Optical control of mesoscopic spin ensembles in gallium arsenide
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Chapter 7

Magnetospectroscopy of the bound trion levels

Abstract

We present high-resolution magneto-optical transmission spectroscopy investigations of the trion levels in n-GaAs. Our results reveal the energy levels and the polarization selection rules of the corresponding transitions.
7.1 Introduction

The basic structure of the donor-bound electron and donor-bound exciton have been touched upon in section 2.1. Here we will proceed with a more detailed treatment and examine the energy levels by a spectroscopic study of these bound states. These results serve as a supplement for the earlier chapters because they show that the system we address is not strictly a three level Λ system, but actually consists of more levels. The results can be used to assess the accuracy of the Λ-system model and to judge the goodness of the three-level approximation in various experimental situations.

7.2 Motivation for the study

The structure of the donor bound exciton complex has been a subject of study since first proposed by Lampert [1]. In that work it was already pointed out that the energy levels of the complex can resemble those of the diatomic molecule $H_2$. This concept was further developed by Rühle and Klingenstein [2]. They considered the electrons to form a spin singlet with the hole in orbit, the hole angular momentum $J = 3/2$ coupled to the angular momentum of its motion around the impurity site can then explain much of the spectrum. In turn experimental work [3] verified parts of the theory by photoluminescence experiments.

Our intention was to add to this body of work our transmission spectroscopy results on this system. However, after doing a series of measurements presented in this chapter, it was realized that we could not improve on existing literature using this data and an improved approach is needed. Instead we present the results here in support of earlier chapters and focus on the method used to obtain the spectra.

7.3 Complementary spectroscopy

To identify the position of the energy levels of the bound exciton complex we make use of the fact that the differential transmission spectroscopy as presented in Section 8.4 can be performed in four distinct ways, these are
depicted in Fig. 7.1. One laser is fixed at resonance and intensity modulated by a chopper in the beam path, this laser is indicated by the dashed arrow in the figure. For its position, two strong transitions are chosen that can easily be found. The polarization of the fixed laser matches the transition that it is resonant with, as indicated in the figure. In addition to the fixed laser there is a scanning laser, which is not modulated and whose polarization is either H(orizontal) or V(ertical). Alternating between the position of the fixed laser and polarization of the scanning laser gives the four different types. In each case, as the scanning laser moves over a transition a dip will show in the transmission of the fixed laser, this modulated signal is recorded by a lock-in amplifier.

This type of two-laser spectroscopy is needed because the transitions are not resolved in single laser scans due to optical pumping. In Figure 7.2 the single laser spectra are shown at zero magnetic field and in a magnetic field of 6.4 T. At zero magnetic field various $D^0X$ states are indicated by red arrows. In our range of interest we find two of them which are distinguished, according to the model of Ruhle and Klingenstein by the orbital angular momentum of the hole. The inset shows a magnification of that region with the two resonances, here $\ell$ refers to the hole’s orbital motion in the trapping potential.

Out of the four types, the ones that have the fixed laser coupled to a transition from the $|\downarrow\rangle$ state are complementary to the ones where it couples to a transition from $|\uparrow\rangle$. Complementary is meant in the sense that they reveal the same energy levels, the states of the $D^0X$ system, only these will appear at an energy shifted by the electron spin Zeeman energy. We use the correlation between the spectra to identify level positions, as shown in Fig. 7.3.

