Modelling the optical control of electron spin dynamics in a quantum dot near a two-dimensional semiconductor

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Abstract. A quantum dot in the Voigt configuration is a basic system with important potential applications in quantum technologies. The spin states of this quantum dot can act as a prototype qubit. By applying laser pulses the dynamics of the quantum dot spin can be controlled and the necessary quantum gates can be achieved. An important problem is the initialization, i.e., the creation of one of the two electron spin states starting from the natural initial state of the system, which is an equal incoherent mixture of the two spin states. The initialization process can be achieved by proper interaction of the quantum dot with laser pulses. Also, it has been realized that the integration of the quantum dot with photonic structures that give preferential Purcell-enhanced decay rate towards the target spin state increases the fidelity of spin initialization. Here, we propose a new coupled quantum dot - nanophotonic structure that may give high initialization fidelity in short times by coupling the quantum dot with a tungsten disulfide (WS$_2$) monolayer.

For the modelling of the spin dynamics we combine quantum dynamics calculations with electromagnetic calculations. Specifically, we model the interaction of the quantum dot with the applied laser field with density matrix equations. Also, the spontaneous decay rates that enter in the density matrix equations are obtained by electromagnetic calculations based on the electromagnetic Green’s tensor, which is calculated with the scattering superposition method. We first show that the spontaneous decay rates for the quantum dot near a WS$_2$ monolayer are enhanced by the Purcell effect and are anisotropic for quantum dot dipole moments parallel and perpendicular to the layer. We then use the most common method for initialization, optical pumping and show that a preferential Purcell-enhanced decay rate towards the target spin state increases the fidelity of spin initialization in short times, in comparison to the case that the quantum dot is placed in an isotropic photonic environment.

1. Introduction
Semiconductor quantum dots (QDs) are favourable candidates for quantum information technologies. An important QD structure is based on the spin states in the Voigt geometry [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. The natural initial state of the QD in the Voigt geometry is an incoherent mixture with equal populations of the two electron spin states. Therefore, for quantum information processing applications [13], initialization is needed, where a specific electron spin state is created. The most usual approach for initialization is based on the optical pumping process, which is strongly benefited by a preferential Purcell-enhanced deexcitation towards the desired state (anisotropic Purcell effect) by coupling the QD with microphotonic and nanophotonic structures [5, 6, 7, 8, 10, 11, 12].

Here, we place the QD next to a tungsten disulfide (WS$_2$) monolayer. WS$_2$, a transition metal dichalcogenide [14], is an atomically thin, 2D direct band gap semiconductor. It features strong excitonic resonances, high oscillator strengths, supports exciton-polaritons [15, 16], and can modify the spontaneous decay rates for nearby quantum emitters [15, 17]. Below we show that the spontaneous decay rates for a QD near a WS$_2$ monolayer are anisotropic for electric dipoles parallel and perpendicular
Figure 1. (a) Energy level diagram for a QD in the Voigt geometry. The magnetic field induces the Zeeman splitting in the upper and lower levels. The optical field resonant with the $|1\rangle \leftrightarrow |4\rangle$ transition is $x$-polarized. (b) The QD is placed a distance $z = R$ above a WS$_2$ monolayer, which lies on the $x-y$ plane at $z = 0$. The growth axis of the QD is $y$ while the magnetic field is applied along the $x$-axis.

to the layer and the decay rates can also be enhanced by the Purcell effect. Therefore, a WS$_2$ monolayer offers a new platform for modifying the initialization process in a nearby QD in the Voigt geometry.

In this paper, we first obtain the corresponding Purcell factors for the QD next to a WS$_2$ monolayer by calculation of the electromagnetic Green’s tensor, using the scattering superposition method. Then, we use the modified decay rates in the density matrix equations to investigate the dynamics of spin initialization for a coupled QD-WS$_2$ monolayer structure using optical pumping. We also use two different types of optical pulses. Specifically, we apply a continuous wave laser field, which gives a constant Rabi frequency, and a Gaussian laser pulse, leading to a time-dependent Rabi frequency profile, and show that both cases are benefited from the presence of the WS$_2$ monolayer, leading to higher spin initialization fidelities with the WS$_2$ layer than without it.

