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Quantum Optical Control of Donor-bound Electron Spins in GaAs

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Chapter 4

Optical control of three-level systems: Theory

In this chapter I will present a general approach to the mathematical treatment of three-level systems, based on the density matrix formalism. I will focus on the calculation of the optical spectra in the steady state regime, when optical excitations are provided by continuous wave lasers (CW), and explain how it is used to explain the experiment on electromagnetically induced transparency (EIT). Based on the equation of motion of the density matrix I will also calculate the time evolution of the spin ensemble during and after the resonant excitation by a fast optical pulse.

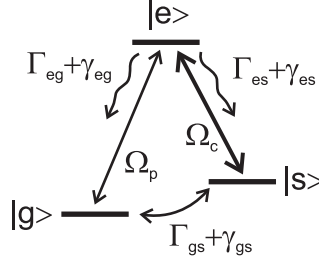


Figure 4.1: Model 3-level system. Two optical fields: probe and control are tuned to excite transitions from the ground $|g\rangle$ to the excited $|e\rangle$ and from the spin-flip $|s\rangle$ to the excited $|e\rangle$ state. Γ_{ij} and γ_{ij} are corresponding population relaxation and dephasing rates.

4.1 Introduction

The three-level system is a key object in the field of quantum electrodynamics. Typically the three-level system consists of a long living ground state $|g\rangle$, long-living "spin-flip" state $|s\rangle$ and an excited state $|e\rangle$, see Fig. 4.1. The crucial property of the system is that quantum coherence between states $|g\rangle$ and $|s\rangle$ is preserved for a time that is long enough on a timescale needed for spin manipulation. In the case of the Λ -system, which will be considered throughout this thesis, optical transitions are only allowed from the ground to the excited state ($|g\rangle \rightarrow |e\rangle$ transition) and from the spin-flip to the excited state ($|s\rangle \rightarrow |e\rangle$ transition). Provided that the coherence time of the ground and the spin-flip state of such a system is long enough, any superpositions and coherent populations transfers from the ground to the spin-flip state are theoretically achievable via designed optical manipulation of this system.

To describe how optical fields interact with a model three-level system, we will turn our attention to the technique involving the density matrix formalism. We are interested in the response of the system to excitations provided by two optical fields, where one field (control field) will be treated exactly, while the other field (probe field) will be calculated only to the first order approximation. We will show how the spectral response of the three-level system changes according to the ratio between the strength of the optical excitation and the rate of the relaxation and decoherence processes in the system.

4.2 Density matrix and equation of motion

The concept of the density matrix is based on the idea that any quantum mechanical system can be described not in terms of its wave function, but rather by using an operator describing both the population of each eigenstate and the correlation between each pair of eigenstates, known as coherence. For the three-level system, the used density matrix operator will be a 3-by-3 matrix:

$$\hat{\rho} = \begin{pmatrix} \rho_{gg} & \rho_{gs} & \rho_{ge} \\ \rho_{sg} & \rho_{ss} & \rho_{se} \\ \rho_{eg} & \rho_{es} & \rho_{ee} \end{pmatrix}, \quad (4.1)$$

where the diagonal elements are the populations of the ground, spin-flip and excited states respectively, while off-diagonal elements are coherences.

The density matrix operator obeys the operators equation of motion with relaxation term:

$$\frac{d\rho}{dt} = -\frac{i}{\hbar} [\hat{H}_{total}, \hat{\rho}] + \hat{L}(\hat{\rho})_{relax}, \quad (4.2)$$

where the first term describes the dynamics of the density matrix operator driven by \hat{H}_{total} - a complete Hamiltonian of the system, while the second term of this equation is a superoperator, which describes all the relaxation and decoherence processes.

The total Hamiltonian consists of the Hamiltonian of the bare three-level system \hat{H}_0 and the interaction Hamiltonian \hat{H}_{int} . The three-level system Hamiltonian is, as expected, just a 3-by-3 matrix with diagonal elements being the energy of the ground, spin-flip and excited state, while the off-diagonal elements are equal to zero.

