

John von Neumann Institute for Computing (NIC)

Quantum Simulations of Complex Many-Body Systems: From Theory to Algorithms

edited by

Johannes Grotendorst

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Alejandro Muramatsu

Winter School, 25 February - 1 March 2002

Rolduc Conference Centre, Kerkrade, The Netherlands

Poster Presentations

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Universität Stuttgart

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Preface

This Winter School continues a series of schools and conferences in Computational Science organized by the John von Neumann Institute for Computing (NIC). The topics of the School, Quantum Monte Carlo and Quantum Molecular Dynamics, play an outstanding role in many NIC research projects which use the supercomputing facilities provided by the Central Institute for Applied Mathematics (ZAM) of the Research Centre Jülich. The programme of the Winter School covers modern quantum simulation techniques and their implementation on high-performance computers, in particular on parallel systems. The focus clearly is on numerical methods which are tailored to treat large quantum systems with many coupled degrees of freedom ranging from superfluid Helium to chemical reactions. Among others, the following topics are treated by twenty-five lectures:

- Diffusion and Green's function Monte Carlo
- Path integral Monte Carlo and Molecular Dynamics
- Car-Parrinello / ab initio Molecular Dynamics
- Real-time quantum dynamics for large systems
- Lattice and continuum algorithms
- Exchange statistics for bosons and fermions / sign problem
- Parallel numerical techniques and tools
- Numerical integration and random numbers

This strongly interdisciplinary School aims at bridging three “gaps” in the vast field of large-scale quantum simulations. The first gap is between chemistry and physics, the second one between typical graduate courses in these fields and state-of-the-art research, and finally the one between the Monte Carlo and Molecular Dynamics communities. The participants will benefit from this School by learning about recent methodological advances within and outside their field of specialization. In addition, they get insight into recent software developments and implementation issues involved, in particular in the context of high-performance computing.

The lecturers of this Winter School come from chemistry, physics, mathematics and computer science and this is true for the audience as well. Participants from thirty mainly European countries attend the NIC Winter School, and more than eighty contributions have been submitted for the poster sessions. This overwhelming international resonance clearly reflects the attractiveness of the programme and demonstrates the willingness of the participants to play an active role in this high-level scientific School.

The scientific programme was worked out by Johannes Grotendorst (Research Centre Jülich), Dominik Marx (Ruhr-Universität Bochum), and Alejandro Muramatsu (Universität Stuttgart). The programme structure consists of overview lectures on various important fields, focus lectures on Quantum Monte Carlo and Quantum Molecular Dynamics methods, and special lectures on numerical and computational techniques.

Many organizations and individuals have contributed significantly to the success of this Winter School. Without the financial support of the European Commission within the framework of the specific research and training programme “Improving Human Research Potential” this one-week School on quantum simulation methods would not have been possible. We are grateful for the generous financial support by the Federal Ministry for Education and Research (BMBF) and by the Research Centre Jülich as well as for the help provided by its Conference Service and its Central Institute for Applied Mathematics.

We are greatly indebted to the local organization committee at Forschungszentrum Jülich who did the bigger part of the preparing work, namely Rüdiger Esser (Finance), Bernd Krahl-Urban (Accommodation and Registration) and Monika Marx (Web Management, Proceedings), and last but not least the conference secretaries Yasmin Abdel-Fattah, Elke Bielitz and Anke Reinartz. Special thanks go to Monika Marx for her tireless commitment concerning the editing and realization of this book. Furthermore, we appreciate the work of Stephan Brück who supported the difficult typesetting with great care. Finally, we would like to thank both the Ruhr-Universität Bochum and the Universität Stuttgart for their support of this activity in the area of high-end scientific education.

This book of abstracts of the poster presentations is also published in the internet at <http://www.fz-juelich.de/nic-series/volume11>.

Jülich, Bochum, and Stuttgart
February 2002

Johannes Grotendorst
Dominik Marx
Alejandro Muramatsu

Programme

Quantum Simulations of Complex Many-Body Systems: From Theory to Algorithms

NIC Winter School
25 February - 1 March 2002
Rolduc Conference Centre, Kerkrade
The Netherlands

Monday, 25 February 2002

- 9.00-17.30 **Registration**
- 9.30-10.00 **Opening**
Friedel Hoßfeld, NIC, Forschungszentrum Jülich
Dominik Marx, Ruhr-Universität Bochum
- 10.00-11.00 *Marius Lewerenz, Université Pierre et Marie Curie, France*
Monte Carlo Methods: Overview and Basics
Chair: Alejandro Muramatsu
- 11.00-11.30 **Coffee Break**
- 11.30-12.30 *Wolfhard Janke, Universität Leipzig*
Pseudo Random Numbers: Generation and Quality Checks
Chair: James B. Anderson
- 12.30-14.30 **Lunch Break**
- 14.30-15.30 *James B. Anderson, The Pennsylvania State University, USA*
Diffusion and Green's Function Quantum Monte Carlo Methods
Chair: Fakher F. Assaad
- 15.30-16.30 *Godehard Sutmann, Forschungszentrum Jülich*
Classical Molecular Dynamics
Chair: Christian Lubich
- 16.30-17.00 **Coffee Break**
- 17.00-18.00 *Mark E. Tuckerman, New York University, USA*
Path Integration via Molecular Dynamics
Chair: Rafael Ramírez

Tuesday, 26 February 2002

- 9.00-10.00 *David M. Ceperley, University of Illinois, USA*
Path Integral Monte Carlo
Chair: Marius Lewerenz
- 10.00-11.00 *David M. Ceperley, University of Illinois, USA*
Simulations of Fermions and the Sign Problem
Chair: Sandro Sorella
- 11.00-11.30 **Coffee Break**
- 11.30-12.30 *Bernd Mohr, Forschungszentrum Jülich*
Parallel Programming Models, Tools and Performance Analysis
Chair: Johannes Grotendorst
- 12.30-14.30 **Lunch Break**
- 14.30-16.30 **Visit to Research Centre Jülich**
- 18.00-19.00 **Reception and Dinner at the See-Casino**

Wednesday, 27 February 2002

- 9.00-10.00 *Eberhard K. U. Gross, Freie Universität Berlin*
Static and Time-Dependent Many-Body Effects via Density-Functional Theory
Chair: Dieter Vollhardt
- 10.00-11.00 *Mark E. Tuckerman, New York University, USA*
Ab Initio Molecular Dynamics and Ab Initio Path Integrals
Chair: Reinhold Egger
- 11.00-11.30 **Coffee Break**
- 11.30-12.30 *Christian Lubich, Universität Tübingen*
**Integrators for Quantum Dynamics:
A Numerical Analyst's Brief Review**
Chair: Paul Gibbon
- 12.30-14.30 **Lunch Break**
- 14.30-15.30 **Poster Session A**
- 15.30-16.30 *Uwe Manthe, Technische Universität München*
Quantum Molecular Dynamics with Wave Packets
Chair: Nikos L. Doltsinis
- 16.30-17.00 **Coffee Break**
- 17.00-18.00 *Bernard Bernu, Université Pierre et Marie Curie, France*
**Exchange Frequencies in 2D Solids: Example of Helium 3
Adsorbed on Graphite and the Wigner Crystal**
Chair: David M. Ceperley

Thursday, 28 February 2002

- 9.00-10.00 *Fakher F. Assaad, Universität Stuttgart*
Quantum Monte Carlo Methods on Lattices
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- 10.00-11.00 *Fakher F. Assaad, Universität Stuttgart*
Determinantal Methods on Lattices
Chair: Wolfhard Janke
- 11.00-11.30 **Coffee Break**
- 11.30-12.30 *H. Martin Bücker, RWTH Aachen*
Iteratively Solving Large Sparse Linear Systems on Parallel Computers
Chair: Johannes Grotendorst
- 12.30-14.30 **Lunch Break**
- 14.30-15.30 **Poster Session B**
- 15.30-16.30 *Sandro Sorella, SISSA, Italy*
Effective Hamiltonian Approach for Strongly Correlated Lattice Models
Chair: David M. Ceperley
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- 17.00-18.00 *Dieter Vollhardt, Universität Augsburg*
The LDA+DMFT Approach to Materials with Strong Electronic Correlations
Chair: Eberhard K. U. Gross

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- 9.00-10.00 *Wolfhard Janke, Universität Leipzig*
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Chair: Saverio Moroni
- 10.00-11.00 *Nikos L. Doltsinis, Ruhr-Universität Bochum*
Nonadiabatic Dynamics: Mean-Field and Surface Hopping
Chair: Uwe Manthe
- 11.00-11.30 **Coffee Break**
- 11.30-12.30 *Paul Gibbon, Forschungszentrum Jülich*
Long-Range Interactions in Many-Particle Simulation
Chair: Godehard Sutmann
- 12.30-14.30 **Lunch Break**

- 14.30-15.30 *Saverio Moroni, INFN, Italy*
Reptation Quantum Monte Carlo
Chair: Wolfhard Janke
- 15.30-16.30 *Rafael Ramírez, CSIC, Spain*
Dynamic Properties via Fixed Centroid Path Integrals
Chair: Mark E. Tuckerman
- 16.30-17.00 **Coffee Break**
- 17.00-18.00 *Reinhold Egger, Heinrich-Heine-Universität Düsseldorf*
**Relieving the Fermionic and the Dynamical Sign Problem:
Multilevel Blocking Monte Carlo Simulations**
Chair: Alejandro Muramatsu
- 19.00-22.00 **Organ Recital and Farewell Dinner**