2. Basic Equations

We study a typical QD in the Voigt configuration [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. We consider a singly-charged self-assembled QD grown along the $y$-axis, where an external magnetic field has been applied in the Voigt geometry, along the $x$-axis, and lifts the degeneracy of the electron/hole levels. We name the ground spin levels as $|1\rangle = |\downarrow_x\rangle$ and $|2\rangle = |\uparrow_x\rangle$ and the two excited trion states as $|3\rangle = |\downarrow_x\uparrow_x\uparrow_x\rangle$ and $|4\rangle = |\downarrow_x\uparrow_x\downarrow_x\rangle$. Here, $\uparrow (\downarrow)$ and $\uparrow (\downarrow)$ denote heavy hole and electron spins, respectively. For the level scheme, see Fig. 1(a). Here, the vertical transitions ($|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |3\rangle$) give $x$-polarized dipole matrix elements and the cross transitions ($|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |4\rangle$) give $z$-polarized dipole matrix elements. In this work, we aim to create state $|2\rangle$. We explore the method of optical pumping and study the case that the QD interacts with a $x$-polarized laser field applied at exact resonance with the $|1\rangle \leftrightarrow |4\rangle$ transition.

The Hamiltonian describing the interaction between the laser field and the QD structure, in the dipole and rotating wave approximations, is given by

$$H = \sum_{n=1}^{4} \hbar \omega_n |n\rangle \langle n| - \hbar \left[ V(t) e^{-i\omega_0 t} |4\rangle \langle 1| + V(t) e^{-i\omega_1 t} |3\rangle \langle 2| + \text{H.c.} \right],$$

where $\hbar \omega_n, n = 1, 2, 3, 4$, is the energy of state $|n\rangle$ and $V(t)$ is the generally time-dependent Rabi frequency of the applied field. The field is resonant with the $|1\rangle \leftrightarrow |4\rangle$ transition, thus we have chosen $\omega_0 = \omega_4 - \omega_1 = \omega_{41}$. 


Using the above Hamiltonian we can obtain the equations of motion for the slowly varying parts of the density matrix elements of the QD:

\begin{align*}
\dot{\sigma}_{11} &= \gamma_{41}\sigma_{44} + \gamma_{31}\sigma_{33} - 2\nu\sigma_{11}^I, \\
\dot{\sigma}_{22} &= \gamma_{32}\sigma_{33} + \gamma_{42}\sigma_{44} + 2\nu\sigma_{22}^I, \\
\dot{\sigma}_{33} &= -\left(\gamma_{31} + \gamma_{32}\right)\sigma_{33} - 2\nu\sigma_{33}^I, \\
\dot{\sigma}_{44} &= -\left(\gamma_{41} + \gamma_{42}\right)\sigma_{44} + 2\nu\sigma_{44}^I, \\
\dot{\sigma}_{11}^I &= -\frac{\gamma_{41} + \gamma_{42}}{2}\sigma_{44}^I + \mathcal{V}(\sigma_{11} - \sigma_{44}), \\
\dot{\sigma}_{23}^R &= -\frac{\gamma_{31} + \gamma_{32}}{2}\sigma_{23}^R + (\omega_{21} + \omega_{43})\sigma_{21}^I, \\
\dot{\sigma}_{23}^I &= -\frac{\gamma_{31} + \gamma_{32}}{2}\sigma_{23}^I - (\omega_{21} + \omega_{43})\sigma_{21}^R + \mathcal{V}(\sigma_{33} - \sigma_{22}),
\end{align*}

where \(\sigma_{23} = \sigma_{23}e^{-i(\omega_{41} + \omega_{43})t}\) and the superscripts \(R, I\) denote real and imaginary parts, respectively. The density matrix equations have also incorporated the spontaneous emission from the upper to the lower energy levels, with

\[
\gamma_{41} = \gamma_{32} = \gamma_x = F_x\gamma, \quad \gamma_{42} = \gamma_{31} = \gamma_z = F_z\gamma
\]

being the radiative decay rates of the corresponding QD transitions modified by the coupling between the QD and the WS\(_2\) layer, where \(\gamma\) is the decay rate in free-space and \(F_j\), with \(j = x, z\), is the corresponding Purcell factor. Throughout this paper the free-space relaxation rate is taken to be \(h\gamma = 1.2\ \mu\text{eV}\) [3, 9].

