Electromagnetically induced transparency with electron spins bound to neutral Si donors in GaAs

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Abstract

In this thesis a demonstration of electromagnetically induced transparency (EIT) with an ensemble of donor-bound electrons is presented. Gallium arsenide doped with Silicon was used and measurements were performed at liquid Helium temperature in the presence of a magnetic field.

First magneto-photoluminescence and transmission experiments were performed to identify the optical transitions between the spin states of an electron bound to a neutral donor ($D_0^+$) to the states of an exciton bound to a neutral donor ($D_0^+X$). Using pump-assisted spectroscopy the Λ-system, that is based on the Zeeman split electron spin states of $D_0^+$ and the ground state of $D_0^+X$, was identified. This system was then used to demonstrate EIT as function of the optical detuning and intensity of the control field. Optical driving of the system up to $\Omega_c \approx 16$ GHz is shown and it was found that the electron spin dephasing time $T_2^* \approx 2$ ns was limited by hyperfine coupling to nuclear spins.

Reducing these nuclear spin fluctuations will therefore be the main aim of future research.
Chapter 1

Introduction

Electromagnetically induced transparency (EIT) is the phenomenon that a medium, that is otherwise opaque, can become transparent under the application of an optical field. For this a so-called three-level Λ-system is needed, see Fig. 1.1. The system containing one electron consists of a ground state $|g\rangle$ and a metastable state $|s\rangle$ that are both coupled by fast optical transitions to a common short-lived excited state $|e\rangle$. When an optical field is applied to one of the transitions, absorption, like in any two-level system, will occur. However when a second optical field is applied such that both fields are equally detuned from the excited state (a condition known as two-photon resonance), an increase in transparency is observed. The difference with the nonlinear effect of Autler-Townes splitting [1] is that it relies on driving of a system which possesses a long coherence time.

![Figure 1.1: Λ-system used for EIT. The two lowest states $|g\rangle$ and $|s\rangle$ are connected through the excited state $|e\rangle$ and two applied fields $E_1$ and $E_2$. EIT occurs when the two fields are equally detuned from resonance by $\Delta$.](image)

Optical non-linearities, which occur under EIT conditions, reduce the group velocity of light propagating through the medium. Slowdown by up to 7 orders of magnitude has been reported [2][3]. This phenomenon known as 'slow light’ can be used to store and retrieve light pulses and is the basis for a quantum memory application [4][5][6]. Long distance quantum communication protocols such as the DLCZ-scheme also rely on Λ-systems [7][8][9][10][11]. The two lowest states can be used to store a quantum state, and fast optical transitions to the excited state make fast coherent preparation, manipulation and coupling between quantum states possible. Demonstration of EIT in a Λ-system is a prerequisite for the implementation.
Coherent population trapping (CPT) of an electron in the two lowest states, a precursor to EIT, was first demonstrated by Alzette et al. in 1976 using Natrium vapors [12]. The first experimental demonstration of EIT was performed in Strontium vapor by Boller et al. in 1991 [13]. We use a solid state system since this offers key advantages above vapors for future applications, such as the possibility of fabrication in optical fibers and a larger scalability.

Gallium arsenide (GaAs) doped with Silicon (Si) is used, where the two ground states are formed by the Zeeman split spin states of an electron bound to a neutral donor (D⁰). The short lived excited state is formed by an exciton bound to a neutral donor (D⁰X, also called trion state). Lightly doped Si:GaAs is used, such that the different donor sites are well separated and show little inhomogeneity, but the medium still inhibits a high optical density. It has been shown that CPT in such a system is possible [14][15] and optical control of the spin state has been demonstrated [16][17]. Our goal is now a demonstration of EIT.

Photoluminescence and transmission spectroscopy experiments were performed at liquid Helium temperature on a 10 \( \mu \)m thick Si:GaAs sample with dopant concentration \( N_{Si} = 3 \times 10^{13} \) cm\(^{-3}\). The D⁰-D⁰X transitions were identified. Using pump-assisted single laser spectroscopy it was possible to discriminate the D⁰-D⁰X transitions that form \( \Lambda \)-systems. EIT was then demonstrated as function of the control field detuning and power. It was possible to verify theoretically expected behavior.
Chapter 2

Experimental setup

2.1 Photoluminescence and transmission experiments

A schematic drawing of the experimental setup is shown in Fig. 2.1. It is designed to perform two kinds of experiments. In photoluminescence experiments excitation light is shone on the sample and the photoluminescent light is collected by the spectrometer. In transmission experiments transmitted light from one or two lasers is collected by a photodiode behind the sample. See text for more detailed description.
2.1.1 Photoluminescence experiment

In photoluminescence experiments the sample is optically excited and the photoluminescent light is collected and analyzed using the spectrometer. Laser light from a CW Coherent MBR-110 Ti:sapphire laser is tuned to a typical wavelength $\lambda = 805$ nm, since the sample is then excited across the bandgap of GaAs. The exact wavelength is measured using the wavelength meter (MK Photonics, Lambdameter L-M-007, resolution $10^{-5}$ nm). The light intensity is set on the optical table, then it is coupled into the polarization maintaining single mode fiber (Shafter and Kirchoff, PANDA type, $NA = 0.13$, mode-field diameter $5 \mu m$ and cut-off wavelength $700$ nm) and brought to the sample. The sample is located inside a He bath cryostat in the presence of a magnetic field (superconducting magnet, inner Bohr radius 60 mm, Cryogenics Ltd.).

Light leaving the fiber is focussed on the sample making use of a optical microscope in a confocal geometry. First the propagation direction is diverted by a dielectric prism mirror (Thorlabs MRA05-E02), such that the $\vec{k}$-vector of the light is orthogonal to the direction of the magnetic field (Voigt geometry) and no Faraday rotation occurs in the optical elements. An aspheric lens (Thorlabs 350430, focal distance $f = 5.0$ mm, numerical aperture $NA = 0.15$ and clear aperture $CA = 1.5$ mm) is used to first collimate the light, then the light is focussed on the sample by a second aspheric lens (Thorlabs 350140, $f = 1.45$ mm, $NA = 0.55$ mm, $CA = 1.5$ mm). The prism mirror and the two lenses are located inside a DuPont Vespel-SP1 housing.

The sampleholder with the sample can be moved using a stack of three Attocube slip-stick piezo-motors (ANP101, 5 mm range in all three directions), such that different parts of the sample can be addressed and the sample surface can be placed in or out of focus (move it along $y$-axis, see Fig. 2.1). When in focus the amount of reflected light that is coupled back into the fiber is maximum (confocal geometry), and the amount of collected photoluminescence is maximum. Using a fiber beam splitter (OZ optics) the photoluminescence light is separated and after passing through a bandpass filter (cutoff wavelength $\lambda_{co} = 820$ nm, $\Delta \lambda = 10$ nm), the light is analyzed using a spectrometer (Acton 750i monochromator equipped with nitrogen cooled CCD camera, resolution 0.03 nm).

2.1.2 Transmission experiments

In transmission experiments the transmission of one or two lasers through the sample is studied. The light intensity and linear polarization of the CW laser(s) is set on the optical table and coupled into the fiber. In order to apply a lock-in detection scheme, light is modulated using a chopper (Optical Chopper, Thorlabs, MC 1000 A) at $\sim 6$ kHz. In the case of two laser experiments one laser beam is modulated such that a distinction can be made between an AC and a DC signal component of the collected light. The intensity of the beam(s) is measured using a photodetector (Thorlabs PDA36A-EC, Si Amplified detector) that is connected to the beam splitter. This signal is also used to correct for (fast) time and wavelength dependent fluctuations in the intensity of the lasers.

Light is polarized linearly and coupled into the fiber according to one of the two polarization axis of the fiber. When it leaves the fiber it has a linear polarization which is, after the mirror, either orthogonal (H-polarization) or parallel (V-polarization) to the applied magnetic field. H-polarized light is seen at the sample as a superposition of $\sigma^+$ and $\sigma^-$ polarization and can induce transitions in the sample with a change of $\pm 1$ angular momentum, while
V-polarized light is equivalent to $\pi$ polarized light and does not change the angular momentum. For the photoluminescence experiments the used polarization for excitation was found to make no difference.

Transmitted light through the sample is collected by a photodiode (Hamamatsu, pin, 5106) behind the sample. The signal from the photodiode is pre-amplified (DL Instruments 1211, current pre-amplifier), and electrically filtered by a high-pass and a low-pass filter (cutoff frequencies $f_{hp} = 3$ kHz, $f_{lp} = 10$ kHz) and amplified (Stanford Research Systems, SR560, low-noise amplifier). The signal is then analyzed with a lock-in amplifier (SRS, SR830 DSP Lockin Amplifier) which uses the signal from the chopper as a reference signal. Since the used lasers have a linewidth of 1 MHz, corresponding with $2 \times 10^{-6}$ nm, the resolution in the transmission experiments is determined by the wavelength meter ($10^{-5}$ nm).

Transmission experiments are typically performed with the sample surface out of focus in order to prevent standing waves between the sample surface and the tip of the fiber. A detailed analysis of the dependence of the spotsize on the amount of defocusing was performed and is presented in Appendix A.