Saturday, 2 March 2002

- 9.00 **Departure**
Bustransfer to Aachen Railway Station

NIC Winter School 25.02. – 01.03.2002

	Monday	Tuesday	Wednesday	Thursday	Friday
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10:00-11:00	KEYNOTE Monte Carlo Methods: Overview and Basics M. Lewerenz	MC FOCUS Simulation of Fermions and the Sign Problem D. M. Ceperley	MD FOCUS Ab Initio Molecular Dynamics and Ab Initio Path Integrals M. E. Tuckerman	MC FOCUS Determinantal Methods on Lattices F. F. Assaad	MD FOCUS Nonadiabatic Dynamics: Mean-Field and Surface Hopping N. L. Doltsinis
11:00-11:30	Coffee Break				
11:30-12:30	NUMERICS Pseudo Random Numbers: Generation and Quality Checks W. Janke	NUMERICS Parallel Programming Models, Tools and Performance Analysis B. Mohr	NUMERICS Integrators for Quantum Dynamics: A Numerical Analyst's Brief Review Ch. Lubich	NUMERICS Iteratively Solving Large Sparse Linear Systems on Parallel Computers M. Bucker	NUMERICS Long-Range Interactions in Many-Particle Simulation P. Gibbon
12:30-14:30	Lunch break				
14:30-15:30	MC OVERVIEW Diffusion and Green's Function Quantum Monte Carlo Methods J. B. Anderson	EXCURSION Visit to Research Centre Jülich	POSTER PRESENTATIONS (A1 - A40)	POSTER PRESENTATIONS (B1 - B41)	MC FOCUS Reptation Quantum Monte Carlo S. Moroni
15:30-16:30	KEYNOTE Classic Molecular Dynamics G. Sutmann	EXCURSION Visit to Research Centre Jülich	MD FOCUS Quantum Molecular Dynamics with Wave Packets U. Manthe	MC FOCUS Effective Hamiltonian Approach for Strongly Correlated Lattice Models S. Sorella	MD FOCUS Dynamic Properties via Fixed Centroid Path Integrals R. Ramírez
16:30-17:00	Coffee Break				
17:00-18:00	MD OVERVIEW Path Integration via Molecular Dynamics M. E. Tuckerman	EXCURSION Visit to Research Centre Jülich	MC FOCUS Exchange Frequencies in 2D Solids: Example of Helium 3 Absorbed on Graphite and the Wigner Crystal B. Bernu	MC FOCUS The LDA+DMFT Approach to Materials with Strong Electronic Correlations D. Vollhardt	MD FOCUS Relieving the Fermionic and the Dynamical Sign Problem: Multilevel Blocking Monte Carlo Simulations R. Egger
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Supporting Car-Parrinello Molecular Dynamics with UNICORE

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UNICORE (UNiform Interface to COmputer REsources)¹ creates a science and engineering Grid² combining resources of supercomputer centers and making them available through the Internet³. The UNICORE user benefits from the seamless access through the graphical *UNICORE Client* to the distributed resources to solve large problems in computational science without having to learn about the differences between execution platforms and environments⁴.

The widely used Car-Parrinello Molecular Dynamics (CPMD) application⁵, an *ab initio* electronic structure and molecular dynamics program⁶, was selected as a first example to demonstrate the capabilities of UNICORE to scientists. Through the graphical interface, developed at Research Centre Jülich, the user can prepare a CPMD job and run it on a variety of systems at different locations. Furthermore the interface provides the users with a intuitive way to specify the full set of configuration parameters (specification of the input and output files, the library for pseudopotentials, etc.) for a CPMD simulation⁷. It allows to run the CPMD simulations, which comprise many steps in a pipeline⁸. In addition, the *CPMD Wizard* makes it easy to configure the full set of the control parameters, cell properties, pseudopotentials and atom positions for the CPMD simulation.

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Quantum Dynamics in Wigner Representation

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The operator time correlation functions $C_{FA}(t) = \langle F(0)A(t) \rangle$ for different dynamic properties are among the most interesting quantities in studying dynamics of quantum particles. On the contrary to the calculation of the time correlation functions for classical systems the problem of numerical consideration of the time correlation function for systems of quantum particles is much more difficult. The approach within Wigner formulation of quantum mechanics we pursue in this work is to overcome these difficulties by combining both molecular dynamics and Monte Carlo methods. The Fourier transform of the product of matrix elements of the dynamic propagators obeys an integral Wigner-Liouville-type equation. Initial conditions for this equation are given by the Fourier transform of the Wiener path integral representation of the matrix elements of the propagators at the chosen initial times. This approach computes numerical traces and spectra of the relevant dynamical quantities¹⁻⁴. Anderson localization at temperature $T = 0$ relies on quantum coherence of electron trajectories and results from interference. The key parameter in the physics of electron interference phenomena is the dephasing time of electrons. At finite temperatures the electron coherence is destroyed by both the electron-electron and the phonon-electron scattering. To study the influence of these effects on kinetic electron properties in a random environment we have simulated the quantum dynamics in a one-dimensional canonical ensemble at finite temperature for both interacting and noninteracting electrons. The main quantities calculated in this work are the temporal momentum-momentum correlation functions, their frequency-domain Fourier transforms and the time dependence of the spatial dispersions. We discovered that the results strongly depend on the electron-electron interaction, clearly demonstrating the delocalizing influence of the many-particle interaction even at finite temperatures. The authors thank the NIC Jülich for computer time.

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Photodissociation of OCIO in Bulk Liquids

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The electronic spectra and the photodissociation dynamics of OCIO on the excited state in bulk water, acetonitrile and ethanol are computed using classical molecular dynamics computer simulations. The trajectories are run on an ab-initio potential energy surface of K. A. Peterson¹, which is fit to a global 3-D analytical surface. The calculated cage escape probability in these liquids seems to correlate with the vibrational relaxation rate of the parent molecule and is in reasonable agreement with experiments in water and acetonitrile, but somewhat overestimates the experimental probability in the case of ethanol.

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^{14}N -NMR Shielding of Cytosine Tautomers: An Ab Initio Study

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Solvent induced effect on nitrogen NMR shielding of two dominant tautomers is calculated using density functional theory combined with polarizable continuum model and using the continuous set gauge transformation. Relative solvent effects are calculated using the corresponding nuclear shielding in cyclohexane as reference. Direct ($\Delta\sigma_{dir}$) and indirect ($\Delta\sigma_{ind}$) solvent effects are obtained with a slight modification of the method used by Cammi et al¹⁻³. Instead of deriving $\Delta\sigma_{ind}$ from the difference of the PCM-optimized shielding and the PCM shielding of the molecule held at the geometry optimized in vacuo⁴⁻⁶, it is obtained from the shielding calculated in vacuo for a molecule that is geometry optimized in the solution. Thus $\Delta\sigma_{dir} = \sigma_{sol}(R_v) - \sigma_{cyc}(R_v)$ and $\Delta\sigma_{ind} = \sigma_{vac}(R_s) - \sigma_{vac}(R_{cyc})$. Where $\sigma_{sol}(R_v)$ is the value of the nuclear shielding computed in solution but with the solute in the geometry optimized in vacuo, and $\sigma_{vac}(R_s)$ is the value of nuclear shielding in vacuo but with the solute geometry optimized in solution. $\sigma_{cyc}(R_v)$ and $\sigma_{vac}(R_{cyc})$ are the corresponding parameters for the calculation with cyclohexane^{7,8}.

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DFT-Calculations on the Electrophilic Reaction with Water of the Guanine and Adenine Radical Cations: A Model for the Situation in DNA

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Using density functional theory (DFT), the H_2O (modeled by $\text{OH}^\cdot$) addition on the C8-site of the guanine and adenine radical cations ($\text{Gua}^{\bullet+}/\text{Ade}^{\bullet+}$) is calculated to be *exothermic* by -75.3 and -77.7 kcal/mol, respectively.¹ In contrast, in the absence of the N1 proton on $\text{Gua}^{\bullet+}$, i.e., in the case of the neutral radical ($\text{Gua}(-\text{H})^\cdot$) the H_2O addition is $+29.4$ kcal/mol *endothermic*. Similarly, in the case of the *neutral* adenine radical ($\text{Ade}(-\text{H})^\cdot$), the N⁶-deprotonated radical cation, the H_2O addition is *endothermic* by $+43.7$ kcal/mol. Related to these observations is the fact that with the radical cations, $\text{Gua}^{\bullet+}$ and $\text{Ade}^{\bullet+}$, the positive charge density on the C8-carbon is higher than with the deprotonated forms. This means that nucleophilic attack is likely to have a lower activation energy in the case of the former than the latter. The protonated radical, $\text{Gua}^{\bullet+}$, simulates the situation in *double-stranded* (ds) DNA where the transfer of the N1 proton to solvent molecules is inhibited due to its base pairing with cytosine.² In contrast, in *single-stranded* DNA and in RNA, $\text{Gua}^{\bullet+}$ is expected to quickly lose its N1 proton to the water phase. In comparison, with $\text{Ade}^{\bullet+}$ in ds DNA the exocyclic N⁶-atom is in contact with water molecules in the major groove of the DNA double helix³ and thus should be able to rapidly lose a proton to a water molecule even when it is paired with thymine. This concept provides an explanation for the experimental observation of 7,8-dihydro-8-oxoguanine (8-OGua) formation only in ds DNA and negligible formation of 7,8-dihydro-8-oxoadenine (8-OAde) in any other form of DNA.^{4–6}