For the electromagnetic calculations of the Purcell factor, we obtain the electromagnetic Green’s tensor, using the scattering superposition method [10, 11, 12]. The geometry we study consists of a dielectric environment with permittivity \(\varepsilon_1\) and a WS\(_2\) monolayer placed in it. As it is shown in Fig. 1(b), the WS\(_2\) monolayer lies on the \(x - y\) plane at \(z = 0\), while the QD is placed a distance \(z = R\) above the layer. The optical response of WS\(_2\) monolayer is given by its surface conductivity, \(\sigma\). For a layer positioned at the \(x - y\) plane at \(z = 0\), in a homogeneous material with dielectric permittivity \(\varepsilon_1\), the generalized Fresnel coefficients will be [18],

\[
R_{1M}^{11} = -\frac{-\alpha k_0}{k_{z1} + \alpha k_0}, \\
R_{1N}^{11} = \frac{\alpha k_0 k_{z1}}{k_1^2 + \alpha k_0 k_{z1}}
\]

where \(\alpha = 2\pi\sigma/c\). For a QD positioned at \(r_{\text{QD}} = (0, 0, R)\), the Purcell factors corresponding to \(z\)- and \(x\)- orientations of the transition dipole moment are obtained by:

\[
F_z = \sqrt{\varepsilon_1} + \frac{3c}{2\omega} \frac{k_s k_{z1}^2}{k_{z1}^2} R_{1N}^{11} e^{2ik_{z1}R},
\]

where \(k_s, k_{z1} = \sqrt{k_1^2 - k_s^2}\) are the in-plane and perpendicular wavevector components, respectively, and \(k_1 = \frac{\omega}{c} \sqrt{\varepsilon_1}\) is the wavenumber in the dielectric medium.
The in-plane conductivity $\sigma$, which encodes the optical response of WS$_2$, is given by the expression $\sigma_{2D} = i\varepsilon_0 \omega d [\varepsilon_{WS_2}(\omega) - 1]$, where $d = 0.65$ nm is the thickness of the WS$_2$ monolayer. Also,

$$\varepsilon_{WS_2}(\omega) = \varepsilon_\infty + \sum_{j=1}^{9} \frac{f_j}{\omega_j^2 - \omega^2 - i0.5\omega\gamma_j},$$

(14)

where the various parameters are extracted by fitting to experimental data [19, 20, 21] The fitting parameters are: $\varepsilon_\infty = 8.76$, $f_j = [1.9, 0.254, 0.146, 0.068, 3.07, 1.17, 0.068, 15.5, 12.7]$ (eV$^2$), $h\omega_j = [2.014, 2.185, 2.250, 2.285, 2.402, 2.575, 2.655, 2.845, 3.047]$ (eV), and $h\gamma_j = [0.029, 0.1, 0.1, 0.1, 0.14, 0.21, 0.21, 0.265, 0.25]$ (eV). The above modelling for $\varepsilon_{WS_2}(\omega)$ corresponds to low temperature response.

### 3. Numerical results

Using the above relations we can calculate the Purcell factors for the system QD-WS$_2$ monolayer. Throughout this work we consider a QD with an emission energy of $\hbar\omega_0 = 1.31$ eV. In Fig. 2 we display the Purcell factors $F_x, F_z$ as functions of the distance $R$ between the QD and the WS$_2$ monolayer, taking that the dielectric environment is free-space ($\varepsilon_1 = 1$). In both cases there is an enhancement of the spontaneous decay rates relevant to the decay rate in vacuum, due to the Purcell effect. Also, the decay rates reduce monotonically as the distance between the QD and the WS$_2$ layer increases. In addition, for every distance the decay rate for a dipole parallel to the WS$_2$ layer (e.g. a $x$-oriented dipole) is always lower than the decay rate for a dipole perpendicular to the WS$_2$ layer (e.g. a $z$-oriented dipole), so the necessary spontaneous emission anisotropy needed for Purcell-enhanced deexcitation process towards the desired state is succeeded for a QD near a WS$_2$ layer.

