

## Final Report

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**Project Title:** Enhanced Materials Based on Submonolayer Type-II Quantum Dots

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## Enhanced Materials Based on Submonolayer Type-II Quantum Dots

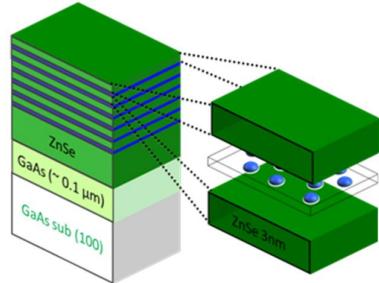
### Introduction and background

During the past few years we have investigated a nanostructured material known as sub-monolayer type-II QDs, made from wide bandgap II-VI semiconductors. Our goal is to understand and exploit their tunable optical and electrical properties by taking advantage of the type-II band alignment and quantum confinement effects.[1] Type-II ZnTe quantum dots (QDs) in a ZnSe host are particularly interesting because of their relatively large valence band and conduction band offsets.[2] During the previous award we advanced our understanding of these novel nanostructures and *our ability to control and manipulate their structural properties*, and we obtained materials with new and enhanced properties. We also developed an experimental approach to characterize the structures with high precision and insight. In the current award we planned to develop new materials based on sub-monolayer type-II QDs that may be advantageous for photovoltaic and spintronics applications. We also proposed to expand the structural characterization of these materials by refining the X-ray diffraction methodologies. During the first year we reported on ZnTe/ZnCdSe type II QDs materials that have ideal properties for the development of intermediate band solar cells (IBSC). We also began to develop a systematic approach to define the size and density of these QDS, and initiated the investigation of their spin dynamics. During the second year we developed new ways to analyze and model high resolution X-ray diffraction (HRXRD) data to extract structural details of the sub-monolayer type-II QD structures, developed models and experimental methods to measure in-plane dimensions of the QDs and to calculate the dependence of QD size and density on the MBE growth conditions, measured the linear polarization of the PL and confirmed the predictions obtained from HRXRD of anisotropic shape of the QDs, and initiated temperature dependent time resolved Kerr rotation (TRKR) studies to explore the spin dynamics of these materials.

During the current reporting period [year three of the award plus one year of no cost extension (NCE)] we have completed the following studies: 1) we have developed a comprehensive approach to describe and model the growth of these ultra small type II QDs. Analysis of the PL evolution, combined with SIMS and other characterization probes allowed us to predict the size and density of the QDs as a function of the growth conditions. 2) A full development and implementation of novel sophisticated HRXRD techniques were applied from which accurate size and shape of the buried type II-QDs could be extracted. It was found that elongated QDs form, with a preferential alignment along a well-defined crystallographic plane. 3) A correlation of the shape anisotropy with polarization dependent PL was obtained, also confirming the detailed shape and providing insight about the effects of this shape anisotropy with the physics of the type II QD systems. 4) A detailed time resolved Kerr rotation investigation has led to the demonstration of enhanced spin lifetimes for the samples with large densities of type II QDs and an understanding of the interplay between the QDS and Te-isoelectroic centers that form in the spacer layers that separate the QDs.

Our team is highly complementary and interdisciplinary. The growth and synthesis by Migration Enhanced-MBE is done at CCNY (Tamargo Lab). Spectroscopic studies using photoluminescence (PL) - cw and time-resolved, PL excitation (PLE), magneto-PL and near-field microscopy is performed at QC (Kuskovsky Lab). Faraday and Kerr rotation measurements are performed at CCNY (Meriles Lab). Electrical and photoelectrical studies include the Hall as well as photo-Hall effect and I-V and photoconductivity measurements (at CCNY and QC). HRXRD is performed at CU, BNL (Noyan Lab) and at CCNY. Students working on this project are trained in-depth in all the aspects of the research, including growth, characterization and modeling.

During the grant period three students (Bidisha Roy, Siddharth Dhomkar and Haojie Ji) obtained their Ph.D.s. They are currently postdoctoral researchers in laboratories at CCNY and in Germany. A fourth student (Vasilios Deligiannakis) will defend his thesis in Fall of 2017.



**Figure 1:** Schematic of the multi-QD ZnTe/ZnSe structure. Blue dots represent the ZnTe QDs.

## Principal results during the grant period (October 1, 2012 to November 14, 2016)

### 1. Zn(Cd)Te/ZnCdSe quantum dot based intermediate band solar cells

A novel material system consisting of stacked ultra-small type-II ZnCdTe quantum dots embedded in  $Zn_{0.51}Cd_{0.49}Se$  that exhibits nearly optimal material parameters for fabrication of an intermediate band solar cell (IBSC) has been grown and investigated. [3,4] Figure 1.1 shows a bandstructure schematic of the proposed IBSC device structure, indicating the mini band formation that leads to IB absorption. The demonstration of this band formation and subsequent absorption of below bandgap photons boosts the claim.

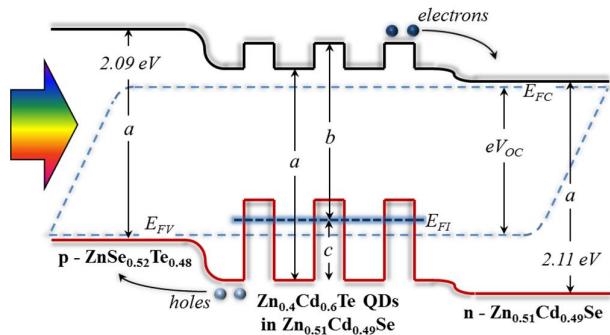


Figure 1.1 : bandstructure schematic for proposed IBSC based on type II QDs

Intermediate band solar cells can potentially have efficiency of 63% under full solar concentration, but the material systems investigated until now are far from optimum and are fraught with growth related issues. The system of ZnCdTe “sub-monolayer” quantum dots embedded in  $Zn_{0.51}Cd_{0.49}Se$  promises to overcome these problems. The physical and material properties of the proposed structure provide a nearly ideal match to the proposed active material for a working intermediate band solar cell.

Self-assembled quantum dots (QDs) embedded in a host semiconductor can, in principle, be used to create the IB, but this approach has not yet been successful due to various difficulties related mainly to unavailability of the appropriate material system and growth related issues.

