

Engineering radiative coupling of excitons in 2D semiconductors: supplementary material

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This document provides supplementary information to “Engineering radiative coupling of excitons in 2D semiconductors,” <https://doi.org/10.1364/optica.6.001443>. Sections 1 and 2 provide descriptions of the methods used to acquire the experimental data, including detailed sample preparation procedures and spectroscopy methods. Section 3 describes how the mirror-to-monolayer distance is calibrated. Section 4 provides an effective Hamiltonian description for our photonic system.

1. Sample preparation

Typical dry transfer method based on polymer stamping is applied to fabricate the encapsulated MoSe₂ and WSe₂ monolayer encapsulated with hexagonal boron nitrides(hBN) flakes. MoSe₂, WSe₂ and hBN were exfoliated from the high-quality crystals on 285nm SiO₂/silicon substrates. A polyethylene terephthalate stamp is used to pick up the layers in sequence at 50°C under microscope operation. The entire stamp along with the encapsulated TMDC is transferred onto a double-side polished sapphire wafer by melting the PET at 110°C. The PET stamp is later dissolved in a methylene dichloride solution.

2. Reflectance and photoluminescence spectroscopy

The encapsulated MoSe₂ or WSe₂ on a sapphire substrate is clamped on a copper plate with a through hole. A 2-by-2 millimeter distributed-bragg-reflector(DBR) with stopband from 640 to 800nm is mounted on a piezo-electric stage and positioned inside the through hole. Before the optical measurements, the DBR is pushed against the sapphire substrate leaving a small distance around 2-3 microns between the encapsulated TMDC and the mirror. During measurements, we step the piezo-electric stage to record the mirror-distance dependence and measure photoluminescence and reflectance spectra at the same sample position. All optical measurements are done in a cryostat with sample temperature at 50 Kelvin.

Differential reflectance and photoluminescence of the encapsulate TMD are measured. A femtosecond Ti:sapphire laser around 750nm was focused onto the sample via a 50× long working distance(0.42 numerical aperture) and the reflected light is selected by an aperture equivalent to ~2μm spot size in diameter. The light was collected into a grating spectrometer with a spectral resolution of 0.2 nm and recorded by a charge-coupled device. The reflectance contrast spectrum was determined by $R_s/R_m(\omega)$, where $R_s(\omega)$ and $R_m(\omega)$ are the reflected intensity from the mirror with and without the encapsulated monolayer. All reflection measurements are done with laser intensity of 50nW/μm² to prevent light-induced nonlinearity. This laser power corresponds ~8×10⁹ excitons/cm² when the exciton absorption is the strongest(anti-node). For photoluminescence, either 532nm or 633nm excitation laser is used to excite the sample. The emission is collected by the same optical path with a color filter to extinct the excitation laser.

3. Mirror distance calibration

To calibrate the monolayer-to-mirror distance for each piezo step, we utilize the interference pattern between the DBR mirror and the sapphire substrate when they are sufficiently far apart. FigureS1A shows the reflection spectra of the sapphire-DBR system normalized to the spectrum measured with sapphire only. The fringes observed within the DBR stopband(640 to 800nm) comes from the interference pattern between the sapphire

reflection and the DBR reflection. The wavelengths of the neighboring reflection minima are related to the mirror distance by the following equation:

$$2d \left(\frac{1}{\lambda_{\min,1}} - \frac{1}{\lambda_{\min,2}} \right) = 1.$$

This allows us to estimate d at different piezo positions for sufficiently large d to allow at least two fringes in the stopband. The calibration results are plotted in Figure S1B, showing a linear relation between d and the piezo step. This allows us to extrapolate the relation to smaller d for measurements shown in the main text.

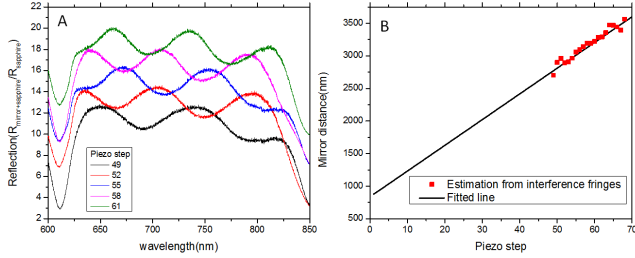


Figure S1. (A) Reflection spectra of the sapphire-DBR system normalized to the spectrum measured with sapphire only. The fringes arise from the interference between the sapphire and DBR reflections. (B) The sapphire-to-DBR distance estimated using the interference fringes in (A) vs. the corresponding piezo step.

