such large values of binding energies of excitons make them dominate the optical properties of 2D TMDCs and allow the observation of various classes of excitons including the neutral, charged, localized, dark, intervalley, excitonic molecules and many other bound states [5]. The optical characteristics of these monolayer TMDCs are widely tunable by varying various parameters like doping, thickness and the dielectric and photonic environment making them excellent choices for optoelectronic devices [4].

For a photon with energy $\hbar \omega$ and $\mathbf{q}_\parallel$ as the projection of momentum on the TMDC plane, the optical selection rules allow the observation of various classes of excitons including the neutral, charged, localized, dark, intervalley, excitonic molecules and many other bound states [5]. The optical characteristics of these monolayer TMDCs are widely tunable by varying various parameters like doping, thickness and the dielectric and photonic environment making them excellent choices for optoelectronic devices [4].

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These transitions result in the formation of the bright ($X_b$) or the optically active excitons, while the forbidden transitions result in dark excitons ($X_d$) [6–8]. Though forbidden, they can be excited by their interactions with bright excitons, an external electric or magnetic field [9]. These dark excitons have relatively long radiative lifetimes compared their bright counterparts due to suppressed direct photon emission, making them ideal candidates for light storage and sensing applications [10]. But, their out-of-plane dipole moment makes their optical accessibility limited. The brightening of these dark excitons has been explored via numerous methods. One such being magnetic brightening where a magnetic field of large magnitude (usually above 10 T) is applied which alters the spin alignment without drastically disturbing the electronic structure [11–20]. An application of an out-of-plane electric field via plasmon polaritons can probe the dark excitons through strong near-field coupling [21, 22]. Photoluminescence collection from the monolayer sample edge [23] and angle resolved measurements [24] were also employed to detect the in-plane emission of dark excitons.

An alternative way to probe these dark excitons is to alter the exciton dynamics using engineered microcavities. The exciton dynamics can be best explained using a two-level quantum model where an excitonic state is described by an excitation frequency and a radiative decay rate. Further refinements include decoherence rate, intervalley scattering and other non-radiative decay channels. It is well known that by engineering the photonic environment of such a quantum emitter one can tune the local optical density of states thereby engineering the radiative decay rate [25, 26]. For instance, Khelifa et al [27] demonstrate waveguide-coupled disk resonators made of hexagonal boron nitride (h-BN) to selectively couple emission from interlayer excitons of a MoSe2–WSe2 heterobilayer, which also show an out of plane dipole component. The high oscillator strengths of intralayer transitions of the two monolayers lead to a high absorption efficiency. On the other hand, the low 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 the vdW disk resonators. Heindel et al [28] exploit deterministically fabricated quantum-dot microlenses to boost photon extraction and access the dark excitons through spin-blockaded metastable biexcitons. Andres-Penares et al [29] propose a versatile spectroscopic method that enables the identification of the out-of-plane component of dipoles due to the selective coupling of light emitted by in-plane and out-of-plane dipoles to the whispering gallery modes (WGM) of spherical dielectric microresonators, in close contact to them. The proposed method relies on the different selective abilities of IP and OP dipolar emission to excite WGM of silica microspheres deposited on top of the emitting layers. While the approaches mentioned above have enabled access to dark excitons, they do not seem feasible for a practical realisation in the fabrication schemes owing to their requirements of cryogenic temperature or large magnetic fields and complex geometry.

We propose to utilise a silica microdisk resonator platform to selectively couple the dark and bright excitons by engineering the resonator geometry. WGM resonators allow light to propagate along their circumference, making evanescent coupling of light in and out of the cavities facile [30, 31]. These are a class of resonators characterised by high $Q$ factors, low modal volume and high free spectral range (FSR). Silica as a choice of resonator material allows for reduced optical loss and easy chip integration [32, 33]. Silica microdisk has one of the simplest geometries of the WGM resonators and is robust to fabrication imperfections [34]. The central idea in our proposal is the engineering of the wedge angle of a tapered microdisk. The tapering of the microdisk is a result of the wet etching process used in the fabrication scheme of the silica microdisk [35]. The resonant modes tend to shift away from the sidewalls of the microcavity with increase in inclination thereby reducing radiation loss. This proves advantageous in increasing the $Q$-factor and Finesse of the cavity [36]. It is also reported that the tapering of microdisks results in compensation of the normal dispersion of the cavity [37].

We model the 2D TMDC excitons as dipoles and study the effect of in-plane (IP) and out-of-plane (OP) dipole orientation on their selective coupling to the WGMs of the tapered microdisk. Thereby the relative contributions of dark and bright excitons of the 2D TMDCs are analysed. The dark and bright excitons have predominantly out-of-plane and in-plane dipole moments respectively.

