Two-laser spectroscopy and coherent manipulation of color-center spin ensembles in silicon carbide
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Chapter 4

Electromagnetically Induced Transparency with c-axis divacancy spin ensembles in silicon carbide

Abstract
Divacancies in 4H-SiC oriented along the growth axis carry $S = 1$ spins with long coherence times, which can be resonantly optically addressed at cryogenic temperatures. Ensembles of these spins (both in as-grown and electron-irradiated samples) show significant inhomogeneous broadening of the optical transitions. Despite this broadening homogeneous sub-ensembles can be addressed, by exploiting optical pumping effects that can occur when coupling five levels with only two laser fields. To achieve coherent interplay between such a sub-ensemble and light, Electromagnetically Induced Transparency (EIT) can be a powerful tool, opening the way to the slowing and storing of quantum states of light in SiC ensembles. In this chapter we investigate EIT in effective five-level systems, in the presence of both transition-frequency and laser-intensity inhomogeneity, in c-axis divacancy ensembles in 4H-SiC. We show it is possible to isolate a single EIT feature by applying a weak static magnetic field. By solving a five-level master equation in Lindblad form, averaging over the inhomogeneities, and fitting to a series of EIT measurements, the population relaxation and pure dephasing rates are extracted. Also, the temperature dependence and limits for EIT in these systems are investigated. Finally, divacancy spin ensembles are shown to be promising for quantum memory applications, when integrating divacancy ensembles in single-mode waveguides.
4.1 Introduction

A basic qubit is a two-state quantum system, with a long coherence time. Coupling stationary qubits to photons is an important step in building quantum networks: since photons are fast and long-coherent, they are well suited for shuttling quantum information between qubits, encoded for instance in their energy or polarization. However, since the spacing between two long-lived qubit energy levels is generally well below the optical regime, a third level is required for all-optical control. Many systems containing (at least) three levels of this description have been realized, such as single ions caught in laser cooling traps [5], atomic vapors in transparent canisters [56], and carefully grown quantum dots [7]. More recently, color centers in transparent crystals were also shown to be functional qubits. These are electronic spins bound to semiconductor lattice defects, with all the requisite levels deep in the band gap. Besides the most widely studied color center, the NV$^-$ center in diamond [9], several point defects in silicon carbide have now also been shown to host such spins with very long coherence times [10, 11, 57, 58]. These centers in SiC have a variety of distinct properties, making mixing and matching qubits based on their intended use a possibility. Combining these naturally forming qubits with the scalability of SiC lithography and device processing may provide a technology for quantum integrated circuits.

When optically manipulating such a spin, it can be key to avoid putting it in a position where it can spontaneously lose energy before its task has been accomplished. When this happens, the system irretrievably decoheres, erasing its quantum state. Typically, population relaxation from the optically excited state is such a fast destructive process, and so when using optical fields for quantum control, the system should remain in its ground state levels. This can be achieved by coupling both ground states to the excited state simultaneously, using two laser fields. Over a narrow window of detuning between the lasers, the excitations cancel due to quantum interference in the system’s dynamics, and the system becomes transparent to light. This effect is termed electromagnetically induced transparency (EIT) [20]. EIT has been instrumental in many areas, such as the slowing of light [16], cooling atoms all the way to their ground state [17], and retrieval of a stored pulse of only a few photons [18, 19]. In such applications, it can be appealing to work with an ensemble of defects. When a small light pulse propagates through a spin ensemble, it can experience gain through efficient absorption and directional stimulated emission [3], amplifying the signal. This type of operation would avoid the need for technically-demanding optical cavities,
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and has been shown to work in for instance rubidium vapours [56].

In this work, an ensemble of $10^{15}$ to $10^{17}$ c-axis divacancy spins in 4H-SiC is addressed simultaneously, which suffers from inhomogeneous broadening of the optical transitions. Driving the divacancies involves six energy levels (an $S = 1$ triplet ground and excited state, see chapter 3). To nonetheless address homogeneous sub-ensembles, spin-related absorption (SRA) features are used, which entails the driving of more than two transitions with only two lasers. Finally in such ensemble measurements, the optical fields do not always have homogeneous intensity (for example, they can have a TEM$_{00}$ Gaussian profile). We show that in spite of the additional levels, couplings, and inhomogeneities, robust EIT in a homogeneous sub-ensemble can be realized. Through careful analysis using a master equation formalism in Lindblad form, values for the decay and dephasing rates are extracted. Overall, we show that the system can be engineered and controlled to be suitable for quantum applications in optical integrated circuits, while needing only two lasers for EIT.

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4.2.1 EIT in SRA features

Figure 4.1a shows the energy level structure of c-axis divacancies. The ground state and excited state are $S = 1$ spin triplets, with sublevels $|g_i\rangle$ and $|e_j\rangle$, respectively (we again use the labeling $i, j = \text{lower}, \text{middle}, \text{upper}$). Note that the full excited state may be more complex, but this excited state triplet is at least an isolated subset of the full excited state (see chapter 3). The energy levels have been shifted by an external magnetic field applied at a particular angle $\varphi$ to the c-axis, in order to make the $|g_m\rangle$-$|e_m\rangle$ transition equal to the $|g_u\rangle$-$|e_u\rangle$ transition. Because of this, a single laser field of frequency $\omega_p$ (probe, green) drives these ground states simultaneously. A second laser field of frequency $\omega_c$ (control, orange) drives the spin between $|g_l\rangle$ and $|e_m\rangle$, with its frequency detuned from the exact transition frequency by $\Delta$. In this driving arrangement the lasers drive optical transitions while avoiding optical pumping into any of the levels $|g_i\rangle$. This is the essence of addressing homogeneous SRA features in the presence of inhomogeneous broadening (see chapter 3). Note that one excited state is not involved (in this case $|e_l\rangle$), and so the SRA features are the result of driving transitions in effective five-level systems.
Figure 4.1: Schematic of 5-level EIT in PL2 SiC ensembles. a) Under SRA conditions, three ground state levels and two excited state levels are coupled by a control and probe laser field (Rabi frequencies $\Omega_c$ and $\Omega_p$, respectively). Population relaxation from any $|a\rangle$ to $|b\rangle$ occurs with a rate $\Gamma_{a,b}$, while pure dephasing occurs with a rate $\gamma_{a,b}$. Inset shows probe absorption when scanning probe detuning $\delta'$, with the EIT dip at two-photon resonance ($\delta' = 0$ MHz). Detunings $\Delta$ and $k$ are control laser detuning, and detuning from optimal SRA (simultaneous resonance of the probe for $|g_m\rangle$-$|e_m\rangle$ and $|g_u\rangle$-$|e_u\rangle$), respectively. b) With SRA optimized ($B > 12$ mT and the angle $\varphi$ between $B$ and $c$-axis near 57$^\circ$), besides the SRA system shown in a ($O_1$), a second SRA system ($O_2$) is addressed simultaneously (see chapter 3, section 3.3). Its excited state $|e^*\rangle$ is of higher energy (relative energies with respect to $O_1$ indicated by horizontal arrows), so the lower two instead of the upper two levels are addressed. If $O_1$ exists in the ensemble, so does $O_2$, due to inhomogeneous broadening of the optical transition (see also Fig. 4.2).