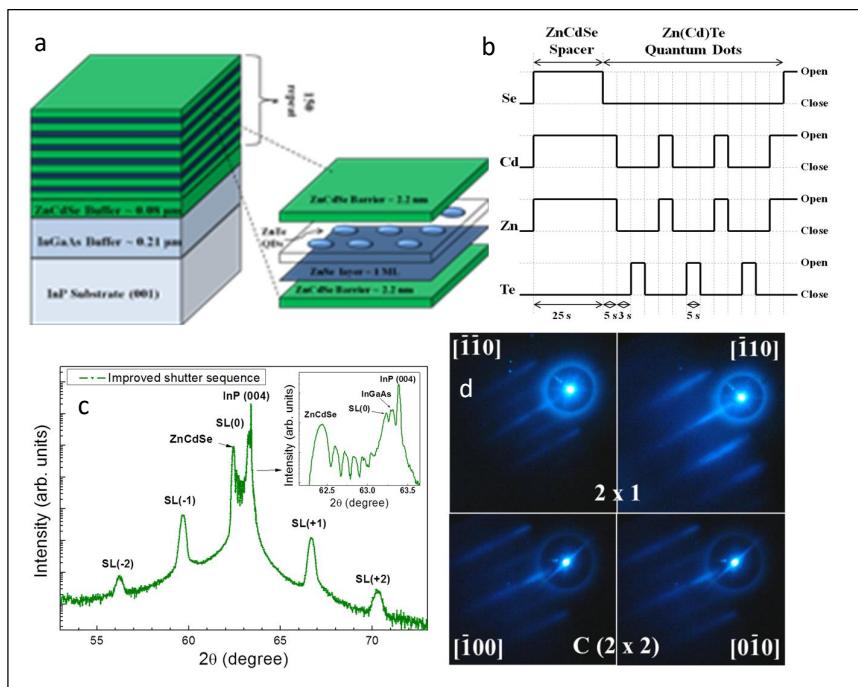


Figure 1.2 : a) Schematic of proposed layer structure for IBSC. b) Shutter sequence needed to achieve high quality materials by ME-MBE, c) HRXRD scan for a structure grown using the shutter sequence, d) RHEED patterns for the sample during ME-MBE growth.

Our team has grown novel multilayer structures comprising of as many as 150 layers containing sub-monolayer ZnCdTe QDs embedded in a  $Zn_{0.51}Cd_{0.49}Se$  matrix. [5,6] The ultra-small QDs form without producing wetting layers. Figures 1.2 (a-d) show the proposed layered structure, the optimized shutter sequence needed to grow the structure by ME-MBE, the high resolution x-ray diffraction scan, and the observed RHEED pattern during growth, indicating excellent materials quality. We have shown that this system possesses material and physical parameters that are very close to optimal for an IB solar cell. Apart from the right band gaps and band offsets, the observed vertical

correlation of the QDs, illustrated in Figure 1.3a) produces sufficient overlap of wavefunctions of confined charge carriers, holes in this case, in order to form an IB, as shown schematically in Figure 1.3b).

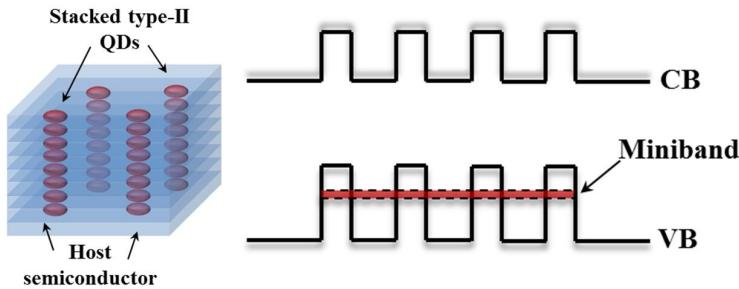


Figure 1.3 a) schematic illustrating stacked QD configuration b) miniband formation in the stacked QDs.

The type-II band alignment is expected to avoid undesired voltage reduction and assist in the carrier extraction process due to reduced radiative recombination within the QDs. It also should suppress non-radiative Auger recombination, owing to the spatially indirect nature of the excitons. Furthermore, the absence of wetting layers insures preservation of the open circuit voltage and should enhance the efficiency of an IBSC substantially.

## 2. Determination of size and distribution of type-II ZnTe/ZnSe stacked-submonolayer quantum dots.

Samples	A	B	C	D	E	F
Te flux in $\times 10^{-7}$ torr	2	1.2	1.3	1.9	2	8
Number of Te MEE cycles	1	3	3	3	3	2
SL period in nm	1.7	2.5	1.7	2.5	1.7	2.8
Number of periods	100	100	100	250	100	120
Strain along growth direction	0.00303	0.00193	0.00432	0.00116	0.00612	0.00303
Te content from SIMS (XRD) in %	0.1 (-)	0.2 (-)	0.3(0.1)	0.3(0.2)	0.5(0.3)	3.2 (2.6)

Table 2.I. List of important parameters for all the samples investigated

For practical applications of self-assembled semiconductor quantum dots (QDs), it is important to control their densities and sizes, but these parameters are difficult to obtain, specifically in case of smaller QDs. By combining a series of characterization tools, we obtain accurate estimates of the dot densities, and their size as we vary the Te flux and the number of Te cycles during their growth via MEE. [7] The parameters of the investigated samples are summarized in Table 2.I.

### a) Photoluminescence and magneto-photoluminescence results

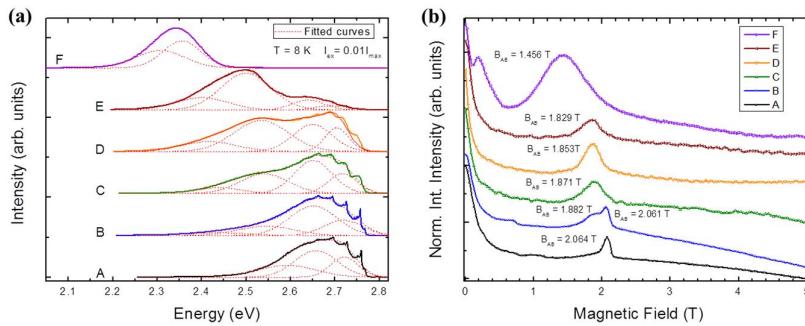


Figure 2.1. (a) PL spectra for all the samples fitted with four gaussian lineshapes, (b) Magneto-PL data for all the samples showing Aharanov-Bohm peak.

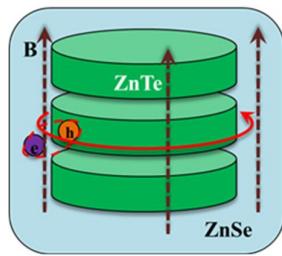
in all the other peaks (see figure 2.1(a)).

Confirmation of the presence of type-II QDs was obtained from excitation intensity dependent PL analysis supported by the observation of robust Aharonov-Bohm (AB) oscillation in the magneto-PL spectra. The PL spectra were fitted with four gaussian lineshapes. The near band edge peak mostly originates from the emission from Te isoelectronic centers, while the contribution from QDs dominates

PL peak position redshifts with increasing  $\tau$  (Te flux\* MEE duration) indicating increase in average QD thickness, which was then estimated by solving 1-D Schrondinger equation. Diameters of these disc-shaped stacked QDs were precisely calculated by means of spectral analysis of optical signature of the AB excitons (see figure 2.1 (b)), discussed in the following section.

### b) Model to calculate diameters of disc-shaped stacked submonolayer QDs

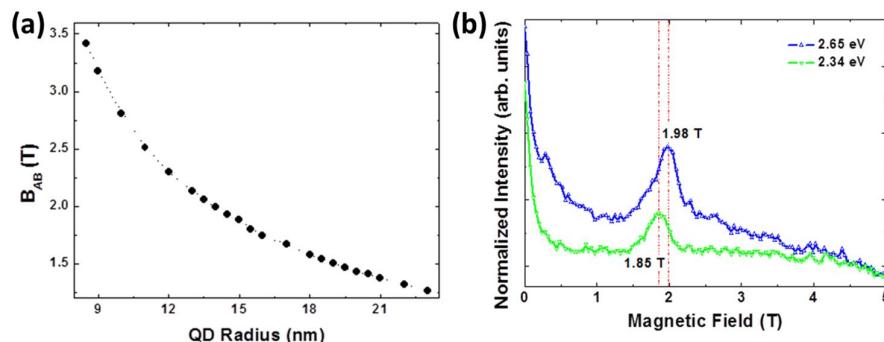
The submonolayer QDs are known to form vertical stacks due to high vertical correlation. The ZnSe spacers between the QD layers were less than 3.0 nm thick, much smaller than the lateral separation between QDs. Because of the vertical confinement, the electrons, which are located in the ZnSe barrier, will reside at the sides of the stacked QDs.