The interaction Hamiltonian consists of the elements that describe the interaction of the optical fields with the system. We will treat the optical fields classically and define the control field ϵ_c and the probe field ϵ_p as the product of the time periodic functions and the complex amplitudes:

$$\begin{aligned} \epsilon_c(t) &= \frac{1}{2} E_c e^{-i\omega_c t} + c.c. \\ \epsilon_p(t) &= \frac{1}{2} E_p e^{-i\omega_p t} + c.c. \end{aligned} \quad (4.3)$$

Here E_c and E_p are the complex amplitudes of the control and probe fields and *c.c.* stands for complex conjugate.

If we consider only dipole optical transitions in the system, the interaction Hamiltonian \hat{V} is written as:

$$\hat{V} = -\hat{\mu}\vec{\epsilon}(t) = -\hat{\mu}[\vec{\epsilon}_c(t) + \vec{\epsilon}_p(t)], \quad (4.4)$$

where $\hat{\mu}$ is the matrix dipole operator of the three-level system. Assuming only optical transitions from the ground to the excited and from the spin-flip to the excited state are possible, we can write the dipole matrix operator as:

$$\hat{\mu} = \begin{pmatrix} 0 & 0 & \mu_{ge} \\ 0 & 0 & \mu_{se} \\ \mu_{eg} & \mu_{es} & 0 \end{pmatrix} \quad (4.5)$$

Combining all these operators together, the total Hamiltonian can be written as:

$$\hat{H}_{total} = \begin{pmatrix} \hbar\omega_g & 0 & V_{ge} \\ 0 & \hbar\omega_s & V_{se} \\ V_{eg} & V_{es} & \hbar\omega_e \end{pmatrix} \quad (4.6)$$

where V_{ij} are products of the optical fields with the corresponding dipole operators and $\hbar\omega_g, \hbar\omega_s, \hbar\omega_e$ are the energies of the ground, spin-flip and excited state respectively.

Now, after calculating all the necessary commutators and neglecting for a while the relaxation operator, we can write down the equation of motion for each component of the density matrix operator:

$$\begin{aligned} \frac{d\rho_{gg}}{dt} &= -\frac{i}{\hbar} [V_{ge}\rho_{eg} - \rho_{ge}V_{eg}] \\ \frac{d\rho_{ss}}{dt} &= -\frac{i}{\hbar} [V_{se}\rho_{es} - \rho_{se}V_{es}] \\ \frac{d\rho_{eg}}{dt} &= -\frac{i}{\hbar} [V_{eg}(\rho_{gg} - \rho_{ee}) + \rho_{eg}\hbar\omega_{eg} + V_{es}\rho_{sg}] \\ \frac{d\rho_{es}}{dt} &= -\frac{i}{\hbar} [V_{es}(\rho_{ss} - \rho_{ee}) + \rho_{es}\hbar\omega_{es} + V_{eg}\rho_{gs}] \\ \frac{d\rho_{sg}}{dt} &= -\frac{i}{\hbar} [\rho_{sg}\hbar\omega_{sg} + V_{se}\rho_{eg} - \rho_{se}V_{eg}] \end{aligned}$$

4.3 Rotating frame transformation and rotating wave approximation (RWA)

Since off-diagonal elements of the density matrix operator are fast time varying functions, it is more easy to search for the solution of the equation by applying a rotation frame transformation. This is done by writing each component of the density matrix as the product of a slow and a fast time varying functions:

$$\begin{aligned}\rho_{eg} &= \sigma_{eg} e^{-i\omega_p t} \\ \rho_{es} &= \sigma_{es} e^{-i\omega_c t} \\ \rho_{sg} &= \sigma_{sg} e^{-i(\omega_p - \omega_c) t}\end{aligned}$$

Here σ_{eg} , σ_{es} , σ_{sg} are slowly varying amplitudes and ω_p, ω_c are the frequencies of the probe and the control field respectively. Diagonal elements of the density matrix operator do not have any fast time varying part and therefore remain the same.