The situation where both lasers are equally detuned from their mutual excited state ($|e_m\rangle$ in Fig. 4.1a) by $\Delta$ is called two-photon resonance. In our further studies the probe laser frequency is scanned, and its frequency is referenced with respect to its detuning $\delta'$ from two-photon resonance. Note that $\delta'$ is only defined with respect to the control laser frequency, and not the excited state, so two-photon resonance is not dependent on inhomogeneous broadening (see Fig. 4.1a). The probe absorption as a function of $\delta'$ is shown in the inset (aligned with the excited state levels for clarity). The frequency with which the transitions are driven is given by the Rabi frequencies $\Omega_c$ and $\Omega_p$, which are proportional...
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to the square root of laser intensity, and the optical dipole of the transition (see appendix 4.8 for details and derivations). The width of the broad overall absorption line is governed by $\Omega_c$ and $\Omega_p$, the population relaxation rates $\Gamma_{a,b}$, and the pure dephasing rates $\gamma_{a,b}$. Additional broadening can be caused by the detuning $k$ (defined in Fig. 4.1a), which equals $\Delta$ plus the difference between the $|g_m\rangle\rightarrow|e_m\rangle$ and $|g_u\rangle\rightarrow|e_u\rangle$ transitions ($k$ can in principle be tuned to zero by setting the direction and magnitude of the external magnetic field). The reason these defects are of interest for quantum information science are the slow dephasing rates between the ground states $\gamma_{g_i,g_j}$, which have been shown for a single spin to be more than a millisecond using spin echo techniques ($T_2^*$), and still well over a microsecond when averaged over a macroscopic ensemble ($T_2^*$) [10].

Suppose $|g_u\rangle$, $|e_u\rangle$ and $|e_l\rangle$ are absent. In such a three-level system, exactly on two-photon resonance (probe and control fields equally detuned from $|e_m\rangle$ by $\Delta$), a sharp dip in absorption occurs. This is the well-studied EIT feature, where the system does not absorb the light due to quantum interference of the excitation pathways. In this situation, the system becomes trapped in a coherent dark state, which for this example (assuming ideal spin coherence) is $|\Psi\rangle \propto \Omega_p |g_l\rangle - \Omega_c |g_m\rangle$. The EIT lineshape is determined by the Rabi frequencies and dephasing rates, with a strong dependence on the slow pure dephasing rate $\gamma_{g_l,g_m}$ between ground state levels (the population relaxation between them is much slower still, and will be assumed negligible in the remainder of this chapter). This three-level arrangement is commonly referred to as a $\Lambda$ system.

In the effective five-level system under investigation here (which we will also call $\Lambda$ system for convenience) EIT also occurs, and the corresponding coherent dark state is $|\Psi\rangle \propto \Omega_p |g_l\rangle - \Omega_c |g_m\rangle + 0 |g_u\rangle$. Due to suppressed excitation from $|g_l\rangle$ and $|g_m\rangle$ by quantum interference, any population initially present in $|g_u\rangle$ is quickly pumped into this dark state.

Despite the complexity of these five-level systems, it is possible to derive analytical expressions for the absorption. When solving the master equation in Lindblad form to obtain the steady-state density matrix for the optically driven system (see appendix 4.8), the key point is that each ground state level is driven by a single laser frequency, and that the lasers have a fixed relative phase and detuning. Under these conditions it is always possible to choose a basis rotating in time with the dynamics, and a steady-state solution can be found, from which the absorption then follows. This remains true regardless of how many transitions are coupled by the optical fields (see for example Fig. 4.6a), even when more energy levels are involved. This is very useful when extracting the system parameters
from measurements of EIT, as will be done in section 4.4.2.

As depicted in the inset of Fig. 4.1, the EIT feature can shift from the overall absorption line center due to non-zero $\Delta$ and $k$, but the depth of the EIT feature with respect to the overall absorption line (i.e. the suppression of absorption) stays the same to within a few percent, i.e. the EIT is robust against imperfect magnetic field alignment for SRA.

### 4.2.2 Transition frequency and laser field inhomogeneity

In the macroscopic ensembles of c-axis divacancies that we studied, the optical transition between $|g_i\rangle$ and $|e_j\rangle$ is broadened by over $10^3$ times the homogeneous transition linewidth (see chapter 3, section 3.2). The consequence of this for SRA systems is illustrated in Fig. 4.2. Probe (green) and control (orange) laser fields enter from the left, encountering several divacancies (dumbbells). Divacancy I (red) has a particular optical transition frequency between ground and excited state, indicated by $f_0$. Together with the control laser frequency $f_C$, this sets the detuning $\Delta$. Divacancy II (purple) is identical to I, except the excited state levels lie at higher energies. The control laser is identical, so $\Delta_{II} > \Delta_I$ : this is the broadening. Divacancy III (blue) has its excited state levels shifted to even higher energies, so that the lasers couple different transitions. This way, several different coupling schemes can in principle be driven simultaneously. Note that only the spacing between ground and excited state changes: the $S = 1$ level spacings within each remain the same (see also chapter 3, section 3.5.2). This broadening necessitates the use of SRA features for addressing a homogeneous spin sub-ensemble. Additionally, the laser is not of uniform intensity, but rather a TEM$_{00}$ mode (so a transverse Gaussian intensity distribution), with near-constant width throughout the sample.
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Figure 4.2: Optical transition inhomogeneity. Illustration of inhomogeneous broadening. The optical fields (green and orange, left) encounter three divacancies, which have distinct optical transition frequencies. This leads to different detunings from two-photon resonance (I-II), or driving of multiple SRA systems (III).

