Electromagnetically induced transparency with localized impurity electron spins in a semiconductor
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Chapter 3

High-resolution magneto-optical transmission spectroscopy on the donor-bound-exciton complex in GaAs

Abstract

We present high-resolution magneto-optical spectroscopy studies of the donor-bound exciton \((D^0 X)\) states in \(n\)-GaAs. In our approach we resonantly excite transitions from one of the two spin states of localized donor electrons \((D^0 \text{ systems})\) to specific levels of the \(D^0 X\) complex, and derive spectroscopic signals from laser fields transmitted through the material. Our goal was to characterize the \(D^0 X\) levels in terms of their quantum numbers, level splittings, and the purity of polarization selection rules of the \(D^0-D^0 X\) transitions. While we can partly link our analysis to earlier experimental studies of this system (mainly photoluminescence results, PL), our conclusion is that we cannot fully identify the correspondence between these PL results and our transmission results. We also conclude that the existing theoretical models for the \(D^0 X\) system can only partly describe our observations.

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This chapter is based on Ref. 5 on p. 127.
3.1 Introduction

As introduced in Chapter 1, the research presented in this thesis aims to investigate how quantum-optical control techniques can be implemented and optimized with a solid-state material system, for the long-term goal of realizing quantum-information processing. Optimally using the spin coherence of the $D^0$ system via the $D^0$-$D^0X$ optical transitions requires knowledge of the $D^0X$ levels, including their behavior in an applied magnetic field. In particular, an approach based on Electromagnetically Induced Transparency (EIT, see Chapters 1, 2) works optimal with strong optical transitions that obey clear polarization selection rules, to a $D^0X$ level that is energetically well separated from other levels. Knowledge of the selection rules for the various $D^0X$ levels, and how they shift when applying a magnetic field in a particular direction, is then indispensable. While purely phenomenological data on this is already of great value, the goal of the research presented in this chapter is to also identify the quantum numbers of the observed $D^0X$ states, and to confirm the correspondence between our magneto-optical $D^0X$ observations and theoretical modeling. Given that extensive earlier work by others in this direction had only limited success (see below), our expectations for success should be moderate. Our motivation to nevertheless study again it is twofold. Most earlier studies used signals from photoluminescence, while our approach may have an advantage from direct resonant excitation of transitions, and studying signals from this in transmission. Further, we need to establish to what extent the predicted behavior of the models really occurs for our approach.

Earlier work by our team on the GaAs $D^0$ system gave specific additional motivations for such a spectroscopic study. For one, we found that in applied magnetic fields between 7 T and 8 T, experimentally finding the $D^0$-$D^0X$ transitions and realizing EIT was typically much harder (and for several samples impossible) as compared to such work below 7 T or above 8 T. Understanding of this observation is till now fully lacking. Further, for Dynamic Nuclear Polarization (DNP, see also Chapters 1, 2, 4), direct electron-nuclear spin flip-flops are energetically forbidden when their Zeeman splittings differ significantly. More complex mechanisms then dominate the process, possibly mediated by optical transitions [1]. Understanding what mechanisms could here be at play clearly needs insight into the spin and orbital-momentum quantum numbers of the involved $D^0X$ levels, as well as the associated selection rules.

The $D^0X$ system in GaAs and similar semiconductors has already been studied for more than 50 years (further reviewed in the next section, see also Chap-
3.1 Introduction

This line of research (in particular the theoretical description) left several open questions, since the \( D^0X \) system is rather complex. The relevant energy splittings are rather small, such that it is at the edge of what can be reliably addressed with numerical approaches such as Density Functional Theory. An analytical approach is also challenging. The \( D^0X \) system is an atom-like system in a solid-state environment, where three charge carriers (two electrons and a hole) are in orbit around the donor (Si) core ion (which has in this description a charge of \( +|e| \)). Since it concerns a shallow donor, the state has to obey the symmetries of the atomic lattice and the \( \sim10\text{-nm-radius} \) (in zero field spherical) envelope wave function, while this latter symmetry is distorted towards cylindrical cyclotron states when a magnetic field is applied. In addition, the three charge carriers have interactions at an energy scale similar to that of the confinement potentials, and spin-orbit couplings are significant. A theoretical model that captures all aspects of this system’s behavior has not been presented yet, and will be very complex. Approximations for this, and theoretical concepts underlying the complete behavior, will be introduced in Section 3.2 and throughout the main text.

A further outline of this chapter is as follows. Section 3.3 presents the approach and results for single-laser spectroscopic studies, and Section 3.4 presents this for two-laser spectroscopic studies. The results are collected and analyzed in Section 3.5, and Section 3.6 presents the conclusions. Section 3.7 is an appendix where we present an overview of various notations used in the literature, since the use of these has not been consistent, and we ran into several (apparent) errors or points of confusion. Two more appendices present details of our analysis.

3.1.1 Materials and experimental methods

We will only provide here a brief summary of the \( n \)-GaAs material and experimental methods used for the work presented in this chapter, since most aspects are identical to those presented in Chapter 2 and Chapter 5. Specific details of how single- and two-laser spectroscopy were carried out for the results presented in this chapter are discussed in the sections where these results are presented.