2.2 Sample and preparation

The sample that is studied is GaAs doped with Si that was epitaxially grown on a GaAs substrate by the group of D. Reuter and A. Wieck from the University of Bochum. Multiple samples with different donor concentrations were grown. The donor concentration was determined by measuring the concentration of charges present, which is equal to the concentration of donors. The lowest doped sample that was used in the experiments however has a charge concentration too small to be measurable. The second lowest doped sample has a concentration $N_{Si} = 1 \times 10^{14}$ cm$^{-3}$. In transmission experiments it was seen that the lowest doped sample is about half as absorbing as the second lowest doped sample, and also fitting experimental results to a model suggest that $N_{Si} \simeq 3 \times 10^{13}$ cm$^{-3}$ for the lowest doped sample (see Chapter 5).

The 10 $\mu$m thick Si:GaAs film was removed from the substrate using an epitaxial lift-off (ELO) process and transferred to a wedged sapphire substrate [18] to which it is fixed by Van der Waals forces. The sapphire is a good thermal conductor and is in turn mounted on a copper cold finger, such that good heat sinking is assured. The dimensions of the sample are typically on the order of 1 mm, and multiple samples can be placed next to each other and analyzed sequentially.
A sample of Si doped GaAs was used for experiments. In this chapter first a brief overview is given of neutral donor bound electrons (D\textsuperscript{0}), excitons (X) and neutral donor bound excitons (D\textsuperscript{0}X), and how they are influenced by a magnetic field. Magneto-photoluminescence spectroscopy (MPL) was used to learn more about these systems and other transitions that occur just below the bandgap of GaAs. Single laser spectroscopy (SLS) gives the most accurate information about the D\textsuperscript{0}.D\textsuperscript{0}X transitions, and using the so-called pump-probe technique the Λ-system is identified that is used for further EIT experiments in Chapter 5.

3.1 Electrons bound to neutral donors

When describing the donor bound electron (D\textsuperscript{0}) system not only the electron and the Si\textsuperscript{+} atom should be taken into account, but also all of the lattice atoms. A simplification of this problem is provided by the effective mass theory (EMT). The total wavefunction \(\psi(r)\) is by described by the product of a Bloch wavefunction \(\phi(r)\) and a slowing varying EMT envelope function \(F(r)\) [19]. Assuming the impurity Coulomb potential field is sufficiently weak compared to the lattice potential, the wavefunction \(\phi(r)\) will resemble the Bloch wavefunction of unperturbed electrons at the conduction band minimum. The envelope function describing the electron wavefunction satisfies the hydrogen effective mass equation:

\[
\left(-\frac{\hbar^2 \nabla^2}{2m^*_e} - \frac{e^2}{4\pi\varepsilon_0\varepsilon_r r}\right)F(r) = EF(r),
\]

where \(m^*_e = 0.067m_e\) [20] is the effective electron mass in GaAs, \(m_e\) is the electron mass, \(e\) is the elementary charge, \(\varepsilon_0\) is the vacuum permittivity constant and \(\varepsilon_r = 12.56\) [21] is the relative permittivity. The system forms Hydrogen like energy levels, where the energy of the \(n\)\textsuperscript{th} level relative to the ionization limit is given by:

\[
E_n = \frac{m^*_e}{m_e} \frac{1}{\varepsilon_r^2} \frac{R_H}{n^2},
\]

where \(R_H\) is the Rydberg constant of the hydrogen atom (13.6 eV). The \(n = 1\) groundstate forms a 1s like orbital and has an energy \(E_1 = 5.8\) meV below the bandgap of GaAs \((E_g\)
1.519 eV at 0 K \(^{[22]}\)). To prevent thermal ionization of the ground state, the sample has to be cooled below \(T \simeq \frac{E_n}{k_B} = 67\) K, where \(k_B\) is the Boltzmann constant.

The Bohr radius \(r_n\) of the \(n\)th level can be approximated by:

\[
r_n = \frac{m_e}{m_e^*} \varepsilon r_n^2 a_H,
\]

where \(a_H = 5.29177 \times 10^{-11}\) m is the Bohr radius of the hydrogen atom. This gives \(r_1 = 9.9\) nm. The first excited (\(n=2\)) state has a binding energy of 1.4 meV below the bandgap and a Bohr radius \(r_2 = 396\) Å. This state (labeled as the two-electron satellite (TES) state by Kai-Mei Fu \(^{[15]}\)) has an energy difference, corresponding with a difference in the absorption wavelength of the D\(^0\)-D\(^0\)X transition, \(\Delta \lambda = 2\) nm with respect to the ground state.

Since GaAs has a lattice constant of \(l_{\text{GaAs}} = 0.564\) nm (at \(T = 300\) K) \(^{[23]}\) and contains two atoms per unit cell, a sphere with radius \(r_1\) is spread out over \(\sim 5 \times 10^4\) nuclear atoms.

In case of a fully random distribution in space of the donors, the average nearest-neighbor distance \(r_{n.n.} = 0.55 \rho^{-1/3}\) \(^{[24]}\), where \(\rho\) is the concentration. As in our lowest doped sample the donor concentration was estimated to be \(\rho = 3 \times 10^{13}\) cm\(^{-3}\), corresponding to \(r_{n.n.} = 177\) nm, about 18 times the Bohr radius.

### 3.1.1 Donors in a magnetic field

When no magnetic field is applied, the \(n=1, 1s\) ground state is two-fold degenerate due to the spin of the electron. When a magnetic field \(B\) is applied this degeneracy is lifted and a two level system is formed. The energy of the spin up state \(|\uparrow\rangle\) is lower than the spin down state \(|\downarrow\rangle\) by an amount of:

\[
\Delta E = g_e \mu_B B,
\]

where \(g_e = -0.44 \pm 0.02\) \(^{[27]}\) is the bound electron g-factor and \(\mu_B\) is the Bohr magneton. For \(B = 9\) T (the maximum field that we can reach with our magnet), \(\Delta E = 0.23\) meV, corresponding to a splitting of \(\Delta \lambda = 1.2\) Å. It would require a temperature below \(T \simeq \Delta E/k_B = 2.7\) K, to thermally depopulate the spin down state \(|\downarrow\rangle\). Since we operate at the temperature of liquid Helium \(T = 4.2\) K and both states are populated in about a 2:1 ratio.

### 3.2 Excitons

A free exciton (X) can be described as a weakly bounded electron-hole pair. Using the weighted average of the heavy and light hole effective masses in GaAs \(m_h = 0.2 m_e\), it follows that the reduced electron-hole mass \(\mu = (1/m_e^* + 1/m_h)^{-1} = 0.05 m_e\) \(^{[26]}\). Using Eqn. 3.2 and Eqn. 3.3 it follows that the \(n = 1\) exciton has a binding energy \(E_X = 4.3\) meV and Bohr radius \(r_X = 13.3\) nm. The energy to excite a free exciton is therefore the bandgap energy \(E_g\) minus \(E_X\), corresponding to \(\lambda_X \simeq 818\) nm.

### 3.2.1 Excitons in a magnetic field

When a magnetic field is applied to an exciton, the wavefunction gets perturbed and the binding energy decreases \(^{[26]}\), meaning the energy needed to excite an exciton increases. In weak fields the magnetic field is treated as a perturbation to the unperturbed exciton. This
3.3 Excitons bound to neutral donors

leads to a diamagnetic shift that scales with the magnetic field as:

$$\Delta E_d = +\frac{e^2}{12\mu r_X^2}B^2$$  \hspace{1cm} (3.5)

For the above given values this leads to an energy shift $\Delta E_d \sim 0.052$ meV/T².

For strong fields, the interaction between holes and electrons with the field is stronger than their mutual Coulomb interaction. The electrons and holes form Landau levels to which the Coulomb interaction acts as a perturbation. Since the Landau levels are also diamagnetic, the energy shift is now given by the cyclotron energy:

$$\Delta E_c = +\frac{\hbar e}{\mu}B$$  \hspace{1cm} (3.6)

This leads to an energy shift $\Delta E_c \sim 2.3$ meV/T. The transition between the weak and strong field occurs when the cyclotron energy $\Delta E_c$ is larger than the binding energy ($E_X = 4.3$ meV), so the crossover occurs around $B = 2$ T.

The average shift in diamagnetic energy of the exciton can be described using these two equations. A more detailed description would also have to take into account the difference between light and heavy hole masses and the Zeeman effect for light/heavy holes and electrons (that all have different g-factors). A complete picture of the energy shift of excitons is therefore rather complicated and beyond the scope of this thesis.

3.3 Excitons bound to neutral donors

The neutral donor complex forms an attractive potential to which an exciton can be bound. A four body complex is formed, consisting of the Si⁺⁺ atom, two electrons in a singlet state and a hole. This neutral donor bound exciton complex (D₀X) was first predicted by Lampert [27] and experimentally observed by Haynes [28]. The energy to excite the D₀ to the D₀X is equal to the energy to excite an exciton minus the binding energy of the exciton to D₀. The binding energy for the exciton ground state to the D₀ ground state at $B = 0$ T is approximately 0.9 meV (corresponding to a difference between the exciton and D₀-D₀X excitation wavelength $\Delta \lambda = 0.5$ nm) [29] [30].