4. Renormalization of exciton resonance energy and linewidth

The general Hamiltonian describing the collective excitations of Wannier excitons in quantum wells is considered in [1,2]. In this reference, the renormalized resonance energy and linewidth due to the coupling between free-space photons and the collective excitations are shown as Ω_{qnm} and γ_{qnm} in the equation (27) in [1]. Here, we provide an effective Hamiltonian including an excitonic transition and light-matter interactions when the thin film is placed in front of a perfect mirror:

$$H = \hbar\omega_0 b^\dagger b + \frac{1}{2} \int dr (|E(r)|^2 + |B(r)|^2) - d \cdot (E(L)b^\dagger + E^\dagger(L)b)$$

where $b^\dagger, b$ are creation and annihilation operators for an exciton, $\hbar\omega_0$ the excitonic resonance energy, d the transition dipole for the exciton, $E(r)$ and $B(r)$ the electric and magnetic field operator at position r , and L the location of the exciton dipole moment. The electric field in front of a mirror forms a standing wave pattern, which is also imposed on the vacuum fluctuations. Therefore the electric field operator $E(z) = i \int dk \alpha \sin(kz) a_k$, $k > 0$. Note that we assume α , the normalization constant, is independent of k since only the mode around exciton resonance is considered. Inserting the electric field operator into the Hamiltonian, we obtain equation (1) with $g = \alpha|d|/\hbar$. To solve the Hamiltonian, we make the ansatz of wavefunction:

$$|\psi(t)\rangle = X(t)|\text{exc}, 0\rangle + \int dk P_k(t) |g, k\rangle$$

where $|\text{exc}\rangle, |g\rangle$ denote the one exciton and ground state of the monolayer, $|0\rangle, |k\rangle$ denote the vacuum state and one photon in

mode k state of the radiation field, and $X(t), P_k(t)$ are the corresponding coefficients. The time-dependent Schrodinger equation, $i \frac{\partial |\psi(t)\rangle}{\partial t} = H|\psi(t)\rangle$, gives the dynamics of $X(t), P_k(t)$:

$$\begin{aligned} \dot{X}(t) &= -\frac{g}{\hbar} \int dk \sin(kL) \tilde{P}_k(t) e^{-i(\omega_k - \omega_0)t} \\ \dot{\tilde{P}}_k(t) &= -\frac{g}{\hbar} \sin(kL) \tilde{X}(t) e^{i(\omega_k - \omega_0)t}, \end{aligned}$$

where $\tilde{X}(t), \tilde{P}_k(t)$ are the excitonic and photonic wavefunction in the rotating frame. Solving the differential equation:

$$\begin{aligned} \dot{X}(t) &= -\frac{g^2}{\hbar^2} \int dk \sin(kL) \int_0^t dt' \tilde{X}(t') e^{-i(\omega_k - \omega_0)(t' - t)} \\ &= \frac{\pi g^2}{2c\hbar^2} \int_0^t dt' \tilde{X}(t') e^{-i\omega_0(t' - t)} [\delta(t' - t + \tau) - 2\delta(t' - t) \\ &\quad + \delta(t' - t - \tau)] \\ &= -\frac{\gamma}{2} \tilde{X}(t) + \frac{\gamma}{2} e^{i\omega_0\tau} \tilde{X}(t - \tau) \Theta(t - \tau) \end{aligned}$$

where $\tau = 2L/c$, $\gamma = \frac{\pi g^2}{c\hbar^2}$ and $\Theta(t)$ is the heavyside step function. For steady state, we can ignore $\Theta(t)$ and focus on the asymptotic solution for $t \rightarrow \infty$. The differential equation becomes a simple exponential decay $\dot{X}(t) = -\frac{\gamma}{2} (1 - e^{i\omega_0\tau}) \tilde{X}(t)$ in this limit, leading to a solution for excitonic wavefunction:

$$\begin{aligned} X(t) &= X(0) \exp[-i\tilde{E}_0 t/\hbar] \exp[-\frac{\tilde{\gamma}}{2} t] \\ |X(t)|^2 &= |X(0)|^2 \exp[-\tilde{\gamma} t] \end{aligned}$$

with $\tilde{E}_0, \tilde{\gamma}$ defined in equation 3(a) and (b).

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