We will also write down explicit expressions for the components V_{ij} of the interaction Hamiltonian in terms of the product of the dipole matrix operator and the optical field:

$$\hat{V}(t)_{ij} = -\hat{\mu}_{ij} \left[\frac{1}{2} E_c e^{-i\omega_c t} + c.c. + \frac{1}{2} E_p e^{-i\omega_p t} + c.c. \right] \quad (4.7)$$

After substituting explicit form of the interaction Hamiltonian it is easy to see that many time dependencies are canceling each other, while those that are left can be neglected as soon as the following condition is satisfied:

$$|\omega_p - \omega_c| \ll \omega_p, \omega_c \quad (4.8)$$

This is the essence of the rotation wave approximation (RWA) - it neglects contributions from off-resonance terms. In addition to that we will introduce Rabi frequencies for the probe and control field, which represent the strength of the interaction between the optical fields and the three-level system:

$$\begin{aligned}\Omega_c &= \mu_{es} E_c / \hbar \\ \Omega_p &= \mu_{eg} E_p / \hbar\end{aligned}$$

Having done all this and after introducing the frequency detuning of the probe and control fields with corresponding optical transitions: $\Delta_c = \omega_c - \omega_{es}$ - the control laser single photon detuning, $\Delta_p = \omega_p - \omega_{eg}$ - the probe laser single photon

detuning and $\delta = \Delta_p - \Delta_c$ - the two photon detuning, we obtain the following set of equations:

$$\begin{aligned}
 \frac{d\rho_{gg}}{dt} &= -\frac{1}{2}\Omega_p^*\sigma_{eg} + \frac{1}{2}\Omega_p\sigma_{eg}^* \\
 \frac{d\rho_{ss}}{dt} &= -\frac{1}{2}\Omega_c^*\sigma_{es} + \frac{1}{2}\Omega_c\sigma_{es}^* \\
 \frac{d\sigma_{eg}}{dt} &= -\Delta_p\sigma_{eg} - \frac{1}{2}\Omega_p(\rho_{gg} - \rho_{ee}) - \frac{1}{2}\Omega_c\sigma_{sg} \\
 \frac{d\sigma_{es}}{dt} &= -\Delta_c\sigma_{es} - \frac{1}{2}\Omega_c(\rho_{ss} - \rho_{ee}) - \frac{1}{2}\Omega_p\sigma_{sg}^* \\
 \frac{d\sigma_{sg}}{dt} &= -\delta\sigma_{sg} - \frac{1}{2}\Omega_c^*\sigma_{eg} + \frac{1}{2}\Omega_p\sigma_{es}^*
 \end{aligned}$$

4.4 Lindblad relaxation operator

Now we will introduce the relaxation Hamiltonian $\hat{L}(\rho)$ where we will account for all possible relaxations and dephasing processes in the system:

$$\hat{L}(\hat{\rho}) = \left(\begin{array}{l}
 -\Gamma_{gs}\rho_{gg} + \Gamma_{sg}\rho_{ss} + \Gamma_{eg}\rho_{ee} \\
 -\left(\frac{\Gamma_{gs} + \Gamma_{sg}}{2} + \gamma_s\right)\rho_{sg} \\
 -\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \Gamma_{es}}{2} + \gamma_{ea,eb}\right)\rho_{eg} \\
 -\left(\frac{\Gamma_{gs} + \Gamma_{sg}}{2} + \gamma_s\right)\rho_{gs} \\
 \Gamma_{gs}\rho_{gg} - \Gamma_{sg}\rho_{ss} + \Gamma_{es}\rho_{ee} \\
 -\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \Gamma_{es}}{2} + \gamma_{ea,eb}\right)\rho_{eg} \\
 -\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \Gamma_{es}}{2} + \gamma_{ea,eb}\right)\rho_{ge} \\
 -\left(\frac{\Gamma_{sg} + \Gamma_{eg} + \Gamma_{es}}{2} + \gamma_{ea,eb}\right)\rho_{se} \\
 -(\Gamma_{eg} + \Gamma_{es})\rho_{ee}
 \end{array} \right) \quad (4.9)$$

To see how this Hamiltonian describes relaxations it is instructive to see how it appears for different populations and coherences.