3.3.1 Excitons bound to neutral donors in a magnetic field

Since the energy to excite an exciton increases with the magnetic field, the energy of the D₀-D₀X transition increases at higher magnetic field. Also Zeeman splitting occurs, fully determined by the spin of the $J = 3/2$ hole, since the electrons form a singlet state. Each D₀X level therefore splits into 4 Zeeman levels. A schematic picture of the two lowest D₀X levels that together with the two 1s D₀ groundstates form a Λ-system, is shown in Fig. 3.3 a) on page 13.

Since experiments are done at a fixed magnetic field, the shift in energy of the X and D₀-D₀X transitions are of no direct importance. They can however be of interest to determine the magnetic field that was applied in an indirect way. The shift in energy of the X transition follows about the diamagnetic shift $\Delta E \sim 0.052$ meV/T² and the shift of the D₀-D₀X transitions is about $\Delta E \sim 0.028$ meV/T² [14]. Since the Zeeman splitting between the groundstates can
be calculated using Eqn. 3.4, the strength of the magnetic field can be confirmed in three independent ways. This was found to be very useful when one time the superconducting magnet was probably broken.

### 3.4 Magneto-photoluminescence spectroscopy

Magneto-photoluminescence (MPL) spectroscopy was used in order to study the behavior of optical transitions, related to excitons, donor bound excitons and other systems, just below the bandgap of GaAs in presence of a magnetic field. The sample was excited across the bandgap and the photoluminescence was measured as function of the magnetic field. The result is shown in Fig. 3.1. Many peaks are visible and a general trend is seen of peaks moving to higher energy as function of the magnetic field. The exciton peak is easily identified since it has the highest energy. It splits into multiple peaks due to different amounts of diamagnetic shift for light/heavy holes and Zeeman splitting between excitons with different total spin. The D⁰X-D⁰ transitions (labeled as D⁰X) are visible, as well as transitions that have a slightly higher energy due to the orbital angular momentum \( L = 1, 2, \ldots \) of the exciton. A comparison with earlier reported measurements [14] shows that the transitions with a lower energy than the D⁰X transitions, are not TES-lines (transitions from D⁰X to D⁰ TES-states), as they are separated by less than 2 nm from D⁰X. They are most likely due to transitions from excitons bound to ionized Silicon donors (D⁺X-D⁺) or acceptors (A⁰X-A⁰) such as Carbon, that can be present in the sample as impurities.
3.5 Single laser spectroscopy

In order to study the transitions more accurately single laser spectroscopy (SLS) was performed. The resolution is limited by the wavelength meter ($10^{-4}$ Å) and is better than of the spectrometer (0.3 Å). Also one of the disadvantages of photoluminescence is that it probes mostly the surface of the sample, where the depletion layer is present.

SLS has been performed for different magnetic fields, see Fig. 3.2. Two different polarizations have been used, corresponding with $\pi$ polarization (V-polarization), that doesn’t induce any change in angular momentum, and $\sigma$ polarization (H-polarization), that does induce a change in angular momentum $\pm 1$. For energies larger than the bandgap all light is absorbed and the transmission is zero. The free exciton $X$ and $D^0D^0X$ transitions. (*) The value of the magnetic fields has been corrected since we were dealing with a broken superconducting magnet.

![Figure 3.2: Single laser spectroscopy performed for different magnetic fields. The transmission is measured, and clear absorption peaks can be seen for the free exciton ($X$) and $D^0$. $D^0X$ transitions. (*) The value of the magnetic fields has been corrected since we were dealing with a broken superconducting magnet.](image)
Fabry-Perot interference inside the sample. This means the transmission is highest (lowest) when constructive (destructive) interference occurs inside the sample and the spacing between peaks $\Delta \lambda$ is given by:

$$\Delta \lambda \approx \frac{\lambda^2}{2nl},$$

where $n$ is the refractive index of the sample and $l$ is the thickness. The chirped wavelength dependency is due to the wavelength dependent refractive index around the strong absorption of the exciton transition.

### 3.6 Pump-assisted SLS

Using SLS the $D^0$-$D^0X$ transitions can be determined, but some are only weakly absorbing or are difficult to observe since the probing itself causes optical pumping, that bleaches the transition. Pump-assisted SLS is an useful method to increase the absorbance of these transitions and confirm which transitions form a $\Lambda$-system.

When an electron is optically excited from the ground (metastable) state to an excited state, it decays to the ground or metastable state typically at a rate $\Gamma \sim 1(\text{ns})^{-1}$ \textsuperscript{[15]}. The ratio of electrons that decay to the ground state to those that decay to the metastable state depends on the ratio between the dipole moments $\mu_{ge}: \mu_{se}$, where $\mu_{ge}$ ($\mu_{se}$) is the dipole moment of the $|g(s)\rangle \leftrightarrow |e\rangle$ transition. When $\mu_{ge} < \mu_{se}$ ($\mu_{se} < \mu_{ge}$) most electrons decay to the metastable (ground) state. The relaxation rate between the two lowest states (typically $\Gamma \sim 1(\mu s)^{-1}$ \textsuperscript{[15]}) will then limit the optical re-excitation, and thus the absorbance.

When a second ‘pump’ laser is fixed on a transition from the metastable (ground) state to an excited state, the electron is optically excited typically at a rate $\Omega \sim 1$ GHz. By relaxation from this excited state the electron is ‘pumped back’ to the ground (metastable) state at a faster rate than the relaxation rate between the two lowest states alone. Therefore the absorbance of transitions from the ground (metastable) state to excited states increases. The absorbance of the transitions from the metastable (ground) state to an excited state decreases.

Transitions that form a $\Lambda$-system can be identified since they are separated in energy by the Zeeman splitting (Eqn. 3.4), and also pumping one transition increases the absorbance of the other. In Fig. 3.3 the two $\Lambda$-systems that have the lowest excited states are identified. The transitions $A$ and $A^*$ are used for EIT experiments in Chapter 5.
3.6. Pump-assisted SLS

Figure 3.3: a) In the presence of a magnetic field $B = 6.4$ T, the lowest $D^0$ 1s state splits into a spin-up and a spin-down state due to Zeeman splitting. Also the lowest (L=0) state of the $D^0X$ complex splits into four levels. The transitions to the lowest excited state are labeled $A/A^*$, the ones to the second lowest excited state $B/B^*$. b) Pump and probe technique applied. Pumping in the $A$ ($A^*$) transition enhances absorption of the probe in the $A^*$ and $B^*$ ($A$ and $B$) transitions, a confirmation of the scheme in a). When the $A$ transition was pumped no other transitions on the red side of it appeared, confirming it’s the lowest energy $D^0$-$D^0X$ transition. For the EIT experiments we drive the $A$ and $A^*$ transitions.
CHAPTER 3. Characterization of the $D^0$-$D^0_X A$-system
Chapter 4

Introduction to electromagnetically induced transparency

Using the Λ-systems that was identified using pump-assisted SLS (see previous chapter), it is possible to perform EIT experiments (see next chapter). In this chapter a theoretic model is presented how optical fields interact with a Λ-system, and the physical principle behind coherent population trapping and EIT is explained. A model to calculate the transmission of the probe laser in EIT experiments is discussed and some examples of calculated transmission scans are shown.

4.1 Theoretical description of a Λ-system

4.1.1 Density matrix and equation of motion

A schematic drawing of two optical fields applied to a model Λ-system is shown in Fig. 4.1. The Λ-system consists of the ground state \( |g\rangle \), the long living and highly coherent metastable state \( |s\rangle \) and the excited state \( |e\rangle \). The system can be described by the density-matrix:

\[
\hat{\rho} = \begin{pmatrix}
\rho_{gg} & \rho_{gs} & \rho_{ge} \\
\rho_{sg} & \rho_{ss} & \rho_{se} \\
\rho_{eg} & \rho_{es} & \rho_{ss}
\end{pmatrix}
\]

(4.1)

The diagonal elements describe the population in the ground, metastable or excited state, while the off-diagonal components represent the correlation (or coherence) between wave-functions of each pair of states. Since we are looking for steady-state solutions, we are looking for a solution of the equation:

\[
\frac{\partial \hat{\rho}}{\partial t} = -\frac{i}{\hbar} [\hat{H}_{\text{total}}, \hat{\rho}] + L(\hat{\rho}) = 0,
\]

(4.2)

where \( \hat{H}_{\text{total}} \) is the Hamiltonian of the system and \( L(\hat{\rho}) \) is the Linbladian operator describing the relaxation and decoherence processes in the system. The total Hamiltonian \( \hat{H}_{\text{total}} \) consists of the bare Λ-system Hamiltonian \( \hat{H}_0 \) and the interaction Hamiltonian \( \hat{H}_{\text{int}} \). \( \hat{H}_0 \) is given by:

\[
\hat{H}_0 = \begin{pmatrix}
\hbar \omega_g & 0 & 0 \\
0 & \hbar \omega_s & 0 \\
0 & 0 & \hbar \omega_e
\end{pmatrix}
\]

