

Correlations in semiconductor quantum dots

by

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Thesis

presented to the Faculty of Graduate and Postdoctoral Studies
of the University of Ottawa

in partial fulfilment of the requirements for the Degree of
DOCTOR OF PHILOSOPHY IN PHYSICS

June 8, 2004

Ottawa – Carleton Institute of Physics
University of Ottawa
Ottawa, Ontario, Canada

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Abstract

In this Thesis, I present a theoretical study of correlation effects in strongly interacting electronic and electron-hole systems confined in semiconductor quantum dots. I focus on three systems: N electrons in a two-dimensional parabolic confinement in the absence and in the presence of a magnetic field, an electron-hole pair confined in a vertically coupled double-quantum-dot molecule, and a charged exciton in a quantum-ring confinement in a magnetic field.

To analyse these systems I use the exact diagonalisation technique in the effective-mass approximation. This approach consists of three steps: construction of a basis set of particle configurations, writing the Hamiltonian in this basis in a matrix form, and numerical diagonalisation of this matrix. Each of these steps is described in detail in the text.

Using the exact diagonalisation technique I identify the properties of the systems due to correlations and formulate predictions of how these properties could be observed experimentally. I confront these predictions with results of recent photoluminescence and transport measurements.

First I treat the system of N electrons in a parabolic confinement in the absence of magnetic field and demonstrate how its properties, such as magnetic moments, can be engineered as a function of the system parameters and the size of the Hilbert space.

Next I analyse the evolution of the ground state of this system as a function of the magnetic field. In the phase diagram of the system I identify the spin-singlet $\nu = 2$

phase and discuss how correlations influence its phase boundaries both as a function of the magnetic field and the number of electrons.

I also demonstrate that in higher magnetic fields electronic correlations lead to the appearance of spin-depolarised phases, whose stability regions separate the weakly correlated phases with higher spin. Further on, I consider electron-hole systems. I show that the Coulomb interaction leads to entanglement of the states of an electron and a hole confined in a pair of vertically coupled quantum dots.

Finally I consider the system of two electrons and one hole (a negatively charged exciton) confined in a quantum ring and in the presence of the magnetic field. I show that the energy of a single electron in the ring geometry exhibits the Aharonov-Bohm oscillations as a function of the magnetic field. In the case of the negatively charged exciton these oscillations are nearly absent due to correlations among particles, and as a result the photoluminescence spectra of the charged complex are dominated by the energy of the final-state electron. The Aharonov-Bohm oscillations of the energy of a single electron are thus observed directly in the optical spectra.

Sommaire

Une étude théorique des corrélations électroniques dans des points quantiques semi-conducteurs où les électrons et les trous interagissent fortement entre eux est présentée. Trois systèmes en particulier sont considérés: N électrons confinés dans un potentiel parabolique bi-dimensionnel en absence et en présence d'un champ magnétique, une paire électron-trou isolée dans deux points quantiques couplés verticalement, et un complexe excitonique formé dans un anneau quantique soumis à un champ magnétique externe.

Une technique de diagonalisation exacte dans l'approximation de la masse effective est employée pour analyser ces systèmes. Cette approche se divise en trois étapes : la construction d'une base d'états dans l'espace des configurations possibles pour les particules, l'écriture en forme matricielle du Hamiltonien dans la base construite, et la diagonalisation numérique de la matrice. Chacune de ces étapes est expliquée en détails dans cette thèse.

En utilisant cette technique, nous étudions les effets des corrélations sur les propriétés électroniques des systèmes en question et formulons des prédictions théoriques permettant l'observation expérimentale de ces effets. Ces prédictions sont confrontées à des résultats récents obtenus par mesures optique et de transport électrique dans ces systèmes.

On démontre tout d'abord comment les propriétés électroniques de N électrons confinés en deux dimensions dans un potentiel parabolique, comme par exemple le moment magnétique, peuvent être ajustées, en absence de champ magnétique, en fonction des paramètres du système et de la taille de l'espace d'Hilbert.