Always two SRA systems
When tuning the magnetic field orientation to optimize SRA for fields greater than 20 mT, two SRA systems are realized simultaneously (see chapter 3, section 3.3). The first (labeled $O_1$) is drawn in Fig. 4.1a, and the second ($O_2$) is drawn in Fig. 4.1b. System $O_1$ and $O_2$ are identical, except the states $|e_j\rangle$ lie higher in energy, the shift equalling the splitting between $|e_l\rangle$ and $|e_m\rangle$ (horizontal arrows show relative positions of the energy levels between Fig. 4.1a and b). This overlap is a coincidence, due to the particular zero-field splittings of the divacancies. Their SRA features overlap, though they may not be identical ($k^*$ is not necessarily equal to $k$, and the optical dipoles are generally different, since the five levels at play for $O_1$ and $O_2$ are not the same). EIT also occurs at the same detuning for both, since its position depends only on the ground state splittings. Since in chapter 3 it was found that SRA systems involving the $|g_l\rangle$-$|e_l\rangle$ transition are suppressed, and because signals from $O_1$ and $O_2$ are similar and fundamentally hard to separate, the signal contribution of $O_2$ will not be separately analyzed in this chapter. Still, it is good to realize this contribution to SRA exists, lest it come as a surprise in further research using this approach, and it will be mentioned again later on when appropriate.

Transition frequency broadening of SRA
The second consequence of transition inhomogeneity is that a continuum of subensembles with a range of detunings $\Delta$ is addressed simultaneously. In Fig. 4.3a a series of probe absorption traces is shown in the bottom panel, with $\Delta$ vary-
Figure 4.3: Effects of inhomogeneities on lineshapes. a) Due to inhomogeneous broadening of the optical transition in an ensemble of defects, the control laser addresses divacancies with different detunings $\Delta$. The bottom panel shows probe absorption when $\delta'$ is scanned, for a range of spins having different $\Delta$ ($k = 0$ MHz, only one direction of broadening shown for clarity). The top panel shows the sum of these traces (red), compared to absorption of a single defect with $\Delta = 0$ MHz (black). b) The bottom panel shows probe absorption for spin ensembles at various distances from a Gaussian laser spot center. The top panel compares the cumulative absorption (red) and single defect absorption (black); the Gaussian spot makes the overall absorption line narrower, and washes out EIT.

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TEM$_{60}$ laser mode

Because the laser field intensity is not spatially uniform, the Rabi frequency experienced by defects in the center of the beam is different from those at a non-zero distance $r$ from the spot center. The absorption from sub-ensembles over a range of $r$ is shown in the bottom panel of Fig. 4.3b (assuming perfect overlap of probe and control beams). Near $r = 0$ the intensity is high, making both the EIT and the overall lineshape broad. However, only a relatively small portion of the divacancies in the laser beam experiences these laser intensities. Further from the laser center the intensity is lower, and so the EIT and overall lineshape grow narrower and shallower. However, since there are more defects at this radius than at the center, the total absorption from these defects is higher. The top panel shows a comparison of the sum of these contributions (red) to the response of a single spin (black) experiencing only the intensity at the laser spot center. Intensity inhomogeneity severely narrows the overall lineshape, and suppresses EIT. From here on out, the Rabi frequencies listed are those in the center of a Gaussian laser spot, unless specified otherwise.

4.3 Transmission spectroscopy

The measurement setup is identical to that described in chapter 3 (see chapter 3, section 3.4). In short, a divacancy ensemble of 2 mm in length is addressed by two near-parallel laser fields focussed to a diameter of 70 $\mu$m, resonant with the PL2 zero-phonon line near 1130.6 nm. The control field is switched on and off by a 270 Hz chopper wheel. We detect the transmission of the probe laser field, and this signal gets a 270 Hz component (detected using lock-in techniques) when optical pumping of divacancies is affected by both the control and probe-field resonances. The magnitude of the lock-in signal $R$ is proportional to the probe absorption. EIT is thus seen as a sharp dip in $R$, inside the broader overall absorption dip. An external static magnetic field (below 50 mT) optimizes SRA (see chapter 3, section 3.3), and the temperature can be set anywhere between 2 and 12 K.

Of special importance for EIT is that the control and probe field come from the same laser. For doing this, a portion of the control beam is split off, and gets frequency sidebands added at fixed detunings by an electro-optic modulator (EOM). One of the sidebands passes through a tunable cavity, blocking the fundamental frequency and other sideband, and is called the probe beam. Any
laser frequency jitter is therefore present in both probe and control, leaving the
detuning constant to well below 1 kHz, much less than the absorption features
presented here. Similarly, the phase between the lasers is fixed, so EIT is not
limited by the laser coherence.

The sample was not optically thick: the maximum SRA shown in this chapter
corresponds to a total absorption of nearly 3%. This absorption was observed on
a sample with an enhanced density of divacancies, achieved by irradiating an as-
grown sample with a 2 MeV electron beam (dose $8 \times 10^{18}$ cm$^{-2}$), and subsequently
annealing it at 750 °C for 15 minutes.

4.4 Experimental results and modelling

4.4.1 A single, robust, and isolated SRA Λ system

In chapter 3, section 3.5.3, two SRA Λ systems were identified by magneto-
spectroscopy, for a magnetic field at an angle $\varphi = 57^\circ$ with respect to the c-axis.
Fig. 4.4a shows this magneto-spectroscopy of the SRA of the Λ systems, Λ$_1$ and
Λ$_2$. For a range of magnetic fields, fine scans of two-laser detuning (resolution
1 MHz) for Λ$_1$ versus lock-in $R$ are shown as black traces. These have been
normalized and shifted vertically, to coincide with the magnetic field value where
they were measured. The two-laser detuning in these plots (the frequency differ-
ce between control and probe fields) equals $\delta'$ plus the ground state splitting
between $|g_l\rangle$ and $|g_m\rangle$. There is a single, sharp EIT feature visible for all magnetic
fields, as opposed to the basal-plane oriented divacancies discussed in chapter 2,
which does not visibly split or broaden. A single, isolated SRA Λ system is ad-
dressed, which is desirable for EIT applications: any stray transitions aborbing
light at the EIT frequency are detrimental.

In Fig. 4.4b a scan of two-laser detuning versus lock-in $R$ (normalized to one)
is shown. The magnetic field was moved away from optimal SRA, resulting in
a level shift $k = -23$ MHz, causing the sharp EIT dip to be shifted from the
center of the absorption line. In the background a Lorentzian lineshape (plus a
background and a small slope) is drawn in blue, which fits the absorption well,
away from two-photon resonance. To the low-detuning side of the sharp EIT dip,
clearly visible due to the EIT being off-center, is an increase in absorption with
respect to the Lorentzian lineshape.