For populations it takes the following form:

$$\frac{d\rho_{gg}}{dt} = -\Gamma_{gs}\rho_{gg} + \Gamma_{sg}\rho_{ss} + \Gamma_{eg}\rho_{ee}, \quad (4.10)$$

which indicates that the population in the ground state can change (increase) due to the relaxation of the excited state $\Gamma_{eg}\rho_{ee}$ and due to competing relaxation-excitation processes between ground and spin flip state $-\Gamma_{gs}\rho_{gg} + \Gamma_{sg}\rho_{ss}$.

The same can be written for spin-flip state:

$$\frac{d\rho_{ss}}{dt} = \Gamma_{gs}\rho_{gg} - \Gamma_{sg}\rho_{ss} + \Gamma_{es}\rho_{ee} \quad (4.11)$$

Relaxation of the excited state population can be also derived from the first principle, but also from the property of the density matrix which implies that for any closed system the trace of the density matrix must be equal to unity.

For coherences:

$$\frac{d\sigma_{eg}}{dt} = -\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \Gamma_{es}}{2} + \gamma_{ea,eb}\right)\rho_{eg} \quad (4.12)$$

Coherence ρ_{eg} is lost due to direct dephasing of the excited eg states $\gamma_{ea,eb}$ and due to population relaxations of ground and excited states. Same arguments hold for the rest of coherences.

The last step is to make the following substitutions: for populations we can write that $\rho_{ee} = W - \rho_{gg} - \rho_{ss}$, where $W = 1$ in most of the cases, but keeping this in a general form we can use it later for describing dynamics of any open systems, where relaxations can happen outside the three-level system. We will introduce new complex frequency detuning that include the relaxations terms: $\tilde{\Delta}_c = \Delta_c + i\left(\frac{\Gamma_{sg} + \Gamma_{eg} + \Gamma_{es}}{2} + \gamma_{ea,eb}\right)$ - control laser single photon detuning, $\tilde{\Delta}_p = \Delta_p + i\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \Gamma_{es}}{2} + \gamma_{ea,eb}\right)$ - probe laser single photon detuning, $\tilde{\delta} = \delta + i\left(\frac{\Gamma_{gs} + \Gamma_{sg}}{2} + \gamma_s\right)$ - two photon detuning.

The resulting set of algebraic equations reads as follows:

$$\begin{aligned}
i\Gamma_{eg}W &= -\frac{1}{2}\Omega_p^*\sigma_{eg} + \frac{1}{2}\Omega_p\sigma_{eg}^* - i(\Gamma_{gs} + \Gamma_{eg})\rho_{gg} - i(\Gamma_{eg} - \Gamma_{sg})\rho_{ss} \\
i\Gamma_{es}W &= -\frac{1}{2}\Omega_c^*\sigma_{es} + \frac{1}{2}\Omega_c\sigma_{es}^* - i(\Gamma_{sg} + \Gamma_{es})\rho_{ss} - i(\Gamma_{es} - \Gamma_{gs})\rho_{gg} \\
-\frac{1}{2}\Omega_p W &= -\tilde{\Delta}_p\sigma_{eg} - \frac{1}{2}\Omega_p(2\rho_{gg} + \rho_{ss}) - \frac{1}{2}\Omega_c\sigma_{sg} \\
-\frac{1}{2}\Omega_c W &= -\tilde{\Delta}_c\sigma_{es} - \frac{1}{2}\Omega_c(2\rho_{ss} + \rho_{gg}) - \frac{1}{2}\Omega_p\sigma_{sg}^* \\
0 &= -\tilde{\delta}\sigma_{sg} - \frac{1}{2}\Omega_c^*\sigma_{eg} + \frac{1}{2}\Omega_p\sigma_{es}^* \\
-\frac{1}{2}\Omega_p^* W &= -\tilde{\Delta}_p^*\sigma_{eg}^* - \frac{1}{2}\Omega_p^*(2\rho_{gg} + \rho_{ss}) - \frac{1}{2}\Omega_c^*\sigma_{sg}^* \\
-\frac{1}{2}\Omega_c^* W &= -\tilde{\Delta}_c^*\sigma_{es}^* - \frac{1}{2}\Omega_c^*(2\rho_{ss} + \rho_{gg}) - \frac{1}{2}\Omega_p^*\sigma_{sg} \\
0 &= -\tilde{\delta}^*\sigma_{sg}^* - \frac{1}{2}\Omega_c\sigma_{eg}^* + \frac{1}{2}\Omega_p^*\sigma_{es}
\end{aligned} \tag{4.13}$$