On analyse ensuite l'évolution en champ magnétique de l'état fondamental. Une phase de spin singulet $\nu = 2$ est identifiée dans le diagramme de phase et l'influence des corrélations sur cette phase est étudiée en fonction du champ magnétique et du nombre d'électrons. On démontre aussi qu'à plus fort champ magnétique, les corrélations électroniques induisent de nouvelles phases dépolarisées en spin dont les régions de stabilité séparent les phases polarisées mais moins corrélées.

On montre, en second lieu, comment l'interaction de Coulomb enchevêtre l'état d'un électron avec celui d'un trou, tous deux confinés dans une paire de points quantiques couplés verticalement.

Finalement, on considère un complexe excitonique composé de deux électrons et d'un trou (exciton chargé) formé dans un anneau quantique en présence d'un champ magnétique. On montre que les oscillations d'Aharanov-Bohm, présentes dans le spectre à une particule, disparaissent dans celui de l'exciton chargé. Cet effet, dû aux corrélations entre les trois particules, implique que le spectre de photoluminescence de l'exciton chargé est dominé par l'énergie de l'électron non apparié. Les oscillations d'Aharanov-Bohm de cet électron peuvent être alors observées optiquement.

Acknowledgements

I would like to express my deepest gratitude to all the people that made the completion of this thesis possible.

First and foremost, I thank Dr. Pawel Hawrylak, my research supervisor, for making my Ph.D. studies in Canada possible, teaching me everything I know about how to do research, and putting up with me for all these years. His personal involvement led to the creation of a unique group of coworkers, with a harmonious atmosphere and enough challenges to ensure the growth of all of its members.

I thank all my colleagues, past and present. They are, in order of appearance: Dr. Shuhei Yoshida, Dr. Andreas Wensauer, Dr. Jordan Kyriakidis, Dr. Brandon van Zyl, Dr. Shun-Jen Cheng, Dr. Weidong Sheng, Dr. Marian Florescu, Alexandra Olaya Castro, Rowan Thomson, Juan Ignacio Climente Plasencia, Wojciech Dybalski, Dr. Ramin Abolfath. You have always been fantastic coworkers and friends. It is amazing how such very different people can find ways of working together so smoothly and entirely without conflicts, which, especially in the scientific community, is not that easy to achieve.

I also thank our experimentalist colleagues: Dr. Andy Sachrajda, Dr. Mariusz Ciorga, Michel Pioro-Ladrière, who endured patiently my theoretical ramblings on many occasions. I thank Michel for his help with the French version of the abstract of this Thesis.

I extend my gratitude to the Institute for Microstructural Sciences, National Research Council of Canada, with which I was affiliated as a visitor during the four years of my studies. All the results described in this work were obtained using the Institute's com-

puting facilities, and hereby I wish to thank Claude Cantin and Dr. Geof Aers for their patience and understanding in this context (they know best what I mean).

And, last but not least, I thank my Parents, Anna and Jacek Korkusiński, and my Brother Aleksander, for their unwavering support over these thousands of kilometres of land and sea between us. Unfortunately, my Father's time in this world ended before this thesis was submitted. I wish to dedicate this work to Him and to all my Family.

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List of Acronyms

2DEG	two-dimensional electron gas
CI	configuration interaction (method)
CM	centre of mass
DFT	density functional theory
DOS	density of states
FD	Fock-Darwin (spectrum)
IMS	Institute for Microstructural Sciences, National Research Council of Canada
IQHE	integer quantum Hall effect
FQHE	fractional quantum Hall effect
LL	Landau level
LLL	lowest Landau level
MDD	maximum-density droplet
PL	photoluminescence
QD	quantum dot
QDIP	quantum-dot infrared photodetector
QDMC	quantum diffusion Monte Carlo
QMC	quantum Monte Carlo
QWIP	quantum-well infrared photodetector
SAD	self-assembled dot
SDFT	spin density functional theory
SET	single-electron transistor
WL	wetting layer

