ELECTRON-ELECTRON INTERACTIONS, SCREENING AND POLARIZABILITY

GRAPHENE AND SEMICONDUCTOR QDOTS

IN SEMICONDUCTOR AND

GRAPHENE QDOTS

I.OZFIDAN(1,5), P. POTASZ(1,3), A.D.GUCLU(1,4), M. KORKUSINSKI(2), O. VOZNYY(1,6), P. HAWRYLAK(1)

DEPARTMENT OF PHYSICS, UNIVERSITY OF OTTAWA, OTTAWA, CANADA
 EMERGING TECHNOLOGIES DIVISION, NRC, OTTAWA, CANADA
 INSTITUTE OF PHYSICS, WROCLAW UNIVERSITY OF TECHNOLOGY, POLAND
 INSTITUTE OF PHYSICS, IZMIR INSTITUTE OF TECHNOLOGY, IZMIR, TURKEY
 5 DEPARTMENT OF PHYSICS, UNIVERSITY OF ALBERTA
 6. EC, UNIVERSITY OF TOTONTO



SEMICONDUCTOR AND GRAPHENE QUANTUM DOTS

InAs QD IN InP NWIRE CONTROL OF HEIGHT, DIAMETER, SHELL

> dot

nanowire core PHOTONS FOR QCOMMUNICATION

HgTe QDOTS: QDOTS IN TOPOLOGICAL INSULATORS ROBUST EDGE STATES-SENSORS



PRINTABLE



GRAPHENE QDOTS – ATOMIC CONTROL OF HEIGTH:







OUTLINE

INTRODUCTION

SCREENING IN QUANTUM DOTS (cRPA)

GRAPHENE QDOTS:

BANGAP, EXCITONS AND BIEXCITONS

SUBLATTICE ENGINEERING-MAGNETIC MOMENT AND E-E CORRELATIONS



2D ELECTRON GAS

ELECTRON-ELECTRON INTERACTIONS? 2DEG OF SCHRODINGER FERMIONS

 $H = \sum_{i} \frac{1}{2m} \left(-\frac{\partial^{2}}{\partial \vec{r}_{i}^{2}} \right) + \sum_{i < j} \frac{e^{2}}{\varepsilon |\vec{r}_{i} - \vec{r}_{j}|}$ $H = Ry\left(\sum_{i} \left(-\frac{\partial^{2}}{\partial \vec{r}_{i}^{2}} \right) + \sum_{i < j} \frac{2}{|\vec{r}_{i} - \vec{r}_{j}|} \right)$ $H = \left(\sum_{i} \frac{1}{r_{s}^{2}} \left(-\frac{\partial^{2}}{\partial \vec{r}_{i}^{2}} \right) + \frac{1}{r_{s}} \sum_{i < j} \frac{2}{|\vec{r}_{i} - \vec{r}_{j}|} \right)$ $H = \frac{1}{r_{s}^{2}} \left(\sum_{i} \left(-\frac{\partial^{2}}{\partial \vec{r}_{i}^{2}} \right) + r_{s} \sum_{i < j} \frac{2}{|\vec{r}_{i} - \vec{r}_{j}|} \right)$

e- *r*s ee- e- e-

uOttawa L'Université canadienne Canada's university

PERTURBATION THEORY IN RS

ELECTRON-ELECTRON INTERACTIONS

2D ELECTRON GAS

2DEG OF SCHRODINGER FERMIONS-PERTURBATION THEORY

 $E_{k} = \xi_{k}^{e} + \Sigma_{e}(k, \xi_{k}^{e})$ $Im\Sigma_{e}(k, \xi_{k}^{e}) = \int_{0}^{\infty} \frac{d \, qq^{\Omega}}{2\pi} \int_{0}^{(k,q)} \frac{d\omega}{2\pi} \frac{\{1 - f(\xi_{k}^{e} - \omega) | \{-2 \, Im W_{ee}(q, \omega)\}\}}{2\pi \sqrt{[\omega - \Omega_{-}(k,q)][\Omega_{+}(k,q) - \omega]}}$ SCREENED INTERACTION W $RPA: Im(W(q, \omega)) = Im\{W^{0}/(1 - W^{0}\Pi^{0})\}$ $LOCAL FIELD CORRECTION W_{0} \Rightarrow W_{0}(1 - G)$ VNS: Im(W(q, \omega))

$$= \operatorname{Im} \{ W^{0}(1-G) / [1-W^{0}(1-G)\Pi^{0}] \}$$

١.