**Figure 2.2.** Schematic diagram of a lateral tightly-bound exciton in type-II ZnTe/ZnSe stacked QDs.

approach. This approach takes into account the lateral wavefunction of the hole in analysis of the optical emission in the type-II ZnTe/ZnSe stacked submonolayer QDs. The schematic diagram of the proposed model is shown in figure 2.2. The hole is localized on a circular trajectory inside the QD, while the electron is localized on a trajectory outside the QD. Within this model, we can separate the radial wavefunctions of electron and hole, which can be obtained by solving the single-particle equations in the effective mass approximation. We use COMSOL and MatLab to solve the equations self-consistently using an iterative procedure. The result shows that for the QDs with large lateral size ( $R_s > 9$  nm), the trajectory radii of electron and hole increase linearly with the increase of QD stack radius,

The magnetic field value of the AB oscillation peak depends on the QD stack radius,  $R_s$ , this allows calculating  $B_{AB}$  as a function of  $R_s$ , which we plot in figure 2.3. Employing this dependence, we can probe the lateral sizes of ZnTe QDs from the experimentally observed AB oscillation peaks.



**Figure 2.3.** (a) The transition AB effect peak,  $B_{AB}$ , as a function of the QD stack radius,  $R_s$ . (b) The corresponding emission intensities as functions of magnetic field for two different emission energies of sample C, shown as an example of spectral analysis of AB peaks.

the  $B_{AB}$  and the  $R_s$  (Fig. 2.3 (a)), we obtained the QD stack radii corresponding to the emission energies across the PL spectra for all the samples.

Our results showed that, in sample A, grown with the smallest Te flux and with a single Te-MEE cycle, QDs have a fairly uniform radius of about 13.4 nm, giving the smallest lateral size of ZnTe submonolayer QDs in this particular ZnTe/ZnSe multilayer system. In sample F, grown with the highest Te flux and with two Te-MEE cycle, the QD radius varies gradually across the PL spectrum, from 18.6 to 20.9 nm, the largest variation among all the samples. In samples B through E, the QD radii take several discrete values, confirming the vertical correlation and stacked nature of the submonolayer QDs, and the ability to tune the QD radius by the Te flux during growth.

The excitonic AB effect [8,9] presents itself as a transition between different angular momentum states of the excitons (see Ref. 10 and references therein). This transition from bright to dark excitonic state changes the PL intensity due to optical selection rules. [9]. The magnetic field value,  $B_{AB}$ , of the AB transition is dependent on the trajectories of electrons and holes. We applied a lateral tightly-bound exciton model for the type-II ZnTe/ZnSe QDs to calculate the electron and hole trajectories as a function of the QD stack radius. Thus, we have obtained the dependence of  $B_{AB}$  on the QD stack radius,  $R_s$ .

The wavefunction of the hole is “attracted” to the lateral boundary of the QDs, and the trajectory of hole is as important as that of electron to determine the magnetic flux for the excitonic AB effect. Therefore, we modified our original model and applied a lateral tightly-bound exciton

approach. This approach takes into account the lateral wavefunction of the hole in analysis of the optical emission in the type-II ZnTe/ZnSe stacked submonolayer QDs. The schematic diagram of the proposed model is shown in figure 2.2. The hole is localized on a circular trajectory inside the QD, while the electron is localized on a trajectory outside the QD. Within this model, we can separate the radial wavefunctions of electron and hole, which can be obtained by solving the single-particle equations in the effective mass approximation. We use COMSOL and MatLab to solve the equations self-consistently using an iterative procedure. The result shows that for the QDs with large lateral size ( $R_s > 9$  nm), the trajectory radii of electron and hole increase linearly with the increase of QD stack radius,

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To investigate the lateral size distribution of QDs in each sample, we plotted the PL intensities as functions of magnetic field and located the  $B_{AB}$  for different emission energies (an example of two emission energies for sample C is shown in Figure 2.3 (b)). Using one-to-one mapping between

### c) Estimation of QD density

The results obtained from PL and magneto-PL analysis in conjunction with high resolution x-ray diffraction and secondary ion mass spectrometry (SIMS) data of the Te concentrations, were then used to evaluate the QD density (see figure 2.4). We were able to provide the dependence of average QD volume and density as a function of Te flux as well as number of Te MEE cycles (or  $\tau$ ) and show that these are the key parameters to control the QD dimension and distribution. Moreover, it was found that the dot density increases much faster than the QD size with respect to the increase in Te flux and the number of Te MEE cycles.

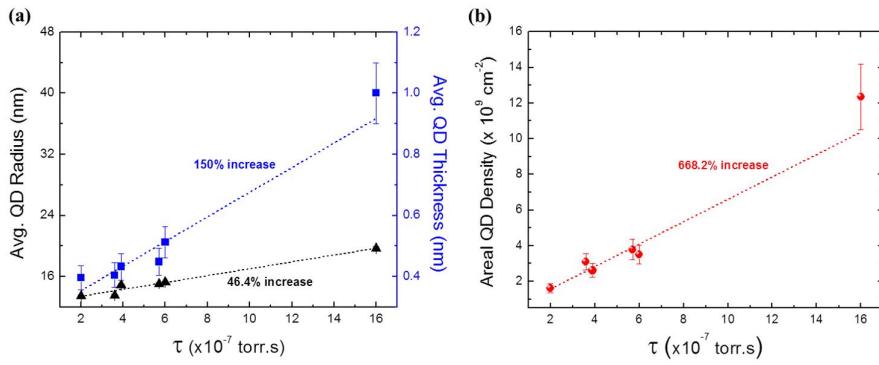


Figure 2.4. (a) Average QD radius and thickness and (b) Average QD density as a function  $\tau$ .

### 3. Determination of shape anisotropy in embedded low contrast submonolayer quantum dot structures

As diffuse x-ray scattering is sensitive to dot shape and size distribution, we utilize high-resolution in-plane reciprocal space map (RSM) to quantify any preferential elongation of submonolayer (SML) quantum dots (QDs) along crystallographic directions. [10] The experimental data indeed exhibits a strong anisotropy in the diffuse scattering signal [Figure 3.1(a)] but one needs to perform the simulations to extract the useful information. Thus, to explain the experimental observation, in-plane diffuse scattering has been computed using kinematic sum and realistic QD shape and size distribution. One of the important input parameters required for the simulations, the QD size distribution, is obtained via the magneto-photoluminescence (PL) signal observed from these type-II structures.