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Effect of Strain-Induced Electronic Topological Transitions on the Superconducting Properties of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ Thin Films

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We propose a Ginzburg-Landau phenomenological model for the dependence of the critical temperature on microscopic strain in tetragonal high- T_c cuprates¹. Such a model is in agreement with the experimental results for LSCO under epitaxial strain², as well as with the hydrostatic pressure dependence of T_c in most cuprates. In particular, a nonmonotonic dependence of T_c on hydrostatic pressure, as well as on in-plane or apical microstrain, is derived. Moreover, we recover a monotonic dependence of T_c on the hopping ratio t'/t , as derived from LDA calculations for several hole-doped cuprates³. From a microscopic point of view, such results can be understood as due to the proximity to an electronic topological transition (ETT)^{4,5}. In the case of LSCO, we argue that such an ETT can be driven by a strain-induced modification of the band structure, at constant hole content, at variance with a doping-induced ETT, as is usually assumed^{1,2}.

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Femtocurrents in Chemical Reactions

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We study the bond-breaking, bond-forming process in chemical reactions using an ab-initio molecular dynamics method. We use a newly developed method, which is based on the optimization of a suitably defined action in order to obtain the reactive trajectories. The Hellmann-Feynman forces are needed to optimize the action and have been calculated within density-functional theory. We present the concerted [4+2] cycloaddition of cyclopentadiene and ethylene and the nonconcerted [2+2] cycloaddition of two ethylene molecules. During the reactions microscopic electric currents occur which can be quantified in its duration (≥ 10 fs) and intensity. Furthermore the current pattern discriminates between concerted and nonconcerted reaction mechanisms. The time scale of the spikes of current makes them accessible to femtosecond time-resolved spectroscopy.

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Reaction Path Sampling of Acid Catalyzed Amide Hydrolysis

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The mechanism of the nucleophilic attack of a water molecule to the carbon atom of an amide is explored by means of reaction path sampling Car-Parrinello molecular dynamics simulations. The process is investigated in aqueous solution. The solvent was found to play a critical role for this reaction. Our simulations show that the reaction occurs as a two-step process. The addition of the hydroxyl group to the amide carbon atom involves the association of a water molecule transferring one of its protons to the water phase. This was followed by N-protonation as a separate reaction step.

Fast Multipole Method

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The fast multipole method (FMM)^{1,2} calculates the potentials arise in various scientific applications. The direct method for evaluating these potentials requires $\mathcal{O}(\mathcal{N}^2)$ work regarding the number of particles $\mathcal{N}$. The fast multipole method achieves linear scaling and enables the definition of error bounds.

The fast multipole method consists of several steps. At first the particle coordinates are scaled so that all particles are enclosed by a box with coordinate ranges $[0, 1] \times [0, 1] \times [0, 1]$. The parent box is divided in half along each Cartesian axis to yield a set of 8 smaller child boxes. The child boxes are subdivided again creating children of the children. The particles are sorted by box numbers using the radix sort³ algorithm which scales linearly. The charges contained within each lowest level box are expanded in multipoles about the center of the box. The multipole expansions are translated to the center of the parent boxes. On each level the multipole expansions are transformed into local Taylor expansions. The parent's Taylor expansions are translated to the centers of the parent's children until the lowest level is reached. Each lowest level box contains a Taylor expansion representing the effects of all particles located in the far field region. The final step evaluates the interaction of all particles with the particles in the same box and in the boxes of the near field region. Usually the fast multipole method shows $\mathcal{O}(\mathcal{L}^4)$ scaling with regard to the length of the multipole expansions $\mathcal{L}$. We have reduced the $\mathcal{O}(\mathcal{L}^4)$ scaling to $\mathcal{O}(\mathcal{L}^3)$ using the rotation based FMM⁴. For systems with more than a million of particles the fast multipole method is not only substantially faster but also more accurate compared to the direct method. Calculations up to several billions of particles seem possible.

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Fundamental Investigation of the Influence of Defects on the h-BN to c-BN Transformation

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Cubic Boron Nitride is synthesized from hexagonal graphitic phase. The physical nature of the synthesis can be affected by chemical or intrinsic defects. This defect dependence is investigated using an ab-initio local density approach. The energy difference between the c-BN and h-BN related defect structures depends upon defect induced bonding changes including lattice deformation. This energy difference is related to the defect formation energies. Suggestions are made regarding the importance of some simple chemical defects in inducing the h-BN to c-BN transformation.

Phase Transitions of Classical and Quantum Hard Disks in External Periodic Potentials: A Path Integral MC Study

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Hard and soft disks in external periodic potentials show rich phase diagrams including freezing and melting transitions when the density of the system is varied. We report on extensive Monte Carlo simulations for detailed finite size scaling analyses of various thermodynamic quantities like the order parameter, its cumulants etc. in order to map the phase diagram of the system for various values of the density and the amplitude of the external potential. For hard disks we find clear indication of a reentrant liquid phase over a significant region of the parameter space. Our simulations therefore show that the system of hard disks behaves in a fashion similar to charge-stabilized colloids which are known to undergo an initial freezing, followed by a re-melting transition as the amplitude of the imposed, modulating field produced by crossed laser beams is steadily increased. Detailed analysis of our data shows several features consistent with a recent dislocation unbinding theory of laser-induced melting. The differences and similarities of systems with soft potentials (DLVO, $1/r^{12}$) are discussed.

In order to quantify the quantum effect of the finite particle masses on the phase diagram, we investigated the system by PIMC. We find a increased reentrance region which can be even larger than the freezing region. A possible explanation of this effect, based on quantum delocalisation in conjunction with the external potential, is presented.

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A Six-Dimensional Variational Calculation of the Bending Energy Levels of HF Trimer and DF Trimer

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The bending energy levels of HF trimer and DF trimer were calculated variationally using a six-dimensional kinetic energy operator^{1,2} derived by constraining the stretch degrees of freedom to their equilibrium values. A basis of direct products of spherical harmonics is used to represent the wavefunction. As many as 17 million basis functions are used to converge the low-lying levels. The symmetry adapted Lanczos method is used to calculate levels of all symmetries from one sequence of matrix-vector products. Two efficient ways of implementing the symmetry adapted Lanczos method^{3,4} are presented. To determine the concerted hydrogen bond breaking tunneling splitting in HF trimer and DF trimer one needs to use the D_{3h} point group. By doing D_{3h} calculations we find that the splitting is negligible for the low-lying levels. If the tunneling is unfeasible the appropriate group is C_{3h} . Our calculation using the most accurate SO-3+HF3BG potential developed by Quack, Stohner, and Suhm⁵ indicates that two gas phase overtone bending bands of HF trimer⁶ should be reassigned, and that the noble gas matrix spectra of the bending fundamental bands are significantly shifted from their gas phase counterparts.

A recent new development is to use Lebedev grid to do potential quadrature in the spherical harmonic basis. Compared with the direct product Gauss-Legendre and Gauss-Chebyshev quadratures, use of Lebedev grid reduces the size of grids by a factor of 3 and therefore the memory cost is considerably reduced.

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Dynamics in Ethene: A Study of Non-Adiabatic Couplings

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The photodynamics of polyenes reflects that of many biologically relevant systems involved for example in the vision mechanisms. Despite repeated investigation of the spectra obtained for different polyenes during the last 60 years, many features remain unassigned¹. The reasons of these failures are linked to the properties of the excited states. The presence of valence states and of low-lying Rydberg states as well as the large geometry differences between ground and excited states complicate the analysis of the spectra.

The study of ethene, as the fundamental unit of polyenes, is the logical starting point for the understanding, the simulation, and the modeling of more complex polyenes. The absorption spectrum of the $\pi\pi^*$ excited state is broad and diffuse², and an ultrafast internal conversion to the ground state dominates the photophysical dynamics of this system.

Our approach consists in an accurate description of the potential energy surfaces of the 3 lowest valence states (π^2 , $\pi\pi^*$ and π^{*2}) as a function of selected internal coordinates. A parametrized model is then used to construct a diabatic representation of the electronic Hamiltonian. For the resolution of the nuclear Schrödinger equation on these 3 coupled surfaces, we use the *Multi Configurational Time Dependent Hartree* (MCTDH) method³ which allows us to treat efficiently the time evolution of a wave packet with up to 5 vibrational degrees of freedom.

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Molecular Dynamics Simulation of ZSM-5

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We present our preliminary results from a simulation of zeolite ZSM-5. The initial crystallographic positions were taken from the X-ray diffraction data of Olson et al.¹. We studied systems with 3456 particles, which were produced by equilibration of the zeolite framework via constant-energy molecular dynamics simulation with periodic boundary conditions and using velocity-Verlet algorithm². The atomic interaction potentials used in our simulation were fitted by Vashishta et al.³ and Kramer et al.⁴. Results for structural (i.e. partial and total pair-correlation functions, bond angle distribution), dynamic (i.e. density of states) and relaxational (i.e. participation ratios) properties are presented.