(4.3)
Figure 4.1: Schematic drawing of the model Λ-system. The system consists of three states \( |g\rangle, |s\rangle, |e\rangle \) with energies \( \hbar \omega_g, \hbar \omega_s, \hbar \omega_e \). A probe (control) field is applied with frequency \( \omega_p, \omega_c \) and Rabi frequency (driving strength) \( \Omega_p, \Omega_c \). The population relaxation rates are denoted by \( \Gamma \), the decoherence rates by \( \gamma \). Because of large energy separation and low temperature, no relaxation is assumed from the ground states \( |g\rangle \) or \( |s\rangle \) to the excited state \( |e\rangle \).

where \( \hbar \omega_g, \hbar \omega_s, \hbar \omega_e \) is the energy of an electron in state \( |g\rangle \) (|s\rangle, |e\rangle). \( \hat{H}_{int} \) is given by:

\[
\hat{H}_{int} = -\hat{\mu} E(t),
\]

where \( \hat{\mu} \) is the dipole operator and \( E(t) \) describes the optical fields. Since optical fields can only induce transitions between \( |g\rangle \) or \( |s\rangle \) and \( |e\rangle \), \( \hat{\mu} \) is given by:

\[
\hat{\mu} = \begin{pmatrix}
0 & 0 & \mu_{ge} \\
0 & 0 & \mu_{se} \\
\mu_{eg} & \mu_{es} & 0
\end{pmatrix}
\]

\( E(t) \) is given by:

\[
E(t) = \frac{1}{2} E_c e^{-i \omega_c t} + \frac{1}{2} E_p e^{-i \omega_p t} + c.c.
\]

Here \( E_c \) and \( E_p \) are the amplitudes of the control and probe electric fields and c.c. stands for complex conjugate. The Rabi frequency is defined as the strength of the interaction of the optical field with the system:

\[
\Omega_c = \frac{\mu_{es} E_c}{\hbar}
\]

\[
\Omega_p = \frac{\mu_{eg} E_p}{\hbar}
\]

To remove the time dependence and simplify the result, a rotating frame transformation is applied. The off-resonant terms are neglected which is called the rotating wave approximation.
4.1. Theoretical description of a Λ-system

(RWA). The off-diagonal components of the density matrix can be described as the product of a slow and a fast varying component:

\[ \rho_{eg} = \sigma_{eg} e^{-i\omega_p t} \]  \hspace{1cm} (4.9)  
\[ \rho_{es} = \sigma_{es} e^{-i\omega_c t} \]  \hspace{1cm} (4.10)  
\[ \rho_{sg} = \sigma_{sg} e^{-i(\omega_p - \omega_c) t} \]  \hspace{1cm} (4.11)  

where \( \sigma_{eg}, \sigma_{es}, \sigma_{sg} \) represent the slowly varying amplitudes. To simplify the result, \( \rho_{eg} (\rho_{es}, \rho_{sg}) \) and c.c. are replaced by \( \sigma_{ge} (\sigma_{es}, \sigma_{sg}) \) and c.c. to obtain the density matrix \( \hat{\rho}' \):

\[ \hat{\rho}' = \begin{pmatrix} \rho_{gg} & \sigma_{sg}^* & \sigma_{eg}^* \\ \sigma_{sg} & \rho_{ss} & \sigma_{es}^* \\ \sigma_{eg} & \sigma_{es} & \rho_{ss} \end{pmatrix} \]  \hspace{1cm} (4.12)  

The equation of motion of the density matrix \( \hat{\rho}' \) is now given by

\[ \frac{\partial \hat{\rho}'}{\partial t} = -\frac{i}{\hbar} [\hat{H}'_{\text{total}}, \hat{\rho}'] + L(\hat{\rho}') = 0, \]  \hspace{1cm} (4.13)  

where the Hamiltonian is now given by:

\[ \hat{H}'_{\text{total}} = \hbar \begin{pmatrix} 0 & 0 & 0 \\ 0 & \omega_s - \omega_g - (\omega_p - \omega_c) & -\Omega_p^r \\ \Omega_p^r & -\Omega_c^r & \omega_e - \omega_g - \omega_p \end{pmatrix} \]  \hspace{1cm} (4.14)  

4.1.2 Coherent population trapping and EIT

In order to get some insight in the physical principle behind coherent population trapping and EIT, \( \hat{H}'_{\text{total}} \) is evaluated when the fields are applied in resonance with the transitions (so \( \omega_p = \omega_e - \omega_g \) and \( \omega_c = \omega_e - \omega_s \)):

\[ \hat{H}'_{\text{total}} = \hbar \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \Omega_p^r \\ \Omega_p^r & \Omega_c^r & 0 \end{pmatrix} \]  \hspace{1cm} (4.15)  

This system has the following eigenstates:

\[ \lambda_{\text{dark}} = 0, |\Psi\rangle = \frac{1}{\sqrt{[\Omega_p^r]^2 + [\Omega_c^r]^2}} (\Omega_e^r, -\Omega_p^r, 0) \]  \hspace{1cm} (4.16)  

\[ \lambda_a = -\sqrt{[\Omega_p^r]^2 + [\Omega_c^r]^2}, |\Psi\rangle = \frac{1}{\sqrt{[\Omega_p^r]^2 + [\Omega_c^r]^2}} (\Omega_c^r, \Omega_p^r, -\sqrt{[\Omega_p^r]^2 + [\Omega_c^r]^2}) \]  \hspace{1cm} (4.17)  

\[ \lambda_b = \sqrt{[\Omega_p^r]^2 + [\Omega_c^r]^2}, |\Psi\rangle = \frac{1}{\sqrt{[\Omega_p^r]^2 + [\Omega_c^r]^2}} (\Omega_c^r, \Omega_p^r, \sqrt{[\Omega_p^r]^2 + [\Omega_c^r]^2}) \]  \hspace{1cm} (4.18)  

It is now seen that a 'dark state' eigenstate (Eqn. 4.16) is obtained, which surprisingly does not contain any components of the excited state. So even though the system is resonantly driven on both \( |g\rangle \rightarrow |e\rangle \) and \( |s\rangle \rightarrow |e\rangle \) transitions, there is an eigenstate that is the superposition of only the two lower states. Destructive quantum interference thus prevents population of the excited state.
The name 'dark state' arose from photoluminescence experiments, since the photoluminescent light from the excited state to lower energy states, is proportional to the population in the excited state. The photoluminescence therefore decreases when the system is coherently trapped. Destructive interference also prevents population from the groundstates to get transferred to the excited state. This means the absorption decreases, resulting in an increase of the transmission.

When also relaxation and decoherence is taken into account, it is found that such a 'dark state' still occurs, but only if the system is sufficiently driven such that \( \Omega_c \geq \sqrt{\gamma_c^* \gamma_s} \), where \( \gamma_s \) is the decoherence rate of the excited state and \( \gamma_c \) is the decoherence rate between the ground states. Also the system has to be driven in the so-called two-photon resonance (TPR) condition: \( \omega_p - \omega_{eg} = \omega_c - \omega_{es} \). When \( \omega_{p,\text{res}} \) satisfies this TPR condition, EIT occurs when \( \omega_{p,\text{res}} - \Omega_c/2 \leq \omega_p \leq \omega_{p,\text{res}} + \Omega_c/2 \).

Examples of simulated probe transmission spectra are shown in Fig. 4.2. When the TPR condition is satisfied an increase in transmission, on top of a Lorentzian absorption dip, is observed. The influence of \( \Omega_c \) and \( T_2^* \) is also illustrated.

### 4.1.3 Relaxation and decoherence operator

The relaxation and decoherence operator \( \hat{L}(\tilde{\rho}) \) is given by:

\[
\hat{L}(\tilde{\rho}) = \begin{pmatrix}
-\Gamma_{gs}\rho_{gg} + \Gamma_{sg}\rho_{ss} + \Gamma_{eg}\rho_{ee} & -\left(\frac{\Gamma_{gs} + \Gamma_{eg}}{2}\right) + \gamma_s\rho_{gs} & -\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \gamma_{es}}{2}\right) + \gamma_l\rho_{ge} \\
-\left(\frac{\Gamma_{gs} + \Gamma_{eg}}{2}\right) + \gamma_s\rho_{sg} & -\Gamma_{gg}\rho_{gg} + \Gamma_{ss}\rho_{ss} + \Gamma_{ee}\rho_{ee} & -\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \gamma_{es}}{2}\right) + \gamma_l\rho_{es} \\
-\left(\frac{\Gamma_{gs} + \Gamma_{eg}}{2}\right) + \gamma_s\rho_{eg} & -\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \gamma_{es}}{2}\right) + \gamma_l\rho_{es} & -\left(\frac{\Gamma_{eg} + \gamma_{es}}{2}\right) + \gamma_l\rho_{ee}
\end{pmatrix},
\]

