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Spontaneous Emission Spectrum of a WS_2 Monolayer under Strong Coupling Conditions †

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Abstract: We study the spontaneous emission (SE) spectrum of a two-level quantum emitter (QE) near a WS_2 layer, in which case the Purcell factor of the QE can take values up to 10^4. We study the Rabi splitting in the SE spectrum at room temperature for QE free-space decay times in the 10–500 ps range. We observe that at close distance of the QE to the WS_2 layer the Rabi splitting lies between 0.25 eV and 0.05 eV, for increasing free-space decay times, indicating strong coupling conditions for the light-matter interaction between the QE and the WS_2 layer. As the distance between the QE and the layer increases, the light-matter interaction coupling enters the weak coupling regime, which leads to vanishing Rabi splitting in the SE spectrum for free-space decay times larger than a few tenths of ps.

Keywords: Quantum emitter; two-dimensional material; WS_2 layer; spontaneous emission spectrum; Rabi splitting; strong coupling

1. Introduction

Two-dimensional materials allow for extreme light confinement, thus becoming important candidates for all optical application platforms. Monolayers of transition-metal dichalcogenides (TMDs) are direct band gap semiconducting 2D materials featuring bandgaps in the visible and near-IR range, strong excitonic resonances, and high oscillator strengths, among other properties [1], as well as support for exciton polaritons [2]. These materials have been investigated extensively by theory and experiment in recent years [3–5]. TMDs are usually used as emission sources or absorbing layers, in which case their properties have been tested. Additionally, the optical properties of QEs such as molecules or quantum dots near single or multilayer TMDs have been investigated, in which case the relaxation rate of the QE either increases [6] or decreases [7]. The studies on the coupled QEs/TMD monolayers remain so far in the weak light-matter coupling regime.

In this work, we focus on the SE properties of a QE, modelled as a two-level system, in proximity to a WS_2 layer. We start by calculating the Purcell factor of the QE; we then show that the combined system approaches the strong light-matter coupling regime [8–13], when the separation of the QE to the WS_2 monolayer is small. We find that the strong coupling is manifested by Rabi splitting in the SE spectrum of the QE, which strongly depends on the free-space decay rate τ_0 of the QE, since the light-matter coupling between the emitter and the monolayer is inversely proportional to τ_0, as well as on the distance between the QE and the WS_2 layer.

2. Theory

An excited QE, modelled by a two-level system, as shown on the left side in Figure 1, interacts with its environment through the electromagnetic field and when the interaction is in the weak coupling regime, it relaxes from its excited state to the ground state by emitting a photon. In that case, the SE rate of the QE is $\Gamma_i(\omega, \mathbf{r}) = \frac{2\omega^2\mu^2}{\hbar\varepsilon_0 c^2}\hat{\mathbf{n}}_i \cdot \mathrm{Im}\,\mathcal{G}(\mathbf{r},\mathbf{r},\omega)\cdot\hat{\mathbf{n}}_i$, where $\hat{\mathbf{n}}_i$ is a unit vector along the i ($i = z, x$) direction of the transition dipole moment, $\boldsymbol{\mu}$, and $\mathcal{G}(\mathbf{r},\mathbf{s},\omega)$ is the Green's tensor, corresponding to the response of the geometry under consideration to a point-like excitation. In order to quantify the influence of the environment on the QE emission, we investigate the directional Purcell factor given by $\lambda^i(\omega,\mathbf{r}) = \frac{\Gamma_i(\omega,\mathbf{r})}{\Gamma_0}$, ($i = z, x$), where Γ_0 is the free-space spontaneous decay rate.