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Visualization of MD and MC Simulations for Atomistic Modeling

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Visualization plays an important role in atomistic modeling of all types. The MC and MD calculations made in the Computational Physics Group at the Technion include modeling of diamond/graphite systems and aluminium/alumina interfaces, as well as studying defects in copper and vanadium.

Examples of AViz' applications such as modeling the creation of interstitial defects in diamond, and the behavior of a 50,000 atom drop of aluminium on an alumina surface will be presented. Some tricks for 3d visualization, such as highlighting atoms with changed coordination numbers and bonds as well as their implementation in AViz will be described. Visualization of spin systems is also possible with AViz, as is the preparation of animations and movies. AViz is Open Source (GNU license) and is downloadable from our website in .rpm for LINUX. We have begun development of visualizations for shapes other than spheres and for polymer chains and are interested in developing a smoke visualization scheme for electronic clouds.

Dynamics of Water-Water and Anion-Water Hydrogen Bonds in Aqueous Electrolyte Solutions

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We study the dynamics of hydrogen bonds between two water molecules and also between a water molecule and a negative ion in aqueous electrolyte solutions by means of molecular dynamics (MD) simulations. We have performed a series of sufficiently long MD simulations of aqueous NaCl and NaBr solutions at different concentrations, ranging from 0.0M to 3.3M at room temperature. The SPC/E model is used for water¹, the ions are modelled as charged Lennard-Jones particles² and the hydrogen bonds are defined by using a set of geometrical criteria^{3,4}. The dynamics of anion-water hydrogen bonds is found to be slower than that of water-water hydrogen bonds. The extent of slowing down of the anion-water hydrogen bonds increases with decrease of ion size and also with increase of salt concentration of the aqueous solutions. We also calculate the diffusion coefficient and the orientational relaxation times of water molecules in the hydration shells of the anions and in the bulk. The results of hydrogen bond dynamics are rationalized in terms of a slower diffusional and orientational motion of water molecules in the hydration shells of the halide ions as compared to those of bulk water molecules⁵. The simulation results are compared with those of recent experiments⁶.

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MD-Simulations of the Dynamic Properties of Non-Ideal Plasmas

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To investigate the dynamic properties of strongly coupled Coulomb systems a proper treatment of correlations is essential. Based on the numerical integration of the equations of motion, molecular dynamics (MD) simulations provide a fully microscopic and dynamic description of a classical many-body system containing all correlations.

To approximate an extended System the actual simulation box is periodically continued in all directions where the long range part of the Coulomb interaction is treated by a Ewald summation. To guarantee a stable equilibrium state in a classical attractive two-component plasma we use a effective potential, which modifies the Coulomb potential at small distances, e.g.:

$$V_{\alpha\beta}(r) = \frac{Z_{\alpha}Z_{\beta}e^2}{4\pi\epsilon_0} \frac{1}{r} \left[1 - \exp\left(-\frac{r}{\lambda_{\alpha\beta}}\right) \right] \quad (1)$$

$\lambda_{\alpha\beta}$ are external parameters, the indices α, β denote electrons or ions.

We present and discuss this numerical treatment which we used to study non-ideal one and two component plasmas. Special attention is payed on the dynamic structure factor which contains information about the dynamical properties of the plasma. We compare data obtained the by numerical simulation with several theoretical approaches.

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Diagrammatic Quantum Monte Carlo for Many-Body Problems: Applied to Excitons

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A novel method for the precise numeric solution of the irreducible two-body problem is presented and applied to excitons in solids. The approach is based on the Monte Carlo simulation of the two-body Green function specified by Feynman's diagrammatic expansion. The method does not rely on the specific form of the electron and hole dispersion laws and is valid for any attractive electron-hole potential. Limits of validity of the Wannier (large radius) and Frenkel (small radius) approximations are established, accurate data for the intermediate radius excitons are presented, and evidence for the charge transfer nature of the monopolar exciton in mixed valence materials is given.

Adsorption Study of Hydrogen in Single Walled Carbon Nanotubes Containing Argon via Grand Canonical Monte Carlo Simulations

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The adsorption of hydrogen molecules in the case of Single Walled Carbon Nanotubes (SWNT) containing an Argon atom has been investigated. Grand Canonical Monte Carlo technique was employed to study the variation of the adsorbed Hydrogen molecules in the (9,9) SWNT as a function of various external conditions. The Temperature range was from 77.5K to 293.5K whilst Pressure was between 1.0 and 10 MPa. The properties that we focussed on, were the Gravimetric and Volumetric Densities of Hydrogen in the particular system. The Molecular Simulations results obtained are presented and discussed.

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Molecular Dynamics Simulation Studies of Supercritical Carbon Dioxide Using Available Potential Models: Investigation of the Bulk Thermodynamical, Transport and Dynamical Properties

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The properties of the pure supercritical carbon dioxide were investigated by means of canonical (NVT) molecular dynamics simulation techniques. Previously developed potential models^{1,2} for the system were employed to simulate the fluid at different state points and their properties were obtained and discussed. The results obtained were compared with available experimental data. The simulations have shown that the models used, to a higher or lower degree, provide realistic descriptions of the supercritical fluid in a wide range of thermodynamic state points.

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Molecular Dynamics Simulations of the Temperature- and Pressure-Induced Denaturation of Staphylococcal Nuclease

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The temperature- and pressure-induced unfolding of staphylococcal nuclease (Snase) was studied by a variety of experimental techniques.^{1,2} In particular, the pressure-induced unfolding was investigated using synchrotron small-angle X-ray scattering (SAXS) and Fourier-transform infrared (FT-IR) spectroscopy, which monitor changes in the tertiary and secondary structural properties of the protein upon pressurization.³ For detailed understanding of the structural, dynamic and thermodynamic properties of the unfolded states of the protein we have carried out a series of nanosecond constant pressure and temperature simulations of Snase. Molecular dynamics simulations were performed using the AMBER force field.⁴ We investigated the changes in secondary structure of Snase upon application of pressure (1 bar to 5 kbar) and temperature (300 K to 350 K). Structural and dynamic differences in the pressure and temperature denatured states are discussed.

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Auxiliary Field Quantum Monte Carlo Study of the Degenerate Fermi-Gas

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The properties of the interacting degenerate Fermi-gas are studied by the Auxiliary Field Quantum Monte Carlo method¹ in the grand-canonical ensemble with fixed particle number. We used a schematic quadratic interaction between the fermions in order to construct a separable Hamiltonian and thereby avoiding the sign-problem². Thermodynamical properties⁴ were analogous to results obtained by contact interactions in the low interacting regime. However, inspection of the highest eigenvalue of the two-particle density matrix⁵ revealed that Fermi condensation³ is impossible, due to the effective repulsive mean field contribution of the quadratic interaction, which becomes even more dominating when the interaction is made stronger. Hereby we confirm the results obtained by Brosens et al.⁷ who performed an analytic calculation on a path-integral formalism.

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Using NMR-Data in a Reverse Monte Carlo Approach for Modelling Solids

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We present an extension of the Reverse Monte Carlo approach for modelling amorphous solids which allows the use of NMR-data as an additional part of the cost function. This ansatz should improve the reliability of the resulting structural model. Reverse Monte Carlo approaches suffer from ambiguities that arise because all diffraction data are 1D-projections of the 3D-structure. Therefore, it is advisable to utilise as many different experimental data as possible. Recent experience has shown that the isotropic ^{15}N -NMR-shifts for B/N-systems may be parameterised as a function of the first two coordination spheres around an N¹. These correlation functions have been implemented in an RMC routine. To test the feasibility of our approach, we have applied it to clusters which had been generated by cutting out atoms from both hexagonal and cubic BN.

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Correlated Electrons in Quantum Dots

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We present results for the ground-state of interacting electrons in 2D parabolic quantum dots. We focus on the strongly correlated regime, when the electrons begin to crystallize and form a so-called Wigner molecule. For this purpose we have performed unrestricted Hartree-Fock (UHF) and path integral Monte-Carlo (PIMC) simulations.

UHF calculations¹ give qualitatively good results for the ground-state energies, even for strong interaction, especially for the spin polarized states. Considerable caution must be taken when interpreting other quantities obtained from the UHF Slater determinant: The UHF symmetry breaking as shown by the charge densities does not directly reflect the Wigner crystallization. Therefore, we have compared the UHF results with exact PIMC data².

Furthermore, we have performed PIMC calculations with an attractive impurity, which deforms the parabolic potential of the dot. The new Multilevel Blocking (MLB) algorithm³ allows to circumvent the fermion sign problem to a certain degree and to study higher electron numbers or lower temperatures. We show the effect of the single impurity on the ground-state energies and spin and charge densities. We discuss the phenomenon of 'bunching' (vanishing addition energies for some electron number in the dot) which has been observed in the experiment and the formation of a localized spin.