(4.19)

where \( \Gamma_{sg} \) (\( \Gamma_{gs} = \Gamma_{sg} e^{\hbar (\omega_c - \omega_g)/kT} \) is weighted by the Boltzmann factor) is the longitudinal relaxation rate from \( |g\rangle \rightarrow |s\rangle \) (\( |s\rangle \rightarrow |g\rangle \)), \( \Gamma_{eg} \) (\( \Gamma_{es} \)) is the radiative relaxation from \( |e\rangle \rightarrow |g\rangle \) (\( |e\rangle \rightarrow |s\rangle \)), \( \gamma_l \) is the level \( |e\rangle \) (which is also called the trion-state) dephasing rate and \( \gamma_s \) is the transverse relaxation rate between \( |g\rangle \) and \( |s\rangle \). Since the off-diagonal components \( \rho_{eg} \) (\( \rho_{es} \), \( \rho_{sg} \)) are replaced by \( \sigma_{eg} \) (\( \sigma_{es} \), \( \sigma_{sg} \)), the following equations are used to obtain \( \hat{L}(\tilde{\rho}) \) (which follow from Eqn. 4.9, 4.10 and 4.11):

\[
\frac{d\sigma_{eg}}{dt} = -i\frac{\hbar}{\lambda} [\hat{H}_{\text{total}}, \tilde{\rho}]_{eg} + L(\rho_{eg}) e^{i\omega_p t}
\]

(4.20)

\[
\frac{d\sigma_{es}}{dt} = -i\frac{\hbar}{\lambda} [\hat{H}_{\text{total}}, \tilde{\rho}]_{es} + L(\rho_{es}) e^{i\omega_p t}
\]

(4.21)

\[
\frac{d\sigma_{sg}}{dt} = -i\frac{\hbar}{\lambda} [\hat{H}_{\text{total}}, \tilde{\rho}]_{sg} + L(\rho_{sg}) e^{i(\omega_p - \omega_l) t}
\]

(4.22)

### 4.1.4 Final result

In order to simplify the result the following substitutions are made: \( \omega_{eg} = \omega_e - \omega_g \), \( \omega_{es} = \omega_e - \omega_s \), \( \omega_{sg} = \omega_s - \omega_g \), complex control laser single photon detuning \( \Delta_c = \omega_c - \omega_{es} + i\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \gamma_s}{2} \right) + \gamma_l \), complex probe laser single photon detuning \( \Delta_p = \omega_p - \omega_{eg} + i\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \gamma_s}{2} \right) + \gamma_l \) and the complex two photon detuning \( \delta = \omega_p - \omega_{eg} - (\omega_c - \omega_{es}) + i\left(\frac{\Gamma_{gs} + \Gamma_{eg} + \gamma_s}{2} \right) \). Finally the total population \( W \) of the system is given by: \( \rho_{gg} + \rho_{ss} + \rho_{ee} = W \), where \( W = 1 \) is the case when
4.2 The transmission of the probe laser

In order to calculate the transmission of the probe laser, it is necessary to calculate the absorption coefficient. First the expectation value of the dipole matrix operator is found using:

$$\langle \hat{\mu}(t) \rangle = \text{tr}(\hat{\rho}(t)\hat{\mu}(t)) = \rho_{eg}\mu_{ge} + \rho_{es}\mu_{se} + \text{c.c.}$$ \hspace{1cm} (4.24)

This expression gives the value of the electric dipole of a single oscillator. The polarizability $\bar{P}(\omega)$ is found by multiplying this value with the total number of oscillators $N$. From the general expression for the polarizability it is then possible to calculate the electric susceptibility $\chi(\omega_p)$ with the RWA:

$$\bar{P}(\omega_p, t) = N \langle \hat{\mu}(t) \rangle = \varepsilon_o\chi(\omega_p)\bar{E}(\omega_p, t) = \varepsilon_o\chi(\omega_p)\frac{1}{2}E_p e^{-i\omega_p t}$$ \hspace{1cm} (4.25)

$$\chi(\omega_p) = \frac{N}{\varepsilon_0 \frac{1}{2} E_p} \sigma_{ge}(\omega_p) = \frac{N|\mu_{ge}|^2}{\varepsilon_0 \frac{1}{2} \Omega_p} \sigma_{eg}(\omega_p)$$ \hspace{1cm} (4.26)

The absorption coefficient is now calculated using:

$$\alpha(\omega_p) = \frac{\omega_p}{c} Im[\chi(\omega_p)]$$ \hspace{1cm} (4.27)

And the transmitted intensity $I(\omega_p)$ is given by:

$$I(\omega_p) = I_0 e^{-\alpha(\omega_p)L},$$ \hspace{1cm} (4.28)

where $I_0$ is the transmitted intensity without absorption and $L$ is the thickness of the sample.

4.2.1 Analytical solution of the no population transfer approximation

When the system is driven such that $\Omega_c \gg \Omega_p$, the population is approximately entirely trapped in the ground state $|g\rangle$, so $\rho_{gg} \simeq 1$ and $\rho_{ss} \simeq \rho_{ee} \simeq 0$. By neglecting all terms non-linear in $\Omega_p$, but taking all orders of $\Omega_c$, it is found that:

$$\sigma_{eg} = \frac{\Omega_p/2}{(-\tilde{\Delta}_p + \frac{\Omega_c/2\delta}{\delta})},$$ \hspace{1cm} (4.29)
Note, when taking $\Omega_c = 0$ or when the system is driven far from the TPR condition $\delta \gg |\Omega_c/2|^2$, the absorption coefficient $\alpha$ is given by:

$$\alpha \propto \frac{1}{1 + \left(\frac{\omega_p - \omega_{eg}}{\Gamma}\right)^2},$$

(4.30)

where $\Gamma = \frac{\Gamma_{gs} + \Gamma_{eg} + \Gamma_{es}}{2} + \gamma_t$. This is the result for a Lorentzian absorption profile with half-width at half-maximum equal to $\Gamma$, as one would also expect when driving a two-level system. Note also that when $\Omega_p \ll \Omega_c$, the absorption coefficient is no longer a function of $\Omega_p$.

### 4.3 Some properties of EIT

![Figure 4.2](image_url)

**Figure 4.2:** Examples of EIT in the transmission spectra when the probe is scanned along the $|g\rangle \leftrightarrow |e\rangle$ transition and a control field is applied at the $|s\rangle \leftrightarrow |e\rangle$ transition with a certain detuning $\Delta_c = \lambda_c - \lambda_{eg}$. $\gamma_t = 20$ GHz, $L = 10$ $\mu$m and every subsequent graph has an 0.5 offset. In a) and b) the strength of the control field $\Omega_c$ is varied while $\gamma_s = \frac{1}{T^*_2}$ is kept constant. In c) and d) $\Omega_c$ is fixed while $T^*_2$ is varied.
In Fig. 4.2 some examples of calculated EIT in transmission spectra is shown. The relaxation rates and dipole moments are taken such that they are equal to values earlier reported [14][31]. The decoherence rate $\gamma_t = 20 \text{ GHz}$ corresponds with what is found in the experimental results (see next chapter). $\Omega_c$ and $\gamma_s = \frac{1}{T_2^*}$ are varied for different detuning $\Delta_c = \lambda_c - \lambda_{res}$.

In Fig. 4.2 a) and b) it is seen that a pronounced EIT peak occurs when approximately $\Omega_c \geq \sqrt{\gamma_t \gamma_s}$. Increasing $\Omega_c$ increases the height of the EIT peak but also the distance between the minima in transmission. In Fig. 4.2 c) and d) $\Omega_c$ is kept constant while $\gamma_s$ is varied. Now it is seen that an increase in $T_2^*$ only induces an increase in the height of the EIT peak.
Chapter 5

Experimental demonstration of electromagnetically induced transparency

In chapter 3 it was demonstrated how a Λ-system was identified (see Fig. 2.3). This system was now used to perform EIT experiments. A control field was applied to the A transition, which is lowest in energy, in order to prevent pumping electrons to the second lowest excited state. A (much weaker) probe field was then scanned along the A* transition and the transmission is measured. The magnetic field \( B = 7.5 \) T and the temperature \( T = 4.2 \) K. EIT as function of the detuning and the power of the control field is presented. The experimental results were fitted to a model, this will first be discussed. The results correspond to the theoretically expected behavior or can be explained qualitatively. Finally some suggestions are made for future experiments.

5.1 Fitting

In the performed EIT experiments, the wavelength of the control laser \( \lambda_c \) is fixed while the probe laser (\( \lambda_p \)) is scanned along the A* transition. Using Eqn. 4.23 and Eqn. 4.28 the transmission as function of \( \lambda_p \) can be calculated using all the parameters of the three level system. Most of these parameters where taken from [15]: \( \Gamma_{sg} = 0.38 \mu s^{-1} \), \( \Gamma_{gs} = \Gamma_{eg} e^{\frac{-\hbar \omega_{eg}}{k_BT}} \approx 0.24 \mu s^{-1} \) (at \( T = 4.2 \) K and \( B = 6 \) T), \( \Gamma_{eg} = 0.08 \text{ ns}^{-1} \), \( \Gamma_{es} = 1 \text{ ns}^{-1} \), \( \mu_{ge} = 0.75 \) e nm and \( \mu_{se} = 0.07 \) e nm. \( \gamma_t = 4.6 - 22 \text{ GHz} \) was found to depend on the strength of the coupling field [15], so we use this as a fitting variable.