All spectroscopy results were obtained from laser transmission studies on a \( 10\mu\text{m} \) thick GaAs layer that contained silicon atoms as donor impurities at a concentration of \( \sim3 \times 10^{13} \text{ cm}^{-3} \). The material was MBE grown along the \([001]\) crystal direction (orthogonal to the layer). All results were obtained at 4.2 K. The magnetic field was applied parallel to the \([110]\) crystal direction, with magnitudes...
from $B = 0$ T to $B = 9.4$ T. The propagation of the laser fields was along the growth axis, and the fields reached the sample with linear polarization that was either parallel (denoted as V[ertical]) or orthogonal (denoted as H[orizontal]) to the direction of the magnetic field. This chapter only presents results where shifts of the $D^0X$ levels due to strain in the material are not significant (conclusion based on comparison with results in the literature [2] and related results from our team [3]).

3.2 Properties and models of the $D^0X$ system in GaAs

Building on the basic descriptions of the $D^0$ and $D^0X$ systems in Section 1.3, we will present here further properties and models of the $D^0X$ system in GaAs. This system has already been studied for decades (with many corrections on the way), and state-of-the-art results and an authoritative review are provided by Ref. [4], and more recent extensions of this work are reported in Ref. [5]. As a basis for using these reports, we also used several earlier publications [6–23].

![Figure 3.1: Orbiting-particle illustrations of the $D^0X$ system with two electrons and one hole bound by the Si-donor core ion. Theoretical modeling assumed as starting points (a) that an exciton (bound electron-hole pair) is in orbit around a non-ionized Si donor, or (b) that the Si core ion has two electrons in a singlet state tightly bound to it, and that this system has—in turn—a hole in orbit around it. Further refinements of the modeling considered superpositions of the two cases.](image-url)

The physical picture that emerges is that the $D^0X$ has two electrons and one hole localized in a bound state around the core ion of the Si donor. For the lowest energy levels, the two electrons are always in a singlet state (the triplet state has much higher energies, never observed as a stable state). In a magnetic
field, the $D^0X$ system still shows a set of at least 6 different energy eigenstates (discrete $D^0X$ levels) that are easily observed. These levels result from two different degrees of freedom.

First, there are the Zeeman sub-levels of the hole. The hole states at the top of the valence band have a wave function (at the scale of the lattice) that has a total angular momentum characterized by quantum number $j = \frac{3}{2}$, and quantum numbers $m_j = \pm \frac{1}{2}, \pm \frac{3}{2}$ for the component along the field. These four eigenstates are often described as the four spin (or quasi-spin) levels of the hole, even though they are in fact eigenstates of the spin-orbit coupled electronic state.

Second, there are at low energies different states for the angular momentum of the envelope wave function of the orbiting complex with two electrons and one hole (which are for the bound state strongly interacting particles). This envelope wave function has azimuthal quantum number $L$, and magnetic quantum number $M_L$. The lowest optically active states have $L = 0$ (and thereby $M_L = 0$), or $L = 1$ and $M_L = 0$. The literature never discusses the states with $L = 1$ and $M_L = \pm 1$ (and only on a few occasions $L = 2, 3$, mainly for $B = 0$ T [5, 17]), presumably because they have much higher energy, or because selection rules associated with the envelope wave function prohibit transitions into these states from the $D^0$ ground state.

As discussed in the previous section, detailed modeling of the $D^0X$ states in magnetic field is still beyond the capabilities of modern theoretical physics. Various approximations have been tried (see e.g. Ref. [12], illustrated in Fig. 3.1), but even the very extensive work of Ref. [4] still concludes that it can only link part of the observed levels to theoretical descriptions.

For our goal to assign $m_j$ quantum numbers to $D^0X$ levels, a proper approach should account for the fact that in strong magnetic fields along crystal directions that differ from <100> (in our studies we use [110], building on the results of Ref. [5]), $m_j$ is no longer a good quantum number ($m_j$ states get mixed). We introduce this better in Appendix 3.8. At this stage we did not perform a complete analysis ourselves for expected behavior of this effect. Further, we find that a significant part of the literature discusses $m_j$ values for the $D^0X$ hole while (seemingly) fully ignoring this mixing of $m_j$ states. We carried out and report our analysis in a manner that can link our results to the literature, and also expect that at the field values used in our study the levels are still dominated by one particular $m_j$ state.
3.3 Single-laser spectroscopy

For our studies in magnetic fields between $B = 0$ T and 2 T we used transmission studies with a single scanning laser. The photodiode was directly behind the GaAs layer in the cryogenic measurement volume. For improving signal-to-noise, the laser beam was chopped at 6 kHz and the photodiode signal was recorded with a lock-in amplifier. The laser intensities were typically a factor 10 lower than those used in Chapter 2 for the EIT studies. The signals were corrected for small drifts in the laser power by normalizing them to a reference signal (here, and for all other results in this chapter).

Figure 3.2(a) presents results of such measurements for $B = 0$ T. The results for the laser field with H and V polarization were identical. At spectral positions far away from material absorptions, the transmittance (defined as the ratio between transmitted and incident optical powers) of the system shows Fabry-Pérot oscillations that modulate the transmittance between values of $\sim 0.4$ and $\sim 0.9$ (for a detailed analysis see Ref. [3]). The Fabry-Pérot effects are mainly due to reflections at the front and back side of the GaAs layer. At the frequency where we resonantly excite the $X_{n=1}$ free exciton, the transmittance is close to zero. In our report here we further mostly present lock-in signals in arbitrary units, as a generic transmission signal. If desirable, the actual values for the corresponding transmittance can be estimated by looking up the spectral lines in Fig. 3.2(a).