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Monte Carlo Modelling of First Order Quantum Effects in Deep Submicron HEMTs

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Pseudomorphic high electron mobility transistors (PHEMTs) are scaled into deep decanano dimensions in order to retain an improvement in the device DC and RF performance¹. A reduction of the gate-to-channel separation and rapid variations of the electric field in the scaled devices enhance the impact of the quantum mechanical (QM) confinement effects on the device performance³. QM effects may be introduced at low cost in Monte Carlo (MC) device simulations using the effective potential (EP) approach². The EP smooths the classical potential and accounts for quantum effects like displacement of the mobile charge centroid away from the heterojunction interface between the channel and the supply layer.

A classical potential P_{class} , obtained by solving the Poisson equation, is smoothed by Gaussian distribution $G(\mathbf{x})$ to get an EP, P_{eff} , as²

$$P_{\text{eff}} = \int d\mathbf{r}' P_{\text{class}}(\mathbf{r} + \mathbf{r}') G(\mathbf{r}'), \quad G(\mathbf{x}) = \frac{1}{a\sqrt{2\pi}} \exp\left(-\frac{x^2}{2a^2}\right), \quad (1)$$

where a is the standard deviation that is approximated by $a = \hbar/\sqrt{8mk_B T}$ ⁴. However, the standard deviation of the Gaussian (1) is adjusted to match a difference between the classical Poisson and self-consistent Poisson-Schrödinger solutions considering a 1D model of the PHEMT structure. P_{eff} is then used in the MC propagation routine at each time step required by self-consistent approach. A smoothing of P_{class} by the Gaussian (1) is performed only for the electron transport in the Γ valley. We assume that the electron transport in L and X valleys is not affected by a carrier confinement at the AlGaAs/InGaAs heterointerface because these valley are energetically much higher than the Γ valley.

The quantum confinement has a detrimental effect on both single and double⁵ doped scaled PHEMTs resulting in a decrease in the drive current and transconductance. As the devices are scaled from 120 nm gate length to 90, 70, 50 and 30 nm gate lengths the impact of the QM confinement increases.

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Is Locally Enhanced Sampling Variant of Molecular Dynamics Suitable for Parallel Computing?

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In the locally enhanced sampling (LES) variant of the molecular dynamics method selected part of a large biomolecular system is multiplied. Simultaneous evolution of many non-interacting copies of the subsystem allows for a faster screening of the conformational space¹. In principle a parallel computer is ideally suited for solving LES equations. The LES method has been already implemented in codes MOIL² and AMBER³. Parallel version of stochastic path calculations was also developed⁴. In the poster possibilities of using a Linux cluster for LES MD calculations will be discussed. An example of performance of the LES method used in calculation of photooxidation of gammaB-crystallines will be presented. These proteins are involved in the eye lens cataract formation.

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Helium Molecules in Two- and Quasi-Two Dimensions

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The ground state properties of helium molecules in infinite and restricted 2- and quasi 2-dimensional (2D) space are studied. Using Monte Carlo calculation in two successive stages, VMC (simple variational Monte Carlo) and DMC (diffusion Monte Carlo) we find that all studied molecules (except $^3\text{He}_3$ with spin-3/2) are bound in 2D. It is discovered that in infinite 2D space ^3He trimer prefers the structure of one dimer and one separate particle while a composition of two separate dimers is preferable for ^3He tetramer. In holding potentials with well adjusted parameters there is a drastic increase of binding for trimer and tetramer in both sorts of helium atoms.

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Molecular Dynamics Simulations of Self-Assembly in Monolayers

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We have done molecular dynamics simulations on series of the semifluorinated monolayer systems, which are of similar structural motif with respect to smectic liquid crystal molecules. It is found that the fraction of rigid part is critical for monolayer ordering. We analyzed the conformational disorder of the system which showed the movement of the gauche defects as a function of time. We have also studied another molecular systems of smectic topology, which has conjugated rigid part such as benzene in the middle of the molecule.

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Quantum and Semiclassical Wavepacket Correlation Methods for Nonadiabatic Reactive Scattering

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A time-dependent formalism of quantum mechanical scattering theory for electronically nonadiabatic reactions is developed based on wavepacket correlation function theory. For a simple single surface model, both quantum and semiclassical methods are applied based on wavepacket correlation function theory. Then, for electronically nonadiabatic two surface reactions model, quantum method is applied. Multi-state diabatic wavepackets, which correlate with particular adiabatic states in the asymptotic region, are propagated by the direct exponentiation method for the Hamiltonian matrix in the split operator formalism. This method is applied to two model reactions of the one-dimensional reactive curve-crossing problem.

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Proton Transfer in Aqueous KOH Solution: OH⁻ Diffusion Mechanism

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We present an *ab-initio* molecular dynamics study of the solvation and diffusion of OH⁻ in KOH solutions. We show that these properties depend strongly on the concentration. In the dilute limit (3 M) we recover the results of D. Marx et al.¹. At higher concentration (14 M), the picture is rather different. We validate our results by a favourable comparison with the available spectroscopic data. Our calculation establishes unambiguously that contrary to what has often been assumed, OH⁻ properties cannot simply be derived from those of H⁺ by polarity exchange.

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Extreme—Orbit Approach to Rate Constant Calculations by LSC-IVR

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The LSC-IVR method is based on classical trajectories using sampling initial conditions weighted by a Wigner distribution function, which allows the incorporation of quantum effects. The integration of these trajectories over the initial conditions leads to the flux—position function, which is readily related to the rate constants. In the approach presented here, we extend the method to take into account anharmonic effects on the reaction coordinate. We found out that for any one—dimensional potential it is possible to evaluate the Wigner distribution function by steepest descent, so that every matrix element is dominated by a particular extreme orbit. As an example, we apply the method to the inverse parabola potential.

Product Energy Partitioning in the Dissociation of 1,2-Difluoroethylene by Direct Classical Trajectories

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Product energy distributions for the dissociation of 1,2-difluoroethylene are computed by classical trajectories. The trajectories are integrated by using energies and forces taken directly from AM1 calculations with specific reaction parameters. The trajectories are initiated at the barrier using several sampling models. The computed translational energies and the vibrational populations of the HF product are compared with the available experimental data.

Product Energy Partitioning in the Dissociation of Ethylene by Direct Classical Trajectories

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Classical trajectories are used to compute product energy distributions for the dissociation of ethylene and several deuterated derivatives at 193 and 157 nm. The trajectories are integrated “on the fly” by taken the forces directly from AM1 calculations with specific reaction parameters. Several barrier sampling models are employed to initiate the trajectories. The computed product energy distributions are compared with available experimental data.

A Linearized Semiclassical-Initial Value Representation Study of the Forward and Reverse $\text{Cl} + \text{CH}_4 \rightarrow \text{ClH} + \text{CH}_3$ Reaction and H/D Kinetic Isotope Effects

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The rate constants for the $\text{Cl} + \text{CH}_4 \rightarrow \text{ClH} + \text{CH}_3$ forward and reverse reactions were computed by the linearized semiclassical-initial value representation (LSC-IVR) method. Also the H/D kinetic isotope effects for the forward reaction were calculated. Both the thermal rates and the kinetic isotope effects were found to be in good agreement with experiment and with those obtained by other methods based on the transition state theory.

Glycine Dimer: Empirical and High-Level Non-Empirical Ab Initio Calculations and Experimental Study

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Amino acids and proteins are among the most important biological systems. The simplest amino acid, glycine, is known to exist in the solution and crystalline phase as the $\text{NH}_3^+ - \text{CH}_2 - \text{COO}^-$ zwitterion, while in the gas phase it appears in the non-ionized form $\text{NH}_2 - \text{CH}_2 - \text{COOH}$.^{1–3} The specific behavior of glycine (as well as of any other amino acids) is determined by its great flexibility, which is due to the free rotation around the three axes defined by the N-C, C-C and C-O bonds.⁴ The situation is even more complicated in the case of the glycine dimer because the global minimum can be realized not only via interaction of monomers in their lowest energy structure but also through interaction of monomers in any conformeric structure with higher energy. The energy increase can be compensated by a gain in more favourable interaction energy. Evidently, very efficient searching procedure needs to be adopted to scan the whole PES, and it is impossible to rely on chemical experience or intuition only. These searching techniques are usually based on computer simulations and it is mainly the molecular dynamics which is being used. Localization of all stationary points at the PES is beyond the capabilities of *ab initio* techniques and must be done using empirical potential methods. *Ab initio* calculations are then used to verify the quality of empirical force field.

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Conductance of the Single-Electron Transistor: A Comparison of Experimental Data with Monte Carlo Calculations

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We report on experimental results for the conductance of metallic single-electron transistors¹ as a function of temperature, gate-voltage and dimensionless conductance. In contrast to previous experiments^{2,3} our transistor layout allows for a direct measurement of the parallel conductance. Thus we can make a comparison between our data and path integral Monte Carlo calculations^{4,5} without any adjustable parameter. The theoretical results for the conductance were obtained by a combination of imaginary-time path integral Monte Carlo and the maximum entropy analytical continuation method^{6,7}.