### 2. Results and discussion

WGM microdisk resonators have significant morphological dependence. Radius ($r$), thickness ($t$) and wedge angle ($\theta$) of the sidewall of a microdisk (figure 2(a)) are important parameters that alter the propagation of light within the cavity. In the following, we fix the radius of the silica microdisk at 5 $\mu$m, though the analysis can be easily extended to larger disk sizes. The TMDC layer is deposited on top of the microdisk and covers part of the tapered region as shown in figure 1.

#### 2.1. Thickness parameter

The thickness ($t$) of the microdisk is varied between 200 nm and 1200 nm and the permissible modes corresponding to each of these thicknesses are analysed using the finite-difference eigenmode (FDE) solver. Figure 2(d) shows the variation of the effective refractive index ($n_{eff}$) for these allowed modes of the microdisk of varying $t$. The microdisks of $t < 900$ nm support only the fundamental TE$_{00}$ and TM$_{00}$ which are shown in figure 2(b) thus making the thickness range 200 nm < $t$ < 900 nm constitute the single-mode operational region of the microdisk. While the microdisk with $t \geq 900$ nm supports the higher-order modes such as the TE$_{01}$ and TM$_{01}$ as shown in figure 2(b) clearly making it the multi-modal region of the microdisk. The electric field profiles of the fundamental TE$_{00}$ mode illustrate that the mode is clearly evanescent for smaller
values of $t$ such as $t = 200$ nm as plotted in figure 2(c). While the mode remains bounded within the disk boundary in case of $t = 800$ nm. We thus choose $t = 800$ nm which supports bounded mode with the reduced modal competition.

2.2. Tapering for selective coupling

The dependence of $\theta$ (angle subtended by the sidewall and the bottom side of the microdisk with $\theta = 90^0$ representing a normal sidewall and $\theta < 90^0$ representing a tapered sidewall as shown in figure 2(a)) on the excitation of WGMs of the microdisk is quantified using Purcell factor (PF) which is a measure of the interaction strength of excitons with light. The decay rate of an emitter oscillating at a frequency $\omega$ in free space with a dipole moment $d$ is given by $\Gamma_0$ [38]:

$$\Gamma_0 = \omega_0 |d|^2 / \left(12 \pi \varepsilon_0 \mu_c \omega^3 \right). \quad (1)$$

The resonant cavity alters the decay rate of the emitter without perturbing the frequency of emission, since the Lamb shift is usually quite small compared to the resonance frequency...
Fermi’s Golden rule state that $\Gamma$ is proportional to the local density of states (LDOS) which is dependent on the position, orientation and transition frequency of the emitter dipole. $PF$ defines the ratio of decay rate of the emitter in the near field of the cavity ($\Gamma_c$) to that in free space ($\Gamma_0$) as given in equation (2):

$$PF = \frac{\Gamma_c}{\Gamma_0} = 1 + \frac{6 \pi \varepsilon_0}{|d|^2 k^3} |d\cdot E_s(r_e)|,$$

where $\varepsilon_0$ is the vacuum permittivity, $k$ is the wave number in free space and $E_s$ is the scattered electric field of the emitter positioned at $r_e$. $PF$ can also be expressed as the enhancement of total power emitted by an exciton either radiated to the far-field $P_{rad}$ or dissipated into the resonator or in this case that which is coupled to the WGMs of the microdisk $P_{dis}$:

$$PF = (P_{rad} + P_{dis}) / P_0,$$

with $P_0$ being the source power. Thus the Purcell factor can be divided into radiative and non-radiative or simply the dissipated terms as:

$$PF = PF_{rad} + PF_{dis}.$$
In terms of the eigenmodes of the system $E_n$, the Purcell factor is governed by the local optical density of states $ho(e_d, r, \omega)$ which can be written in the form [40]:

$$\rho(e_d, r, \omega) \, d\omega = \sum_n \delta(\omega - \omega_n) |e_d \cdot E_n(r)|^2,$$

where the dipole is oriented along $e_d$. Though it is evident from the above equations that the Purcell factor is also dependent on the emitter location and orientation, we first determine the effect of $\theta$ on $PF$ with the help of integrated field strength in order to consider the average effect over the entire typical 2D TMDC flake size. We deduce the horizontal ($E_h$) and vertical ($E_v$) components of electric field which are expressed by a combination of the electric field components along $x$, $y$, and $z$ axes incorporating $\theta$. The electric field is obtained using the FDE solver where the field intensity is normalized to unity. The $E_x$, $E_y$, and $E_z$ components each constitute as a subset of the electric field. The region marked between $r_1$ and $r$ exhibits significant electric field strength, defined here by $r_1 = 3 \mu m$ for $r = 5 \mu m$, whereas $r_2$ denotes the start of the tapering of the sidewall. The diffusion length of monolayer TMDC materials (typically $<0.5 \mu m$) [41] maybe of concern to determine the value of $r_1$ but we find that the electric field of the optical mode is negligible beyond $x = 3 \mu m$ in the radial direction and hence will not contribute to the LDOS and thereby the Purcell factor. The $E_h$ and $E_v$ values are integrated over the disk edge $C_1$ with $r_1 \leq r \leq r_2$ and the taper edge $C_2$ ($r > r_2$) (figure 2(a)), as given in equations (6) and (7) below where $l$ represents the lines demarcated by the disk boundaries $C_1$ and $C_2$:

$$E_{h_{\text{int}}}^2 = \left(\int_{C_1} |E_h|^2 \, dl + \int_{C_2} |E_h|^2 \, dl\right)/\int dl,$$  

$$E_{v_{\text{int}}}^2 = \left(\int_{C_1} |E_v|^2 \, dl + \int_{C_2} |E_v|^2 \, dl\right)/\int dl.  \tag{7}$$