4.5 Analytical solution in no-population transfer approximation

This set of equations can be significantly simplified if we assume that no population transfer happens in the system. This is for example the case in systems where initially all the population is trapped in the ground state and the probe field is much weaker than the control field. Mathematically it can be described by the following inequalities: $\Omega_p \ll \Omega_c$, $\rho_{gg} \simeq 1 = W$, $\rho_{ss} \simeq \rho_{ee} \simeq 0$. When we also neglect all the terms nonlinear in Ω_p we obtain the following solution for σ_{eg} :

$$\sigma_{eg} = \frac{\frac{1}{2}\Omega_p}{\left(-\tilde{\Delta}_p + \frac{|\frac{1}{2}\Omega_c|^2}{\tilde{\delta}}\right)} \tag{4.14}$$

If we will set $\Omega_c = 0$ we obtain the expected solution for the two-level system, where the absorption spectra follows a Lorentzian profile.

4.6 Full solution of the density matrix equation

The 8-by-8 matrix equation 4.13 can be solved analytically, but the result is quite bulky and therefore we will solve it numerically on the desired frequency domain.

Before we proceed to the result, it is instructive to explain how to calculate photoluminescence excitation and absorption spectra of the system using the density matrix.

Optical photoluminescence in the three-level system can occur from the excited state and therefore is proportional to the population ρ_{ee} of this state.

On the other hand, to obtain absorption spectra we have to focus on the off-diagonal elements of the density matrix. First, we have to apply the inverse rotating frame transformation:

$$\rho_{eg} = \sigma_{eg} e^{-i\omega_p t} \quad (4.15)$$

Then we calculate expectation value of the dipole matrix operator by using the following relation:

$$\langle \hat{\mu}(t) \rangle = \text{tr} [\hat{\rho}(t) \hat{\mu}(t)] \quad (4.16)$$

Explicitly it is written in the following form:

$$\langle \hat{\mu}(t) \rangle = \text{tr} (\hat{\rho}(t) \hat{\mu}(t)) = \rho_{eg} \mu_{ge} + \rho_{es} \mu_{se} + c.c. \quad (4.17)$$

This expression gives us the value of the expectation value of the electric dipole of a single oscillator. To obtain the full polarization $\vec{P}(\omega_p)$ of the media we have to multiply it by the concentration of the dipoles. By using the general expression of the polarizability of the media, we can calculate the electric susceptibility $\chi(\omega)$:

$$\vec{P}(\omega) = N \langle \hat{\mu}(t) \rangle = \varepsilon_0 \chi(\omega_p) \vec{E}(\omega_p) \quad (4.18)$$

$$\chi(\omega_p) = \frac{2N}{\varepsilon_0 \vec{E}(\omega_p)} \langle \hat{\mu}(\omega_p) \rangle = \frac{2N \mu_{ge}}{\varepsilon_0 \vec{E}(\omega_p)} \sigma_{eg}(\omega_p) = \frac{2N |\mu_{ge}|^2}{\varepsilon_0 \hbar \Omega_p} \sigma_{eg}(\omega_p) \quad (4.19)$$

Note that we got rid of time dependence since the coherence of the density matrix is following the same time evolution as the probe field. We also neglected the effect of the ρ_{es} coherence, since it follows from the RWA, or in other words, it is off-resonant with the probe field.