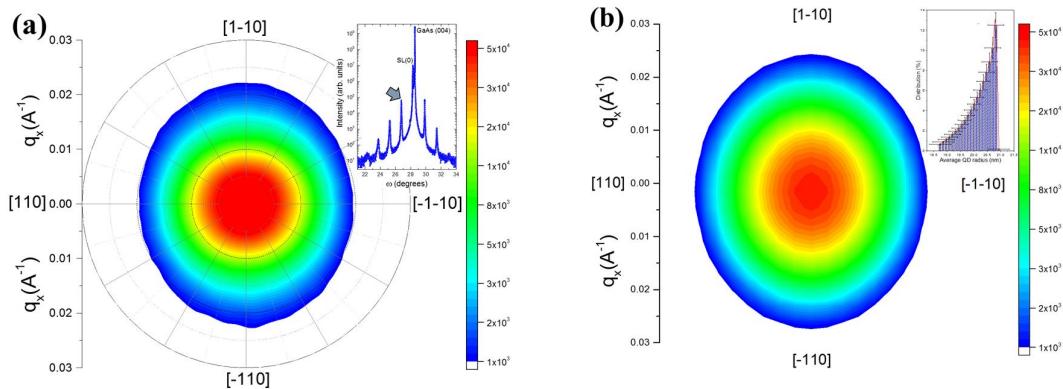
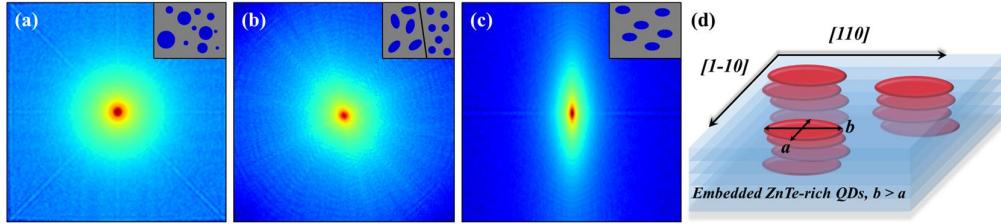


Figure 3.1 (a) In-plane reciprocal space map around satellite SL(-1) of (004) Bragg peak depicting clear anisotropic shape. Inset: x-2h scan along (004) showing periodic satellite SL peaks. The arrow indicates the specific satellite peak under investigation. (b) Simulated rocking curves as a function of azimuthal angle for a known size dispersion of elongated QDs. Inset: QD lateral size dispersion as calculated from magneto-PL analysis.

A set of different QD configurations has been computed, as shown in Figure 3.2(a)–(c), in order to reproduce the experimental results. The optimized simulations, as shown in Figure 3.1(b), present clear evidence of the anisotropically shaped QDs elongated along [110] orientation, illustrated in Figure 3.2(c and d) and the best match with the experimental data was obtained for the QD aspect ratio of  $1.3 \pm 0.05$ .

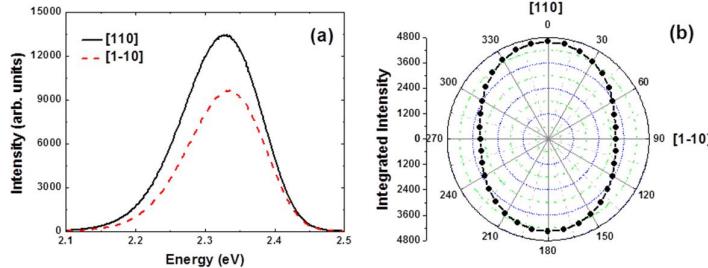


**Figure 3.2.** (a)–(c) Simulated RSMs of QDs with various shapes and orientations, the insets show the corresponding QD configuration. (d) Schematic diagram of the disc-shaped embedded SML QDs elongated along [110] orientation.

As the anisotropy in the QD shape can manifest itself in the PL emission we perform linear polarization dependent PL experiments to test our findings (see section 4). As expected, the PL emission presents strong linear polarization along [110] axis and the aspect ratio corresponding to the observed degree of polarization found to be 1.27, which is in excellent agreement with the one obtained, using the high-resolution x-ray diffraction simulations. Thus, our results clearly demonstrate that this combination of characterization techniques is uniquely suited to elucidate the morphology of these hard-to-image SML QD structures. A secondary advantage of our formalism is the non-destructive nature of the individual techniques and the ease of sample preparation. Our approach provides a novel, fast, accurate and precise way of characterizing embedded ultra-small QD arrays.

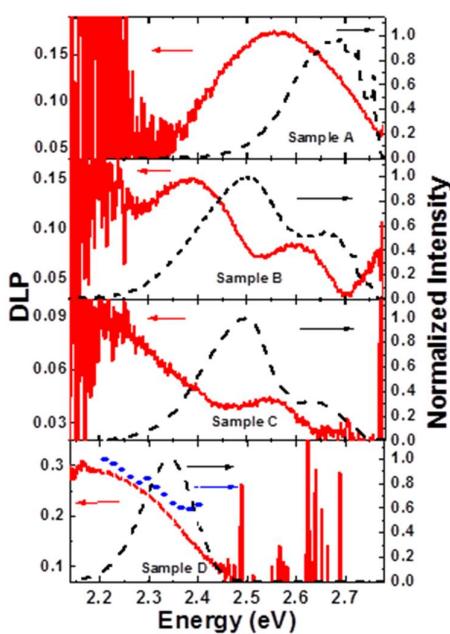
#### 4. Optical Anisotropy in Type-II ZnTe/ZnSe Submonolayer Quantum Dots

As mentioned above, linearly polarized photoluminescence was observed for type-II ZnTe/ZnSe submonolayer quantum dots (QDs) (Figure 4.1). [11]



**Figure 4.1.** (a) The photoluminescence emission of sample D polarized along [110] (solid line) and [1-10] (dash line) crystal axes. (b) The integrated intensity of the photoluminescence of sample D as a function of the angle between axis of the linear polarizer and the [110] crystal axis.

The value of anisotropic exchange splitting for bright excitonic states is found to be  $\sim 200 \mu\text{eV}$  from the measurement of the degree of circular polarization as a function of the magnetic field, which is fitted to :

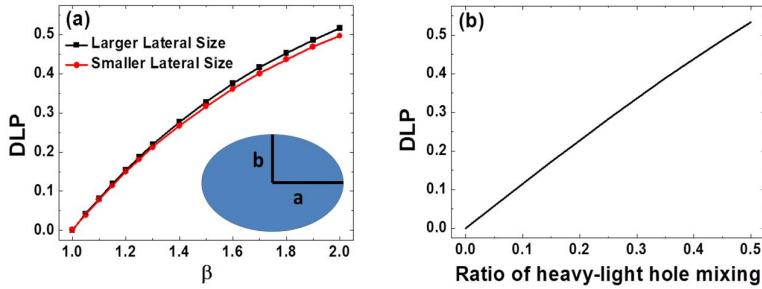


$$DCP(B) = \frac{B^2}{B^2 + B_1^2} [P_c^0 + \tilde{P}_c^0 \frac{B^2}{B^2 + B_2^2}]$$

The comparison of spectral dependence of the degree of linear polarization (DLP) among four samples, shown in Figure 4.2, indicates that the optical anisotropy is mostly related to the elongation of ZnTe QDs, instead of the interfacial symmetry lowering or the anisotropic strain relief and defects.

