Chapter 3

Measurements of the local nuclear spin distribution by electromagnetically induced transparency

Abstract

We demonstrate how nuclear spin polarization can be induced and detected by optical interaction with donor-bound electrons in GaAs. Resonant addressing of a Λ-system formed by the Zeeman-split bound-electron spin states and a donor-bound exciton state results in nuclear spin polarization either along or opposite to an external magnetic field. The coherent population trapping effect in the Λ-system is used to detect spin polarization of nuclei at the donor location. We present how this technique is used as a tool to monitor the local build-up and relaxation of nuclear spin polarization.
3.1 How the EIT lineshape reflects the nuclear spin state

The lineshape of the EIT resonance contains information on the spin polarization of the nuclei near the donors. In two-laser spectroscopy the measured transmission of each of the lasers through the GaAs sample is determined by the amplitude transfer function $T(\omega_i, \Omega_i|\omega_j, \Omega_j) = \exp(i\omega_i nd/c\chi(\omega_i, \Omega_i|\omega_j, \Omega_j)/2)$, where $d$ is the thickness of the medium, $n$ is the refractive index of GaAs and $c$ is the speed of light, $i, j \in 1, 2$ with $i \neq j$ labels the laser frequencies $\omega$ and powers expressed as Rabi frequency $\Omega$, as also introduced in Fig. 3.1. The notation of separating the variables in $T$ and $\chi$ by a vertical line is meant to indicate that we consider the transfer function with variables $\omega_i, \Omega_i$ conditional on $\omega_j, \Omega_j$ which then assume the role of parameters. The susceptibility $\chi$ is obtained by considering the polarization density of a medium filled with donors with concentration $\rho$, each represented as a $\Lambda$-system as described in Chapter 2. The dependence of the transmittance on the decay and dephasing parameters of the $\Lambda$-system is implicit. The susceptibility is made up from the polarizabilities of individual systems $\beta$ which can, and generally will, show small differences, thus forming an inhomogeneous ensemble. We focus on the inhomogeneity arising from the nuclear spin polarization. This inhomogeneity gives rise to a distribution of Overhauser shifts $P(\delta)$ and we express the susceptibility for the medium accordingly as

$$\chi(\omega_i, \Omega_i|\omega_j, \Omega_j) = \rho \int P(\delta)\beta(\omega_i, \Omega_i|\omega_j, \Omega_j, \delta)d\delta. \tag{3.1}$$

The polarizability $\beta$ exhibits an EIT resonance ($\beta$ decreases as the transmission increases). When this resonance is narrow as compared to $P(\delta)$ (such that it can be approximated by a Dirac delta function), Eq. 3.1 implies that the transmission near EIT resonance takes the shape of $P(\delta)$. We now show that in this GaAs system the EIT lineshape indeed changes as we induce a nuclear spin polarization by optical pumping. For this experiment we use the setup as described in Sec. 8.2 where two laser beams co-propagate through the sample and are collected immediately behind the sample on a photodiode. One of the laser beams is modulated by a chopper at 6 kHz and we isolate the modulated part of the total transmission signal by lock-in detection (for details see Sec. 8.4).
3.2 Detection of optically induced dynamic nuclear polarization

Dynamic nuclear polarization (DNP) resulting from optical pumping is detected by comparing the initial EIT lineshape, with the nuclear spins at thermal equilibrium, to the EIT lineshape after pumping. Figure 3.2a shows the initial EIT resonance measured in the transmission signal of a 10 µm thick film of n-GaAs. The EIT lineshape contains information of...
the underlying nuclear spin distribution (Eq. 3.1), so any modification of the nuclear spin state by optically induced dynamic nuclear polarization (DNP, as explained in Section 2.4.3) will be reflected in the EIT resonance. When the same scan is taken again after 30 minutes of optical pumping, with a single pump laser resonant on the A∗ transition (defined in Fig. 3.2a), the EIT resonance has shifted and broadened. This is visible in Panel b where the two EIT scans are compared. Since the EIT shift relates to the underlying nuclear spin distribution as described in the previous section, we can associate the shift with a change in the average of the distribution $P(\delta)$ and hence a net buildup of nuclear spin polarization. The shift of 4µeV depicted in the figure corresponds to an average Overhauser field of approximately 0.2 T. The broadening indicates an increased inhomogeneity of the Overhauser field. As we discuss in the next section, there is a dependence of the Overhauser shift on pump laser intensity. Due to the experimental setup we cannot address the ensemble within the laser spot with equal intensity. The first reason for this is that the spot is Gaussian and therefore the pump laser intensity for individual donors decreases with increasing radial distance from the spot center. Another factor is that because of reflections at the GaAs film surface a standing wave intensity distribution forms inside the sample. The former effect could be mitigated by shaping the profile of the pump beam and the latter could be improved by applying anti-reflection coating on the GaAs film.

3.3 Timescale of optically induced DNP buildup

In Fig. 3.3 we observe the EIT lineshape after a duration of pumping sufficiently long for the DNP process to saturate, i.e. when a balance is established between the polarizing effect of the pump laser and the depolarizing effect of spin diffusion. However it is also possible to monitor the evolution of the system during optical pumping to get insight into the timescale at which the DNP process takes place. This is shown in Fig. 3.3. Panel a shows repeated EIT scans which were taken while the pump laser was on. The stronger laser that is fixed on resonance with the A∗ transition fullfills the function of optical pumping during most of the
3.4 Bidirectionality of DNP in the $\Lambda$-system

The $\Lambda$-system allows for optical orientation of the electron spin along or against the magnetic field. We verify in Fig. 3.4 that polarization of the electron spin by pumping on the A transition reverses the direction of the EIT shift. This is done by first pumping for 30 minutes on either the...
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Figure 3.3: (a) Build up of nuclear spin polarization measured by taking EIT scans repeatedly, using the strong pump laser (6 W cm$^{-2}$ on A*) as the control. A weak probe laser (0.1 W cm$^{-2}$) periodically scans the A-transition (see time labels). Time $t = 0$ is the moment where the pump laser is switched on after keeping the sample in the dark for an hour (see inset of (b)). (b) The Overhauser shift as fitted from the data in (a) as a function of time (dots). The solid line is a fit for exponential build-up, with a time constant $\tau_{\text{build}} \approx 340$ s.