In Fig. 3 we present calculations for the time evolution of the population of the four quantum states under the application of a continuous wave laser field with Rabi frequency $\mathcal{V}(t) = \mathcal{V}_0$. In all the calculations in this work the quantum dot system starts from an initial incoherent mixture of the two electron-spin states, so $\sigma_{11}(0) = 1/2$, $\sigma_{22}(0) = 1/2$, $\sigma_{33}(0) = \sigma_{44}(0) = 0$, and $\sigma_{nm}(0) = 0$ with $n \neq m$. In Fig. 3(a), we consider that the QD is in an isotropic photonic environment (specifically, here, free-space), so the excited trion states decay to the ground spin states with the same population decay rate $\gamma$. We note that high population transfer to the spin state $|2\rangle$ can occur that increases with the development of time, specifically $\sigma_{22}(1 \text{ ns}) = 0.790273$, $\sigma_{22}(2 \text{ ns}) = 0.915198$, and $\sigma_{22}(3 \text{ ns}) = 0.964372$. We then consider that the QD is placed near a WS$_2$ monolayer. Then, the decay rates become asymmetric and are determined by the results of Fig. 2. Here, we take three different distances between the QD and the WS$_2$ monolayer, $R = 5$ nm in Fig. 3(b), $R = 10$ nm in Fig. 3(c), and $R = 15$ nm in Fig. 3(d). In all cases the time evolution of the population changes, as different decay rates are used for the different distances. Also, the population transfer to the spin changes $|2\rangle$ is succeeded with higher fidelities than when the QD is

![Figure 2](image-url)
In free-space vacuum. Specifically, $\sigma_{22}(1 \text{ ns}) = 0.928327$, $\sigma_{22}(2 \text{ ns}) = 0.980307$, $\sigma_{22}(3 \text{ ns}) = 0.985932$ for $R = 5 \text{ nm}$, $\sigma_{22}(1 \text{ ns}) = 0.9725$, $\sigma_{22}(2 \text{ ns}) = 0.996774$, $\sigma_{22}(3 \text{ ns}) = 0.997837$ for $R = 10 \text{ nm}$, and $\sigma_{22}(1 \text{ ns}) = 0.884518$, $\sigma_{22}(2 \text{ ns}) = 0.973891$, $\sigma_{22}(3 \text{ ns}) = 0.992869$ for $R = 15 \text{ nm}$. Obviously, the highest fidelity from these examples is obtained for $R = 10 \text{ nm}$.

In Fig. 4 we present calculations for the time evolution of the population of the four quantum states under the application of a Gaussian laser pulse with Rabi frequency $\mathcal{V}(t) = \mathcal{V}_0 e^{-(t-t_f/2)^2/t_p^2}$, where $t_p$ determines the width and $t_f/2$ the center of the laser pulse. In Fig. 4(a), we consider that the QD is in free-space vacuum. Efficient population transfer to the spin state $|2\rangle$ occurs that leads to $\sigma_{22}(t_f) = 0.867084$ at the end of the pulse (here $t_f = 3 \text{ ns}$). We then study the case with the QD near the WS$_2$ monolayer for three different distances between the QD and the WS$_2$ monolayer, $R = 5 \text{ nm}$ in Fig. 4(b), $R = 10 \text{ nm}$ in Fig. 4(c), and $R = 15 \text{ nm}$ in Fig. 4(d). In all cases the time evolution of the population changes, as different decay rates are used for the different distances. Also, the population transfer to the spin state $|2\rangle$ is succeeded with higher fidelities than when the QD is in an isotropic photonic environment. Specifically, $\sigma_{22}(t_f) = 0.917693$ for $R = 5 \text{ nm}$, $\sigma_{22}(t_f) = 0.981043$ for $R = 10 \text{ nm}$, and $\sigma_{22}(t_f) = 0.929604$ for $R = 15 \text{ nm}$. In this case, too, the highest spin initialization fidelity from these examples is obtained for $R = 10 \text{ nm}$. Also, the corresponding results for the continuous wave laser field gives higher fidelities than the Gaussian laser pulse.

Figure 3. The time evolution of the population, $\sigma_{nn}(t)$ with $n = 1 - 4$, of states $|1\rangle$ (solid curve), $|2\rangle$ (dashed curve), $|3\rangle$ (dotted curve) and $|4\rangle$ (dash-dotted curve) for a continuous wave laser field with $\mathcal{V}_0 = 7.5 \text{ ns}^{-1}$. In (a) the QD is placed in vacuum and in (b), (c), and (d) it is placed near a WS$_2$ monolayer. In (b) $R = 5 \text{ nm}$, (c) $R = 10 \text{ nm}$, and (d) $R = 15 \text{ nm}$.