Numerical calculations based on the occupation probabilities of holes in  $p_x$  and  $p_y$  orbitals are performed to estimate the lateral aspect ratio of the QDs, and it is shown (Figure 4.3) that it varies between 1.1 and 1.4, while thicker QDs are more elongated and have stronger heavy-light hole mixing.

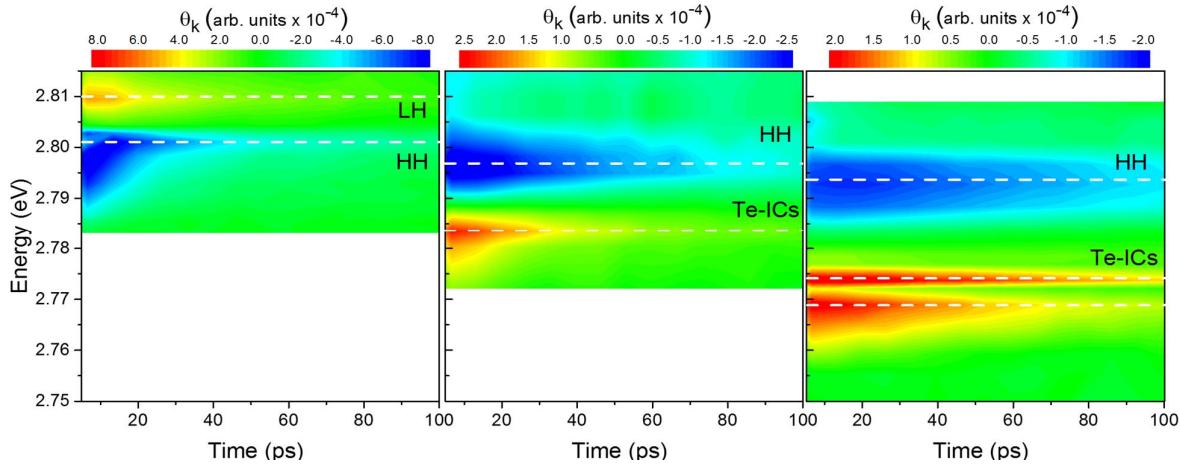
**Figure 4.2.** Spectral dependence of the degree of linear polarization for all samples overlaid over corresponding normalized spectra (dashed lines). The blue dots are the spectral dependence of normalized anisotropic exchange splitting for sample D.



**Figure 4.3.** (a) Calculated  $DLP$  as a function of aspect ratio of the elongated QDs with larger lateral size (squares) and smaller lateral size (circles) as described in text. The inset shows a lateral elongated QD with aspect ratio  $\beta = a / b$ . (b)  $DLP$  as a function of the ratio of heavy-light hole mixing.

##### 5. Probing spin dynamics of type-II sub-monlayer Quantum Dots:

Systems of reduced dimensionality, such as quantum dots (QD) have been proposed as promising candidates for spintronic applications. We have investigated the spin properties of our type-II submonlayer quantum dots using time resolved Kerr Rotatoion (TRKR). Our results indicate that significantly longer electron spin lifetimes are achieved in the samples with larger densities of ZnTe QDs. We were also able to directly probe the spin dynamics of tellurium isoelectronic centers (Te-ICs) present in the ZnSe matrix, which have not previously been reported. [12]

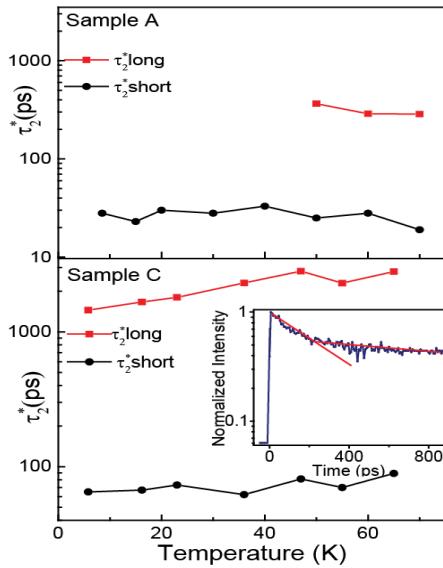


**Figure 5.2** Spectral dependence of the TRKR signal for samples A, B and C. Dashed lines indicate the position of the excitonic levels associated with ZnSe spacer layers. Energies corresponding to direct excitation of Te-ICs are also labeled. Regions with no color indicate the absence of a TRKR signal.

Figure 5.1 compares the TRKR contour plots for three samples studied (A, B and C) with increasingly more Te content each, measured at  $\sim 10$ K. Sample A (with lowest Te), shown in Figure 1a, pumping directly at energies corresponding to the Te-IC centers (blue band) and QDs (green band), did not show any TRKR signal. Keeping the pump helicity fixed, a spin flip is observed going from the ZnSe heavy hole (HH) to light hole (LH) excitation energies, indicated by the Kerr rotation signal changing from positive to negative at 2.795eV and 2.810eV, respectively. The spin lifetimes observed in this sample were very short, on the order of tens of picoseconds. Similar TRKR lifetimes were measured by us (not shown) and have been observed by others for bulk undoped ZnSe at low temperatures.

Samples B and C, with a larger amount of Te (0.2% and 0.3%, respectively), behave quite differently from sample A (which has 0.1% Te content). In both samples the TRKR signals persist as we probe deeper in energy into the region of the blue band, which originates from the Te-ICs. We interpret these observations by considering that we are able to probe the Te ICs in these samples that have increased Te content. Direct excitation of Te-bound excitons has been observed as a phonon-broadened peak below the band edge in photoluminescence excitation experiments in other II-VI materials.

Sample C exhibits long spin lifetimes of up to 1 ns at low temperatures, in sharp contrast to the lifetimes measured in sample A. In addition, we observe a spin flip near the energies corresponding to the Te isoelectronic centers for both samples C and B. Probing at energies around the LH to free exciton (FE) transition of ZnSe the TRKR signal is significantly diminished. In addition, we do not observe the spin flip seen



**Figure 5.3** Temperature dependent spin relaxation lifetimes measured by TRKR pumped and probed at 2.78 and 2.75 eV for samples A and C, respectively. Typically a biexponential decay is seen in the TRKR, shown in the inset for sample C at 65K.

A similar behavior to that of sample C has been observed in undoped type-II quantum wells, which also exhibit spin lifetimes in excess of 1 ns at low temperatures.

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in bulk ZnSe and in sample A due to the non-degenerate LH and HH. We tentatively explain the latter behavior for samples B and C as being due to the capture of light holes by the presence of the type-II QDs within the samples. These ZnTe-based QDs form a staggered band alignment with the ZnSe matrix producing a potential well for holes.

To better characterize the increase in spin lifetime observed for the samples with higher Te content, we investigated the temperature dependence of the Te-IC center related spin lifetimes in samples A and C. In order to probe the Te-IC centers directly we used the lowest energies at which we observed TRKR signal for each sample. Figure 5.2 shows the temperature dependence of the TRKR lifetime for samples A and C. In both cases, the TRKR measurements typically exhibit a bi-exponential decay which is denoted by a long  $\tau^*_2$  and a short  $\tau^*_2$  on the graphs.