To analyze the traces, a five-level master equation in Lindblad form was solved
for a range of both laser detunings $\Delta$ and laser intensities. The resulting traces
Figure 4.4: Single EIT features in a robust SRA $\Lambda$ system. a) Absorption scans of the probe laser, as a function of detuning between the lasers. Magneto-spectroscopy (1000x lower laser powers) placed in background for reference (darker is more absorption). The lock-in traces showing EIT dips (black) are shifted vertically to coincide with their respective magnetic fields. All along the SRA feature $\Lambda_1$, at the magic angle of $\varphi = 57^\circ$ (see chapter 3, section 3.3), a single sharp EIT feature appears. b-c) Absorption measurements showing off-center EIT (b) due to magnetic field misalignment, and central EIT (c) after correcting the field for optimal SRA. EIT width and height are not affected strongly by the magnetic field change. In b, a Lorentzian lineshape (plus background and slope) is drawn in light blue for comparison, illustrating that the experimental trace has an excessa amplitude in the line center with respect to a standard Lorentzian lineshape. The red line is our model fit (see main text), capturing the inhomogeneous EIT physics. Fitting this model to single traces does not yield unique coherences and lifetimes: stronger dipole couplings can compensate for lower relaxation and dephasing rates, producing nearly identical fits.
were summed and normalized, to account for optical transition inhomogeneity and laser intensity inhomogeneity (as shown for both inhomogeneities separately by the red curves in Fig. 4.3a and b). The resulting trace embodies absorption by an optically thin ensemble, and was fit to the data. To keep the number of free parameters small, both excited states were assumed to have equal decay \( \Gamma_e \) and pure dephasing \( \gamma_e \) rates. Only two distinct values were allowed for the optical dipoles between ground state levels \( |g_i \rangle \) and excited state levels \( |e_j \rangle \): \( \mu_{ij} \) and \( \mu_{i\neq j} \). The Rabi frequencies were then calculated as \( \Omega_{ij} = \Omega_0 \mu_{ij} \), where \( \Omega_0 \) is proportional to the electric field amplitude of the laser fields in the center of the beam. Lastly, the pure dephasings between any two ground states were set equal to \( \gamma_g \). The overall Lorentzian lineshape, EIT dip, and relative absorption increase are all captured by the model. The relative increase is found to be due to a combination of unequal transitions dipoles, a possible signal contribution from the \( O_2 \) SRA system, and laser inhomogeneity, and is thus partially an effect of interaction with an ensemble. In Fig. 4.4c a similar measurement result is shown, but with the magnetic field corrected to optimize SRA, shifting the EIT feature back to the middle. The EIT dip depth and width are hardly changed as compared to Fig. 4.4b, showing that EIT is indeed robust against such misalignment. The background is due to leakage from the control laser to the photodiode, from imperfect separation of the beams in our experiment.

The problem with these fits, however, is that they are not unique: by increasing the optical dipole values a little (for example a factor 1.1), and decreasing all dephasing and decay rates (by a factor 10), a nearly identical fit can be obtained. So, for a fit to a single absorption measurement, we can essentially trade an increase in Rabi frequency for a decrease in decay and dephasing rates, and not extract reliable system parameters. This is overcome by measuring absorption traces over a range of laser intensities.

It is known for three-level systems, and we verified it to be the case for five-level systems as well, that EIT becomes significant for control-laser Rabi frequencies

\[
\Omega_c > \sqrt{(\Gamma_e + \gamma_e)(\Gamma_g + \gamma_g)}.
\]  

(4.1)

When measuring for laser intensities that show how EIT evolves from weak to strong, \( \Omega_c \) crosses this critical point set by the decay rates of the system, and so the ratio of the dipoles and rates is uniquely determined over this range of control field intensities.
4.4 Experimental results and modelling

Figure 4.5: Power dependence of double EIT. a) At low magnetic field two Λ systems are driven simultaneously, at any angle \( \varphi \). This results in an absorption line with two EIT dips, split by \( k_g \). b) Lock-in measurement traces showing double EIT, for both laser powers varying simultaneously between 0.1 and 1 mW. The six solid lines are from one single fit (model described in text), yielding unique values for relaxation and dephasing rates.

4.4.2 Extracting decay and dephasing rates and dipoles

In Fig. 4.5a, the laser couplings for SRA are shown for very low magnetic field strength (<10 mT). For such weak fields \( |g_m\rangle \) and \( |g_u\rangle \), as well as \( |e_m\rangle \) and \( |e_u\rangle \), have not been shifted apart by more than the homogeneous transition linewidths. As a result, both the \( \Lambda_1 \) and \( \Lambda_2 \) systems are driven simultaneously, resulting in the double EIT depicted in the inset. Since only one frequency laser addresses each ground state level, the absorption spectrum can still be calculated analytically and fit to the data (see section 4.2.1). By measuring double EIT traces, in addition to extracting the population relaxation and pure dephasing rates, we gain insight in the relative quality of EIT in \( \Lambda_1 \) and \( \Lambda_2 \), and the influence of driving additional transitions near EIT conditions.

Figure 4.5b shows the probe absorption as a function of two-laser detuning,
for laser powers ranging between 100 µW and 1mW (both lasers powers equal). The angle \( \varphi \) was rotated away from the magic angle of 57°, but clear SRA is still realized as long as the magnetic field remains below 10 mT, so that \( |g_m\rangle \) and \( |g_u\rangle \) are still closer to each other than the homogeneous transition linewidths.

The traces are offset for clarity, but their relative amplitudes are unaltered. Behind the topmost trace a Lorentzian is drawn in grey, illustrating the two EIT dips, and an increase in absorption between them with respect to a standard Lorentzian absorption lineshape. The six black curves are the result of a single fit through all six datasets simultaneously (all traces contribute to the total error equally). The power dependence, EIT, and the increase in absorption near line center are well captured by the model. The fit parameters (now indeed unique) found are \( \Omega_0 = (7.4 \pm 0.6) \text{ MHz} \), \( \Gamma_e = (2.7 \pm 0.4) \text{ MHz} \), \( \gamma_e = (0 \pm 0.15) \text{ MHz} \), and \( \gamma_g = (0.23 \pm 0.06) \text{ MHz} \). Assumed was perfect overlap of the TEM\(_{00}\) control and probe beams inside the sample: an imperfect overlap would alter the fit values, but not by more than a factor 2 (and in fact would only bring \( \gamma_g \) further down). The ground state dephasing rate implies an ensemble coherence time \( T_{2}^* = (4.3 \pm 0.5) \mu s \), which is slightly longer than that found in microwave experiments on PL2 defect ensembles. This shows that optical fields cause no additional spin dephasing.