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Real-Time Monte Carlo Simulations for Electron Transfer Processes

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Dissipative electron transfer (ET) processes play an important role in a wide range of physical, chemical and biological systems and can be readily investigated within the framework of the celebrated spin-boson (SB) model¹. In large areas of the respective parameter space the donor population $P(t)$ exhibits an exponential decay whose decay rate can be directly obtained from the antisymmetrized spin-spin correlation function $\text{Im}\{C(0, t)\} = (2i)^{-1}(\langle\sigma_z(0)\sigma_z(t)\rangle - \langle\sigma_z(t)\sigma_z(0)\rangle)$. Unfortunately, a straightforward quantum Monte Carlo (MC) simulation of these dynamical quantities within the SB model is severely handicapped by the notorious “dynamical sign problem”², i.e. an exponentially decaying signal-to-noise ratio rendering conventional real-time simulations numerically unstable. However, the recently proposed “Multilevel Blocking” (MLB) algorithm for path-integral MC simulations³ is capable of solving this problem without a loss of accuracy. Furthermore, unlike other approaches which can only be applied in certain limiting cases, e.g. weak damping⁴, the MLB algorithm allows for studying the dynamical properties of ET processes within the whole parameter space⁵. Within this framework we present numerically exact results for $P(t)$ and $C(0, t)$. Employing a correction procedure accounting for the different initial conditions (i.e. factorized and fully correlated, respectively) allows for obtaining both $P(t)$ and $C(0, t)$ from the same MC trajectories while a suitable change in the MLB algorithm enables the calculation of both local and non-local real-time functions (e.g. $P(t)$ and $C(0, t)$, respectively). Thus investigating the whole parameter space we find a new low-temperature regime, a phenomenological extension of the classical Marcus prediction⁶ and a crossover from incoherent to coherent dynamics.

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Classical Electrons in Laterally Coupled Diatomic Two-Dimensional Artificial Molecule

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Quantum dots (or artificial atoms) are nanoscale semiconductor structures where a small number of electrons are confined into a small spatial region.¹ The electron motion is usually further restricted to two dimensions. There is strong theoretical evidence for the existence of a limit where the electron system crystallises to Wigner molecules, which is seen as the localisation of the electron density around positions that minimise the Coulomb repulsion¹. In the limit of weak confinement (low density) or a very strong magnetic field the quantum effects are quenched or obscured and the classical electron correlations start to dominate the properties of the system. The ultimate limit is a purely classical system where only the Coulomb repulsion between the electrons defines the ground state. The problem reduces to finding the classical positions of electrons that minimise the total energy of the system.

There is growing interest in calculating and measuring the properties of coupled quantum dots¹. Due to the 2D nature of quantum dots the two-atom system is different whether the quantum dots are coupled in the plane in which the electrons are confined (*laterally* coupled) or in the perpendicular direction (*vertically* coupled). Especially for *laterally* coupled quantum dots only a limited number of studies have appeared. Classical studies serve as a good starting point for more demanding quantum mechanical calculations. Moreover, the study of classical electrons in *vertically* coupled artificial atoms has revealed interesting structural transitions in the ground state electron configurations as a function of the distance between the atoms¹.

In this study¹, structural properties of a finite number ($N = 2 - 20$) of point charges (classical electrons) confined laterally in a two-dimensional two-minima potential are calculated as a function of the distance (d) between the minima. The particles are confined by identical parabolic potentials and repel each other through a Coulomb potential. Both ground state and metastable electron configurations are discussed. At zero distance previous results of other calculations and experiments are reproduced. Discontinuous transitions from one configuration to another as a function of d are observed for $N = 6, 8, 11, 16, 17, 18, 19$.

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Finite Element Techniques Applied to Wave Packet Dynamics in Rydberg-Atoms and Atoms in Very Strong Magnetic Fields

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We investigated the dynamics of wave packets in Rydberg atoms with static and time dependent external fields in field regimes where the classical counterparts of these systems exhibit chaotic behaviour¹. For the quantum simulations we developed effective computational methods using finite element techniques and combinations of finite elements and the discrete variable method². The same discretization scheme has been applied to compute wavelengths and oscillator strengths via Hartree Fock studies of many-electron atoms in extremely strong magnetic fields ($B > 10^8$ T). These very strong magnetic fields occur in the atmosphere of neutron stars³.

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Path Integral Molecular Dynamics Algorithms for Bosons and Fermions

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We present path integral molecular dynamics (PIMD) methods for identical particle systems. In our methods, the effect of indistinguishability of particles due to Bose-Einstein or Fermi-Dirac statistics is cast in a form of potential function^{1,2}. We refer to it as a permutation pseudopotential. The pseudopotential is formulated using the standard primitive approximation and extended using a more efficient discretization scheme, a pair density matrix approximation³. On the basis of the pseudopotential technique, molecular dynamics algorithms are constructed as in the standard PIMD⁴. Model calculations have been carried out to test our methodology. Furthermore, a method to generate the trajectory of the centroid variables for identical particles⁵ is discussed. A path integral hybrid Monte Carlo (PIHMC) method for bosons⁶ is also presented. Application of the PIHMC to liquid helium is demonstrated.

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Coexistence of S -Wave Superconductivity and Antiferromagnetism

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We consider a half-filled attractive Hubbard model supplemented by pair-hopping processes and a Heisenberg term. This model shows various phase transitions including a charge-density wave (CDW) to superfluid transition (SF) as well as superfluid to antiferromagnetic (SDW). We show the existence of a phase where, due to symmetry, all above mentioned phases (SDW+CDW+SF) coexist. In this model, phase boundaries are characterized by higher symmetry. The coexistence phase has $SO(4)$ symmetry¹ plus an additional particle-hole symmetry. Those results are obtained on the basis of zero temperature auxiliary field QMC simulations using a new efficient algorithm for calculating time-displaced correlation functions².

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Phase Transition in Strongly Degenerate Hydrogen Plasma

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Hydrogen at high pressures remains the subject of many investigations as the many interesting phenomena, such as a metal–insulator transition (MIT), Mott effect and a plasma phase transition (PPT) have been predicted. They occur in situations where both, quantum and Coulomb effects are important, making a theoretical analysis difficult. Among the most promising theoretical approaches to such systems is the path-integral quantum Monte Carlo (PIMC) method^{1,2} which has seen remarkable recent progress, e.g.². However, for Fermi systems, these simulations are substantially hampered by the so-called fermion sign problem. Additional assumptions with uncontrol accuracy³ such as fixed node and restricted path concepts have been introduced to overcome this difficulty².

Recently, we have presented a new path integral representation which avoids additional approximations (direct path integral Monte-Carlo, DPIMC) which has successfully been applied to strongly coupled hydrogen⁴⁻⁷. The method is applied to compute the pressure, energy and pair distribution functions of a hydrogen plasma in the region of *strong coupling and strong degeneracy*. Our numerical results allow to analyze atom and molecule formation and break up and predict, at high density, proton ordering and pairing of electrons. We apply also the DPIMC method to the analysis of dense liquid hydrogen in the region of the hypothetic plasma phase transition. Computing the equation of state and the internal energy, we find clear indications for the existence of the PPT – its first confirmation by a *first-principle* method. It is shown that the PPT manifests itself by the formation of large metallic droplets which are crucial for the plasma transport properties. The authors thank the NIC Jülich for computer time.

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An Efficient Data Compression Method for the Davidson Subspace Diagonalization Scheme: New Possibilities in Computational Science

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The Davidson Subspace Diagonalization Scheme¹⁻³ is an important method to compute the lowest eigenvalues and the corresponding eigenvectors for high dimensional diagonal dominant matrices. It became a standard method in computational physics and chemistry. The idea is to approximate the eigenvectors by linear combinations of expansion vectors. The processing of large amounts of data which arise in high dimensional sparse eigenvalue problems has always been a severe bottleneck for the Davidson method. On the other hand the expansion and product vectors contain information which does not contribute to the requested accuracy of the eigenvalues and eigenvectors.

A highly flexible and very efficient data compression method⁴ for both, expansion and product vectors is presented within the Davidson diagonalization method. The data compression method is based on an error analysis of the eigenvalues in terms of the expansion and product vectors and on compression schemes for representing floating point numbers with a variable length of the mantissa.

Depending on the sparsity of the eigenvalue matrix saving factors between 10 and 100 can be achieved. Because the data is always processed in compressed form the interprocess communication and the access to secondary storage is dramatically reduced.

This new approach allows to solve diagonal dominant eigenvalue problems with a dimension up to 10^{11} on massively parallel architectures.

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The Iron(III) Catalysed Michael Reaction

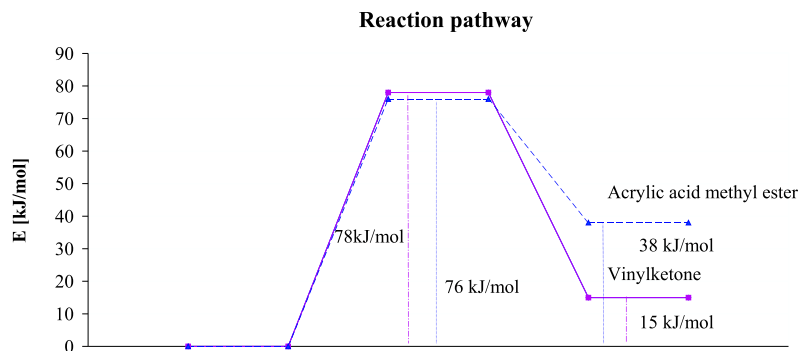
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Recent work by Christoffers et al.¹ shows iron(III) chloride hexahydrate to be a very efficient transition metal catalyst for Michael reactions between 1,3-dicarbonyl compounds and enones. The experiments are simple to do, conversion is fast and clean, solvents are generally unnecessary and chemoselectivity of the iron(III) catalysts is sometimes even better than that of base catalysts. Unfortunately, explanation of the reaction pathways is problematic as characterisation of the hypothetical intermediates is not possible. Furthermore it is not understood why acrylic acid alkyl esters do not react as Michael acceptors under these conditions.