A* - or A-transition, the resulting datasets are color-coded blue and red respectively. The shift of the EIT resonance immediately after pumping indicates the presence of a non-zero Overhauser field, which is opposite in sign for the blue and red cases. To confirm this observation we continue taking EIT scans to observe relaxation of the center of EIT resonance towards its initial value. In this manner it also becomes apparent on which timescale the nuclear spin polarization decays: The solid lines are exponential fits yielding decay times, $\tau_d$ of 600 s after pumping on the A transition, and 480 s after pumping on the A* transition.

The presence of bidirectionality also excludes that the EIT shift is due to heating of the sample during the optical pumping, which would give a shift in only one direction.
3.4 Bidirectionality of DNP in the Λ-system

Figure 3.4: Bi-directionality and relaxation of the induced Overhauser shift. (a) A sequence of scans taken after 30 minutes of optical pumping on A* with intensity of 6 W cm\(^{-2}\) showing the evolution of the EIT resonance over time during the relaxation of prepared DNP. EIT scans are taken with control (2.37 W cm\(^{-2}\)) on the A-transition and scanning probe (0.109 W cm\(^{-2}\)) on the A*-transition (see inset of (c)). (b) Same sequence of EIT scans after optical pumping on A. (c) The Overhauser shift as derived from the data in (a) and (b) as a function of time (dots). The solid lines are fits for exponential decay, with time constants \(\tau_d\) of 600 s (after pumping on the A-transition) and 480 s (after pumping on the A*-transition).
Figure 3.5: The invasive character of EIT measurement on DNP. (a) The difference in EIT peak position for the control laser on the A- (red) and A*-transition (blue), taken with similar intensities for the control laser (2 W cm$^{-2}$) in the two cases, and weaker probe lasers (see main text). (b) Relaxation of the nuclear spin polarization. Datapoints show fitted EIT peak positions for different times after optical pumping on A* (6 W cm$^{-2}$). Gray triangles: fitted EIT peak positions from scans taken after a dark time following 30 minutes of optical pumping on A* with intensity of 6 W cm$^{-2}$ (pumping was repeated for each datapoint). Black dots: fitted EIT peak positions of a sequence of scans taken after 30 minutes of optical pumping on A*. Corresponding lines are exponential fits.

3.5 Invasiveness of the detection method

The optical pumping is closely related to the EIT probing. Therefore we investigate how EIT measurements influence the DNP process. This is shown in Fig. 3.5. The difference between taking EIT with control on the A or A* transition is shown in Panel a. There is a minor shift of 1 µeV. Panel b shows how EIT measurement influences the relaxation of the nuclear spins. When the system is kept in the dark the spins relax faster. We attribute this to the inhomogeneous Knight shift which protects against spin diffusion when the electron spin is (partially) polarized.
3.6 Dependence on pump laser intensity

Figure 3.6: Intensity dependence of DNP. All data sets were acquired in the same way as for Fig. 3.3b. The intensity is increased in steps from $I_0 = 1 \text{ W cm}^{-2}$ to $8I_0$.

3.6 Dependence on pump laser intensity

The dependence on pump laser intensity is presented in Fig. 3.6, with a series of measurements performed in the same way as was done for Fig. 3.3b. The intensity of the pump laser is from $I_0 = 1 \text{ W cm}^{-2}$ to $8I_0$. For intensities over $8I_0$ it was not possible to obtain these measurements because of increased noise levels in the detection setup and a stronger broadening of the shifted EIT peak. However, the fact that the steady state Overhauser field increases roughly linear with pump laser intensity (rather than quickly saturating), as shown in Fig. 3.6, needs additional explanation. The long $D^0$ spin relaxation times suggest that the pump laser completely polarizes the electron spin, already for pump intensities far below $8I_0$. It has been shown that for pump laser intensities as low as
10 mW cm\(^{-2}\) the electron spin polarization is already well over 50\% [1].

This discrepancy can be explained by unintended pumping, such as shown in Fig. 3.7, which causes the dependence of the maximum Overhauser shift on the optical pump intensity to increase up to high intensities. The mechanism behind this is as follows: when the pump laser is tuned to resonance with the transition \(|\uparrow\rangle-|A\rangle\) there may be unintended coupling to the \(|\downarrow\rangle-|B\rangle\). This has as effect that the optical excitation rate (and with it the electron spin flip rate) increases linearly up to high intensities. Without the unintended pumping this rate is limited by \(1/T_2\) because the electron has spin has to decay before the next optical cycle can be initiated; with the unintended coupling the system is optically excited from both ground states, this increases the fluctuation rate of the electron spin. In the calculation for the plot we used \(T_2 = 10\,\mu\text{s}\), corresponding to a maximum optical excitation rate of 0.1 MHz. The excitation rate is seen to extend beyond \(1/T_2\) and the hyperfine flip rate \(\Gamma_h\) will increase with it as described in Sec. 2.4.3.

References

Figure 3.7: (a) When the pump laser is tuned to resonance with the transition $|\uparrow\rangle - |A\rangle$ there can be unintended coupling to the $|\downarrow\rangle - |B\rangle$. (b) This has as effect that the optical excitation rate (and with this the effective electron spin flip rate) increases linearly up to high intensities.
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