The most prominent spectral lines in Fig. 3.2(a) are the broad dips for the $X_{n=1,2,3,...}$ free exciton resonances. The associated modulation of the index of refraction of the GaAs layer is near these resonances so strong that it chirps the Fabry-Pérot oscillations. The spectral distance between the $X_n$ lines for $n = 1, 2, 3...$ is well described by the hydrogen-like series levels for the free exciton [21] (for $n = 2, 3$ we observed them more clearly as separate lines in in other data sets). Spectral lines from $D^0-D^0X$ transitions are indicated with red arrows. These have been identified before [4, 5], and the lowest two lines (see also inset) are transitions to a degenerate set of $D^0X$ levels with $L = 0$ (lowest energy) and $L = 1$.

Figure 3.2(b) presents similar measurements for $B = 1$ T, taken with H and V polarization for the laser field. The inset focusses again on transitions to the $D^0X$ levels with $L = 0$ (lowest energies) and $L = 1$. As compared to the data for $B = 0$ T, all features of the spectrum show a significant shift to higher energies, predominantly due to diamagnetic shifts. For the $D^0-D^0X$ transitions, this has contributions from both the diamagnetic shift of the $D^0$ systems and the $D^0X$...
Figure 3.2: (a) Transmission spectrum of a GaAs layer from a single-laser scan at zero magnetic field (results identical for V and H polarization). The spectrum carries signatures of $D^0-D^0X$ transitions (six red arrows, see main text), excitation of free excitons ($X_n$, with $n = 1, 2, 3, ...$), and oscillations from the Fabry-Pérot effect in the GaAs layer (with a chirped wavelength dependence, in particular chirped around the very strong $X_{n=1}$ transition due to the associated wavelength dependence of the refractive index). The transmittance of the system varies between near-zero at the $X_n$ dips and a value of $\sim 0.5$ where only the Fabry-Pérot effect modulates the transmission. The inset zooms in on the two lowest $D^0-D^0X$ transitions, which are for $L = 0$ and $L = 1$. (b) Similar transmission spectrum recorded for an applied magnetic field of 1 T, for V polarization (red) and H polarization (blue), again with an inset zooming in at the lowest two $D^0-D^0X$ transitions.
system.

For the $D^0-D^0X$ transitions, both the $L = 0$ line and $L = 1$ line are in fact a manifold of multiple lines that have level splittings that are smaller than the width of these lines. Thus, the energy distance between the $L = 0$ line for the red and blue data cannot be simply interpreted as the Zeeman splitting for the $D^0$ electron. The various levels (for example levels for different $m_j$ values) that belong to the $L = 0$ and $L = 1$ manifolds of $D^0X$ also have Zeeman shifts, and the transition strength into each of them (from a particular $D^0$-electron spin state) will not be the same.

Without bringing in further knowledge about the $D^0X$ levels, these results can thus only give a limited contribution to our $D^0-D^0X$ magneto-spectroscopy. For fields below $B \approx 2$ T, the individual levels of the $L = 0$ and $L = 1$ manifolds cannot be resolved, and here these results only provide information about the overall diamagnetic shift for the associated transitions. At $B \approx 2$ T, the single-laser approach also stops working well, since optical pumping into one of the two $D^0$ spin levels causes a loss of signal.

### 3.4 Two-laser spectroscopy

This section presents results from a two-laser spectroscopy approach to studying the $D^0-D^0X$ transitions, for fields $B > 2$ T. The main benefit as compared to the single-laser approach is that it remedies the loss of signal due to optical pumping into one of the two $D^0$ spin levels. Before presenting the results, we first introduce the experimental method, and present high-resolution measurements of the Zeeman splitting of the $D^0$ electron. The latter is needed for deriving the energies of $D^0X$ levels from measured values of $D^0-D^0X$ transition energies.

#### 3.4.1 Experimental method

Our studies with this two-laser approach were again transmission studies with the photodiode directly behind the GaAs layer. For improving signal-to-noise, one of the laser beams was chopped at 6 kHz and the photodiode signal was recorded with a lock-in. One laser was fixed at a frequency where it counteracts optical pumping into one of the two spin states of the $D^0$ system (suitable transitions for this were identified beforehand, using the results of Chapter 2, see also Fig. 3.3). The optical frequency of the other laser was scanned over the range where $D^0-D^0X$ transitions occur. Unlike the results in Chapter 2, the chopper
3.4 Two-laser spectroscopy

![Energy-level schematics](image)

**Figure 3.3:** Energy-level schematics (not to the scale, and with $D^0X$ levels labeled $A$, $B$, $C$, ..) for illustrating how with two lasers a pump-assisted spectroscopy (PAS) technique was used for identifying $D^0$-$D^0X$ transitions. Four different configurations are labeled T1, T2, T3, and T4. The colored arrows represent laser fields with H (blue) and V (red) polarization. For the configuration T1, a V-polarized laser field is fixed in frequency to efficiently drive transition from $|\downarrow\rangle$ to one of the lowest levels (labeled $A$) of $D^0X$, such that it counteracts any optical pumping into $|\downarrow\rangle$. An H-polarized laser field scans across $D^0$-$D^0X$ transitions, mainly causing signal when transitions out of $|\uparrow\rangle$ are addressed with this polarization. The PAS signal arises from lock-in detection synchronized to chopping of the fixed laser beam (see main text). For configurations T3 and T4, the fixed laser efficiently drives transitions from $|\uparrow\rangle$ to level $B$ of $D^0X$. 

was in the fixed laser beam. Due to this, \(D^0-D^0X\) transitions are observed as transmission dips in a nearly flat background (see Fig. 3.5(a), much less influence of the Fabry-Pérot effect is seen, for a detailed analysis see Ref. [3]). A signal occurs when both the two lasers drive a transition out of one of the \(D^0\) spin levels (competitive optical pumping, or cross modulation). We call this method pump-assisted spectroscopy (PAS). The laser intensities were again typically a factor 10 lower than those used in Chapter 2 for the EIT studies (see also Ref. [3]).