ELECTRON-ELECTRON INTERACTIONS 2DEG OF SCHRODINGER FERMIONS-PERTURBATION THEORY

2D ELECTRON GAS

 $E_k = \xi_k^e + \Sigma_e(k, \xi_k^e)$



Local field G

Renormalization of Plasmon energy

Renormalization of el Plasmon coupling

WITH JEFF YOUNG



2D ELECTRON GAS

ELECTRON-ELECTRON INTERACTIONS? 2DEG OF SCHRODINGER FERMIONS

$$H = \sum_{i} \frac{1}{2m} \left(-\frac{\partial^2}{\partial \vec{r}_i^2}\right) + V_{lattice}(\vec{r}_i) + \sum_{i < j} \frac{e^2}{\varepsilon |\vec{r}_i - \vec{r}_j|}$$

Lattice dominates



$$K^*p$$
$$H = \sum_{i} v_F \vec{\sigma} \left(-\frac{\partial}{\partial \vec{r}_i}\right) + \sum_{i < j} \frac{e^2}{\varepsilon |\vec{r}_i - \vec{r}_j|}$$



ELECTRON-ELECTRON INTERACTIONS? 2DEG OF DIRAC FERMIONS Kotov, Castro-N

2DEG OF DIRAC FERMIONS

$$H = \sum_{i} v_F \vec{\sigma} \left(-\frac{\partial}{\partial \vec{r}_i}\right) + \sum_{i < j} \frac{e^2}{\varepsilon |\vec{r}_i - \vec{r}_j|}$$

Kotov,Castro-Neto, Uchoa,Vozmedano, Guinea,DasSarmaí í .

$$H = \sum_{i} \frac{1}{r_s} v_F \vec{\sigma} \left(-\frac{\partial}{\partial \vec{r_i}}\right) + \frac{1}{r_s} \sum_{i < j} \frac{e^2}{\varepsilon |\vec{r_i} - \vec{r_j}|}$$

$$H = \left(\frac{1}{r_s}v_F\right)\left(\sum_i \vec{\sigma}\left(-\frac{\partial}{\partial \vec{r_i}}\right) + \frac{e^2}{2\varepsilon v_F}\sum_{i< j}\frac{2}{|\vec{r_i} - \vec{r_j}|}\right)$$



INDEPENDENT OF RS – DEPENDS ON SCREENING



OUTLINE

INTRODUCTION

SCREENING IN QUANTUM DOTS (cRPA)

GRAPHENE QDOTS:

BANGAP, EXCITONS AND BIEXCITONS

SUBLATTICE ENGINEERING-MAGNETIC MOMENT AND E-E CORRELATIONS





Qdot Hamiltonian

$$\hat{H}_0 = \sum_i \varepsilon_i c_i^+ c_i^- + \sum_{i,j} t_{j,i} c_j^+ c_i^-$$

may include Pz (relevant) and Sigma (irrelevant) orbitals Qdot energy spectrum and wavefunctions $H_0\varphi_i^0 = \varepsilon_i^0\varphi_i^0$

Introduce small *total* perturbation V $H = H_0 + \delta \cdot V$

Calculate perturbatively wavefunctions and energy levels of H

$$H\varphi_i = \mathcal{E}_i \varphi_i$$

$$\varphi_i = \varphi_i^0 + \delta \varphi_i^1 + \dots \qquad \qquad \varepsilon_i = \varepsilon_i^0 + \delta \varepsilon_i^1 + \dots$$