Therefore the values for \( I_0, \gamma_t, \gamma_s, \Omega_c, \omega_{eg}, \omega_{es} \) and \( \Gamma_{es} \) are now the variables while fitting. The value of \( \Omega_p \) is taken arbitrarily small, since when \( \Omega_p \ll \Omega_c \) the exact value does not matter for the transmission spectrum. Fitting was done making use of the self-made Matlab programme 'eit_gui.m', of which further details are provided in App. B.

5.1.1 Influence of spot intensity profile

For fitting is was assumed that the three-level system as in Fig. 4.1 is not coupled to other systems. Since every measurement point was taken during typically \( t_{meas} \sim \) ms, which is much longer than the relaxation and decoherence times, the steady-state solution was calculated
using Eqn. 4.21. The last assumption made during fitting is that the intensity of the control field on the sample $I \propto \Omega_c^2$ is constant. This is of course not the case since the laser spot on the sample follows a Gaussian intensity profile which is given by:

$$I \propto \Omega_{c,0}^2 (r) = 2\Omega_{c,0}^2 e^{-r^2/W^2},$$

where $\Omega_{c,0}$ is the Rabi frequency of the constant control field, $r$ is the distance from the center of the spot and $W$ is the spot size. The total power of the Gaussian spot is normalized such that it is equal to the power of a constant field $\Omega_{c,0}$ falling on a spot with radius $W$. A comparison between the two models is shown in Fig. 5.1.

$$\Delta c= 0 \text{ nm} \Omega_c=5 \text{ GHz}$$
$$\Delta c= -0.1 \text{ nm} \Omega_c=15 \text{ GHz}$$
$$\Delta c= -0.1 \text{ nm} \Omega_c=25 \text{ GHz}$$

**Figure 5.1:** Comparison between two models where $\Omega_c$ is kept constant and where $\Omega_c$ resembles a Gaussian beam shape (Eqn. 5.1). This done as function of the detuning of the control field ($\Delta_c$) and of $\Omega_c$. Only for detuning and $\Omega_c > 15$ GHz a clear difference is seen.

A difference is only clearly visible when the control field is detuned from resonance and $\Omega_{c,0} > 15$ GHz. Sharp 'EIT-features' then seem to get 'rounded off'. In the control field detuning experiments $\Omega_c < 10$ GHz. When the control field power was varied, $\Omega_c < 20$ GHz and the control field was set to zero detuning. The difference is smaller than the typical noise which was also present during experiments, so the model with constant $\Omega_c$ was used.

### 5.2 Control field detuning

In control field detuning experiments, the transmission spectrum is measured for different values of $\lambda_c$, see Fig. 5.2. The position of the EIT peak $\lambda_p - \lambda_{eg}$ is equal to the detuning of
5.3. Control field power variation

The control field $\lambda_c - \lambda_{es}$. It can be seen that the relative height of the EIT peak decreases towards red detuning. This can either be due to an increase of $\gamma_s$, a decrease of $\Omega_c$ or possibly both. In the fitting $\gamma_s = (2.2 \text{ ns})^{-1}$ was fixed and $\Omega_c$ was varied, see Fig.5.2 b). The inhomogeneous dephasing rate $\gamma_t = 17 - 30 \text{ GHz}$ was varied since a fit is was made to an asymmetric dipshape.

Figure 5.2: c) EIT spectra, where the transmission is measured as function of $\lambda_{p}$, for different values of $\lambda_c$. An offset is added to every trace. a) The position of the EIT peak $\lambda_p - \lambda_{cg}$ is equal to the detuning of the control field $\lambda_c - \lambda_{es}$. b) $\Omega_c^2$ decreases when tuning $\lambda_c$ from the blue to the red side of the A transition. The fitting was performed with $\gamma_s = (2.2 \text{ ns})^{-1}$, $\gamma_t = 17 - 30 \text{ GHz}$ and the parameters described in Section 5.1.
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Figure 5.3: c) EIT spectra were taken for different values of the control field intensity $I$, an offset is added to every trace. 

b) $\Omega_c$ obtained by fitting as function of $\sqrt{I}$. The values for $\sqrt{I} = 240 \text{ (nW)}^{1/2}$ and $\sqrt{I} = 290 \text{ (nW)}^{1/2}$ fall outside the graph. a) $T^*_{c2}$ obtained by fitting decreases when $I$ is increased. 

$\gamma_t = 14 - 24 \text{ GHz}$ and the parameters described in Section 5.1 are the other parameters that were used.

5.3 Control field power variation

Next the control field is tuned to $\lambda_c - \lambda_{es} \approx 0 \text{ nm}$ and EIT spectra are measured as function of the control field intensity $I$, see Fig. 5.3. The intensity was varied from low power, where EIT is just visible, to a high level where the height of the EIT-peak seems to no longer increase.
although the noise started to increase significantly. $\Omega_c$ used in fitting is plotted as function of $\sqrt{I}$. $T_2^*=1/\gamma_t$ is also plotted and it decreases for higher intensities.

5.4 Discussion

In the detuning experiments the power $I$ of the control field was kept constant but $\Omega_c^2 \propto I$ seems to decrease towards the red side of the $A$ transition. This can be due to Fabry-Perot interference in the sample, causing the transmission and thus the intensity in the sample to be higher at the blue side of the $A$ transition, rather than at the red side.

The electron spin dephasing time was determined to be $T_2^* \approx 2$ ns from fitting in the detuning experiment. This is very close to the expected value for dephasing by hyperfine coupling to fluctuating nuclear spins [16][14]. The fluctuation in the nuclear spins $\Delta B$ can be estimated from:

$$\Delta B = \frac{B_{\text{max}}}{\sqrt{N}},$$

where $B_{\text{max}} = 5.3$ T [32] is the field of total polarized nuclei and $N = 5 \times 10^4$ is the number of nuclei contained in the 99 \text{Å} Bohr radius of the D$^0$ electron. The fluctuation in the nuclear field is thus $\Delta B \approx 24$ mT. $T_2^*$ can then be estimated using:

$$T_2^* \approx \frac{h}{g\mu_B \Delta B}$$

This gives $T_2^* \approx 1$ ns. The slightly higher value we find can mean $\Delta B$ is slightly smaller than in this approximation.

$\gamma_t$ had to be varied while fitting because a fit had to be made to an asymmetric absorption shape. This asymmetry also arises from Fabry-Perot interference. The average value that was found $\gamma_t \approx 22$ GHz corresponds well with the value previously reported [15].

When increasing the control field power, for $\sqrt{I} = 25 - 49$ (nW)$^{1/2}$, $\Omega_c$ increases linearly. This is expected since $\Omega_c = \frac{\mu_a E}{h}$, where $E$ is electric field. For the lowest power an overestimation of $\Omega_c$ seems to occur. This can be due to the EIT peak having the same 'size' as noise fluctuations, making fitting inaccurate.

$\Omega_c$ determines how fast the system can be driven. The timescale $\tau$ that an electron can be carried from the $|s\rangle$ to the $|e\rangle$ state is given by:

$$\tau = \frac{2\pi}{\Omega_c}$$

Since the system was driven at a rate of at least $\Omega_c = 12$ GHz, $\tau \leq 0.5$ ns was achieved. For higher intensities however $\Omega_c$ seems to saturate around $\Omega_c = 16$ GHz. Also $T_2^*$ seems to decrease when the power is increased. The origin is unknown.

5.5 Future suggestions

In the detuning experiments, the EIT peak seems to decrease towards red detuning. To determine whether there is any dependency of $T_2^*$ on detuning, $\Omega_c$ will have to be kept constant. This can be easily done in future experiments since the transmission of the control field can be measured by analyzing the DC signal of the photodiode behind the sample as well. By keeping the transmission constant Fabry-Perot interference can be compensated.
To exclude heating of the sample as the reason why $\Omega_c$ seems to be limited, the sample can be given more time to cool down between each EIT measurement when a high control field power is used. Also the position where light is shone on the sample can be changed after every measurement.

$T_2^* \approx 2$ ns corresponds to a distance of only 60 cm that light can travel before coherence in the $\Lambda$-system is lost. To make long distance quantum communication possible, $T_2^*$ will have to be increased by reducing the nuclear field fluctuations. Fully polarizing all $5 \times 10^4$ nuclei to which electrons are coupled by means of cooling down will require temperatures below $T \approx \frac{g_n \mu_n B}{k_B \ln(5 \times 10^4)}$, where $\mu_n \approx \frac{\mu_B}{2000}$ is the nuclear magneton and $g_n$ the g-factor of nuclei. For $B = 9$ T and $g_n = 1$, this requires a temperature below $T = 0.5$ mK, which is experimentally challenging. However a different way to polarize nuclei is by dynamical nuclear polarization (DNP). DNP is the transfer of angular momentum from spins to lattice nuclei by means of the hyperfine interaction \cite{33,54,35,56}. Addressing DNP in order to reduce $\Delta B$ is a promising approach and must be attempted in future experiments.
Chapter 6

Conclusion

In this thesis a measurement setup is presented that was used for photoluminescence and transmission experiments. Measurements were done with a 10 µm thick Gallium Arsenide sample doped with Silicon ($N_{Si} = 3 \times 10^{13} \text{ cm}^{-3}$) at liquid Helium temperature in the presence of a magnetic field.