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Plan of the Thesis

and Statement of Originality

In this Thesis I present a theoretical study of correlation effects in strongly interacting electronic and electron-hole systems confined in semiconductor nanostructures. The problem of interest can be posed rigorously by writing the Hamiltonian of a system of N particles confined in a potential $V(x, y, z)$ of a nanostructure:

$$\hat{H} = \sum_{i=1}^N \left[\frac{1}{2m_i^*} \left(\hat{\mathbf{p}}_i - \frac{q_i}{c} \mathbf{A}_i \right)^2 + V_i(\mathbf{r}_i) \right] + \sum_{i \neq j}^N \frac{q_i q_j}{\varepsilon |\mathbf{r}_i - \mathbf{r}_j|}. \quad (1)$$

Here, $\hat{\mathbf{p}}_i$, \mathbf{r}_i , q_i , and m_i^* are respectively: the momentum and position operators and the charge and effective mass of the i -th particle, \mathbf{A} is the magnetic vector potential, c is the speed of light, and ε is the dielectric constant of the material.

The first term in this Hamiltonian describes the problem of a single particle moving in the potential well V of the nanostructure in the presence of an external magnetic field. This problem is at most three-dimensional, and in most cases it can be solved exactly, using analytical or numerical methods.

What makes the Hamiltonian difficult to analyse is the second term, which describes the Coulomb interactions. Now the particles are no longer independent and one needs to build the basis of the Hilbert space for the problem out of configurations of all N particles. The size of this basis grows factorially with the number N . This makes the problem of interacting particles too difficult to solve exactly on present-day computers.

Approximate methods have been developed to approach the problem. For example, in mean-fields treatments, such as the Hartree-Fock theory, the wave function of the system is postulated in the form of a single Slater determinant:

$$\Psi(\mathbf{r}_1\sigma_1, \dots, \mathbf{r}_N\sigma_N) = \begin{vmatrix} \phi_1(\mathbf{r}_1)\chi_{\sigma_1}(\mathbf{r}_1) & \dots & \phi_1(\mathbf{r}_N)\chi_{\sigma_1}(\mathbf{r}_N) \\ \vdots & & \vdots \\ \phi_N(\mathbf{r}_1)\chi_{\sigma_N}(\mathbf{r}_1) & \dots & \phi_N(\mathbf{r}_N)\chi_{\sigma_N}(\mathbf{r}_N) \end{vmatrix}, \quad (2)$$

built out of effective single-particle orbital and spin wave functions $\phi_i(\mathbf{r})\chi_{\sigma_i}(\mathbf{r})$. The total energy is then formulated as a functional of these effective orbitals, and then minimised self-consistently by adjusting these orbitals one by one until convergence. This means in practice that the system is modelled on a single-particle level, but now each particle moves not only in the potential $V(x, y, z)$ of the nanostructure, but also in the mean effective field created by all other particles.

Note that in this approach, allowing to approximate only the ground-state energy of the system, each particle interacts with all other particles not as with point-like objects, but rather through a smeared-out effective potential, which accounts for the total Coulomb direct and exchange interactions. The mean-field theories usually work well in weakly interacting systems, i.e., systems, where the first term of the Hamiltonian (1) plays the dominant role, and the second term is just a perturbation. In such systems the ground state can indeed be well approximated by a single Slater determinant. However, the confinement $V(x, y, z)$ of semiconductor nanostructures can be chosen in such a way that the characteristic energy scale of the single-particle problem is the same as that of interactions, or even smaller. To show this, let us examine the two-dimensional parabolic confinement potential, used frequently in this work:

$$V(r) = \frac{1}{2}m^*\omega_0^2r^2, \quad (3)$$

with the characteristic frequency ω_0 . I will prove later on that the energy gaps between the levels of its single-particle spectrum are of order of $\hbar\omega_0$, while interactions scale as