<table>
<thead>
<tr>
<th>Label</th>
<th>Polarization of scan laser</th>
<th>Signal due to pumping out of</th>
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<tbody>
<tr>
<td>T1</td>
<td>H</td>
<td>(</td>
</tr>
<tr>
<td>T2</td>
<td>V</td>
<td>(</td>
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<td>T3</td>
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</tr>
<tr>
<td>T4</td>
<td>V</td>
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</tbody>
</table>

**Table 3.1:** Summary of the essence of the T1, T2, T3 and T4 measurement scheme.

Figure 3.3 presents in more detail how the above scheme was implemented. In particular, we carried out measurements that focussed on transitions out of the \(|\uparrow|\) and \(|\downarrow|\) state of the \(D^0\) system, and studied this with H- and V-polarization for the scanning laser. This gives four measurement configurations, that we label T1, T2, T3 and T4 (see Fig. 3.3). For easy referencing, we summarize the main features of each configuration in Table 3.1.

### 3.4.2 Measuring the \(D^0\) Zeeman splitting via EIT

Figure 3.4 presents results for determining the Zeeman splitting for the \(D^0\)-1s electron with high accuracy via EIT measurements as presented in Chapter 2 (we verified that for these measurements the EIT-peak position had no significant shift from DNP effects). A fit of the observed Zeeman splitting \(\left(E_{\text{Zeeman}}\right)\) in this figure to the function \(E_{\text{Zeeman}} = \gamma_1 B + \gamma_2 B^2\) yields \(\gamma_1 = 6.346 \pm 0.004\) GHz T\(^{-1}\) and \(\gamma_2 = -0.0796 \pm 0.0005\) GHz T\(^{-2}\). The (field-dependent) effective Landé g-factor \(g = E_{\text{Zeeman}}/\mu_B B = h/\mu_B \cdot (\gamma_1 + \gamma_2 B)\) can thus be determined with an accuracy of about one part in thousand (\(\mu_B\) is the Bohr magneton). We use these values in our analysis of the \(D^0-D^0X\) transition energies in the remainder of this chapter. As an indication for the values of the electron g-factor that result from this expression, we present them here for three values of \(B\) (minus sign taken from the literature):

\[
g = -0.4534 \pm 0.0003 \text{ for zero field},
\]
3.4 Two-laser spectroscopy

Figure 3.4: The Zeeman splitting for the $D^0$-$1s$ electron, determined with high precision by deriving it from the EIT resonance (measured as in Chapter 2), as a function of applied magnetic field. The red dashed line going through the origin is a guide to the eye for linear dependence, and shows that the measured Zeeman splitting has a weakly nonlinear dependence on magnetic field. The green curve is a quadratic fit, see main text.

$$g = -0.4170 \pm 0.0006 \text{ at a field of } B = 6.4 \text{ T, and}$$

$$g = -0.3965 \pm 0.0007 \text{ at a field of } B = 10 \text{ T.}$$

These observations are in good agreement with earlier high-resolution studies of the $D^0$-electron Zeeman splitting [4, 5].

3.4.3 Two-laser spectroscopy of $D^0X$ levels

Figure 3.5(a) presents results from the two-laser spectroscopy schemes T1..T4, introduced in this section. Resonances with $D^0$-$D^0X$ transitions appear as dips. The results for T3 and T4 have been shifted by $E_{Zeeman}$ to higher energies, such that dips for a transition to a particular $D^0X$ level appear at the same optical frequency as for T1 and T2 results. The traces are normalized to a ~0.1 reduction of the signal as in Fig. 3.2(a).

The traces in Fig. 3.5(a) show prominent dips for known transitions (Chapter 2), and various smaller features which can either result from weaker $D^0$-$D^0X$
Figure 3.5: (a) Normalized pump-assisted spectroscopy (PAS) results from configurations T1-T4 (see main text, traces offset for clarity). Resonances with $D^0-D^0X$ transitions appear as dips in the transmission signal. Results from T3 and T4 are displayed after shifting them to higher frequencies over a distance that corresponds to $E_{\text{Zeeman}}$, such that resonance frequencies (dip positions) for specific $D^0X$ levels observed in T3 and T4 traces coincide with the corresponding resonances in T1 and T2 traces. The occurrence of dips thus identify the energy of $D^0X$ level with respect to the energy of the $|\uparrow\rangle$ level. (b) Traces that represent the product of trace T1 and T3 of panel (a) (baseline subtracted and inverted), and similar cases as labeled. Peaks in the T1×T3 trace only occur when both the T1 and the T3 trace contain a dip from a transition to a particular $D^0X$ level, from $|\uparrow\rangle$ and $|\downarrow\rangle$ respectively. This representation shows peaks (labeled A, B, C,...) that reflect $D^0-D^0X$ transitions while uncorrelated noisy structure in the original T1 and T3 traces is suppressed. It also helps with evaluating the polarization dependence of the transitions, see main text.