Calculate perturbatively wavefunctions and energy levels of H

$$\varphi_i(r) = \varphi_i^0(r) + \sum_{j \neq i} A_j^i \varphi_j^0(r)$$

$$\varphi_i(r) = \varphi_i^0(r) + \sum_{j \neq i} \frac{\langle j | V | i \rangle}{\varepsilon_i - \varepsilon_j} \varphi_j^0(r)$$

Calculate charge density induced by perturbation V (there are N electrons or N occupied states)

$$\rho(r) = \sum_{i=1}^{N} \varphi_i^*(r) \varphi_i(r)$$



Calculate charge density induced by perturbation V

$$\rho(r) = \sum_{i=1}^{N} \varphi_i^*(r) \varphi_i(r)$$

$$\rho(r) = \sum_{i=1}^{N} (\varphi_i^0(r) + \sum_{k \neq i} \frac{\langle k | V | i \rangle}{\varepsilon_i - \varepsilon_k} \varphi_k^0(r)) * (\varphi_i^0(r) + \sum_{j \neq i} \frac{\langle j | V | i \rangle}{\varepsilon_i - \varepsilon_j} \varphi_j^0(r))$$

induced charge density is given by

$$\delta n(r') = \sum_{i,k;i\neq k} \left(\frac{f(\varepsilon_i) - f(\varepsilon_k)}{\varepsilon_i - \varepsilon_k} < i | V | k > \varphi_k^0(r') * \varphi_i^0(r') \right)$$
polarizability
$$\delta n(r') = \sum_{i,k;i\neq k} \left(\prod_{ik}^0 < i | V | k > \varphi_k^0(r') * \varphi_i^0(r') \right)$$

U Ottawa L'Université canadienne Canada's university

induced charge density is proportional to applied total but weak potential

$$\delta n(r') = \sum_{i,k;i\neq k} \left(\prod_{ik}^{0} < i \, | \, V \, | \, k > \varphi_{k}^{0}(r') * \varphi_{i}^{0}(r') \right)$$

Induced charge density produces induced potential dV

Total potential V is a sum of external potential and induced potential

 $V(r) = V^{ext}(r) + \delta V(r)$

$$V(r) = V^{ext}(r) + \int dr' V^0(r,r') \delta n(r')$$

V0 may include image charges



induced charge density is proportional to applied total weak potential $V(r) = V^{ext}(r) + \int dr' V^0(r,r') \delta n(r')$

$$\delta n(r') = \sum_{i,k;i\neq k} \left(\prod_{ik}^{0} < i | V | k > \varphi_{k}^{0}(r') * \varphi_{i}^{0}(r') \right)$$

Take matrix elements of total potential, we end up with integral equation

$$\langle i | V | j \rangle = \langle i | V^{ext} | j \rangle + \sum_{k,l} \langle ik | V^0 | lj \rangle \prod_{kl}^0 \langle k | V | l \rangle$$

$$V = V^{ext} + V^0 \prod^0 V$$

$$V - V^0 \prod^0 V = V^{ext}$$
Screened RPA interaction
$$V = \frac{V^{ext}}{1 - V^0 \prod^0} \qquad V = \frac{V^{ext}}{\varepsilon}$$



OUTLINE

INTRODUCTION

SCREENING IN QUANTUM DOTS (cRPA)

GRAPHENE QDOTS:

BANGAP, EXCITONS AND BIEXCITONS

SUBLATTICE ENGINEERING-MAGNETIC MOMENT AND E-E CORRELATIONS





GOAL FOR GRAPHENE QUANTUM DOTS

DEMONSTRATE CARBONONICS:

ELECTRONICS, PHOTONICS AND SPINTRONICS IN A SINGLE MATERIAL USING

LATERAL SIZE, SHAPE, EDGE, SUBLATTICE AND NUMBER OF LAYERS ENGINEERING OF GRAPHENE

INTEGRATE THESE FUNCTIONALITIES AT THE NANOSCALE GRAPHENE INTEGRATED <u>QUANTUM</u> CIRCUIT

u Ottawa

I.Ozfidan, D.Guclu, P.Potasz, M.Korkusinski, í PH



GRAPHENE QUANTUM DOTS SELECTED REFERENCES

1. P. Hawrylak, "Surface Plasmons in Intercalated Graphite", Solid State Com. <u>63</u>, 241 (1987).

2. A.D. Guclu, P. Potasz, O. Voznyy, M. Korkusinski, P. Hawrylak,ö Magnetism and correlations in fractionally filled degenerate shells of graphene quantum dotsö, Phys.Rev.Letters, **103**, 246805 (2009).

1. Oleksandr Voznyy, Alev Devrim Güçlü, Pawel Potasz, Pawel Hawrylak, õEffect of edge reconstruction and passivation on zero-energy states and magnetism in triangular graphene quantum dots with zigzag edgesö, Phys.Rev.**B83**, 165417 (2011).

1. P. Potasz, A. D. Güçlü, A.Wojs and P. Hawrylak, õ Electronic properties of gated triangular graphene quantum dots: magnetism, correlations and geometrical effectsö, Phys. Rev. B **85**, 075431 (2012).

1. D.Guclu and P.Hawrylak, õOptical control of magnetization and optical spin blockade in triangular graphene quantum dotsö, Phys. Rev. **B**87, 035425 (2013).

1. D.Guclu, M.Grabowski and P.Hawrylak, õElectron-electron interactions and topology in the electronic properties of gated graphene nanoribbon rings in Mobius and cylindrical configurationsö, Phys.Rev. **B**87, 035435 (2013).

1. D.Guclu, P. Potasz and P.Hawrylak, Zero-energy states of graphene triangular quantum dots in a magnetic fieldö, Phys.Rev. B88 155429 (2013)

2. I.Ozfidan, M. Korkusinski, A.D.Guclu, J.McGuire and P.Hawrylak, õ Micoscopic theory of optical properties of colloidal graphene quantum dot Phys. Rev. B89,085310 (2014).

1. I.Ozfidan, M. Korkusinski and P.Hawrylak, õTheory of Biexcitons and Biexciton-Exciton Cascade in Graphene Quantum Dotsö, Phys.Rev.B91, 115314(2015).

1. Cheng Sun, Florian Figge, I.Ozfidan, M. Korkusinski, Xin Yan, Liang-shi Li, Pawel Hawrylak and John A. McGuire, Biexciton binding in colloidal graphene quantum dotsö, NanoLetters 15,5742(2015).

1. Devrim Guclu, Pawel Potasz, Marek Korkusinski and Pawel Hawrylak,öGraphene Quantum Dotsö, Springer-Verlag (2014).

2. P Hawrylak, F Peeters, K Ensslin, Editors, õCarbononicsóintegrating electronics, photonics and spintronics with graphene quantum dotsö, Focus issue, Physica status solidi (RRL)-Rapid Research Letters 10 (1), 11 (2016).







PHOTONICS WITH GRAPHENE QUANTUM DOTS

MAKING A SEMICONDUCTOR OUT OF A SEMIMETAL

OPENING A GAP



BANDGAP ENGINEERING IN GRAPHENE QUANTUM DOTS

BAND GAP AS A FUNCTION OF SIZE



BANDGAP ENGINEERING IN GRAPHENE QUANTUM DOTS

BANDGAP COMPARISON WITH SEMICONDUCTORS



BANDGAP ENGINEERING WITH COMPOUND SEMICONDUCTORS



ELECTRONIC PROPERTIES OF GRAPHENE QUANTUM

BANDGAP DEPENDS ON SIZE, EDGE AND SHAPE

ITAWA

ELECTRONIC PROPERTIES OF GRAPHENE QUANTUM DOTS

BANDGAP DEPENDS ON e-e INTERACTIONS

ELECTRONIC PROPERTIES OF GRAPHENE QUANTUM DOTS

BANDGAP DEPENDS ON SIZE, EDGE AND SHAPE

COLLOIDAL GRAPHENE QUANTUM DOTS

OPTICAL PROPERTIES OF GRAPHENE QUANTUM DOTS

C132

Broken symmetry

C168

Number of C atoms – 168 Edges – mixed zigzag-armchair Edges – H passivation

Colloidal Graphene Quantum Dots with Bell-Demotion of the second structures and the second structures of the second structures and structures of the second structures and structures and