In order to explain these observations a computational investigation was made of possible reaction paths. Full geometry optimisations of the ground and transition states were carried out with the B3LYP functional and the 6-311G* basis set. The reaction between 3-Oxo butyric acid methyl ester and but-3-en-2-one was slightly endothermic (+14 kJ mol⁻¹) with an activation energy of 78 kJ mol⁻¹. The reaction with acrylic acid methyl esters shows a similar profile ($\Delta E = +35$ kJ mol⁻¹, $E_a = +75$ kJ mol⁻¹), which is not a conclusive explanation for the observed lack-of-reactivity.

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DFT Calculations of Low-Frequency Vibrational Modes in Heme Proteins

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Low-frequency (less than 100 cm^{-1}) vibrational modes are important in energy and information exchange in proteins. We calculate via density functional theory (DFT) the normal modes of several Iron-Porphyrin compounds. These investigations are meant to help characterize low-frequency vibrational modes observed experimentally in femtosecond coherence spectroscopy (FCS) investigations of heme containing biomolecules and model compounds. DFT calculations reveal the lowest frequency modes to be correlated with the motions of the imidazole simulating the proximal histidine. These motions are also found to be coupled to motion of the vinyl peripheral groups. Modes that contain motions of the heme plane (saddling, ruffling and doming) were also found. The potential energy surface is found to be very flat, containing many local minima.

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Bead-Fourier Path Integral Molecular Dynamics

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Path integral methods provide a powerful tool for accurate calculation of quantum properties at finite temperatures. There exists two different families of techniques which are distinguished by the method used to represent the paths in computer simulations. “*Bead*” methods represent paths with finite number of discrete coordinate-space points that are equidistant in imaginary time and are usually referred to as *beads*. “*Fourier*” methods represent paths by a truncated Fourier expansion.

Recently a combined *bead-fourier* method was suggested and implemented within the Monte Carlo approach¹. The latter represents the trajectory between “beads” as a truncated Fourier sine expansion and hence contains both approaches as the limit cases. Here we present the bead-fourier molecular dynamics method in which sampling of the configurational space is made according to the constant-temperature molecular dynamics algorithm. In order to test the method we applied it to well known systems for which properties can be obtained analytically: quantum harmonic oscillator and a hydrogen atom. Usual Noze-Hoover thermostating scheme turned out to be inefficient, that is why a central mass thermostating scheme has been considered. The choice of numbers of beads and Fourier harmonics was examined. Since ergodicity problems seem to be crucially important for systems of such a type, different ergodicity tests have been carried out. Different energy estimators and their applicability were tested. Finally, the comparison between Monte Carlo and molecular dynamics was carried out.

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The Mechanism of Flavoenzyme-Catalysed Oxidation Studied by Ab Initio MD Simulations

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D-amino acid oxidase (DAAO) is the prototype for the oxidase class of flavin-containing enzymes. Its structure and kinetics are frequently investigated by various experimental techniques to understand the general action of flavoprotein oxidases¹. Three different mechanisms have been proposed for the catalytic removal of an hydride equivalent from the D-amino acid substrate, and its subsequent conversion to α -ketoacid. They differ mainly in the first step of the reaction, where some studies have supported the formation of a carbanion intermediate while other ones seem to involve a direct hydride transfer from the substrate to the flavin. We carried out several Car-Parrinello² molecular dynamics simulations of a system including the active site and the substrate. The activated transfer of a hydrogen atom from a D-amino acid to the flavin has been forced by varying a suitable reaction coordinate from values corresponding to reactants to the products. This 'Blue Moon Ensemble' sampling³ allowed to determine the free energy along the reaction path, as well as providing information on the mechanism for this first reaction step. Indeed, the variation of several geometrical parameters for both the substrate and the active site has been followed in the course of the reaction by monitoring their dynamical averages for each value of the reaction coordinate. Moreover, the variation of the total electronic density has been followed in the course of the reaction, providing dynamic information on the instantaneous electronic rearrangements brought about by the process, which could hardly be obtained by other methods.

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Ethylene Epoxidation by H_2O_2 in a Ti-Zeolite From First Principles

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Due to their use as oxidation catalysts, titanium isomorphously substituted zeolites are of great interest from both academic and industrial point of view. Indeed these materials are able to combine the activity of Ti in selective oxidation with H_2O_2 with the shape selectivity of microporous materials. A number of experimental and theoretical works have been done in order to understand the Ti active site structure and the oxydation mechanism¹. We are interested in the study of the mechanism of olefine epoxidation by H_2O_2 in titanium offretite (Ti-OFF) with computer simulation. Offretite is an hexagonal zeolite, with space group $P6\bar{m}2$. Its framework contains three different cavities: hexagonal prism, cancrinite cage, and gmelinite cage. The unit cell² has two distinct tetrahedral sites namely T_1 and T_2 that have different acid properties: the T_2 is more acidic than the T_1 ³. We have investigated the structure containing in the elementary cell 17 silicons, 1 titanium (in T_1 site) and 36 oxygens. The Car-Parrinello ab-initio molecular dynamics methods⁴ has been used to investigate the structure of the system Ti-OFF with different guest molecules inside the cages. The valence electronic wavefunctions are expanded in a planewave basis set and the energy cut-off adopted for the wavefunctions was 60.00 Ry; the time step was 5 a.u.. Norm-conserving non local pseudopotentials were used for Si, Ti, O and H; the electron-electron interaction was calculated by a gradient corrected density functional approximation. Different simulations were performed to study the stability of Ti tetra-coordinated with different guest species. One, with two water molecules, one inside the hexagonal prism and one inside the gmelinite cage, was performed at room temperature for a time of 3.7 ps. Another one was carried on with two water molecules, one inside hexagonal prism and one inside the gmelinite cage, and the hydrogen peroxide, inside the gmelinite cage, for 1 ps. The studied Ti-OFF systems are stable at room temperature and the titanium assume the tetrahedral configuration with no significant distortion of the framework. Work is in progress for a system containing both H_2O_2 and ethylene. We have found that both ethylene and one water diffuse while H_2O_2 and one water remain close to Ti site. The reaction path simulation is in progress.

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Finite Temperature Effects in Quantum Dots

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Quantum Monte Carlo methods allow a direct procedure to determine electronic structures even at finite temperatures¹. We test Path Integral Monte Carlo² against one and three dimensional harmonic oscillator and apply it in evaluation of finite temperature effects in quantum dots.

Our simulations yield correct excited state occupations including degeneracy for one and three dimensional harmonic oscillators. Encouraged by this, we have investigated the finite temperature distribution of one and two electrons in a single and coupled quantum dots and compared our results with those from experiments and other theoretical methods³.

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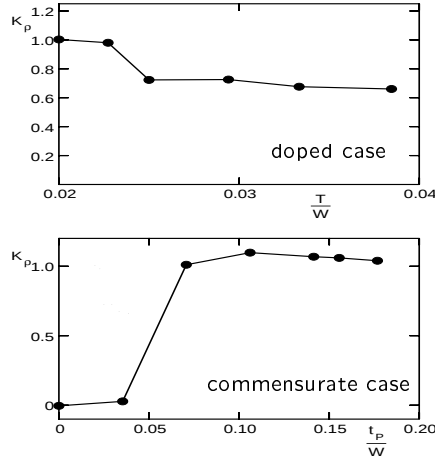
Dynamical Mean Field Approach to Organic Conductors

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Bechgaard salts crystallize in stacks of TMTSF molecules, exhibiting a quasi one-dimensional electronic structure. We model these materials as systems of coupled Hubbard chains and propose a new theoretical method for their numerical treatment: “chain-DMFT” is a generalization of dynamical mean-field theory (DMFT), that treats the 1d chains in a numerically exact manner and the coupling to the neighboring chains in a DMF way, thus leading to a unified description of the one-dimensional and higher dimensional phases of these systems.



In the doped case we investigate the dimensional crossover from a high temperature Luttinger Liquid (LL) phase (with LL parameter $K_\rho \sim 0.7$) to a low temperature Fermi Liquid (FL) ($K_\rho = 1.0$). The figure shows K_ρ as a function of temperature; W is the 1d bandwidth. At half filling, the uncoupled chains are Mott insulators ($K_\rho = 0$); transverse hopping (t_p) of the order of the 1d Mott gap, however, leads to deconfinement of the electrons and FL behavior (see Fig.). We explore the properties of the LL and FL phases, and demonstrate that our chain-DMFT approach cures some artefacts of a simple RPA treatment. Thus, chain-DMFT can be considered to be a tool of choice for treating more general quasi one-dimensional systems.