The dipoles were found to be equal to within a factor 1.7±0.1: this allows the two EIT features to be nearly identical. This is in contrast to predictions of dipoles varying by more than a factor 1000 by a Franck-Condon model for spin, as was found to be applicable for basal plane divacancies (see chapter 2, section 2.8). This supports the conclusion from chapter 3 that the c-axis divacancies required a more complex description than basal plane divacancies to explain the optical dipoles. Finally, the EIT amplitude compared to the overall absorption line is equal, to within 3%, to the EIT amplitude predicted for a three-level system, using the population relaxation and pure dephasing rates just listed for the five-level system. This shows that EIT in a five-level system is insensitive to the driving of additional transitions. Concluding, the robustness of five-level EIT and the relatively long \( T_{2}^* \) of the ground state under optical excitation show promise for c-axis divacancy ensembles in all-optical quantum applications.

### 4.4.3 Temperature dependence of SRA and EIT

Having shown that EIT in SRA features can be isolated, that it is robust against minor misalignments, and does not suffer from additional optical couplings be-
tween ground and excited state, we next investigate how EIT evolves with sample temperature. In Fig. 4.6a, another series of probe absorption versus two-laser detuning is shown, for temperatures between 2 and 12 K.

**Figure 4.6: Temperature dependence of double EIT.** a) Lock-in traces showing the double EIT feature from Fig. 4.5b, for a range of temperatures between 2 and 12 K. The black lines are separate fits. The SRA feature broadens extensively with temperature, though EIT stays clearly visible. b) Pure dephasing rates $\gamma_e$ of the excited state levels, from the fits in a.

As temperature increases, the overall absorption line broadens, and EIT diminishes. Each trace is fit by the master equation model, keeping the optical dipole strengths found before fixed, and allowing the population relaxation rate from the excited state $\Gamma_e$ and pure dephasing rates $\gamma_e$ and $\gamma_g$ to vary. The control laser power was in this experiment a factor 10 higher than before, a change captured well by the model. The traces are offset vertically for clarity, but the relative amplitudes are unchanged. The rates $\Gamma_e$ and $\gamma_g$ are found to not noticeably change over this range: $\gamma_g = (0.18 \pm 0.05) \text{ MHz}$ and $\Gamma_e = (2.6 \pm 0.4) \text{ MHz}$. Above 6 K on the other hand, pure dephasing for the excited state $\gamma_e$ strongly increases, broadening the overall absorption line. Since the required laser power for EIT
goes up with increasing $\gamma_e$ as described in equation 4.1, the power required for EIT goes up sharply above 6 K. This, together with the broadening of the SRA features themselves with respect to the spin splittings, limits the use of EIT in c-axis divacancy SRA features to below 10 K.

### 4.5 Prospect: a quantum memory for light

As mentioned in the introduction, a promising application of such defect ensembles would be to store the quantum information of a pulse of light on the few-photon level (see [18] for a detailed explanation). Initially, the spins need to be prepared in a known ground state, say $|g\rangle$. When writing to the memory, a single photon (playing the role of a weak probe beam) causes a single spin flip, in a manner that it is distributed over the ensemble. The information remains properly stored for the duration of the ensemble coherence time $T_2^*$, and can be read out by applying a strong control beam. This beam flips the collective spin back, causing the emission of a photon with the same quantum state as the original pulse. Because this re-emitted probe light and the strong control light are present simultaneously, EIT occurs, and the weak pulse can travel out of the ensemble without getting re-absorbed, as performed in [56] using a gas of rubidium atoms.

When modelling EIT for this scenario with the extracted system parameters, the inhomogeneous broadening of the optical transition requires us to take some extra care. In Fig. 4.7a probe absorption is shown as a function of the two-laser detuning in the bottom panel (two-photon resonance at 1300 MHz) for a range of detunings $\Delta$, which again represent separate sub-ensembles of defects, addressed simultaneously by the two lasers. $\Omega_p$ is very low (like the small pulse of light that is to be stored), and $\Omega_c$ is strong, ten times higher than required for the onset of EIT for a single defect according to 4.1, with $\gamma_e$ and $\Gamma_g$ set to zero. As a consequence of this high control power, absorption traces with $\Delta \neq 0$ MHz become very asymmetrical (Fano-like lineshapes), with their absorption minima not exactly at two-photon resonance (compare features highlighted in the grey vertical bar in Fig. 4.7a). Due to the high control beam intensity required for EIT, defects very far of resonance still show a small amount of absorption in the intended EIT window. In the top panel the combined absorption in case of an inhomogeneously broadened zero-phonon line of 100 GHz FWHM wide is shown (red), again for an optically thin medium, compared to the absorption of a single defect (black). The absorption by sub-ensembles with $\Delta \neq 0$ MHz completely washes out the EIT, and the retrieval of a stored pulse is rendered impossible for
Figure 4.7: Consequences for quantum memory application. a) The bottom panel shows absorption of a very weak probe ($\Omega_p \ll \Omega_c$) as the probe-control detuning is scanned for a range of inhomogeneously broadened $|e_j\rangle$ levels, resulting in a range of detunings $\Delta$ between two-photon resonance and the optical transition (as defined in Fig. 4.1). Traces are for optically thin samples. Parameters from the fit in Fig. 4.5 are used. The top panel compares absorption by spin ensembles with (red) and without (black) broadening of the optical transition. b) EIT in the presence of both optical transition and intensity inhomogeneity (averaging over an entire 100 GHz ZPL), when further increasing $\Omega_c$. c) As in b, but assuming homogeneous laser intensity. The EIT window broadens and deepens significantly, dipping below 1% for $\Omega_c = 1 \text{ GHz}$, with respect to probe absorption when the control field is off.

This $\Omega_c$. In Fig. 4.7b this combined absorption is shown again, for increasingly higher values of $\Omega_c$, with the lasers again assumed to have transverse Gaussian profiles. The EIT window opens up, requiring more than 140 times higher $\Omega_c$ ($2 \times 10^4$ times higher laser intensity) to reach the same EIT depth as a single defect. In essence, the higher power pushes the asymmetrical lines out to the sides. In Fig. 4.7c these same traces are shown, but for a homogeneous laser intensity (a top-hat profile). The EIT lineshape becomes more rectangular and deeper, with
probe absorption at two-photon resonance for $\Omega_c = 1$ GHz lowered to below 1% of that when no control field is present. Finally, Fig. 4.8 shows the effect on the EIT dip of less inhomogeneous broadening of the ZPL (again for homogeneous laser intensities). For a narrower FWHM of the ZPL, the Rabi frequency $\Omega_c$ due to the control laser required for 1% absorption at maximum EIT drops sharply. A more homogeneous sample, with less strain and other broadening effects, is therefore desirable.

