Spin effects of charged exciton states in electric field tunable quantum dot molecules

Kushal C. Wijesundara¹, Allan Bracker², Dan Gammon², and Eric A. Stinaff³

¹Department of Physics and Astronomy and Nanoscale and Quantum Phenomena Institute, Ohio University, Athens, Ohio 45701-2979, USA
²Naval Research Laboratory, Washington, DC 20375, USA
Email: stinaff@ohio.edu

Abstract — Tuning the relative energy levels in coupled quantum dots with an applied electric field results in controllable spin interactions of bound carriers. These interactions may provide new directions in engineering these systems for optical and spintronic applications. Using polarization resolved photoluminescence experiments we observe spin dependent effects in the spectra which vary as a function of applied field. As we tune the exciton emission we observe variations in circular polarization memory in the neutral exciton and in the singly charged exciton states. We propose possible mechanisms for externally manipulating the spin coupling.

Keywords - Quantum dot molecules; excitons; nano devices; nano photonics

I. INTRODUCTION

It has been shown that the molecular nature of a carrier in a coupled quantum dot system can be tuned with an applied electric field [1]. This is important for quantum information processing as it provides a possible mechanism for manipulating entangled states. However, the molecular nature of the wave function can also introduce new and interesting effects as described in this article. With precise engineering these can lead to nano technological applications for optical and spintronics based devices. In quantum dot molecule (QDM) devices such as these, carrier tunneling may result in the formation of other charge states which is observed as the emergence of additional lines in the photoluminescence (PL) spectra with changing electric field [2], [3]. We observe evidence of such luminescence variations of the excitons that can be understood from carrier tunneling effects. Control of the electron hole exchange interaction in coupled quantum dot systems, via an applied electric field, may also provide a promising technique for spin control [4]. The spin dependent interactions in QDMs are investigated through polarization resolved photoluminescence spectra where we can clearly indentify the neutral exciton states and singly charged states. The effects of the anisotropic exchange interaction can be varied by either the introduction of additional charges resulting in different excitonic states in the QDM or by the spatial change in the carrier wavefunctions with applied electric field. In the single charged states, optically forbidden transitions are in principle useful as two-level systems [5]. These inaccessible states tend to be visible near anticrossings [6], and with an applied external field, variations in spin coupling along with exchange interaction can be observed in the spin fine structure. In the present work, to help understand these effects, we measure the variation of the circular polarization memory as a function of applied electric field for positive trion states which consist of two holes and an electron in the InAs/GaAs QDM system.

II. EXPERIMENTAL DETAILS

For the present study, the samples consist of InAs quantum dots (QD) in a GaAs matrix which has been epitaxially grown through the use of the Stransky-Krastanov technique. Quantum mechanical coupling is achieved via two InAs QD layers and a GaAs barrier between the two dots. Quantization energies of the individual layers of QDs are controlled by the indium flush technique during the growth which provides precise control of the QD heights [7]. The QDM structure has a semi-transparent titanium contact on the top surface and an n⁺-GaAs buffer layer (ohmic contact) which provides both charge and electric field controllability through the Schottky diode structure. Further details of a sample layer sequence can be found elsewhere [1]. Samples were kept in a closed cycle cryostat operated at ~10 K. The excitation was provided by a frequency tunable Ti:Sapphire laser operated in CW mode. The PL was dispersed through a 0.75 m spectrometer and collected with a liquid nitrogen cooled CCD detector. For the polarization resolved measurements we introduced linear polarizers and liquid crystal retarders to the basic PL experiment.

Fig. 1 shows the schematic structure of the QDM embedded in an n+ Schottky diode along with the PL spectra. With an applied electric field along the growth direction (z direction) we observe hole level resonance at reverse biases (high electric field) because the device itself has a relatively larger bottom dot compared to the top dot size [8]. By varying the applied electric field we can shift from a more molecular like state to more atomic like states since the electron and the hole can be spatially separated in indirect states. Similar to charged exciton states observed in single InAs/GaAs quantum dots [9], a positively charged exciton (positive trion; X⁺) which contains an additional hole compared to the exciton (neutral exciton; X⁰) can be observed in QDMs [1] along with a rich X-shaped pattern with several crossings and anticrossings arising from

U.S. Government work not protected by U.S. copyright
tunnelling and spin, of the QDM charged exciton states [Fig. 4(a)]. To gain insight into the details of spin interactions of exciton states, we used the technique of optical pumping [10] with circularly polarized excitation. In QDs, due to strain, the heavy-hole/light-hole degeneracy is lifted and a specific photoexcited carrier spin orientation can be created and controlled by exciting with circularly polarized light. For example circularly polarized light with total angular momentum of $+1\hbar$ results in the creation of a spin-up heavy-hole ($+3/2\hbar$) and a spin-down electron ($-1/2\hbar$).

Information about carrier spins and spin interaction can then be arrived at by measuring the degree of circular polarization memory for the resulting PL, defined as,

$$\rho = \frac{(I^+ - I^-)}{(I^+ + I^-)} \quad (1)$$

with $I^+$ and $I^-$ being the right and left circular polarized luminescence intensity components.