transitions (given the polarization of the scanning laser) or from spurious effects such as a residual influence of the Fabry-Pérot effect in the GaAs layer. To better discriminate these cases, we identify $D^0-D^0X$ transitions (and better identify...
their sensitivity to either H- or V-polarization) by processing these signals as in Fig. 3.5(b). This plot presents products of signals such as $T_1 \times T_3$ (original signals inverted and background set to zero). These product-signals (such as $T_1 \times T_3$) only show peaks when dips at a particular frequency were present in both the $T_1$ and $T_3$ trace, thus removing a portion of the spurious contributions. Further discrimination between peaks of weak transitions and spurious structure on these traces was obtained by following whether the peaks consistently keep appearing when altering the magnetic field in small steps (see Fig. 3.6). By also looking at the height and polarization dependence of the peaks, this approach also confirmed that the order of peaks did not have crossings in the field range $B = 2.5 \text{T}$ to $9.4 \text{T}$. In Fig. 3.5(b), we label peaks as $A$, $B$, $C$,... in a manner that agrees with how we used this labeling in earlier publications from our team. However, as presented in Fig. 3.6(c), we will introduce new generic labeling Line 1..Line 8 for the observed lines with the 8 lowest energies, for an analysis independent of earlier assumptions.

Notably, a peak in Fig. 3.6(c) directly identifies the energy of a $D^0X$ level with respect to the energy of the $|\uparrow\rangle$ state of the $D^0$ electron. In our further analysis as a function of magnetic field, we will always present the energies of $D^0X$ levels with respect to the energy of the $|\uparrow\rangle$ level at each particular magnetic field. The observed shifts in the energy of the $D^0X$ level as a function of field thus have contributions that include Zeeman and diamagnetic shifts from both the $|\uparrow\rangle$ level of $D^0$ and the particular $D^0X$ level.

Energies of $D^0X$ levels derived in this manner (from the set peaks labeled as Line 1..Line 8, Fig. 3.6(c)) were analyzed from data as in Fig. 3.5 obtained at magnetic fields between $B = 2.5 \text{T}$ and $9.4 \text{T}$. The results are presented in Fig. 3.6(a). This figure also presents data for $B < 2 \text{T}$, for the $D^0X$ levels with $L = 0$ and $L = 1$ (for these points we applied a corresponding approach for defining the energies with respect to $|\uparrow\rangle$ [24]). The full data set presented in this manner shows that all transitions associated with Line 1..Line 8 show a strong energy increase with field (about 1000 GHz over 9.4 T) due to diamagnetic shifts, while splittings between $D^0X$ levels increase continuously, up to values on the order of 50 GHz at 9.4 T. The diamagnetic shift shows the expected behavior of a parabolic dependence on $B$ at low fields, crossing over to a linear dependence at high fields [21], with the cross-over at about 5 T [4, 5, 21]. In the next section we aim to assign quantum numbers $L$ and $m_j$ to the $D^0X$ levels associated with Line 1..Line 8, and we aim to link and compare several aspects of this data set to earlier $D^0X$ studies that have been reported.
Figure 3.6: (a) Magnetic-field dependence of the lowest 8 observed $D^0 X$ levels, characterized as a transition frequency with respect to the $|\uparrow\rangle$ level. For fields $B > 2$ T, the level positions are derived from results as in Fig. 3.5(b) (also in panel (c), for $B = 6.4$ T). For fields $B < 2$ T, only two levels can be identified from results as in Fig. 3.2 (also in panel (b), for $B = 0$ T).
3.5 Analysis and discussion

Using various approaches (in part explained here, in part further below), Table 3.3 presents the outcome of our attempt to assign quantum numbers $L$ and $m_j$ to the $D^0X$ levels associated with Line 1..Line 8. We assumed that all of these lowest levels should have quantum number $L = 0$ or $L = 1$ [4, 5]. Assigning a value of $L$ is in part based on checking whether a full fit to a line (details explained in Appendix 3.9) yields a smooth connection between the high-field data and either the $L = 0$ or the $L = 1$ data at low field. A clear case is illustrated in Fig. 3.9: trying a fit (gray) that connects the high-field data for Line 2 to the $L = 0$ low-field data gives a much more convincing result than such a fit (red) to the $L = 1$ low-field data. Further, we studied the splittings between pairs of lines (see for example Line 4–Line 2 and Line 8–Line 6 in Fig. 3.8), for the high-field data ($B > 2.5$ T). In some cases this showed linear behavior (Fig. 3.8(a)), while in another cases it clearly differed from linear behavior (Fig. 3.8(b)). Behavior that strongly differs from linear points to a different diamagnetic shift for the two lines, and thereby a different $L$ value for the two lines.

For assigning an $m_j$ value to a $D^0X$ level, we looked at the polarization dependence of the line. A clear example is Line 2 (see Table 3.3): it has a strong transition from $|↑\rangle$, but only for H polarization, and a strong transition from $|↓\rangle$ for V polarization. This points to a $\Delta m_j = \pm 1$ transition [21] from $|↑\rangle$ and a $\Delta m_j = 0$ from $|↓\rangle$ (see also Chapter 2), and thus to $m_j = -\frac{1}{2}$ for the $D^0X$ level. Another rather clear example is Line 8: here there is only a strong response from $|↑\rangle$ for H polarization ($\Delta m_j = \pm 1$), which points to $m_j = +\frac{3}{2}$ for the $D^0X$ state. However, for several of the lines the polarization dependence does not yield a clear indication for an $m_j$ value that governs the level predominately.