The principal observation is that Sample C exhibits a much longer long  $\tau^*_2$  lifetime at low temperature, exceeding one nanosecond. For sample A the value of the long  $\tau^*_2$  is much shorter below 50K, resulting in single exponential decay behavior. Above 40 K the long  $\tau^*_2$  spin lifetime measured is ~100ps. This behavior is similar to that seen in bulk undoped ZnSe (not shown). This sharp drop off of the lifetime at ~50K has been explained as motional narrowing of excitons. In that mechanism, electron and hole overlap increases at lower temperatures. In sample C, which has a much larger density of type-II quantum dots this decoherence mechanism does not appear to play a dominant role.

**Papers Published with DOE Support 2012 – 2017 (Acknowledgements included)**

1. U. Manna, I. C. Noyan, Q. Zhang, I. F. Salakhutdinov, K. A. Dunn, S.W. Novak, R. Moug, M. C. Tamargo, G. F. Neumark, and I. L. Kuskovsky, Structural properties and spatial ordering in multilayered ZnMgTe/ZnSe type-II quantum dot structures, *J. Appl. Phys.* **111**, 033516 (2012).

"This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE-FG02-10ER46678. Use of the National Synchrotron Light Source at Brookhaven National Laboratory (BNL) was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-98CH10886. One of us (R.M.) acknowledges the support of the National Science Foundation under Award No. HRD-0833180 (CREST-CENSES). The authors would like to thank Dr. Jean Jordan-Sweet, the manager of X20A beamline, and L. Li, M. Treger, S. M. Polvino, and A. J. Ying for useful discussions and assistance during the experiments at BNL."

2. U. Manna, Q. Zhang, S. Dhomkar, I. F. Salakhutdinov, M. C. Tamargo, I. C. Noyan, G. F. Neumark and I. L. Kuskovsky, Radiative transitions in vertically correlated type-II ZnMgTe/ZnSe multilayer quantum dot structure, *J. Appl. Phys.* **112**, 063521 (2012).

"This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE-FG02-10ER46678. Use of the National Synchrotron Light Source, Brookhaven National Laboratory, was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02- 98CH10886. We thank J. Jordon-Sweet, B. Roy, R. Moug, L. Li, and M. Treger for assistance during the experiments and useful discussion. We also thank V. Holy for discussion in interpreting the RSM."

3. S. Dhomkar, I. L. Kuskovsky, U. Manna, I. C. Noyan, M. C. Tamargo, Optimization of growth conditions of type-II Zn(Cd)Te/ZnCdSe submonolayer quantum dot superlattices for intermediate band solar cells, *J. Vac. Sci. Technol. B* **31**, 03C119 (2013).

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4. S. Dhomkar, U. Manna, L. Peng, R. Moug, I. C. Noyan, M. C. Tamargo, I. L. Kuskovsky, Feasibility of submonolayer ZnTe/ZnCdSe quantum dots as intermediate band solar cell material system, *Sol. Energy Mater. Sol. Cells* **117**, 604 (2013). (Cited 14 times)

"This research was supported by the Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. SC003739. Use of the National Synchrotron LightSource, Brookhaven National Laboratory, was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-98CH10886. One of us (RM) acknowledges support from the NSF-CREST CENSES under Award no. HRD0833180."

5. S. Dhomkar, U. Manna, I. C. Noyan, M. C. Tamargo and I. L. Kuskovsky, Vertical Correlation and Miniband Formation in Submonolayer Zn(Cd)Te/ZnCdSe Type-II Quantum Dots for Intermediate Band Solar Cell Application. *Appl. Phys. Lett.* **103**, 181905 (2013). (Cited 11 times)

"This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE- SC003739. Use of the National Synchrotron Light Source, Brookhaven National Laboratory, was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-98CH10886."

6. S. Dhomkar, H. Ji, B. Roy, V. Deligiannakis, A. Wang, M. C. Tamargo, I. L. Kuskovsky, Measurement and control of size and density of type-II ZnTe/ZnSe submonolayer quantum dots grown by migration enhanced epitaxy, *J. Crystal Growth* **422**, 8–14 (2015)

"This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award no. DE-SC003739."

7. S. Dhomkar, N. Vaxelaire, H. Ji, V. Shuvayev, M. C. Tamargo, I. L. Kuskovsky and I. C. Noyan, Determination of shape anisotropy in embedded low contrast submonolayer quantum dot structures, *Appl. Phys. Lett.* **107**, 251905 (2015).

"This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE-SC003739. Use of the National Synchrotron Light Source, Brookhaven National Laboratory, was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-98CH10886."

8. H. Ji, S. Dhomkar, R. Wu, V. Shuvayev, V. Deligiannakis, M. C. Tamargo, J. Ludwig, Z. Lu, D. Smirnov, A. Wang, and I. L. Kuskovsky, Optical Anisotropy in Type-II ZnTe/ZnSe Submonolayer Quantum Dots, *J. Appl. Phys.* **119**, 224306 (2016)

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9. V. Deligiannakis, S. Dhomkar, H. Ji, D. Pagliero, I. L. Kuskovsky, C. A. Meriles, M. C. Tamargo, Spin dynamics of ZnSe-ZnTe nanostructures grown by migration enhanced molecular beam epitaxy, *J. Appl. Phys.* **121**, 115702 (2017)

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#### **Papers Published with partial DOE support (Acknowledgements included)**

10. B. Roy, H. Ji, S. Dhomkar, F.J. Cadieu, L. Peng, R. Moug, M.C. Tamargo, I.L. Kuskovsky, Determination of excitonic size with sub-nanometer precision via excitonic Aharonov-Bohm effect in type-II quantum dots, *Appl. Phys. Lett.* **100**, 213114 (2012). (Partial DOE support) (Cited 14 times)

"This work is supported by the National Science Foundation under Award No. 40A94-0001. The samples used in this study are grown under Department of Energy, Basic Energy Sciences Grant No. DE-FG02-10ER46678. The authors are thankful to Dr. U. Manna and Professors L. Mourokh, A. A. Lisyansky, and I. C. Noyan for helpful discussions and support."

11. B. Roy, H. Ji, S. Dhomkar, F. J. Cadieu, L. Peng, R. Moug, M. C. Tamargo, I. L. Kuskovsky, Distinguishability of stacks in ZnTe/ZnSe quantum dots via spectral analysis of Aharonov-Bohm oscillations, *Eur. Phys. J. B* **86**, 31 (2013). (Partial DOE support)

"This work is supported by the National Science Foundation under Award No. DMR-1006050. The samples used in this study are grown under Department of Energy, Basic Energy Sciences Grant No. DE-

FG02-10ER46678. The authors are thankful to Dr. U. Manna and Profs. L. Mourokh and I.C. Noyan for helpful discussions and support."