![Graph showing the relationship between ZPL FWHM and $\Omega_c$]

**Figure 4.8: Control beam Rabi frequency required for 99% EIT.** The Rabi frequency due to the control laser (proportional to the square root of laser power) required for 99% EIT, for a ZPL FWHM between 100 MHz and 100 GHz, and homogeneous laser intensities. A decrease in FWHM allows for much lower laser powers for the same EIT.

So ideally, more than $1 \times 10^4$ times higher control laser intensity should be used than in these experiments, the laser intensity should be homogeneous, and the ZPL should be narrower. The higher intensity can be in large part realized by concentrating the light into a single mode SiC waveguide on a chip, which would have a diameter on the order of the wavelength, near 1 $\mu$m. This is 70 times smaller than the spot diameter used in these experiments, and since intensity increases quadratically with the beam diameter, this would give about 500 times higher intensity, still using simple diode lasers with power in the mW range. Additionally, in such a waveguide part of the Gaussian-shaped optical mode of the light would fall outside the waveguide, where it does not interact with divacancies. As a consequence the effective laser intensity becomes more homogeneous, further improving EIT as compared to our free-space experiments. Narrowing the ZPL would require future studies on the effects of SiC growth, defect creation, and waveguide fabrication on the ZPL width.
4.6 Summary and conclusions

In this chapter we presented that in an inhomogeneously broadened ensemble of divacancies, a homogeneous Lambda system can be addressed in isolation by means of two-laser control that effectively only addresses a homogeneous sub-ensemble of all divacancies. Despite the broadening, spatially inhomogeneous laser intensity, and the presence of six energy levels and up to six simultaneously optically driven transitions, EIT was realized. Using a master equation formalism, the decay and dephasing rates of the spins were extracted. Looking ahead to potential integration of such ensembles in optical integrated circuits as quantum memories, single-mode waveguides were proposed as ideal systems. These can provide the high and homogeneous laser intensity needed for good EIT, in the presence of inhomogeneous broadening of the optical transitions, which will be hard to fully eliminate for ensembles. This inhomogeneity would then be an asset instead of a hindrance: when using several ensemble memories, they will be able to store and emit photons over a bandwidth defined by the overlap of their broadened zero-phonon lines, and no individual tuning of memory nodes is required.
Supporting Appendix Material

4.7 Appendix: Initial result high-power control beam, low-power probe beam

In order to test the predictions of novel EIT lineshapes due to laser intensity and transition frequency inhomogeneity made in section 4.5, the measurement setup was altered from its description in chapter 3, section 3.4, as used in the rest of this chapter. In order to increase the control beam intensity, the FWHM of the beam inside the sample is decreased to 22 µm (which still keeps the Rayleigh length longer than the optical beam path inside the sample).

![Figure 4.9: EIT from weak probe and strong control beam.](image)

Three traces showing EIT in SRA feature $\Lambda_1$, with the probe beam set to 100 µW (much lower than other EIT traces in this chapter), and the control beam set to the much higher values of 0.8 mW, 4 mW, and 23 mW. The highest control-power trace shows clear EIT of over 40 MHz wide, with a sharp dip in the middle centered at 1683 MHz. The green trace is a fit of a Lorentzian, which is the sharpest possible EIT lineshape predicted by density matrix of a single defect. The black trace is a fit of the model described in section 4.5, including a 3D overlap of the Gaussian probe and control beams, and also a repump beam. The inhomogeneous model clearly fits better, though not perfectly.
To still be able to spatially filter the strong control beam from the weak probe beam, the beams are made to counter-propagate through the sample, while still also spatially separating them. This way, light from the control beam arriving at the photodetector is suppressed by a factor $10^6$, and EIT can be clearly observed in the weak probe beam.

For the lower two control powers, EIT is barely visible, though the SRA feature clearly power broadens for $P_c = 4$ mW. For $P_c = 23$ mW, however, the EIT is very pronounced, more than 20% of the SRA feature height, and over 40 MHz wide. There is a sharp dip in the center at 1683 MHz two-laser detuning, which is atypical of EIT as calculated for a single defect from the density matrix formalism: the sharpest EIT lineshape possible in that model is a Lorentzian [20]. The green line is an attempt to fit a Lorentzian to the dip, which clearly fails to describe the sharp central dip. The black line is a fit using the model described in section 4.5, with the addition that the laser intensities of both probe and control beam were calculated in 3D, taking into account their slightly different orientations by 6°. Also, a repump laser was added to the model, which was used in the experiment to keep the defects from optically bleaching, crossing both probe and control beams from the side (for more about bleaching, see chapter 2, section 2.11). Due to a combination of all the included inhomogeneities, the EIT trace can assume a much sharper shape, clearly fitting the observed data better than the homogeneous model. The sharpness near the peak here is equally due to the Gaussian shape of the intensity and frequency distributions, and the imperfect spatial overlap. A more minutely accurate description of the spatial distributions might yield even better correspondence with our experimental data, but introduced too many uncertainties to be used for fitting in a reasonable time.

### 4.8 Appendix: Master equation in Lindblad form

To obtain an expression for the absorption of an optically thin ensemble of defects, we solve the master equation in Lindblad form:

\[
\frac{d\rho}{dt} = -\frac{i}{\hbar}[H, \rho] + \mathcal{L}(\rho). \tag{4.2}
\]

Here $\rho$ is the density matrix, where the diagonal elements are populations, and the off-diagonal ones are coherences. Both the ground states and excited states are contained in $\rho$: for $E$ excited states and $N$ total states, states 1 through...
$E - 1$ are ground states, and states $E$ through $N$ are excited states. Describing the example system in Fig. 4.10 for example, $|s_1\rangle$ and $|s_2\rangle$ are the first two states in the 4-by-4 matrix $\rho$, and $|s_3\rangle$ and $|s_4\rangle$ are the last two states. $H$ is the Hamiltonian of the electronic system, driven by optical fields. Lastly, $\mathcal{L}(\rho)$ is the Lindblad superoperator, describing decay between the spin levels and pure dephasing. The goal of this appendix is to show under which conditions Eq. 4.2 can be solved analytically, for any number of ground and excited states, and the steps in solving it. The formulas given here are also easily fed into a symbolic programming language. This can be useful, given that the number of coupled linear equations to solve scales as $N^2$ for $N$ states, and hence the matrix to eventually be inverted for solving this system scales contains $N^2$ by $N^2$ elements, which quickly becomes exceedingly tedious and error-prone to construct by hand.