A different approach to finding a match with the physical picture presented in Section 3.2 is to search for a group of four lines with energy splittings between them that behave as the Zeeman splittings of the hole for the lowest $D^0X$ levels (electronic state with $j = \frac{3}{2}$ and $m_j = -\frac{3}{2}, -\frac{1}{2}, +\frac{1}{2}, +\frac{3}{2}$). The expected linear level splittings (in GHz T$^{-1}$) are illustrated in Fig. 3.7. The relevant g-factors for the $m_j = \pm \frac{3}{2}$ and $m_j = \pm \frac{1}{2}$ states are calculated in Appendix 3.8. Table 3.2 presents the observed splittings between Line 1..Line 8, derived from fits as presented in Fig. 3.8. We now search for a correspondence between a part of Table 3.2 and Fig. 3.8(b). Since lines may have crossed before the field reaches 2.5 T, correspondence may appear in a pattern with four lines of Line 1..Line 8 that are not consecutive. Examples that roughly match are formed by:
Table 3.2: This table summarizes how the measured splitting between a Line \(N\) and a Line \(M\) (where \(N, M = 1..8\)) increases as a function of magnetic field. It is analyzed by fitting a linear dependence (through zero). The results are presented as a value in GHz T\(^{-1}\). The uncertainty for each value is about ±1 GHz T\(^{-1}\). The indications *Other diamagnetic shift* for Line 8 indicate that the splitting with the other Lines 1-7 is clearly not linear (see main text).

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<th>Line 4</th>
<th>Line 5</th>
<th>Line 6</th>
<th>Line 7</th>
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<tr>
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<td></td>
<td></td>
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<tr>
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<td>4.0</td>
<td>-</td>
<td></td>
<td></td>
<td></td>
</tr>
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<td>7.5</td>
<td>3.5</td>
<td>-</td>
<td></td>
<td></td>
</tr>
<tr>
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<td>12.2</td>
<td>8.1</td>
<td>4.7</td>
<td>-</td>
<td></td>
</tr>
<tr>
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<td>14.8</td>
<td>10.8</td>
<td>7.3</td>
<td>2.6</td>
<td>-</td>
</tr>
<tr>
<td>Line 7</td>
<td>22.1</td>
<td>18.7</td>
<td>14.7</td>
<td>11.2</td>
<td>6.5</td>
<td>3.9</td>
</tr>
<tr>
<td>Line 8</td>
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<td>23.7</td>
<td>19.7</td>
<td>16.2</td>
<td>11.5</td>
<td>8.9</td>
</tr>
</tbody>
</table>

Other diam.

Other diam.

Other diam.

Other diam.

Other diam.

Other diam.

Other diam.

| Line 1, 2, 4, 5; Line 2, 3, 5, 6; Line 3, 4, 6, 7. However, none of these sets then also show consistent agreement with the values for \(m_j\) as presented in Table 3.3 (not even separately for a pair \(m_j = \pm \frac{3}{2}\) for the outer two levels, or \(m_j = \pm \frac{1}{2}\) for the inner two). We thus conclude that we cannot convincingly identify a set of four (or even two) lines with the behavior of a manifold with \(m_j = -\frac{3}{2}, -\frac{1}{2}, +\frac{1}{2}, +\frac{3}{2}\), that at the same time also all have the same quantum number \(L = 0\) or \(L = 1\).

Despite this limited success for assigning quantum numbers to \(D^0X\) levels, our results do establish –in particular via the strong lines that we labeled \(A\) (Line 2) and \(B\) (Line 3)– the correspondence with related results in the literature [4, 5], both for the polarization selection rules and the magneto-spectroscopic shifts (see Appendix 3.9). There is only one outspoken difference with Ref. [5]: our data favors assigning \(L = 0\) to level \(A\) instead of \(L = 1\).
3.6 Conclusions

In this chapter we presented a full magneto-spectroscopic study of the lowest 8 optically-active $D^0X$ levels, and the associated optical transitions, for fields...
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### Figure 3.7: (a) A schematic illustrating the theoretically expected Zeeman shifts, and the corresponding splittings between levels (with values in GHz T$^{-1}$), for the four $m_j$ levels of a $j = \frac{3}{2}$ hole. (b) In relation to Table 3.2, the theoretically expected Zeeman splittings (in GHz T$^{-1}$) presented in table form.

### Table 3.2: The theoretically expected Zeeman splittings (in GHz T$^{-1}$) for the four $m_j$ levels of a $j = \frac{3}{2}$ hole.