Now we can calculate the absorption coefficient as:

$$\alpha(\omega_p) = \frac{\omega_p}{c} \text{Im} [\chi(\omega_p)] \quad (4.20)$$

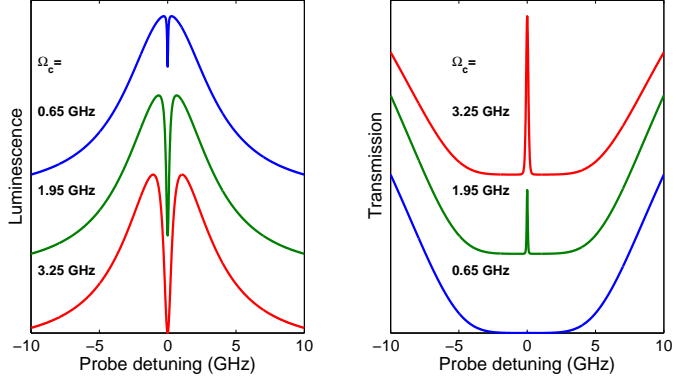


Figure 4.2: Calculated luminescence and transmission spectra of the three level system with interaction parameters close to those previously determined in experiments with donor bound excitons in GaAs [1].

4.7 Optical spectra of n -GaAs in a strong optical driving regime

To demonstrate how to use the method described above we calculate the luminescence and the absorption spectra of the system of donor bound electron spins ensemble in lightly doped GaAs, for which we assume the parameters following [2].

The population relaxation rate from the spin-flip to the ground state $\Gamma_{sg} = (2.6 \mu s)^{-1}$, and is mostly conducted by emitting a phonon. Therefore the inverse process is just the thermal excitation from the ground to the spin-flip state and its rate has to be weighed by the Boltzman factor $\Gamma_{gs} = \Gamma_{sg} e^{-\frac{E_{sg}}{kT}}$.

The radiative population relaxation from the excited to the spin-flip state $\Gamma_{es} = (1 ns)^{-1}$, while radiative relaxation into the ground state $\Gamma_{eg} = 0.08\Gamma_{es}$. Because of the large energy separation we neglect thermal excitations to the excited state and set $\Gamma_{se} = \Gamma_{ge} = 0$.

The pure dephasing rate between the ground and the spin-flip state γ_s is difficult to guess, therefore we use a combined lower level dephasing rate $\gamma_{gs} = \frac{1}{2}(\Gamma_{gs} + \Gamma_{sg}) + \gamma_s = (1.7 ns)^{-1}$ This is the value which has to be used during complex two-photon detuning.

Dephasing γ_e apparently depends strongly on the presence of the control field (probably due to a temperature effect). Therefore we define two dephasing values

$\gamma_{sa} = 4.6 \text{ GHz}$ (no field) and $\gamma_{sb} = 22 \text{ GHz}$ (with control field present). For Rabi frequency of the control field we will use $\Omega_c = 650 \text{ MHz}$. The strength of the probe field is $\Omega_p = 16 \text{ MHz}$.

Results of the spectra calculated using the density matrix equation for the case when control field is exactly on resonance is plotted on Fig. 4.2 and, as was expected, a dip in the luminescence appears and grows larger with increasing Ω_c . This effect is called coherent population trapping (CPT) and the dip in luminescence is attributed to the population being trapped in a coherent superposition of the ground and the spin-flip state, often called a dark state.

In the transmission spectra an increase in the transmission, when the probe and control fields are on resonance, is called electromagnetically induced transparency (EIT). While being a complementary effect to CPT it is usually described as a consequence of destructive interference of the transition amplitudes induced by the control and probe field on the excited state.

More detailed analysis of the properties of EIT spectra as function of the detuning and the strength of the driving field can be found elsewhere [3–5]

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