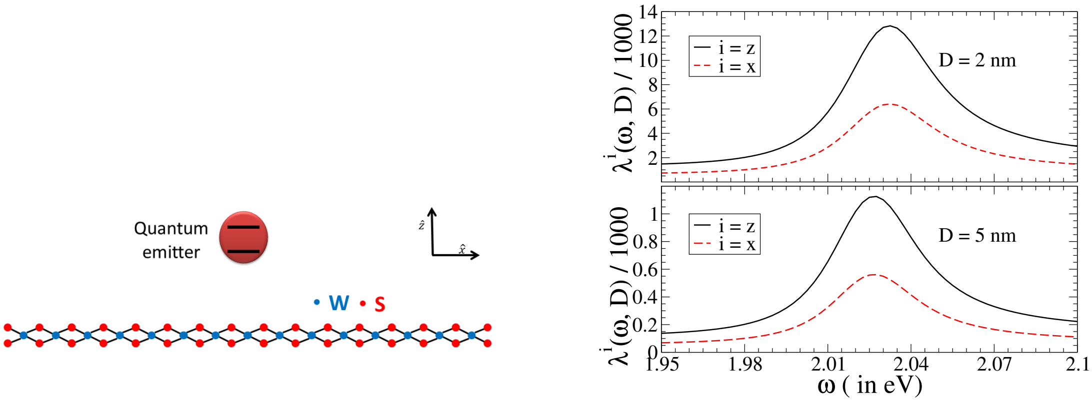

Figure 1. (color online) **Left**: Schematic representation of a two-level QE in proximity to an atomically-thin layer of WS_2. **Right**: Directional Purcell factors of a QE at distance $D = 2$ nm and $D = 5$ nm from an atomically-thin WS_2 layer along the perpendicular (z) and parallel (x) directions.

For the strong coupling regime, we consider the strength of the interaction between the QE and the WS_2 layer given through the spectral density $J(\omega,\mathbf{r})$, which is determined by the Purcell factor, given by [10]

$$J(\omega_1,\omega,\mathbf{r}) = \frac{\Gamma_0(\omega_1)}{2\pi}\lambda_i(\omega,\mathbf{r})\left(\frac{\omega}{\omega_1}\right)^3, \; i = z, x \tag{1}$$

where $\hbar\omega_1$ is the energy difference between the ground and the excited states of the QE. Therefore, the higher the Purcell factor the stronger the coupling of the QE with its environment. Also, the QE dipole strength is crucial for approaching the strong coupling limit; the higher its value the less enhancement of the Purcell factor is needed for entering the strong coupling regime. The SE spectrum of the QE is then given by the expression [8]

$$S(\omega,\mathbf{r}) = \frac{1}{2\pi}\left|\frac{\frac{\mu^2\omega^2}{\varepsilon_0 c^2}\hat{\mathbf{n}}_i\cdot\mathcal{G}(\omega,\mathbf{r},\mathbf{r}_d)}{\omega_1-\omega-\int_0^\infty d\omega' J(\omega_1,\omega',\mathbf{r})\frac{1}{\omega'-\omega}}\right|^2, \tag{2}$$

with $\mathbf{r}_d$ being the position of the signal detection, $\mathbf{r}$ being the QE position, and ω being the emission frequency.

3. Results and Discussion

The Purcell factors for a QE with z- and x-oriented transition dipole moment located at $z = D = 2$ nm and $z = D = 5$ nm from a WS_2 monolayer placed in the xy plane are presented on the right side in Figure 1. More details on the calculation of the Green's tensor and the corresponding Purcell factors are given in ref. [13]. We combine these results with the theory presented in the previous section and present the SE spectrum of a QE with a z-oriented transition dipole moment located at $D = 2$ nm and

$D = 5$ nm from the WS_2 layer for various free-space decay times τ_0. We also discuss the corresponding Rabi splittings in the SE spectrum in each case.