<table>
<thead>
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<th>Line rows</th>
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<td>~10.3</td>
<td>~12.8</td>
</tr>
<tr>
<td>~7.9</td>
<td>~2.5</td>
</tr>
</tbody>
</table>

### Figure 3.8: (a) The observed level splitting (dots) between Line 4 and Line 2 of Fig. 3.6(a). The gray line is a linear fit through the origin. (b) Similarly, the level splitting between Line 8 and Line 6 of Fig. 3.6(a).

between 0 and 9.4 T. We concentrated on the case where the field is applied along the [110] crystal direction. While the experimental results establish at a phenomenological level detailed information about the spectrum and the polarization selection rules, our goal to obtain insight in the quantum numbers $L$ and $m_j$ of the $8 D^0 X$ states had only limited success. The results do establish how the results obtained with our configuration and measurement method link to state-of-the-art results in the literature [4, 5]. The spectroscopic study did not reveal a reason for the large set of results by our team that indicate that observing the EIT effect between 7 T and 8 T is much more difficult than at other magnetic fields. Notably, related work in our team showed that this can also not be at-
3.6 Conclusions

Figure 3.9: Fits (gray, red) of the combined diamagnetic and Zeeman shift for Line 2 (dots, high field) of Fig. 3.6(a), either assumed to link to the $L = 0$ (gray) or $L = 1$ (red) observations for low fields (see main text for details). For the Zeeman shift the fit assumes Line 2 is for a hole with $m_j = -\frac{1}{2}$.

tributed to Fabry-Pérot effects in the GaAs layer [3], and the physics behind this observation thus remains an open problem.
Acknowledgements

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3.7 Appendix: Notes on notations used in the literature

Rather than using the labels Line 1..Line 8 for the observed $D^0X$ levels, in other chapters of this thesis and in related publications from our team we label $D^0X$ levels as $A$, $B$, $C$,.. (see Table 3.3). Here the associated transitions are labeled as $A$ when it is an excitation from $|\downarrow\rangle$, and $A^*$ when it is an excitation from $|\uparrow\rangle$, see Fig. 3.3.

We used this notation following the earlier work by Fu et al. [5, 18], and in particular assign the same label to the transitions $A$, $A^*$ and $B$, $B^*$. Fu et al. also applied the magnetic field along [110]. The difficulty in assigning quantum numbers to $D^0X$ levels is from this body of work also apparent from the fact they initially assigned $L = 0, m_j = -\frac{3}{2}$ to the $A$ level [18], while the later publication of the same data reported it as $L = 1, m_j = -\frac{1}{2}$ [5]. In other chapters of this work, $A$ and $B$ were associated with $L = 0$, while low-energy levels associated with $L = 1$ were labeled as $A_1$, $B_1$, etc.

In turn, Fu et al. used their notation following Karasyuk et al. [4]. The use of $A$, $A^*$ and $A_1$ etc. by Karasyuk et al. is thus very similar (while $a$, $a^*$ and $a_1$ refer to transitions that relax $D^0X$ states to $2p$ levels of $D^0$ rather than the 1s ground state). It should be noted that for most of the studies Karasyuk et al. applied the magnetic field along [100]. This also has consequences for the $g$-factors and order of $m_j$ labeling on Zeeman levels of the $D^0X$ hole (better defined in the next section): $g_{\frac{1}{2}} = -0.32$ is negative while $g_{\frac{3}{2}} = +0.35$ is positive. This notation and $m_j$ assignment is also used in the recent Refs. [19, 23]. Concerning Ref. [4], they use $J$ for the total angular momentum (electronic orbital and spin) of the full $D^0X$ system, while $j$ refers to that of the $D^0X$ hole alone (but not strictly consistent). A later part of the publication uses $I$, $M_I$ for the total angular momentum of the full $D^0X$ system. In other parts, they switch to using the notation $S_{3/2}, P_{3/2}$, etc. (where according to conventional spectroscopic notation $S$ refers to $L = 0$, $P$ to $L = 1$, and the subscript to the value of $J$ as introduced
3.8 Appendix: Derivation of hole g-factors

In this Appendix we present the effective g-factors for the hole states for the situation that applies to our experiments. It should be noted that there are two different conventions in parallel for reporting g-factor values. In particular, the Zeeman energy of a (quasi-)spin level (not a spin splitting), characterized by a particular \( m_j \) value, is either written as \( E_{m_j} = g_{\text{case1}} \mu_B B \) or \( E_{m_j} = m_j g_{\text{case2}} \mu_B B \) (the latter case is commonly used for the electron). For the expression with \( g_{\text{case1}} \) the factor \( m_j \) is incorporated in the g-factor. For the hole g-factors reported here we will use case 1.

For calculating the hole g-factors we use Ref. [7]. This publication gives for the Zeeman contribution \( E_{m_j} \) to the energies of hole states with \( j = \frac{3}{2} \) and \( m_j = -\frac{3}{2}, -\frac{1}{2}, \frac{1}{2}, \frac{3}{2} \) (using Eq. 56a-d of this publication, which applies for \( B \) along [110] and the \( \Gamma_8 \) representation of \( j = \frac{3}{2} \) holes, see also Ref. [23], start of Section III). For introducing symbols and notation see Ref. [25]. This yields

\[
E_{\pm \frac{3}{2}} = \mu_B g_2 B \left[ \pm \Delta \pm \left( \frac{1}{2} \frac{g_1}{g_2} + \frac{7}{8} \right) \right], \quad (3.1)
\]

and

\[
E_{\pm \frac{1}{2}} = \mu_B g_2 B \left[ \pm \Delta \mp \left( \frac{1}{2} \frac{g_1}{g_2} + \frac{7}{8} \right) \right]. \quad (3.2)
\]

Notably, \( m_j \) is for a strong field along [110] no longer a good quantum number (see Ref. [11], top left p. 7015 and top left p. 7017). We can still use \( m_j \) as labels since the levels evolve out of these \( m_j \) states at zero field. This is the reason that we should calculate effective g-factors \( g_{\frac{3}{2}} \) and \( g_{\frac{1}{2}} \) from the material parameters \( g_1 \) and \( g_2 \), which are the bare spherically symmetric and asymmetric g-factors.