III. RESULTS

With an applied electric field we can control the mixing between the direct $\left( ^{7}_{0,0}X^0 \right)$ and indirect $\left( ^{7}_{0,0}X^0 \right)$ excitons, where in general $\left( ^{e_s\pi e_t}X^0 \right)$ is used to denote the exciton state with the number, arrows indicate the number of electrons ($\uparrow$) and holes ($\downarrow$) in either the bottom (B) or top (T) and the spin is indicated by the direction of the arrow. A large wavefunction overlap should result in a low degree of circular polarization memory, such as in the spatially direct exciton, as a consequence of the anisotropic exchange interaction in the direct states [4]. At small energy separations between the direct and indirect excitons (high applied field values) we observe an equal, and relatively lower (~20%), circular polarization memory for both exciton states. At large energy separations we find more atomic like states and expect a lower overlap between the electron and hole wave functions, as shown schematically in Fig. 2. This results in an increase in the circular polarization memory of the indirect exciton and decrease in the direct exciton [Fig. 3].

![Fig. 1. Electric field tunability of the QDM. (a) Schematic view of the QDM embedded in Schottky diode structure along with hole level resonance attained with applied electric field. (b) Photoluminescence spectra showing anticrossing signature corresponding to molecular state of the exciton and the inset provides evidence of having minimum energy splitting between the two excitonic states at the same applied electric field.](image1)

![Fig. 2. Exciton wave function overlap (a) At high electric fields hole level resonances along with electron wave function (blue) symmetric (direct) and asymmetric (indirect) hole wave functions (red). (b) At relatively low electric fields less overlap occurs between the electron and the hole wave functions. The anticrossing signature of the direct and indirect excitons along with corresponding spin configurations are shown in the bottom plot.](image2)

![Fig. 3. Degree of circular polarization memory measurements as a function of applied electric field for direct $\left( ^{7}_{0,0}X^0 \right)$ and indirect $\left( ^{7}_{0,0}X^0 \right)$ excitons. As the applied electric field is increased relative to the exciton anticrossing, circular polarization memory of the indirect exciton increase as a consequence of the reduced electron hole exchange interaction.](image3)
As we further increase the energy separation between the indirect and direct exciton, a variation in the luminescence was observed and additional lines became visible in the PL spectra. This can be attributed to charge carrier tunneling that results in creation of singly charged state (positive trion) as shown in Fig. 4(a) and 4(b). Therefore, by engineering of individual QDs in the QDM system we can design systems where we can controllably create singly charged states useful for two-level systems that can be manipulated with applied electric field. The singly charged positive trion PL (X-shaped pattern), comprises of both crossings and anticrossings and the spin states are identified from the kinetic hole and electron hole exchange interactions that has been discussed elsewhere [6]. Within the spin fine structure of the positive trion state [Fig. 4(b)] we can identify the energy splitting due to the isotropic exchange interaction. This spin fine structure doublet is identified as $X_H^\uparrow, X_L^\uparrow$ where subscripts $H, L$ correspond to the high and low energy components, respectively. In this configuration there is one electron, in the bottom dot, and two holes, one in the bottom dot and one in the top dot. Due to the high electric field dependence we know that the PL lines we observe arise from the recombination of the electron with the hole in the top dot.

From optical selection rules we know that the electron and hole in the top dot have opposite spins, however, the hole in the bottom dot can have spin either parallel (triplet) or anti-parallel (singlet) to the hole in the top dot. The difference in energy between these configurations primarily arises from the isotropic exchange interaction between the electron and hole in the bottom dot. In other words the hole in the top dot allows us to measure the bright-dark exciton splitting without an applied magnetic field typically needed to mix the states and make the dark exciton optically active.

Circular polarization memory measurements on the fine structure doublet indicated an increase in degree of circular polarization memory with applied electric field [Fig. 5]. As we increase the applied electric field we tend to spatially separate the top dot hole thereby reduce the exchange interaction. Therefore we observe increase in circular polarization memory for the spin fine structure doublet with the field similar to that observed for the neutral exciton. One interesting result is that the circular polarization memory of the low energy component is higher than the high energy components as shown in the inset of Fig. 5. This may be due to mixing with the dark states, however it is unclear at this time what the exact mechanism might be that causes this asymmetry in polarization memory.

 Barrier dependent measurements of the positive trion states further indicates the effects of the spatial separation of the hole state as shown in Fig. 6. As we increase the barrier between the top and the bottom dot, the positive trion state with the two holes on separate dots becomes more like a direct neutral exciton. This is because the hole in the top dot is physically more separated from the electron-hole pair in the bottom dot, reducing its overall effect. This is consistent with our observation of lower circular polarization memory for higher barrier QDMs with reduced relative electric field.

Further away from anticrossings, where the hole is atomic-like and no longer tunnels between the two dots, we find the hole in the top dot has a reduced effect on the spin of the exciton in the bottom dot. Therefore, by tuning the applied electric field it may be possible to control the overall spin interaction, effectively turning it on or off controllably.
In summary, we have used polarization resolved PL spectroscopy to identify both neutral exciton and positive trion states of QDMs. With circular polarization memory measurements we were able to identify and control both anisotropic and isotropic exchange interactions in QDMs. We were also able to measure the isotropic electron-hole exchange which gives rise to the positive trion fine structure doublet. Control of the spin interactions in these structures may open new avenues in applied optical and spintronic technologies and in quantum information processing schemes.

ACKNOWLEDGMENT

This work is supported by the Ohio University CMSS program and NSF grant number DMR-1005525.

REFERENCES