$\frac{e^2}{\varepsilon} \sqrt{\frac{2m^*}{\hbar^2}} \sqrt{\hbar\omega_0}$ (\hbar is the Dirac's constant, ε is the dielectric constant of the material, and m^* and e are the electronic effective mass and charge, respectively). Thus the ratio λ of the characteristic interaction and single-particle energies is $\lambda = \frac{e^2}{\varepsilon} \sqrt{\frac{2m^*}{\hbar^2}} \frac{1}{\sqrt{\omega_0}}$, and can be made larger than 1 for ω_0 small enough (for instance, in GaAs, $m^* = 0.067 m_0$ and $\varepsilon = 12.4$, and $\lambda > 1$ for $\hbar\omega_0 < 23.7$ meV, a value easily obtained experimentally). The properties of these strongly interacting systems cannot be fully understood in terms of quasiparticles (particles dressed in interactions), because in this regime the particles are correlated and manifest collective behaviour. To account for these phenomena, the wave function should also contain terms dependent explicitly on differences between electronic coordinates, which is not fully included in the mean-field treatment. This is achieved by writing the eigenstates (ground and excited) of the Hamiltonian (1) not as a single Slater determinant, but rather as linear combinations of many such objects. In order to establish what these linear combinations should be, one needs to move to more sophisticated, nonperturbative approaches. The goal of this Thesis is to use such approaches to examine correlation effects in these systems, and contrast them with those due to the Coulomb direct and exchange terms.

I choose to study the correlation effects in semiconductor nanostructures, because such systems can be realised experimentally, offering a possibility of direct verification of my findings. The nanostructures I focus on in this context are quantum dots and quantum rings. I start this work with a general introduction to these zero-dimensional structures, from fundamental concepts to specific realisations (Chapter 1). In this Chapter I also present a review of the available literature treating the subject of many interacting particles confined in them. As I have pointed out, proper treatment of correlations in these systems cannot be accomplished within the mean-field approaches, but requires sophisticated methods and tools of mathematical and computational physics. My presentation of these tools starts in Chapter 2 with a description of single-particle properties of typical nanostructures. Having done that, in Chapter 3 I formulate the problem of many

interacting particles confined in nanostructures and describe the exact diagonalisation method, capable of capturing the physics of such systems in an exact (nonperturbative) manner. This level of complexity of my theories is necessary to account for the correlation effects in the system, although I acknowledge the existence of other methods, capable of accomplishing this task in an approximate manner. Only then do I proceed to the main body of my work. In Chapter 4 I analyse the properties of systems of many electrons confined in parabolic quantum dots and I show that properties of such systems, such as their magnetic moments, can be engineered. This engineering is accomplished by controlling the Hilbert space of the many-particle system, which, in turn, is achieved by tuning the single-particle properties of the dots. In Chapter 5 I analyse many-electron systems under parabolic confinement and in an external magnetic field, and I show how the electronic correlations modify the evolution of these systems as a function of the number of electrons, the magnetic field, the Zeeman energy, etc. Then I move on to electron-hole systems, and in Chapter 6 I consider a single electron-hole pair confined in the vertically coupled double-quantum-dot molecule. I show that the Coulomb interactions of the carriers lead to the appearance of a special kind of correlation, known as entanglement. I continue the analysis of electron-hole systems in Chapter 7, where I demonstrate how correlations can lead to suppression of Aharonov-Bohm oscillations of an exciton and a negatively charged exciton confined in a quantum ring. I conclude the thesis with a brief summary in Chapter 8.

All the calculations presented in the text were carried out under the supervision of Dr. Pawel Hawrylak, the Group Leader of the Quantum Theory Group at the Institute for Microstructural Sciences, National Research Council in Ottawa. Since many of the methods are numerical in nature, this Group adopted a specific procedure to prove their correctness: apart from the cases where the result was obvious, it had to be reproduced independently by two or more people. Note the word “independently”: in this case it means that each of the persons involved had to parametrise the model in their own

way, write their own computer programs, and prepare their own presentation of results. Therefore I feel that I can, in good faith, claim the results presented herein as my own, however not without crediting the coworkers and colleagues, who have helped to put this research on solid scientific ground. They are:

- Dr. Pawel Hawrylak, of the Institute of Microstructural Sciences, National Research Council of Canada, Ottawa - the Group Leader, who directed all of our research, ultimately supervising and reproducing all the results;
- Dr. Andreas Wensauer, formerly at the Physics Department, University of Regensburg, Germany, now at Framatome Inc., Erlangen, Germany, a visitor at IMS, and a co-developer of the exact diagonalisation tools used to analyze the spectra of lateral gated quantum dot devices;
- Dr. Jordan Kyriakidis, formerly a Research Associate at IMS, now an Assistant Professor at the Physics Department, Dalhousie University, Halifax, NS, who was involved in analyzing the electronic spectra of deformed lateral dots;
- Dr. Shun-Jen Cheng, formerly a Research Associate at IMS, now an Assistant Professor at National Chiao Tung University in Taiwan, who worked on the excitonic spectra of deformed self-assembled dots and dots in high magnetic fields;
- Dr. Weidong Sheng, a Research Associate at IMS, who has worked on microscopic models of electron and hole states in self-assembled quantum dots;
- Juan Ignacio Climente Plasencia, now a Ph.D. Student at the Physics Department, Universitat Jaume I, Castelló, Valencia, Spain, who worked on double-hole spectra of self-assembled dots.

I am indebted to all these coworkers and colleagues.

Most of our original results have already been published in refereed journals (see Appendix A for the full list of papers), which prompted me to build this Thesis as a

collection of articles, rather than write a separate monograph. Out of the full list I have chosen eight papers, which, in my opinion, present a consistent line of research into correlations in nanostructured materials. These papers are:

1. A. Wensauer, M. Korkusiński, and P. Hawrylak, “Configuration interaction method for Fock-Darwin states”, *Solid State Commun.* **130**, 115 (2004);
2. M. Korkusiński, W. Sheng, and P. Hawrylak, “Designing quantum systems in self-assembled quantum dots”, *Phys. Stat. Sol. (b)* **238**, 246 (2003);
3. A. Wensauer, M. Korkusiński, and P. Hawrylak, “Theory of spin-singlet filling factor $\nu = 2$ quantum Hall droplet”, *Phys. Rev. B* **67**, 035325 (2003);
4. M. Korkusiński, P. Hawrylak, M. Ciorga, M. Pioro-Ladrière, and A.S. Sachrajda, “Pairing of spin excitations in lateral quantum dots”, submitted for publication in *Phys. Rev. Lett.*;
5. M. Korkusiński and P. Hawrylak, “Electronic structure of vertically stacked self-assembled quantum disks”, *Phys. Rev. B* **63**, 195311 (2001);
6. M. Korkusiński, P. Hawrylak, M. Bayer, G. Ortner, A. Forchel, S. Fafard, and Z. Wasilewski, “Entangled states of electron-hole complex in a single InAs/GaAs coupled quantum dot molecule”, *Physica E* **13**, 610 (2002);
7. M. Korkusiński, P. Hawrylak, and M. Bayer, “Negatively charged exciton on a quantum ring”, *Phys. Stat. Sol. (b)* **234**, 274 (2002);
8. M. Bayer, M. Korkusiński, P. Hawrylak, T. Gutbrod, M. Michel, and A. Forchel, “Optical detection of the Aharonov-Bohm effect on a charged particle in a nanoscale quantum ring”, *Phys. Rev. Lett.* **90**, 186801 (2003).

As can be seen from this list, all these papers resulted from joint effort. The first four of them are strictly theoretical in nature, and, in the best possible faith, I declare that I have

contributed to their entire content within the collaborative procedure outlined above, with only one exception: the spin density functional calculations presented in the third paper are due to Dr. Andreas Wensauer, and I have not reproduced them on my own. As for the last four papers, they cover both theoretical and experimental aspects of the research, but the emphasis is put on a detailed presentation of the theory, and experimental results are only invoked to confirm it. Being a theorist, I have not contributed to the experimental aspects of these publications, but I declare my full involvement in the theories presented there - again, within the collaborative procedure described earlier.

Searches of available literature have revealed a number of publications tackling problems similar to those presented in this Thesis, and using similar methods; I review some of these publications in order to put our results in the appropriate perspective. I find, however, that in each of these cases my work is sufficiently distinct to be considered unique and original.