The values for GaAs for \( g_1 = -0.40 \) and \( g_2 = +0.33 \) are given in Ref. [4] (Table III, which uses notation \( g_1 \) and \( g_2 \)). When expressing the energies of the
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(quasi-)spin levels as

\[ E_{\pm \frac{3}{2}} = \pm g_{\frac{3}{2}} \mu_B B \]  

and

\[ E_{\pm \frac{1}{2}} = \pm g_{\frac{1}{2}} \mu_B B \]  

the valid hole g-factor values are

\[ g_{\frac{3}{2}} = +0.485, \]  

\[ g_{\frac{1}{2}} = +0.281. \]  

For an applied field along [100] the equations corresponding to the above result in

\[ g_{\frac{3}{2}} = g_1 + 9 \frac{3}{4} g_2 \] and \[ g_{\frac{1}{2}} = g_1 + \frac{1}{4} g_2, \] which results in \[ g_{\frac{3}{2}} = +0.35 \] and \[ g_{\frac{1}{2}} = -0.32 \] \[ \text{(Eq. 19), [4].} \]

3.9 Appendix: Fitting the magneto-spectroscopy results

The traces as in Fig. 3.9 contain energies \( E_{\text{Line}N} \) of \( D^0X \) levels (each associated with a Line \( N \)), measured with respect to the energy of the state \( |\uparrow\rangle \) of the \( D^0 \) system. Thus, to analyze and fit the energy shift in magnetic field, we set up an approach that accounts for four terms,

\[ E_{\text{Line}N} = E_0 + \frac{1}{2} E_{\text{Zeeman}} + E_{m_j} + E_{\text{diam}}. \]  

The first term \( E_0 \), is a phenomenological constant, and is simply the observed energy associated with the \( L = 0 \) or \( L = 1 \) line at \( B = 0 \) T. The second term \( \frac{1}{2} E_{\text{Zeeman}} \) (see Section 3.4.2) accounts for the Zeeman shift of \( |\uparrow\rangle \). The third term (in some cases applied by trial and evaluation for finding a consistent picture) is the Zeeman term for the \( D^0X \) level, which is the term \( E_{m_j} \) as presented in Appendix 3.8. For this we need to use (or try) an \( m_j \) value, for which we use the findings presented in Table 3.3. The fourth term accounts for diamagnetic-shift contributions from both the \( |\uparrow\rangle \) level and the relevant \( D^0X \) level. We follow Ref. [4] and incorporate that into one single phenomenological expression for the two diamagnetic-shift contributions, since taking them separately gives too many free parameters. The expression used is

\[ E_{\text{diam}} = \mu_B \frac{m_e}{m^*} \sqrt{B_0^2 + B_1 B + B^2}. \]  

\[ \text{(3.8)} \]
Here $m_e$ is the free electron mass, $m^*$ is a phenomenological effective mass value that is some mixed value for that of the electron and the hole (indicating the expected order of magnitude).

Thus, we set up least-squares fitting with only three free parameters: $m^*$, $B_0$ and $B_1$. Notably, for traces as in Fig. 3.9 (and after removing the Zeeman contributions), $B_1$ governs the slope at low fields, $m^*/m_e$ governs the asymptotic slope at high fields, and $B_0$ sets the field where the trace crosses over from parabolic to linear. Since the behavior of holes in GaAs is highly anisotropic, the values of $m^*$, $B_0$ and $B_1$ can show a weak dependence on the value of $L$ and $|m_j|$ [4].

The character of this model for fitting yields a strong inter-dependence between the resulting values for $m^*$, $B_0$ and $B_1$, and could yield good fits (similar to the lines in Fig. 3.9) for a range of such values (defining an error-bar range). Here the value for $m^*/m_e = 0.075 \pm 0.005$, with the best fits consistent with the value $m^*/m_e = 0.0739$ reported by Ref. [4], such that we continued with a two-free-parameter approach, and $m^*/m_e$ fixed at $=0.0739$. This did yield $B_0 = 5 \pm 1$ T and $B_1 = 0.8 \pm 0.2$ T, where the error bar is dominated by the fact that similar quality fits can be obtained via inter-adjustment of the two fit parameters. These values also show agreement with the results in Ref. [4], both in value and possible depth and error bar of the analysis.

References


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[24] Here we took the average energy of the two peak positions as observed H- and V-polarization, and added \( \frac{1}{2}E_{Zeeman} \) using the data of Section 3.4.2.
For a complete introduction of notation we refer to the original publication. Notably, in the main text we immediatly write \( \frac{\delta_i}{\delta_0} \) rather than using \( p \), where \( p = \frac{\delta_i}{\delta_0} \). We apply the mentioned assumption that \( s_2 = s_3 = 0 \). This yields for the mentioned factors \( \Delta = \Delta_{\pm} = \frac{1}{4} \sqrt{16p^2 + 68p + 79} \). Also note that for our analysis we can leave out the term \( [s_1 + \frac{5}{4}(s_2 + s_3)]B \) since it is the same for all four \( m_j \) levels.
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