26 - 30 June 2023
Belgrade, Serbia

21. СИМПОЗИЈУМ ФИЗИКЕ КОНДЕНЗОВАНЕ МАТЕРИЈЕ

THE 21st SYMPOSIUM ON CONDENSED MATTER PHYSICS

BOOK OF ABSTRACTS

Institute of Physics Belgrade
Vinca Institute of Nuclear Sciences
University of Belgrade, Faculty of Physics
Serbian Academy of Sciences and Arts
Ministry of Science, Technological Development and Innovation

Conference Chairs

Vladimir Dobrosavljević, *Florida State University, USA*

Zorica Konstantinović, *Institute of Physics Belgrade*

Željko Šljivančanin, *Vinča Institute of Nuclear Sciences*

Organizing Committee

Jelena Pešić, *Institute of Physics Belgrade*- chair

Andrijana Šolajić, *Institute of Physics Belgrade*

Ivana Milošević, *Institute of Physics Belgrade*

Tijana Tomašević-Ilić, *Institute of Physics Belgrade*

Jelena Mitrić, *Institute of Physics Belgrade*

Bojana Višić, *Institute of Physics Belgrade*

Jovan Blagojević, *Institute of Physics Belgrade*

Marko Orozović, *Vinča Institute of Nuclear Sciences*

Mitra Stepić, *Vinča Institute of Nuclear Sciences*

Igor Popov, *Institute for Multidisciplinary Research, Belgrade*

Program Committee

Ivan Božović, *Brookhaven National Laboratory, USA*

Vladimir Dobrosavljević, *Florida State University, USA*

Milan Damnjanović, *Faculty of Physics, University of Belgrade, Serbia*

Vladimir Djoković, *Vinča Institute, University of Belgrade, Serbia*

Gyula Eres, *Oak Ridge National Laboratory, USA*

Laszló Forró, *University of Notre Dame, USA*

Radoš Gajić, *Institute of Physics Belgrade*

Igor Herbut, *Simon Fraser University, Canada*

Zoran Ikonić, *University of Leeds, UK*

Nenad Lazarević, *Institute of Physics Belgrade*

Ivanka Milošević, *Faculty of Physics, University of Belgrade, Serbia*

Milorad Milošević, *University of Antwerp, Belgium*

Milica Milovanović, *Institute of Physics Belgrade*

Stevan Nadj-Perge, *CALTECH, USA*

Branislav Nikolić, *University of Delaware, USA*

Cedomir Petrovic, *Brookhaven National Laboratory, USA*

Dragana Popović, *National High Magnetic Field Laboratory, USA*

Zoran V. Popović, *Institute of Physics Belgrade*

Zoran Radović, *Faculty of Physics, University of Belgrade, Serbia*

Miljko Satarić, *Faculty of Technical Sciences, University of Novi Sad, Serbia*

Marko Spasenović, *Institute of Chemistry, Technology and Metallurgy, Belgrade, Serbia*

Djordje Spasojević, *Faculty of Physics, University of Belgrade, Serbia*

Željko Šljivančanin, *Vinča Institute of Nuclear Sciences*

Bosiljka Tadić, *Jožef Štefan Institute, Slovenia*

Milan Tadić, *School of Electrical Engineering, University of Belgrade*, *Serbia*

Darko Tanasković, *Institute of Physics Belgrade*

Jasmina Tekić, *Vinča Institute of Nuclear Sciences*

21. СИМПОЗИЈУМ ФИЗИКЕ КОНДЕНЗОВАНЕ МАТЕРИЈЕ

THE 21st SYMPOSIUM ON CONDENSED MATTER PHYSICS

26 - 30 June 2023, Belgrade, Serbia

https://www.sfkm2023.ipb.ac.rs/

Institute of Physics Belgrade

Vinca Institute of Nuclear Sciences

University of Belgrade, Faculty of Physics

Serbian Academy of Sciences and Arts

Ministry of Science, Technological Development and Innovation

TABLE OF CONTENTS

1. INVITED TALKS

2. CONTRIBUTED TALKS

3. POSTER SESSION

Effect of Large Quantum Correlations in "Russian Doll" Quantum Dots: Impact on MEG Solar Cells

Stanko Tomić

Vinča Institute for Nuclear Sciences, The University of Belgrade, 11000 Belgrade, Serbia