7.4 Results

The position where resonances (shown as dips in transmission) occur in Fig. 7.3a depicts the distance of the excited state (in frequency units) to $|\downarrow\rangle$. The figure shows the result of the four different types of spectroscopy (labeled T1 to T4) with T3 and T4 shifted by the Zeeman energy so that their resonances overlap with T1 and T2. Because there is some inter-
Figure 7.1: Four complementary types of DTS, labeled T1-4. One laser (dashed arrow) is locked at a known transition while the other scans over the D⁰X region. Polarizations are H or V as indicated. The laser indicated by a dashed arrow is modulated by a beam chopper. Modulated transmission is recorded via lock-in detection as described in Section 8.4.
Figure 7.2: Transmission spectrum of the 10 µm thick GaAs film on sapphire. Upper panel: without external magnetic field. Red arrows indicate the positions of D^0X levels (see main text), X_{n=1,2,...} are the free exciton resonances and the inset shows the two lowest energy levels of the D^0X complex. Lower panel: with external magnetic field of 6.4 T, the red (blue) line is the spectrum for V (H) polarized light. The inset focusses on the region of the D^0X levels. Individual resonances are not well resolved in the presence of noise and interference effects.
Figure 7.3: Differential transmission spectroscopy (DTS) at magnetic field of 6.4 T, using two lasers in the configurations described in Fig. 7.1. Top panel (scan configurations labeled according to Fig. 7.1), the pair of thick lines (T3, T4) has been shifted by the electron spin Zeeman energy (see Fig. 6.3) such that resonances in the plot occur at the energy of \( D^0 X \) levels as measured from the ground state (\(| \uparrow \rangle \)). Lower panel: product of the graphs in the upper panel (as indicated in the legend), baseline subtracted and inverted. Peaks indicate the position of \( D^0 X \) levels in frequency units, measured from \(| \uparrow \rangle \). Relative peak heights of the different colors contain information about the polarization sensitivity of the transition to the particular level (from \(| \uparrow \rangle \) as well as \(| \downarrow \rangle \), see also main text).
ference on the signal and therefore especially smaller dips are difficult to identify, we look at the correlation between spectra as displayed in Fig. 7.3b. These graphs are obtained by subtracting a baseline from the graphs in Panel a, flipping them, and multiplying them as indicated in the legend. This method gives cleaner data in which resonances are more easily observed. Still by merely looking at the data it can remain unclear as to whether labels such as 'D?' truly indicate a resonance. This issue is resolved by looking at the full magnetospectroscopy result, meaning repetition of the measurement of Fig. 7.3 over a range of magnetic fields. For each magnetic field the correlation method is applied and peak positions and corresponding heights are indicated manually. The resulting graph is displayed in Fig. 7.4 where the colored closed dots are datapoints obtained from the two-laser spectroscopy and the open circles are obtained from single laser spectroscopy (which yields sufficient resolution only at fields up to 2 T). The y-axis of the plot indicates the difference of levels. The size of the closed dots indicates the corresponding peak height. Depending on the threshold set for peak selection (i.e. how high should a peak be as compared to the noise and other fluctuations) more candidates for levels can be found. For example a weak line is visible above the B level, more such lines show up when the threshold is reduced. From the relative heights (or dot sizes) of different color for a particular resonance, information about the polarization selection rules of the transition can be obtained. For example the level labeled A, which is mostly used for EIT measurements throughout this thesis can be verified to correspond to the $\ell = 0, m_j = -1/2$ state. This is understood as the two electrons forming a spin singlet and the hole being in it’s lowest rotational state with spin down, which corresponds to e.g. the observations of Karasyuk [3] and Fu [4].

7.5 Conclusion

The observations presented in this Chapter are useful to the earlier presented work on the physics in the Λ-system, because it helps to gauge how good the approximation of a Λ-system actually is (whether other levels might be involved or interfering) for a range of magnetic fields. On
Figure 7.4: Magnetospectroscopy results at low and high magnetic fields combined. D⁰X levels are displayed relative to the one with lowest energy (leftmost peak in lower panel of 7.3). The open circles, up to 2 T, are obtained by single laser spectroscopy (SLS) with V (red) or H (blue) polarized light. The two branches originate from the ℓ = 0 (lower) and ℓ = 1 (upper) levels indicated in Fig. 7.2. Between 2 T and 4 T the levels are not well resolved in both SLS and DTS. Upwards of 4 T the dots show DTS results (color coding as in legend, cf. 7.3). Between 7.5 T and 8 T there is a gap where the DTS signal becomes too noisy to resolve the levels.

The other hand it proved too difficult to derive the exact nature of the D⁰X states based on the data so far collected. The resolution of the spectroscopy method is sufficient but polarization inside the microscope is imperfect, as explained in Chapter 8. Furthermore, extra data should be collected by rotating the sample with respect to the external magnetic field as this changes the observed level structure (this is demonstrated in the experimental work of [3] and the theory of [5]). Finally there is an unidentified feature in the data of Fig. 7.3: The absence of datapoints along a vertical line around 7.5-8 T. Here (and only here) the spectra are
too noisy to observe resonances, meaning the spectroscopy method does not work. The reason for this is not understood so far.

References


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