**Figure 4.10: Example four-level system.** A four-level system, driven by two optical fields. The fields, labelled probe and control, are characterized by Rabi frequencies $\Omega_c$ and $\Omega_p$ (proportional to the electric field amplitudes), and angular frequencies $\omega_c$ and $\omega_p$ (see Eq. 4.8). Relevant decay rates ($\Gamma_{ij}$) are drawn in gray between the states. Detuning of the control laser from resonance is labeled $\Delta$, and detuning of the probe laser from two-photon resonance is labeled $\delta'$. We use the convention that blue-detuning makes the detunings positive.
4.8 Appendix: Master equation in Lindblad form

4.8.1 The Hamiltonian and the Rotating Wave Approximation

We start with the Hamiltonian of the optically driven system:

\[ H = H_0 + V, \]  \hspace{2cm} (4.3)

where \( H_0 \) is the bare Hamiltonian of the system, and \( V \) is the perturbation due to the optical fields. In matrix form, \( H_0 \) is written as

\[
H_0 = \begin{pmatrix}
\hbar \omega_1 & 0 & \ldots & 0 \\
0 & \hbar \omega_2 & \ddots & \\
\vdots & \ddots & \ddots & \\
0 & \ldots & \hbar \omega_N
\end{pmatrix},
\]

(4.4)

with the energies of the \( N \) states of the system on the diagonal (\( N = 5 \) in chapter 4, with three ground states and two excited states), and the other matrix elements zero. The perturbation \( V \) is written as

\[
V = \begin{pmatrix}
0 & V_{12} & \ldots & V_{1N} \\
V_{21} & 0 & \ddots & \\
\vdots & \ddots & \ddots & \\
V_{N1} & \ldots & 0
\end{pmatrix},
\]

(4.5)

The diagonal elements of \( V \) represent permanent dipole moments, which are assumed to be (near) zero. The energy of the off-diagonal terms is

\[ V_{nm} = -\vec{\mu}_{nm} \cdot \vec{E}, \]  \hspace{2cm} (4.6)

where \( \vec{\mu}_{nm} = \vec{\mu}_{mn} \) is the dipole of the transition \( |n\rangle \rightarrow |m\rangle \), and \( \vec{E} \) is the electric field. This is simply the energy of a dipole induced by an electric field, which is valid since the wavelength of the electric field (micrometer) is much larger than the size of the divacancy (Ångström). This is commonly known as the dipole approximation. From here on out, we drop the vector notation, assuming the field and dipole moment are properly aligned with each other. Which dipoles are nonzero (i.e. hence which transitions are driven by optical fields) is established
a priori, by applying knowledge of detunings of the optical frequencies from resonance, and the optical linewidths of the transitions. For example, for the four-level system shown in Fig. 4.10, the dipole matrix is

\[
\mu = \begin{pmatrix}
0 & 0 & \mu_{13} & 0 \\
0 & 0 & \mu_{23} & 0 \\
\mu_{31} & \mu_{32} & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix},
\]

(4.7)
since only two transitions are driven. This does not yet explicitly contain information on which fields drives which transitions: only whether transitions are to be considered or not. The optical fields are described classically:

\[
E(t) = \frac{1}{2} \sum_{i=1}^{Q} E_i e^{i\omega_i t} + c.c.,
\]

(4.8)

Here \(E_i\) is the amplitude of optical field \(i\), \(\omega_i\) is its angular frequency. We ignore the phase of the fields (that is, \(E_i\) is a real number), which is valid if the fields are perfectly in phase, e.g. generated from the same laser by an electro-optic modulator (as is the case in chapters 3 and 4).

This in principle completes the Hamiltonian of the system. However, it is an unwieldy equation with many time-dependent driving terms, for example for the system depicted in Fig. 4.10:

\[
V_{13} = -\frac{\mu_{13}}{2} \left[ E_p e^{i\omega_p t} + E_c e^{i\omega_c t} + E_p e^{-i\omega_p t} + E_c e^{-i\omega_c t} \right].
\]

(4.9)

We can greatly reduce the number of driving terms by realizing not all driving happens on a relevant time scale. To identify these terms, we change to the interaction picture, which serves to shift all time dependence from \(H_0\) onto \(V\). The resulting operator completely governs the time evolution of the system, and will contain terms that oscillate so fast that they will average out very quickly, and hence we can remove them. This is known as the rotating wave approximation (RWA). Moving to the interaction picture requires the unitary operator \(U\), defined as
4.8 Appendix: Master equation in Lindblad form

\[ U = e^{iH_0t/\hbar} = \begin{pmatrix}
  e^{i\omega_1 t} & 0 & \cdots & 0 \\
  0 & e^{i\omega_2 t} & \cdots & \\
  \vdots & \vdots & \ddots & \vdots \\
  0 & \cdots & 0 & e^{i\omega_n t}
\end{pmatrix}, \quad (4.10) \]

and with it we transform the Hamiltonian \( H \) to

\[ H' = UHU^\dagger. \quad (4.11) \]

To illustrate the working of the RWA, the term from Eq. 4.9 in the interaction picture is

\[ H'_{13} = -\frac{\mu_{13}}{2} \left[ E_p e^{i\omega_p t} + E_c e^{i\omega_c t} + E_p e^{-i\omega_p t} + E_c e^{-i\omega_c t} \right] e^{i(\omega_1 - \omega_3)t}. \quad (4.12) \]

Here, we can apply our knowledge of which fields are driving which transitions. Since the control laser in Fig. 4.10 is near-resonant with the transition \(|s_1\rangle \rightarrow |s_3\rangle\), the difference between the control laser frequency \( \omega_c \) and the transition frequency \( \omega_3 - \omega_1 \) is small. Hence, the term \( e^{i(\omega_c+(\omega_1-\omega_3))t} \) from Eq. 4.12 evolves slowly in time. All the other fast-changing terms (representing driving far off-resonance) are dropped, and the transformation from Eq. 4.11 is inverted to move back to the Schrödinger picture. For comparison to Eq. 4.9:

\[ H_{13} = -\frac{\mu_{13}}{2} E_p e^{i\omega_p t}, \quad (4.13) \]

which is much more tractable. Next, in order to eliminate all remaining time-dependence when solving Eq. 4.2, we turn our attention to the density matrix \( \rho \).