OPTICAL PROPERTIES OF GRAPHENE QUANTUM DOTS Methodology: Tb+HF+CI Pz orbitals only $= \sum_{\langle i, j \rangle} t_{ij} c_{i\sigma}^{+} c_{j\sigma}$ t - tunneling $+ \frac{1}{2} \sum_{ijkl \sigma\sigma} \langle ij | V | kl \rangle c_{i\sigma}^{+} c_{j\sigma}^{+} c_{k\sigma} c_{l\sigma}$ e-e interactions

$$< ij | V | kl >= \iint dr_1 dr_2 \phi_i^*(r_1) \phi_j^*(r_2) e^2 \phi_k(r_2) \phi_l(r_1)$$

Slater-Koster orbitals

 κ -screening by sigma electrons and surrounding fluid

D.GUCLU, P.POTASZ, M.KORKUSINSKI, O.VOZNYY, PH, PRL2009

Correlated ground and excited states

$$|\alpha\rangle = (A^{\alpha} + \sum_{i,j} B^{\alpha}_{ij} b^{+}_{i\sigma} b_{j\sigma'} + \sum_{ijkl\sigma\sigma'} C^{\alpha}_{ijkl} b^{+}_{i\sigma} b^{+}_{j\sigma'} b_{k\sigma'} b_{l\sigma} + \sum_{ijklmn\sigma\sigma'\sigma''} D^{\alpha}_{ijkl} b^{+}_{i\sigma} b^{+}_{j\sigma'} b_{k\sigma''} b_{l\sigma''} b_{m\sigma'} b_{n\sigma} +) |GS_{HF}\rangle$$

GROUND STATE OF C168 AS A FUNCTION OF STRENGTH OF ELECTRON-ELECTRON INTERACTIONS

L'Université canadienne Canada's university

OPTICAL PROPERTIES OF C168 IN SEMIMETALLIC REGIME

Ef **Tb and HF gaps almost identical**

VB

Ozfidan.Guclu.Korkusinski.PH

tb+HF

62

64

66

68

70

72

index

74

76

78

80

82

84

Energy (eV)

0

-2 -

-4 + 60

OPTICAL PROPERTIES OF GRAPHENE QUANTUM DOTS

DEGENERACY : VALLEY DEGENERACY / C3 SYMMETRY

$$\begin{split} \Psi_{j}^{m} &= \frac{1}{\sqrt{3}} \sum_{\beta = 0,1,2} e^{i\beta \cdot m \cdot 2\pi/3} |j_{\beta}\rangle \\ & \text{m=0,+1,-1} \\ \text{Degeneracy m=+/-1} \quad \begin{array}{c} \text{or} \\ \text{m=0,1,2} \\ \end{array} \end{split}$$

$$\langle m, i | \vec{E} \cdot \vec{r} | m', j \rangle = \delta_{m', m \pm 1} D_{m, m', i, j}$$

Optical selection rules

 $\Delta m = \pm 1$

OPTICAL TRANSITIONS IN GRAPHENE QUANTUM DOTS

OPTICAL PROPERTIES OF C168 IN SEMIMETALLIC REGIME

SINGLET/TRIPLET EXCITONS AT THE BAND EDGE

OPTICAL PROPERTIES OF GRAPHENE QUANTUM DOTS

OPTICAL PROPERTIES OF GRAPHENE QUANTUM DOTS

L'Université canadienne Canada's university

OPTICAL PROPERTIES OF GRAPHENE QUANTUM DOTS

ABSORPTION Ó THEORY VS EXPERIMENT Singlet-triplet splitting too small?