Abstract. Semiconductor nanocrystals (NCs) are the subject of intensive research due to several novel properties which make them of interest for both fundamental science and technological applications. NCs are of particular interest for solar cell applications due to their ability to increase efficiency via the generation of multiexcitons from a single photon. Theoretical predictions indicate that multi-exiton generation (MEG) has the potential to enhance the efficiency of a single gap cell from 33% to 42% [1]. The efficiency of MEG in colloidal QDs is determined by the competition between MEG and other hot electron-cooling processes [2]. These have characteristic times of τ(MEG) ~ τ(cooling) ~1 ps in the colloidal QDs studied to date but for the high efficiency τ(cool) >> τ(MEG) is required. The "Russian Doll" aka core/shell QDs [3] with type II band alignment offers extra degree of freedom in mediating both the optical dipoles and the Coulomb interaction between charges in such structures. Full realization of this potential requires that the energy threshold for MEG be minimized. An attractive interaction between excitons in QD reduces the threshold by the biexciton binding energy, Bxx, but this has been found to be small (-10 meV) for type I QDs. Here, we show that, by taking into account quantum correlation effects between charges, combinations of core diameter and shell thickness can be found for a CdSe/CdTe core/shell QD that result in very large values of Bxx<0. In our analysis excitonic states were found using the full Configuration interaction (CI) method, that incudes explicitly the effects of Coulomb interaction, exact exchange and correlations between many-electron configurations in QD. In setting up the full CI, particular attention was paid to accurate modeling of the dielectric environment variation through the structure as well as surface polarization effects on core/shell and QD/colloid interfaces. Dielectric constants of constituent CdSe and CdTe around the transition energies poles are predicted using ab initio TDDFT [4]. We map the $1S^{(e)}nS^{(h)}$ (n = 1, 2) exciton correlation energy relative to the strong confinement regime as a function of core radius and shell thickness for non-uniform spatial dielectric environment [5]. We observe how the type-II confinement potentials amplifies the dielectric effect on the wave functions and exciton energies, particularly increasing the correlation energy for QDs in which the corresponding single-particle hole is delocalized [5]. We also find that correlation leads to large changes in the momentum matrix element, particularly for the lowest CdSe/CdTe QD exciton in which it is increased up to one order of magnitude in the presence of dielectric confinement. Overall dielectric confinement affected the exciton properties in CdSe/CdTe QDs more than the inverse heterostructures due to the band alignment, which encourages holes to localize in the shell. We conclude that: (i) it is not possible to predict biexciton binding using the Hartree approximation alone; it can only be predicted with a full CI Hamiltonian [6]; (ii) CI predicts Bxx = -70 meV for structures with 0.5 nm thick shell that agrees with experiment [7]; (iii) by ignoring the dielectric confinement, it is not possible to predict biexciton binding for structures with shell thickness > 0.75 nm; (iv) by changing the solvent's dielectric constant from 1 to 2 the variation in the Bxx binding energy is as

big as 100 meV; (v) a proper prediction of Bxx requires the inclusion of correlations and surface polarization effects but the effect of self-polarization is negligible. The strong biexciton binding found in Russian Doll QD is explained by the stronger reduction in the Columbic repulsion between holes than reduction in the attraction between electrons and holes upon the addition of the CdTe shell layer, Fig. 1. The Aufbau principle and Hund rule reveal this to be a consequence of 4 fold degeneracy of the hole ground state imposed by symmetry of the structure [8].

The peak efficiency in "Russian Doll" NC structures grows to 46% and then to 47% as Bxx increases to −25 and −50 meV, respectively. The largest biexciton interaction energy reported to date is ∼100 meV, although this was repulsive (i.e., antibinding) rather than attractive, it gives an indication of the magnitude of the biexciton interaction energies that can be achieved in "Russian Doll" type-II colloidal QD. For a binding energy of −100 meV, the peak efficiency rises to 50% and would rise to as much as 60% if it was possible to produce a binding of −200 meV. It can also be observed that the value of Eg corresponding to the peak efficiency shifts progressively to lower values as the photocurrent is enhanced first by MEG alone, and then by the combined effects of MEG and the onset of biexciton binding [6].

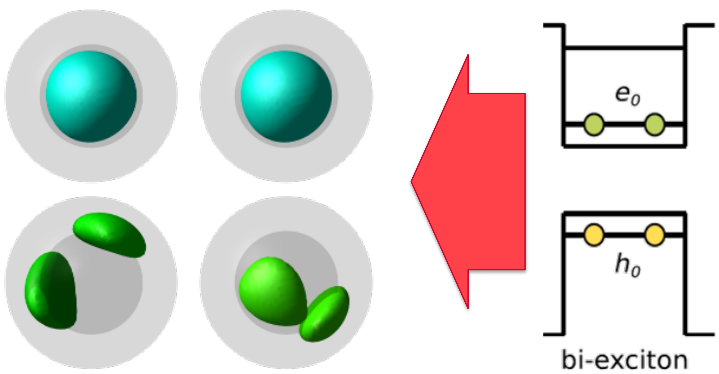

FIGURE 1. Topology of the dominant character of single particle electron/hole states in the ground state bi-exciton.

REFERENCES

1. V.I. Klimov, Appl. Phys. Lett. **89**, 123118 (2006)
2. F.C.M. Spoor, S. Tomić, A.J. Houtepen, L.D.A. Siebbeles, ACS Nano **11**, 6286 (2017)
3. J. Kim, L.-W. Wang, and A. Zunger, Phys. Rev. B **56**, R15541(R) (1997).
4. L. Bernasconi, S. Tomić, M. Ferrero, M. Rérat, R. Orlando, R Dovesi, Phys. Rev. B **83**, 195325 (2011)
5. E.J. Tyrrell and S. Tomić, J. Phys. Chem. C **119**, 12720 (2015)
6. S. Tomić, J.M. Miloszewski, E.J. Tyrrell, D.J. Binks, IEEE J. Photovolt. **6**, 179 (2016)
7. M.A. Leontiadou, E.J. Tyrrell, C.T. Smith, D.Espinobarro-Velazquez, R. Page, J. Miloszewski, T. Walsh, D. Binks, S. Tomić, Sol. Energy Mater Sol. Cells 159, 657 (2017).
8. C.T. Smith, E.J. Tyrrell, M.A. Leontiadou, J. Miloszewski, T. Walsh, M. Cadirci, R. Page, D. Binks, S. Tomić, Sol. Energy Mater Sol. Cells 158, 160 (2016).