4. Conclusions
In this paper we studied the problem of the controlled population dynamics for a QD in the Voigt geometry placed near a WS$_2$ monolayer, under the interaction with a laser field, with emphasis to the problem of spin initialization. We modelled the quantum dynamics of the QD by the density matrix

\[
\rho(t) = \sum_{n,m} \sigma_{nm}(t) |n\rangle \langle m|.
\]
Figure 4. The time evolution of the population, $\sigma_{nn}(t)$ with $n = 1 - 4$, of states $|1\rangle$ (solid curve), $|2\rangle$ (dashed curve), $|3\rangle$ (dotted curve) and $|4\rangle$ (dash-dotted curve) for a Gaussian laser pulse with $V_0 = 8.8625$ ns$^{-1}$, $t_f = 3$ ns, and $t_p = t_f/6$. In (a) the QD is placed in vacuum and in (b), (c), and (d) it is placed near a WS$_2$ monolayer. In (b) $R = 5$ nm, (c) $R = 10$ nm, and (d) $R = 15$ nm.

equations and included the effect of the WS$_2$ layer on the modified, due to the Purcell effect, spontaneous decay rates that enter the density matrix equations. The spin initialization is succeeded by optical pumping using either a continuous wave laser field or a Gaussian laser pulse. We showed that high levels of fidelity, larger than in the case of the QD without the WS$_2$ monolayer, can be quickly obtained due to the anisotropy of the enhanced spontaneous decay rates of the QD near the WS$_2$ layer, depending on the distance between the QD and the WS$_2$ monolayer. We note that these are primary results, and a full parametric study of the spin initialization dynamics for various distances between the QD and the WS$_2$ layer is needed and we have started working on it. Also, one may use optimal control [10, 11, 12] for further increasing the fidelity of the spin initialization.

Acknowledgements
Co-financed by Greece and the European Union - European Regional Development Fund via the General Secretariat for Research and Technology bilateral Greek-Russian Science and Technology collaboration project on Quantum Technologies (project code name POLISIMULATOR). The authors thank Dr. Nikos Iliopoulos for useful discussions and help.

References
[1] Warburton R J 2013 Nat. Materials 12 483
[2] Gao W B, Imamoglu A, Bernien H and Hanson R 2015 Nat. Photonics 9 363.
[3] Emary C, Xu X, Steel D G, Saikin S and Sham L J 2007 Phys. Rev. Lett. 98 047401
[4] Economou S E and Reinecke T L 2007 Phys. Rev. Lett. 99 217401
[5] Loo V, Lanco L, Krebs O, Senellart P and Voisin P 2011 Phys. Rev. B 83 033301
[6] Majumdar A, Kaer P, Bajcsy M, Kim E D, Lagoudakis K G, Rundquist A and Vuckovic J 2013 Phys. Rev. Lett. 111 027402

[7] Antón M A, Carreño F, Melle S, Calderón O G, Cabrera-Granado E and Singh M R 2013 Phys. Rev. B 87 195303

[8] Carreño F, Arrieta-Yáñez F, and Antón M A 2015 Opt. Commun. 343 97

[9] Paspalakis E, Economou S E, and Carreño F 2019 J. Appl. Phys. 125 024305

[10] Stefanatos D, Karanikolas V, Iliopoulos N and Paspalakis E 2020 Physica E 117 113810

[11] Stefanatos D, Karanikolas V, Iliopoulos N and Paspalakis E 2020 Physica E 118 113935

[12] Stefanatos D, Karanikolas V, Iliopoulos N and Paspalakis E 2020 Appl. Sci. 10 1001

[13] DiVincenzo D P 2000 Fortsch. Phys. 48 771

[14] Manzeli S, Ovchinnikov D, Pasquier D, Yazyev O V and Kis A 2017 Nat. Rev. Materials 2 17033

[15] Karanikolas V D, Marocico C A, Eastham P R and Bradley A L 2016 Phys. Rev. B 94 195418

[16] Karanikolas V, Thanopulos I and Paspalakis E 2019 Opt. Lett. 44 2049

[17] Liu H, Wang T, Wang C, Liu D and Luo J-B 2019 J. Chem. Phys. C 123 10087

[18] Hanson G W 2008 J. Appl. Phys. 103 064302

[19] Li Y, Chernikov A, Zhang X, Rigosi A, Hill H M, van der Zande A M, Chenet D A, Shih E-M, Hone J and Heinz T F 2014 Phys. Rev. B 90 205422

[20] Gonçalves P A D, Bertelsen L P, Xiao S and Mortensen N A 2018 Phys. Rev. B 97 041402

[21] Karanikolas V, Thanopulos I and Paspalakis E 2019 Opt. Lett. 44 2049