12. H. Ji, B. Roy, S. Dhomkar, R. T. Moug, M. C. Tamargo, A. Wang, I. L. Kuskovsky, Tuning Between Quantum-Dot- and Quantum-Well-Like Behaviors in Type II ZnTe Submonolayer Quantum Dots by Controlling Tellurium Flux During MBE Growth, *J. Electron. Mater.* **42**, 3297 (2013), (Partial DOE support)

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13. H. Ji, S. Dhomkar, B. Roy, V. Shuvayev, V. Deligiannakis, M. C. Tamargo, J. Ludwig, D. Smirnov, A. Wang, and I. L. Kuskovsky, Determination of lateral size distribution of type-II ZnTe/ZnSe stacked submonolayer quantum dots via spectral analysis of optical signature of the Aharonov-Bohm excitons, *J. Appl. Phys.* **116**, 164308 (2014) (partial support)

"The work is supported by the NSF under Award No. DMR-1006050 and the DoE Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE-SC003739. Specifically, sample preparation and SIMS studies were carried out with the DoE support (SD, VD, MCT and ILK), while calculations and optical investigations were performed with support from the NSF (HJ, BR, and ILK) . The portion of this work performed at the NHMFL is supported by NSF Cooperation Agreement No. DMR-0084173 and by the State University of Florida (JL and DS)."

14. B. Roy, H. Ji, S. Dhomkar, M. C. Tamargo, and I. L. Kuskovsky, Observation of Oscillation in Energy of Aharonov-Bohm Excitons in Type-II ZnTe/ZnSe Stacked Submonolayer Quantum Dots, *Phys. Stat. Sol. C* **11**, 1248-1251 (2014). (Partial DOE support)

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15. I. L. Kuskovsky, L. G. Murokh, B. Roy, H. Ji, S. Dhomkar, J. Ludwig, D. Smirnov, and M. C. Tamargo, Decoherence in semiconductor nanostructures with type-II band alignment: All-optical measurements using Aharonov-Bohm excitons, *Phys. Rev.* (2017) (in press) (partial DOE support)

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#### Contributed Conference Presentations

1. B. Roy, H. Ji, S. Dhomkar, F.J. Cadieu, M.C. Tamargo, and I. L. Kuskovsky "Fine Probing of Distinguishable Stacks in ZnTe/ZnSe Sub-monolayer Quantum Dots," The 2012 US Workshop on the Physics & Chemistry of II-VI Materials, November 27-29, 2012, Seattle, WA, USA

2. S. Dhomkar, U. Manna, M. C. Tamargo, I. C. Noyan, and I. L. Kuskovsky, "Optimization of growth conditions of ZnTe/ZnCdSe submonolayer quantum dots for efficient intermediate band solar cells," The 2012 US Workshop on the Physics & Chemistry of II-VI Materials, November 27-29, 2012, Seattle, Washington, USA
3. H. Ji, B. Roy, S. Dhomkar, R. T. Moug, M. C. Tamargo, A. Wang, and I. L. Kuskovsky, "Tuning between Quantum-Dot and Quantum-Well-Like Behaviors in Type-II ZnTe Sub-Monolayer Quantum Dots by Controlling Tellurium Flux during MBE Growth," The 2012 US Workshop on the Physics & Chemistry of II-VI Materials, November 27-29, 2012, Seattle, WA, USA
4. B. Roy, S. Dhomkar, H. Ji, M.C. Tamargo, and I. L. Kuskovsky, "Effect of built-in electric field in stacked type-II ZnTe/ZnSe submonolayer quantum dots: enhancement and narrowing of Aharonov-Bohm oscillations," APS March Meeting 2013, March 18-22, 2013, Baltimore, MD, USA
5. S. Dhomkar, U. Manna, M.C. Tamargo, I.C. Noyan, and I. L. Kuskovsky, "Study of vertical correlation in type-II ZnCdTe/ZnCdSe submonolayer quantum dots for efficient intermediate band solar cells," APS March Meeting 2013, March 18-22, 2013, Baltimore, Maryland, USA
6. H. Ji, B. Roy, S. Dhomkar, R. T. Moug, M. C. Tamargo, A. Wang, and I. L. Kuskovsky, "Tuning between Quantum-Dot and Quantum-Well-Like Behaviors in Type-II Zn-Se-Te Multilayers by Controlling Tellurium Flux during MBE Growth," APS March Meeting 2013, March 18-22, 2013, Baltimore, MD, USA
7. I. L. Kuskovsky, B. Roy, H. Ji, S. Dhomkar, D. Smirnov, and M. C. Tamargo, "Excitonic Aharonov-Bohm Effect in Stacked Type-II II-VI Quantum Dots," The 16th International Conference on II-VI Compound and Related Materials, September 9-13, 2013, Nagahama, Japan
8. B. Roy, H. Ji, S. Dhomkar, M. C. Tamargo, and I. L. Kuskovsky, "Energy Oscillation of Aharonov-Bohm Excitons in Type-II ZnTe/ZnSe Stacked Submonolayer Quantum Dots," The 16th International Conference on II-VI Compound and Related Materials, September 9-13, 2013, Nagahama, Japan
9. S. Dhomkar, U. Manna, M.C. Tamargo, I.C. Noyan, and I. L. Kuskovsky, "Vertical correlation and miniband formation in submonolayer Zn(Cd)Te/ZnCdSe quantum dots for intermediate band solar photovoltaics," The 16th International Conference on II-VI Compound and Related Materials, September 9-13, 2013, Nagahama, Japan
10. S. Dhomkar, U. Manna, M.C. Tamargo, I.C. Noyan, and I. L. Kuskovsky, "Growth and characterization of submonolayer Zn(Cd)Te/ZnCdSe quantum dots for intermediate band solar cell applications," The 16th International Conference on II-VI Compound and Related Materials, September 9-13, 2013, Nagahama, Japan
11. H. Ji, B. Roy, S. Dhomkar, R. T. Moug, M. C. Tamargo, A. Wang, and I. L. Kuskovsky, "Tuning Between Quantum-Dot and Quantum-Well-Like Behaviors in Type-II ZnTe Sub-Monolayer Quantum Dots by Controlling Tellurium Flux during MBE Growth," The 16th International Conference on II-VI Compound and Related Materials, September 9-13, 2013, Nagahama, Japan
12. U. Manna, S. Dhomkar, I. C. Noyan, Q. Zhang, M. C. Tamargo, K. A. Dunn, S. W. Novak, G. F. Neumark, and I. L. Kuskovsky, "Submonolayer ZnMgTe/ZnSe quantum dots for improved doping and intermediate band solar sell applications: A structural analysis," The 16th International Conference on II-VI Compound and Related Materials, September 9-13, 2013, Nagahama, Japan