In the left panel in Figure 2 we present the SE spectrum for a QE with transition frequency $\omega_1 = 2$ eV and z-oriented transition dipole moment located at $D = 2$ nm from the WS_2 layer for various free-space lifetimes τ_0 in the range between 10 ps and 500 ps. We observe that for small τ_0 values, which correspond to large values for $\Gamma_0 = 1/\tau_0$, several peaks are present in the spectrum. They are not all of the same magnitude. Defining as Rabi splitting the energy difference between the two highest peaks in the spectrum for a given τ_0 value, we read such splittings in the range between 0.25 eV, for $\tau_0 = 10$ ps, and 0.05 eV, for $\tau_0 = 500$ ps, which correspond to $0.125\omega_1$ and $0.025\omega_1$, respectively. We conclude that the light-matter interaction between the QE and the WS_2 layer lies in the strong coupling regime, when $\tau_0 = 10$ ps, while the coupling slowly becomes weak as the Rabi splitting decreases with increasing τ_0. In the middle panel of Figure 2, we show the Rabi splitting in the spectrum for a QE as in the case presented in the left panel, as function of Γ_0, in order to investigate whether there is a simple linear relation between the two quantities; apparently, the existence of such a simple relation is not supported by the findings in the middle panel in Figure 2.

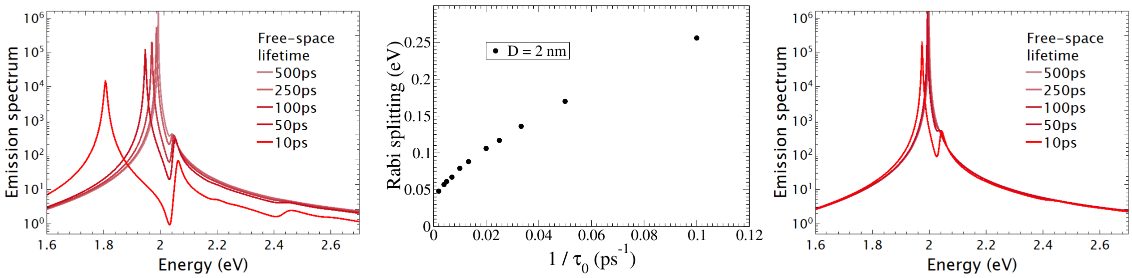

Figure 2. (color online) The SE spectrum of a QE for various free-space life-time values τ_0 with $\omega_1 = 2$ eV and z-oriented transition dipole moment located at $D = 2$ nm (left panel) and at $D = 5$ nm (right panel) from a WS_2 layer. The Rabi splitting in the spectrum of a QE located at $D = 2$ nm as function of $1/\tau_0$ is also shown (middle panel).

Next, in the right panel in Figure 2, we present the SE spectrum for a QE with transition frequency $\omega_1 = 2$ eV and z-oriented transition dipole moment located at $D = 5$ nm from the WS_2 layer for various free-space lifetimes τ_0 in the range between 10 ps and 500 ps. In this case, we find Rabi splitting of 0.069 eV and 0.041 eV, for $\tau_0 = 10$ ps and $\tau_0 = 50$ ps, respectively, while no splitting is observed in all other cases. This finding is understandable since the Purcell factor of the QE at this distance is over an order of magnitude smaller than at $D = 2$ nm, as shown in the right panel in Figure 1.

4. Conclusions

In this work, we studied the spontaneous emission spectrum of a two-level QE near a WS_2 layer, in which case the Purcell factor of the QE can take values up to 10^4. We further studied the Rabi splitting in the SE spectrum at room temperature for a QE with free-space decay times in the 10 ps to 500 ps range. We observe that at close distance of the QE to the WS_2 layer, combined with short decay times, the spectrum can feature several peaks. In such cases, the Rabi splitting lies between 0.25 eV and 0.05 eV, for increasing free-space decay times, indicating strong coupling conditions for the light-matter interaction between the QE and the WS_2 layer. Moreover, no simple relation between the inverse free-space decay time and the corresponding Rabi splitting value has been observed. As the distance between the QE and the layer increases further, the light-matter interaction coupling enters the weak coupling regime, which leads to vanishing Rabi splitting in the SE spectrum for free-space decay times larger than a few tenths of ps.

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Conflicts of Interest: The authors declare no conflict of interest.

Abbreviations

The following abbreviations are used in this manuscript:

SE	spontaneous emission
QE	quantum emitter
TMDs	transition-metal dichalcogenides

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