Magneto-photoluminescence spectroscopy and single-laser spectroscopy was performed in order to identify the transitions between the spin states of an electron bound to a neutral donor (D$^0$) and the states of an exciton bound to a neutral donor (D$^0$X). Using pump-assisted spectroscopy the Λ-system was identified that is based on the Zeeman split electron spin states of D$^0$ and the ground state of D$^0$X. Electromagnetically induced transparency (EIT) was then demonstrated as function of the optical detuning and intensity of the control field. A maximum driving by the control field $\Omega_c = 12 \text{ GHz}$ was achieved and it was found that the spin dephasing time $T_2^* \approx 2 \text{ ns}$ is limited by hyperfine coupling to a fluctuating nuclear field.

These results are a promising first step towards future applications, but an increase in $T_2^*$ will be required. Reducing nuclear field fluctuations will therefore be the main aim of future research in this field.
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Appendix A

Determining the spotsize

In this appendix first a short overview of the basic properties of Gaussian beams, lenses and optical fibers is given. The shape of the beam spot on the sample produced by the microscope was determined as function of the number of steps the sample is moved axially with respect to the light beam by the piezo attocube. The rate with which the attocube moves the sample laterally is determined to be $\sim 0.4 \, \mu m/\text{step}$ and the waist of the beam was determined to be $W_0 \sim 2 \, \mu m$ when the sample is placed in focus. This is larger than the $W_{0,\text{min}} = 0.7 \, \mu m$ spotsize that would be expected based on a model that takes into account the expected geometry of the fiber, dielectric mirror and the two lenses, indicating misalignment of the optical elements.

A.1 Gaussian beam optics

Although spatially confined light cannot travel through space without any angular spread, it can be confined in the form of beams. Waves whose wavefront diverges with a small angle $\theta_0$ with respect to the $z$-axis along which it travels are called paraxial waves. The Gaussian beam is the solution of the paraxial Helmholtz equation, that describes these kind of waves, see Fig. A.1. A good overview is provided by Saleh and Teich [37]. The intensity $I$ of the beam is concentrated along the beam axis and is a Gaussian function:

$$I(\rho, z) = I_0 \left( \frac{W_0}{W(z)} \right)^2 \exp \left(-2\rho^2/W(z)^2\right), \quad (A.1)$$

where $I_0$ is the intensity at the center of the beam, $\rho$ is the distance from the beam center and $W(z)$ is the width depending on the distance $z$ along which the beam travels. $W_0$ is the minimal beam waist when $z = 0$. The dependence of the Gaussian beam width $W(z)$ is given by the hyperbolic function:

$$W(z) = W_0 \sqrt{1 + \left( \frac{z}{z_0} \right)^2}, \quad (A.2)$$

$z_0$ is the depth of focus and is given by:

$$z_0 = \frac{\pi W_0^2}{\lambda}, \quad (A.3)$$

where $\lambda$ is the wavelength of the light. The wavefront of the beam is approximately planar near the beam waist, but ultimately becomes approximately spherical far from the waist. For
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Figure A.1: a) The beam intensity $I$ as function of the distance from the beam axis is a Gaussian function. b) The beam width $W$ as function of the distance $z$ from the beam waist $W_0$. c) A Gaussian beam falling upon a lens results into a modified Gaussian beam. (d) Only light incident within an acceptance angle $\theta_a$ can be guided through the fiber by internal reflections.

A given beam width $W_0$, the angular divergence angle $\theta_0$ with which the beam diverges far from the waist can be approximated by:

$$\tan(\theta_0) \approx \theta_0 \approx \frac{W(z)}{z} = \frac{W_0}{z_0} = \frac{\lambda}{\pi W_0}$$ (A.4)

Say a Gaussian beam falls upon a thin lens with a focal distance $f$. The distance of the waist of the beam to the lens is $z$ and the lens is placed perpendicular to the axis of the beam. The parameters $z'$, $z$ and $W'_0$ of the Gaussian beam that emerges from the lens can be calculated using:

$$M = \left| \frac{f}{z - f} \right| \frac{1}{\sqrt{1 + \left( \frac{z_0}{z - f} \right)^2}}$$ (A.5)

$$W'_0 = MW_0$$ (A.6)

$$z' = f + M^2(z - f)$$ (A.7)

$$z'_0 = M^2z_0$$ (A.8)

Here $M$ is the magnification. In the case of a collimated beam, $z = 0$ and $z_0 = \pi W_0^2/\lambda$, the result is a focussed beam with $z' = f$, $W'_0 = \lambda f/\pi W_0$ and $z'_0 = \lambda f^2/\pi W_0^2$. When light is
coupled into an optical fiber, only light incident within an acceptance angle $\theta_a$ with respect to the axis of the fiber, are guided by internal reflection through the fiber. $\theta_a$ is given by:

$$\theta_a = \sin^{-1}(NA),$$  \hspace{1cm} (A.9)

where NA is the numerical aperture, a property of the fiber. However, when light emerges from a fiber it emerges as a Gaussian beam with a beam waist equal to half the ‘mode field diameter’ (MFD) of the fiber and the divergence angle is calculated using Eqn. A.4.

### A.2 The optical microscope

![Figure A.2: Schematic drawing of the experimental setup used to measure the beam size.](image)

A schematic drawing of the experimental setup used to determine the spot size is shown in Fig. [A.2] A laser diode (Thorlabs LPS-785-FC) is used to shine light with a wavelength $\lambda = 795$ nm on the sample. Light is sent to the sample first using a single mode polarization maintaining fiber (Shafter and Kirchhoff, Panda type, NA = 0.13, MFD = 5 $\mu$m and cut-off wavelength 700 nm. A dielectric mirror (Thorlabs MRA05-E02) is used to reflect the light such that it propagates perpendicular to the magnetic field and no Faraday rotation will occur in the lenses. First an aspheric lens (Thorlabs 350430, NA = 0.15 and clear aperture CA = 1.5 mm) with a focal distance $f_1 = 5.0$ mm, is used to collimate the light. The second aspheric lens (Thorlabs 350140, NA = 0.55 and CA = 1.6 mm) with a focal distance $f_2 = 1.45$ mm
is used to focus the light on the sample. The distance between the lenses is assumed to be 1 \( \mu \)m.

When light emerges from the fiber, the spot size \( D \) when it falls on the first lens follows from:

\[
D = W(z = f_1) = W_0 \sqrt{1 + \left( \frac{f_1}{\pi W_0^2 / \lambda} \right)^2} \approx \frac{f_1 \lambda}{\pi W_0},
\]

where \( W_0 \) is equal to the MFD of the fiber and assuming the distance from the fiber to the lens is equal to \( f_1 \). For the used light source \( \lambda = 795 \) nm, so \( D = 253 \) \( \mu \)m. The depth of focus of the beam after the first lens \( z_0 = \lambda f_1^2 / \pi W_0^2 \), which gives \( z_0 = 250 \) mm. This is much larger than the distance between the lenses, so \( D \) can be assumed to remain constant between the two lenses. When light falls on the second lens it is now focussed to a spot size \( W_{0, \text{min}} \):

\[
W_{0, \text{min}} = \frac{\lambda f_2}{\pi D} = \frac{f_2}{f_1} W_0
\]

It follows that the theoretically smallest achievable spot size \( W_{0, \text{min}} = 0.725 \) \( \mu \)m.

Due to the confocal geometry, the light reflected on the sample is coupled back into the fiber and is collected on the optical table by a photodiode (Thorlabs 350430). Behind the sample transmitted light is collected using another photodiode (Hamamatsu, pin, 5106).

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![Figure A.3: The amount of reflected light collected as function of the number of steps the sample is moved away (positive value) or towards the microscope by the piezo attocube, see also [2]. The spotsize \( W \) at the sample is determined for different amounts of (de)focusing.](image)

When the sample is moved axially (y-axis, see Fig. A.2) the amount of reflected light coupled back into the fiber and collected by the photodiode is shown in Fig. A.3. The reflection profile reaches a maximum when the sample surface is in focus due to the confocal geometry. The reflection profile does not however show one clear maximum, but resembles the superposition of two Lorentzian functions. This is a possible indication of misalignment.
of the optical elements. To determine the radius or spotsize \( W \) of the beam for different amounts of defocusing the following method is used. First the sample is focused or defocused a certain amount of steps. Next the sample is moved laterally (x-axis) and the amount of light collected by the photodiode placed behind the sample is monitored, see Fig. A.3. When the spot is scanned a distance \( x_0 \) along the edge of the sample the amount of light \( T \) collected is:

\[
T = \int_{-\infty}^{x_0} \int_{-\infty}^\infty I_0 e^{\frac{-2(x^2 + y^2)}{W^2}} dxdy = I_0 \frac{\pi}{4} (1 + erf(\sqrt{\frac{2x_0}{W}})) \tag{A.12}
\]

\( x_0 \) can be easily determined since the size of the sample was measured to be 900 \( \mu m \). From this the distance per step the sample is moved by the piezo attocube is found to be \( \sim 0.4 \) \( \mu m/\)step. The derivative:

\[
\frac{dT}{dx} = I_0 \sqrt{\frac{\pi}{2}} W \exp\left(\frac{-2x_0^2}{W^2}\right) \tag{A.13}
\]

is thus a Gaussian function where \( x_0 \) is known and \( W \) is determined from fitting. This way \( W \) is determined for different amounts of defocusing, see Fig. A.3.