 $\kappa = 5, t = -4.2 eV$ Absorption (arb. units) C168 %10 of the highest absorption peak has been assigned to absorption of dark singlets Absorption C132 Absorption (arb. units) **Triplet Levels** Singlet Absorption Peaks Absorption (Experiment) 1.5 2.5 3.0 2.0 3.5 Energy (eV)

BLUE SHIFT

TRANSIENT ABSORPTION-DETECTION OF EXCITED X* AND XX IN GRAPHENE QUANTUM DOTS

J.McGuire et al

DETECTING EXCITED X AND XX STATES

NanoLetters 2015

BIEXCITON-EXCITON CASCADE IN GRAPHENE QUANTUM DOTS XX – X CASCADE FOR ENTANGLED

PHOTON PAIR GENERATION

NO SIGN OF EXCITON CONDENSATION BUT STRONG X_X INTERACTION

OUTLINE

INTRODUCTION

SCREENING IN QUANTUM DOTS (cRPA)

GRAPHENE QDOTS:

BANGAP, EXCITONS AND BIEXCITONS

SUBLATTICE ENGINEERING-MAGNETIC MOMENT AND E-E CORRELATIONS

SPINTRONICS RESTS ON LIEB'S THEOREM

GROUND STATE SPIN OF A HUBBARD MODEL ON BIPARTITE LATTICE S=Na-Nb

SPINTRONICS = SUBLATTICE ENGINEERING

Edge spin Polarization No gap Gap but AF Coupling No net spin BROKEN SUBLATTICE SYMMETRY Ferromagnetic coupling Coupling Max spin

Voznyy, Guclu.. PH PRB2011

GRAPHENE QDOTS

TURNING OFF MAGNETISM WITH GATE(VOLTAGE)

VOLTAGE CONTROL OF CHARGE DENSITY AND MAGNETIC MOMENT

FILLING UP ZERO-ENERGY HF SHELL

TOTAL SPIN OF ELECTRONS ON A DEGENERATE SHELL

HALF FILLED SHELL SPIN POLARISED AS IN DFT/MEAN-FIELD Ezawa; Fernandez-Rossier&Palacios; Kaxiras et al

FILLING UP ZERO-ENERGY HF SHELL

ADDING A SINGLE ELECTRON DEPOLARISES HALF FILLED SPIN POLARISED SHELL!

PHOTONICS AND SPINTRONICS

GRAPHENE QDOTS

TURNING OFF MAGNETISM WITH GATE AND LIGHT

OPTICAL SPIN BLOCKADE

SUMMARY

SCREENING IN SEMICONDUCTOR QUANTUM DOTS (cRPA)

GRAPHENE QDOTS:

BANGAP, EXCITONS AND BIEXCITONS

SUBLATTICE ENGINEERING - MAGNETIC MOMENT, ZERO ENERGY SHELL AND E-E CORRELATIONS, e-e AND VPEG IN WS2

GRAPHENE AND SEMICONDUCTOR QDOTS

ELECTRON-ELECTRON INTERACTIONS, SCREENING AND POLARIZABILITY IN SEMICONDUCTOR AND GRAPHENE QDOTS I.OZFIDAN(1,5), P. POTASZ(1,3), A.D.GUCLU(1,4), M. KORKUSINSKI(2), O. VOZNYY(1,6), P. HAWRYLAK(1)

DEPARTMENT OF PHYSICS, UNIVERSITY OF OTTAWA, OTTAWA, CANADA
 EMERGING TECHNOLOGIES DIVISION, NRC, OTTAWA, CANADA
 INSTITUTE OF PHYSICS, WROCLAW UNIVERSITY OF TECHNOLOGY, POLAND
 INSTITUTE OF PHYSICS, IZMIR INSTITUTE OF TECHNOLOGY, IZMIR, TURKEY
 5 DEPARTMENT OF PHYSICS, UNIVERSITY OF ALBERTA
 6. EC, UNIVERSITY OF TOTONTO