Figure 2(e) depicts the variation of $E_{h_{\text{int}}}$ and $E_{v_{\text{int}}}$ over a range of $\theta$ varying from $30^\circ$ to $90^\circ$. The horizontal and vertical electric field components are clearly distinguishable when $\theta$ is above $85^\circ$ or less than $42^\circ$. Larger values of $\theta$ are difficult to be realised in fabrication since wet etching techniques inherently produce significant tapering of sidewalls. For subsequent discussion, we choose a representative $\theta = 42^\circ$ to study the $PF$ for dipolar emissions varying across orientation and positions.

We use dipolar emissions of varying wavelength equivalent to the emissions of different 2D TMDCs including WS₂,
MoS$_2$, WSe$_2$ and MoSe$_2$ centred at 620 nm, 660 nm, 750 nm and 800 nm. The OP and circularly polarised IP dipoles (figures 3(a) and (b)) placed at different positions numbered 1–3 (figures 3(c)–(e)) along the surface of the microdisk and the taper edge are used to excite the WGMs of the microdisk. The Purcell enhancement obtained for these configurations are numerically analysed using the transmission box of FDTD tool exploiting equation (3). Figure 3 shows the Purcell enhancement obtained for dipolar emission of 800 nm across positions 1, 2 and 3 on the microdisk. Position 1, along the taper of the microdisk, allows for a Purcell enhancement of the OP dipolar emission coupled to the WGM about 5 times greater than that of the IP dipolar emission. While the dipoles on positions 2 and 3, along the top surface of the microdisk, result in weakly coupling to the WGMs of the microdisk. The tapered microdisk clearly couples the near field of the emitter to propagating optical WGM.

Figure 4 shows the PF analysis for dipolar emissions centred at 620 nm, 660 nm and 750 nm. While the FSR and PF value are seen to increase with increase in frequency of emission, the plots reveal a similar trend being followed across different emissions where the taper allows for selective coupling of the Purcell enhanced OP dipole over the IP dipole. A mismatch in polarisation or frequency between the dipole and resonant cavity leads to reduction in the decay rate which may also sometimes result in PF being in less than unity. The amount by which the taper prefers the OP dipole over the IP dipole or the ratio of Purcell enhancement obtained for OP dipole to that of the IP dipole ($PF_{OP}/PF_{IP}$) varies across different 2D TMDCs. This can be attributed to the frequency dependence of PF as described in equation (1). Though the values of ($PF_{OP}/PF_{IP}$) vary, the plots in figure 4 for all the materials clearly depict that the tapering of disk distinguishes the in-plane and out-of-plane modes making the selective optical access of the dark and bright excitons simple and effective through their spectral signatures. This is further validated through field profiles at the resonant peaks of the WGMs 742.3 nm and 743.8 nm corresponding to OP and IP oriented dipoles for an emission at 750 nm. The $E_z$ field profile at 742.3 nm proves the strong coupling of the WGM to the out-of-plane mode while the $E_z$ profile at 743.8 nm illustrates the dominance of in-plane coupled WGM. The relative field magnitudes also depict the preference of out-of-plane mode over the in-plane mode.

3. Conclusion

Dark excitons impact the exciton dynamics significantly in 2D TMDCs [42]. They possess a great potential for light storage, communication and information processing applications owing to their longer radiative lifetime [43]. We have shown how their optical accessibility can be enhanced and tailored by coupling of 2D TMDCs with tapered microdisks. We showed that the WGM in a silica microdisk can selectively couple the Purcell enhanced dark and bright excitons when the 2D TMDC is positioned along the tapered sidewall. Thus, tailoring of the excitonic light emission in 2D TMDCs can be achieved by coupling them with morphologically engineered WGM resonators. In particular, we have shown that engineering of the taper angle in a microdisk is a versatile route to achieving such a Purcell anisotropy for four representative 2D TMDCs. Our work should open up new avenues to explore this microdisk taper platform for studying exotic phenomena such as anisotropic vacuum induced excitonic valley coherence [44, 45] and strong coupling [46, 47] of dark excitons with WGMs and should also serve as a convenient probe for other exotic polaritons in 2D semiconductors, in particular interlayer excitons in heterostructures [48, 49].

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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