**Figure A.4:** The amount of transmission light collected by the photodiode placed behind the sample measured as function of the distance the sample is displaced horizontally by the piezo attocube. (Inset) By taking the derivative of the transmission at the edge, the width \( W \) of the spot can be easily determined.

### A.4 Discussion

In Fig. A.3 it can be clearly seen that the beam width is minimal when the sample is in focus, as is expected from the confocal geometry. The beam waist \( W_0 \sim 2 \mu m \) is however larger.
than the calculated minimal beam waist, $W_{0,\text{min}} = 0.73 \, \mu\text{m}$. Also the fact that we observe the superposition of two Lorentzian reflection profile instead of one indicates misalignment of the optical elements.
Appendix B

Matlab graphical user interface for fitting EIT curves

All fitting in this thesis was performed using the Matlab guided user interface (GUI) `eit_gui.m`. A screen shot of `eit_gui.m` is shown in Fig. B.1. In this appendix first the general principle behind fitting EIT curves is discussed. The function of all the different input fields is explained and a general strategy to fit any arbitrary curve is discussed.

Figure B.1: Screen shot of `eit_gui.m`. Blue is the raw data, the black lines are the values that correspond with 0% and 100% transmission and the red line is the generated fit. Since in this example the data is very asymmetric, it is only possible to obtain a good fit in the region around the 'EIT-peak', not over the entire interval.
B.1 General principle

First an EIT spectrum, in which the transmission intensity is given as function of the probe wavelength $\lambda_p$ and to which one wants to apply fitting, has to be loaded. As was discussed earlier in Chapter 4, the solution of the density matrix can be calculated by solving Eqn. 4.17. $\sigma_{eg}$ is thus calculated as function of the optical driving parameters of the control and probe field ($\Omega_c$, $\lambda_c$, $\Omega_p$, $\lambda_p$), the value of the two optical resonances ($\lambda_{eg}$, $\lambda_{es}$) and the value of the different relaxation ($\Gamma$) and decoherence ($\gamma$) rates in the $\Lambda$-system ($\Gamma_{eg}$, $\Gamma_{es}$, $\Gamma_{se}$, $\Gamma_{es}$, $\gamma_t$, $\gamma_s$). $\sigma_{eg}$ is calculated as function of $\lambda_p$ and fixing all other parameters. The absorption $\alpha$ is calculated using: $\alpha = \frac{2\pi}{\lambda_p} \text{Im} \left[ \frac{N|\mu_{ge}|^2}{\varepsilon_0 \bar{\hbar} \Omega_p} \sigma_{eg}(\lambda_p) \right]$, so also parameters $\mu_{ge}$ and concentration $N$ have to be defined. The transmission $T$ is now calculated using $T = e^{-\alpha L}$, where the length through the sample is assumed to be $L = 10\mu m$. Since $T$ is now a number between 0 and 1, corresponding with 0% and 100% transmission through the sample, also the values of the intensity corresponding with 0% and 100% transmission have to be defined. Now an EIT spectrum can be calculated as function of all the parameters and plotted by pressing the button 'Do EIT!'. The result can then be compared with the loaded file and the parameters can be adjusted in order to obtain a good fit.

B.2 The different panels

The interface of the program has been divided into separate panels.

B.2.1 File you want to upload

Here the name of the file (including the extension '.txt') containing the data to which one wants to obtain a fit has to be filled in. This file has to be located in the same folder as 'eit_gui.m' and has to contain the wavelength of the probe (in Å) in the first column and the intensity in the second.

B.2.2 Background (ax+b)

Here the value of intensity that correspond with 0% and 100% transmission are defined. The value for full transmission is calculated using:

$$T_{100\%} = T_{max} + A \times \frac{\lambda_p - \lambda_{p-start}}{\lambda_{p-end} - \lambda_{p-start}}$$  \hspace{1cm} (B.1)

$T_{max}$ is defined by 'Maximum transmission', $A$ is defined by 'Background Lin. Coeff.', $\lambda_{p-start}$ and $\lambda_{p-end}$ are defined in the box 'Wavelengths'. The value for no transmission $T_{0\%}$ is defined by 'Minimum transmission'. The value of $T_{100\%}$ and $T_{0\%}$ are also displayed in the graph as black lines.

B.2.3 Relaxation/Decoherence rates

Here all the values of the relaxation ($\Gamma$) and decoherence ($\gamma$) rates are defined.
B.3. Fitting strategies

B.2.4 Wavelengths

Here one fills in all the appropriate wavelengths. 'WL (E-G)' is the wavelength of the $|g\rangle \leftrightarrow |e\rangle$ resonance, 'WL (E-S)' of the $|g\rangle \leftrightarrow |e\rangle$ resonance. 'WL Control' is the wavelength of the control field, this wavelength is often measured during experiments. 'WL Probe-start' and 'WL Probe-end' define the interval over which the transmission intensity as function of the probe field is calculated ($\lambda_p^{\text{start}}$ and $\lambda_p^{\text{end}}$).

B.2.5 Other Parameters

Here the other necessary parameters are filled in. 'OmegaProbe' defines the Rabi frequency of the probe field, 'OmegaControl' of the control field. 'mu-ge' is the dipole moment of the $|g\rangle \leftrightarrow |e\rangle$ transition, 'mu-se' of the $|s\rangle \leftrightarrow |e\rangle$ transition. 'Concentration' defines the relative thickness of the sample compared to the assumed thickness of $L = 10\mu m$. However, changing this constant has the same influence as changing the value of the concentration or changing the value of the dipole moment $\mu_{ge}$.

B.2.6 Slider Panel

Using this panel the value of 'OmegaControl', 'GammaT' and 'GammaS' can be changed using a slider. When changing the value of the slider a new EIT spectrum is calculated instantaneously, this is easier and faster than having to type in a new value in one of the fields and pressing the 'Do EIT!' button. When selecting one of these three parameters the slider is automatically set such that 'left' equals zero and 'right' equals $5 \times$ the current value of that specific parameter. The slider can then be changed in steps of 5%. In order to change the slider in smaller steps, the 'left' and 'right' value of the slider can be redefined in the 'Min. Value' and 'Max. Value' fields and by pressing the 'Reset' button.

B.3 Fitting strategies

In order to reduce the time to obtain a good fit and to increase the accuracy, there are two special functions in the 'Fit GammaT' and 'Fit GammaS' panels that can be used. First, of course, it is important to try to already manually obtain a reasonable fit to the data. There are two main thoughts behind the use of the two automatic fitting functions. First, the shape of the overall Lorentzian-like transmission dip minus the region where the 'EIT-peak' occurs, depends mostly on $\gamma_t$, $\lambda_{EG}$, the 'Maximum transmission' and the 'Background Lin. Coeff.' Second, the shape and position of the actual 'EIT-peak', depends on $\gamma_s$, $\Omega_c$ and $\lambda_{es}$. This way, these variables can be all be determined separately.

B.3.1 Fit GammaT

First two intervals have to be defined that are going to be used for fitting: from 'Point1' to 'Point2' and from 'Point3' to 'Point4'. These two intervals should include the general transmission dip, but exclude the region around the actual 'EIT-peak'. The datapoints corresponding to these two intervals can be viewed using the 'Plot' function. When pressing 'Fit', automatically a fit will be created to the selected datapoints, where $\gamma_t$, $\lambda_{eg}$, the 'Maximum transmission' and the 'Background Lin. Coeff.' are the only variables. The result will appear in a separate window. In practice, also $\Omega_c$ affects the 'dip shape', so a good estimate has to
be made in advance, but $\Omega_c$ can be most accurately determined by looking at the 'EIT-peak' itself. However, it was also seen empirically that when $\Omega_c \geq 5GH z$, $\Omega_c$ does not affect the overall Lorentzian-like dipshape anymore.

**B.3.2 Fit GammaS**

Now only one interval has to be defined from 'Point1' to 'Point2'. This interval should include the 'EIT-peak' and can be viewed when pressing 'Plot'. Now when pressing 'Fit', Matlab will make a best fit by only varying $\gamma_s$, $\Omega_c$ and $\lambda_{es}$. This result will also appear in a separate window.

**B.3.3 Final remarks**

In practice it is found useful to repeat the 'Fit GammaT' and 'Fit GammaS' functions multiple times. Also, once a reasonable fit has been obtained, it is found useful to increase the intervals over which these two functions try to make a fit, and to also include the 'EIT-peak' in the 'Fit GammaT' function. It is also found that when $\Omega_p \ll \Omega_c$ the absolute value of $\Omega_p$ is insignificant. As this is also in general the case during experiments, it is sufficient to take any arbitrary small value for $\Omega_p$. Finally, when the transmission dip is very asymmetric, it is often found impossible to obtain a good fit over the entire transmission scan. In this case an first order approximation is not sufficient to completely describe $T_{100 \%}$. Therefore only a good fit can be obtained for a smaller region around the 'EIT-peak'.
Bibliography