13. V. Deligiannakis, S. Dhomkar, H. Ji, B. Roy, D. Pagliero, I. L. Kuskovsky, M. C. Tamargo, and C. A. Meriles, Time Resolved Kerr Rotation Studies on Sub-monolayer ZnTe/ZnSe Quantum Dots, The 30th North American Molecular Beam Epitaxy Conference, October 5-11, 2013
14. I. L. Kuskovsky, H. Ji, S. Dhomkar, J. Ludwig, D. Smirnov and M. C. Tamargo, Millikelvin magneto-photoluminescence of iso electronic bound excitons in type-II quantum dot superlattices, APS March Meeting 2014, March 3-7, 2014 in Denver, Colorado
15. V. Deligiannakis, S. Dhomkar, H. Ji, B. Roy, D. Pagliero, I. L. Kuskovsky, M. C. Tamargo and C. A. Meriles, Time Resolved Kerr Rotation Studies on Sub-monolayer Type-II ZnTe/ZnSe Quantum Dots, APS March Meeting 2014, March 3-7, 2014 in Denver, Colorado
16. S. Dhomkar, H. Ji, B. Roy, I. L. Kuskovsky, A. Wang and M. C. Tamargo, Control over density of submonolayer type-II ZnTe/ZnSe quantum dots grown via migration enhanced epitaxy, APS March Meeting 2014, March 3-7, 2014 in Denver, Colorado
17. Haojie Ji, Siddharth Dhomkar, Maria C. Tamargo, and Igor L. Kuskovsky, Optical Anisotropy in Type-II ZnTe/ZnSe Submonolayer Quantum Dots, March Meeting of the APS, 2014, APS March Meeting 2014, Denver, Colorado
18. B. Roy, H. Ji, S. Dhomkar, L. Murokh, J. Ludwig, D. Smirnov, M. C. Tamargo, I. L. Kuskovsky, Decoherence mechanisms of Aharonov-Bohm excitons in type-II quantum dots, APS March Meeting 2014, March 3-7, 2014 in Denver, Colorado
19. N. Vaxelaire, S. Dhomkhar, I.L. Kuskovsky, M.C. Tamargo, I.C. Noyan, Quantum dot multilayers as probed by High Resolution X-Ray Diffraction, NSLS/NSLS-II & CFN Joint Users' Meeting, Brookhaven, May 19 - 21, 2014.
20. N. Vaxelaire, S. Dhomkhar, I.L. Kuskovsky, M.C. Tamargo, I.C. Noyan Quantum dot multilayers as probed by High Resolution X-Ray Diffraction, Denver X-ray Conference, Big Sky, 28 July-1 August, 2014.
21. H. Ji, S. Dhomkar, J. Ludwig, D. Smirnov, M. C. Tamargo, and I. L. Kuskovsky, Giant Zeeman Splitting in Submonolayer ZnTe/ZnSe Quantum Dots, The 21st International Conference on High Magnetic Fields in Semiconductor Physics, August 3-8, 2014, Panama City Beach, FL, USA
22. N. Vaxelaire, H. Ozturk, H. Yan, Y. Li, I.C. Noyan, Challenges and capabilities of Coherent Bragg Imaging with nanoparticles, International Workshop on Phase Retrieval and Coherent Scattering, Evanston, September 2-5, 2014.
23. S. Dhomkar, N. Vaxelaire, , H. Ji, V. Deligiannakis, J. Jordan-Sweet, M.C. Tamargo, I.L. Kuskovsky, I.C. Noyan, Diffuse X-ray Scattering as a Tool to Characterize Morphology of Multilayered Structures of Ultra-small (Submonolayer) Quantum Dots, APS March Meeting 2015, March 2-6 in San Antonio, Texas.
24. H. Ji, S. Dhomkar, J. Ludwig, D. Smirnov, M.C. Tamargo, I.L. Kuskovsky, g-Factors of Electrons, Holes and Excitons in Type-II ZnTe/ZnSe Submonolayer Quantum Dots, APS March Meeting 2015, March 2-6 in San Antonio, Texas.
25. S. Dhomkar, H. Ji, V. Deligiannakis, J. Ludwig, D. Smirnov, C. Meriles, M. C. Tamargo, I. L. Kuskovsky, Magneto photoluminescence of Tellurium Isoelectronic Bound Ecitons in n-Se-Te in the presence of Type II Submonolayer Quantum Dots, 17<sup>th</sup> International Conference on II-VI Compounds and Related Materials, September 13-18, 2015, Paris, France.

26. V. Deligiannakis, S. Dhomkar, H. Ji, I. L. Kuskovsky, M. C. Tamargo, C. Meriles, Spin Dynamics of Tellurium Isoelectronic Bound Excitons in Zn-Se-Te Nanostructures, 17<sup>th</sup> International Conference on II-VI Compounds and Related Materials, September 13-18, 2015, Paris, France. (*Best Student Poster Award*)
27. I. L. Kuskovsky, L. G. Mourokh, B. Roy, H. Ji, S. Dhomkar, J. Ludwig, D. Smirnov, and M. C. Tamargo, "Decoherence in semiconductor nanostructures with type-II band alignment: all-optical measurements using Aharonov-Bohm excitons," International Conference on Optics of Excitons in Confined Systems (OECS 2015), October 11-16, 2015, Jerusalem, Israel
28. V. Deligiannakis, S. Dhomkar, H. Ji, D. Pagliero, M. C. Tamargo, I. L. Kuskovsky, C. Meriles "Spin Dynamics of Tellurium Isoelectronic Centers Bound Excitons in Zn-Se-Te Nanostructures" APS March Meeting 2016, March 14-18, 2016, Baltimore, MD, USA

#### Invited Conference Presentations

1. I. L. Kuskovsky, "Submonolayer Type-II Quantum Dots: Fundamental Physics and Applications," Nanotechnology Workshop, November 12, 2012, CUNY Graduate Center, New York, NY USA
2. I. L. Kuskovsky, "Stacked Type-II II-VI Quantum Dots: Fundamental Physics and Applications," 2013 EMN West Meeting, Jan. 7 – 10, 2013, Houston, TX USA
3. M. C. Tamargo, "Wide Bandgap II-VI Compounds for High Performance Solar Cells," 9th Seminars of Advanced Studies on Molecular Design and Bioinformatics: Energy, SEADIM9, July 7-12, 2013, Havana, Cuba
4. M. C. Tamargo, "Semiconductor Nanostructures for Novel Device Applications," August 4-9, 2013, MIRTHE Summer Workshop, Princeton, NJ USA
5. M. C. Tamargo, "II-VI nanostructures on III-V substrates for novel applications," Invited Seminar at IBM Yorktown Heights, NY February 14, 2014
6. M. C. Tamargo, (Plenary) "II-VI Semiconductors for Photonic Device Applications: beyond the Blue Laser," 17<sup>th</sup> International Conference on II-VI Compounds and Related Materials, September 13-18, 2015, Paris, France.
7. I. Kuskovsky, "Application and Physics of Type-II Submonolayer Quantum Dots as an Active Medium for Intermediate Band Solar Cells," CLEO 2016, June 5-10, 2016, San Jose, CA
8. M. C. Tamargo, "Heterovalent and Heterocrystalline Epitaxy for Device Applications and New Physical Phenomena," 32nd North American Conference for Molecular Beam Epitaxy, September 18-21, 2016, Saratoga Springs, NY
9. I. Kuskovsky, "Application and Physics of Type-II Submonolayer Quantum Dots as an Active Medium for Intermediate Band Solar Cells", Optical Nanostructures and Advanced Materials for Photovoltaics (PV) at Light, Energy and the Environment 2016, 14–17 November 2016, Leipzig, Germany
10. M. C. Tamargo, "Novel Applications and New Physical Phenomena Based on Heterovalent and Heterocrystalline Nanostructures," The 2016 US Workshop on the Physics and Chemistry of II-VI Materials, October 14-20, 2016, Baltimore, MD

#### Awards

1. M. C. Tamargo, CCNY 2014 President's Award for Outstanding Faculty Service in the Division of Science
2. M. C. Tamargo, Program Committee Chair, 31<sup>st</sup> North American MBE, Mayan Riviera, Mexico 2015
3. M. C. Tamargo, Program Committee Chair, 18<sup>th</sup> International Conference on II-VI Compounds, San Juan, PR 2017

**People that worked on the project**

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