4.8.2 The density matrix in a rotating frame

Since \( H \) contains oscillating driving terms, from Eq. 4.16 the populations and coherences contained in \( \rho \) will also oscillate at those frequencies. However, by adopting a judiciously chosen rotating basis for \( \rho \) rotating along with the driving, Eq. 4.16 can in some cases be solved as a steady-state problem in this rotating frame.
frame: the oscillations are “unwound”, revealing simple behaviour. This slow-changing basis we call \( \sigma \). To relate \( \sigma \) to \( \rho \) we define another unitary operator

\[
R = \begin{pmatrix}
R_{11} & 0 & \ldots & 0 \\
0 & R_{22} & \ddots & \\
\vdots & \ddots & \ddots & \\
0 & \ldots & 0 & R_{nn}
\end{pmatrix},
\]

(4.14)

where

\[
R_{nn} = \begin{cases}
e^{i\omega_L t} & \text{if level coupled by only 1 field of } \omega_L \\
1 & \text{otherwise}
\end{cases}
\]

(4.15)

This operator then relates \( \sigma \) and \( \rho \) by

\[
\rho = R\sigma R^\dagger.
\]

(4.16)

To illustrate how this choice of basis can eliminate time dependence, we write out Eq. 4.2 for \( \rho_{13} \), using the Hamiltonian found by the method in section 4.8.1, and the density matrix from Eq. 4.16:

\[
\frac{d\sigma_{13}}{dt} + i\omega_c \sigma_{13} e^{i\omega_c t} = -i \left[ (\omega_{s3} - \omega_{s1}) \sigma_{13} + \frac{E_c}{2\hbar} \mu_{13}(\sigma_{11} - \sigma_{33}) + \frac{E_p}{2\hbar} \mu_{23} \sigma_{12} \right] e^{i\omega_c t}.
\]

(4.17)

In Eq. 4.17, all terms contain the same time-varying factor \( e^{i\omega_c t} \), which therefore cancels out, removing all time dependence. Also, since due to the RWA only the driving terms close to resonance were kept in the Hamiltonian, and in Eq. 4.14 we define which lasers are resonant with which transitions, all the frequency terms can be gathered into detunings from resonance (in this case, \( \Delta = (\omega_{s3} - \omega_{s1}) - \omega_C \), which is depicted in Fig. 4.10). Finally, for compactness of writing we define Rabi frequencies \( \Omega_{(ab)} = \frac{\mu_{ab} E_{ab}}{\hbar} \) for transitions \( |s_a\rangle \rightarrow |s_b\rangle \), where the \( (ab) \) label can be \( c \) or \( p \), depending on the laser driving the transition, as shown in Fig. 4.10. Applying these steps to Eq. 4.17 yields

\[
\frac{d\sigma_{13}}{dt} = -i \left[ \Delta_{13} + \frac{\Omega_c(\sigma_{11} - \sigma_{33})}{2} + \Omega_p \sigma_{12} \right].
\]

(4.18)
The same cancellation of exponents and gathering of terms into detunings (e.g. $\delta'$ in the example) happens for the other equations which follow from Eq. 4.2, meaning the rotating basis defined in Eq. 4.14 works for the example system.

The requirement for such a rotating basis to exist for any number of states, and any number of lasers, is that all ground states (or all excited states) are driven by single laser frequencies. Mathematically, this is because if more frequencies are coupled to a state, and they were all entered into Eq. 4.14 somehow, the cancellation of exponents seen in Eq. 4.17 does not take place (the $e^{i\omega_c t}$ on the left-hand side would become a sum of two driving terms, while several different driving term combinations appear on the right-hand side). This is why for the example system in Fig. 4.10 no useful transformation can be defined for $R_{33}$ in Eq. 4.14. However, Eq. 4.14 needs to contain information on all driven transitions, in order for them to vanish in the rotating frame. For this information to be included, both states that $|s_3\rangle$ is coupled to need to be used in Eq. 4.14, i.e. they may not be addressed by more than one laser frequency. For the example system this is the case, and hence a rotating basis with no time dependence can be constructed. If for example the transition $|s_2\rangle \rightarrow |s_4\rangle$ is also driven, the transformation will not work, since no information about the probe laser can be usefully included in the rotating basis. Generally, for any number of states that are coupled by optical fields, this requirement is equivalent to having all ground states or all excited states addressed by single laser frequencies. Another way of viewing this is with the notion of beat frequencies. If two states are coupled by a laser, the driving frequency determines the dynamics. If two states are both coupled to a third, as in the example system, the driving between these two states occurs at the difference frequency (or beat frequency) of the two optical fields, and this can be accommodated in a rotating frame. If however one or both of these states is coupled to a fourth state, with a different optical frequency, this interferes with the beat frequency, and this cannot be captured in a rotating frame anymore.

4.8.3 The Lindblad superoperator

The treatment so far concerned a driven system, without decay and dephasing. These are introduced now, in the form of the Lindblad superoperator $\mathcal{L}(\sigma)$, where
\[ \mathcal{L}_{nm} = \begin{cases} \sum_{i=1}^{N} (\Gamma_{in} \sigma_{ii} - \Gamma_{ni} \sigma_{nn}), & \text{if } n = m \\ -\frac{1}{2} \sum_{i=1}^{N} (\Gamma_{ni} + \Gamma_{mi}) + \gamma_n + \gamma_m \sigma_{nm}, & \text{if } n \neq m. \end{cases} \] 

(4.19)

Here \( \Gamma_{ij} \) is decay from state \( |s_i\rangle \) to \( |s_j\rangle \), and \( \gamma_i \) is pure dephasing in state \( |s_i\rangle \), with respect to a reference phase (we use the phase of the lowest ground state for this). The on-diagonal terms can be understood as a sum of all incoherent flows of population into and out of the state. The off-diagonal terms describe loss of coherence due to population exiting either of the two states involved, or pure dephasing in either of these states.

### 4.8.4 Solving the steady-state equations

After successfully applying the steps in the preceding sections, we are left with \( N^2 \) coupled differential equations like Eq. 4.18. Since the right-hand sides do not contain time-dependent terms anymore, and \( \sigma \) is the time-independent envelope of \( \rho \), we can set the time derivatives to zero and solve the equations as a system of coupled linear equations. This system is underdetermined, which can be fixed by replacing an arbitrary equation by the constraint that total population is conserved, i.e. \( \sum_{i=1}^{N} \sigma_{ii} = 1 \). It can then be conveniently solved by casting it in matrix form, and inverting this matrix.

Describing EIT in an optically thick medium would require us to calculate the electric susceptibility \( \chi(\omega) \) from \( \sigma \), and from there the absorption. However, in this thesis only optically thin samples are investigated, with absorption up to a few percent, far from saturation. In this case the absorption per system is proportional to that of the ensemble, and this can be calculated as

\[ A(\omega) \propto \sum_{i=E}^{N} \sigma_{ii}(\omega), \] 

(4.20)

where the sum runs over all the excited states, or in other words: excitation is proportional to absorption.