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The Study of Atomic Shell Approximation Methods for Calculating Molecular Similarity

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In this study the two different methods for analytical evaluation of molecular shape similarity are compared. The main idea of both methods is to use so-called *promolecular approximation* to describe molecular first-order density function, by sum of contributions of atomic electron densities. The first method that was developed by Good et al. proposed the use of linear combination of atom-centered Gaussian functions to approximate electron density. The electron density is simply approximated to the square of STO-3G atomic orbital wavefunctions used in GAUSSIAN *ab initio* quantum mechanical program. The second theoretical model of density fitting, so-called *Atomic Shell Approximation* (ASA), was developed by Ll. Amat et al. The atomic density function is expressed as a linear combination of spherical 1S functions provided with positive definite coefficients. The set of coefficients of the expansion is calculated by minimizing a *quadratic error integral function* between the *ab initio* and ASA density functions.

We found that ASA describe the molecular density shape more accurate than the one proposed by Good. The main inconvenience of both methods is the high concentration of atomic density at atomic nuclei, leading to a poor description of atomic bond formation. We also found that for maximization of similarity index only simple optimisation like simplex method is not sufficient. An elegant algorithm for calculating maximal similarity was proposed by authors of ASA.

Acknowledgments:

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Implementation and Evaluation of MPI-Based Parallel Many-Particle Simulation

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The Message Passing Interface (MPI) based object oriented Particle-Particle Interactions (PPI) library is implemented and evaluated. The library enables a user with little programming knowledge and without any experience in parallel programming to implement a parallel simulation program of any many-particle system. The user provides a C++ class that represents a particle, e.g. an atom, a molecule, a cluster of stars etc. The class should implement the calculation of interaction between pairs of particles and updating particles' attributes, e.g. velocity and position. The resulting parallel program can be run on a wide spectrum of different computer platforms. The library was tested in parallel molecular dynamics simulations of butadiyne and water systems. The basic leapfrog Verlet algorithm was employed as well as the Split Integration Symplectic Method (SISM), based on the analytical calculation of the harmonic part of the particle interactions.

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Bound States in the One-Dimensional Kondo Lattice Model

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We apply recently developed high-order coupling expansion techniques to study zero-temperature properties of 2-hole excitations in the one-dimensional Kondo lattice model around half-filling. Starting from the insulating Kondo spin liquid phase which is the strong-coupling limit of isolated dimers we expand effective Hamiltonians in the subspaces of single-hole and two-hole excitations. This way we can expand the dispersion of a single hole excitation up to 10th order. With growing hopping amplitudes the effective mass of the holes is found to strongly increase. In the two-hole sector we find a continuum of scattering states and two distinct states which separate from the continuum around $k = \pi$ momentum. These 2-hole states can be identified as singlet and triplet combination of two hole excitations. We use series extrapolation techniques to determine the respective binding/antibinding energies.

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Quantum-Classical Liouville Approach to Molecular Dynamics

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In mixed quantum-classical molecular dynamics few but important degrees of freedom of a molecular system are modeled quantum-mechanically while the remaining degrees of freedom are treated within the classical approximation. Such models can be systematically derived as a first order approximation

to the partial Wigner transform of the quantum Liouville-von Neumann equation governing the dynamics of the full system. The resulting adiabatic quantum-classical Liouville equation (QCLE) can be decomposed into three individual propagators by means of a Trotter splitting:

1. Phase oscillations of the coherences resulting from the time evolution of the quantum-mechanical subsystem.
2. Exchange of densities and coherences reflecting non-adiabatic effects in quantum-classical dynamics.
3. Classical Liouvillian transport of densities and coherences along adiabatic potential energy surfaces or arithmetic means thereof.

A novel stochastic implementation of the QCLE is proposed in the present work. In order to substantially improve the traditional algorithm based on surface hopping trajectories, we model the evolution of densities and coherences by a set of surface hopping Gaussian phase-space packets (GPPs) with variable width and with adjustable real or complex amplitudes, respectively.

For the example of a single avoided crossing we demonstrate that convergence towards fully quantum-mechanical dynamics is much faster for surface hopping GPPs than for trajectory-based methods. For dual avoided crossings the Gaussian-based dynamics correctly reproduces the quantum-mechanical result even when trajectory-based methods not accounting for the transport of coherences fail qualitatively.

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Monte Carlo Simulation of the FM Kondo Model Including Coulomb Repulsion

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A new numerical technique for the Kondo lattice Hamiltonian is introduced. The approach combines a Monte Carlo sampling for the classically treated localized spins with a mean field treatment of the electron correlations. The method is applied to one-dimensional chains at low temperatures. Using realistic parameters for manganites, near quarter filling strong tendencies to charge ordering are observed. Spectral functions and correlation functions are calculated.

A Monte Carlo Hybrid Technique for Electron Correlations in Ferromagnetic Kondo Chains

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We present two different methods to tackle the ferromagnetic one-dimensional Kondo problem with additional on-site and nearest neighbor Coulomb repulsion. The electrons interact with localized core spins treated classically. A Markov Chain Monte Carlo scheme for the classical spins is constructed using the partition function of the electronic degrees of freedom as the weight function. Both a mean field approach and a Lanczos routine is used to calculate the fermionic partition function. A comparison of the two methods shows good agreement for realistic parameters.

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Order-N Parallel Tight Binding Molecular Dynamics: Application to the Carbon Nanotubes

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Carbon nanotubes with their remarkable mechanical and electronic properties play a major role in the design of next generations nanoelectronic and nonoelectromechanical devices. Depending on their chirality carbon nanotubes could be conductor, semiconductor as well as insulators. Using Order-N ($O(N)$) parallel tight binding molecular dynamics method we have studied the structural stability and energetics of carbon nanotubes. In classical tight binding calculations standard matrix diagonalization algorithms have a complexity that grows as cube of the system size ($O(N^3)$). In this work we have compared our $O(N)$ algorithm with $O(N^3)$ algorithm on sequential computer. Then parallelization technique is applied to $O(N)$ TBMD program. Our system contains 8 computer having celeron processors with fast ethernet and PVM (Parallel Virtual Machine) library. The results for sequential and parallel runs are compared and assured that they are the same both in numerical accuracy and in physical aspects. Our results show that parallelization technique is beneficial because of the nature of the $O(N)$ algorithm. We will report our results of speed up, efficiency and the system size studies. In present situation, parallelization together with $O(N)$ algorithm proves to be very effective and favorable.

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Treatment of Electron-Phonon and Electron-Electron Interactions Using the Dynamical Cluster Approximation

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I apply the newly developed dynamical-cluster approximation¹(DCA) to models of electron-phonon and electron-electron interaction (Holstein model² and Hubbard model³). DCA is one of a number of techniques which may be used to extend dynamical mean-field theory^{4,5} to include spatial fluctuations (see also e.g.^{6,7}). DMFT can be considered as a course graining of the self-energy across the entire Brillouin zone (BZ). DCA improves on this process by dividing the BZ into a number of course grained areas - thus some momentum-dependence is included. In the limit that the number of course grained areas tends to infinity, the exact momentum dependence is recovered.

An important question relates to the circumstances under which DMFT is a good approximation - i.e. how strongly momentum-dependent is the self-energy? If the self-energy of all course-grained segments is practically identical, then DMFT gives an accurate answer. In this poster I use a range of “iterated perturbation theory” techniques within DCA to assess the limitations of DMFT. In particular the poster will include a critique of Migdal–Eliashberg theory for the Holstein model, and second-order IPT for the Hubbard model.

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Modeling Hydrogen Adsorption onto Carbon Nanotubes from Weak to Strong Limits

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In an attempt to understand hydrogen interaction with carbon nanotubes in the range from physisorbed molecules to chemisorbed atoms, we have used a wide spectrum of molecular dynamics methods, from tight binding, over DFT to diffusion Monte Carlo calculations. Except an explanation of the experimental measurements¹, we also propose new nanotube-like polymers² with promising properties according to our calculations.

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A Reaction Path-Liouville Approach to the Rate Constant for Polyatomic Chemical Reactions

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It has recently been developed a practical algorithm to describe the motion of a molecular system, when is constrained to follow a determined path in the configuration space¹ (i.e. the Intrinsic Reaction Coordinate²). The resulting one-dimensional problem leads to a bi-dimensional phase space where the corresponding Liouville equation for the density of states may be integrated along the reaction path. The integration is performed using a Ritz-Galerkin scheme, as well as expanding the density of states in the phase space as a sum of gaussians. The latter have been conveniently chosen to fit a guessed and slightly perturbed initial Maxwell-Boltzmann distribution corresponding to an initial thermal equilibrium. Once this computation has been done, we can use the new distribution to average the microcanonical flux and, then, compute reaction rates³. The final result is that one may rigorously show that the new reaction path - Liouville procedure establishes a new upper limit for the rate constant value, which is in turn lower than transition state theory predictions⁴. The method has been tested with the bi-dimensional Müller-Brown surface, and is being applied to some interesting reactions.

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Computing Forces in QMC

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Computing forces between nuclei in quantum Monte Carlo opens the door to efficient ab initio molecular dynamics algorithms and to efficient geometry optimizations³. We introduce a simple approach to avoid the infinite variance of the forces, arising both in all-electron calculations and in calculations using pseudo-potentials. For that purpose we use our very recent proposal to compute observables: a generalization of the so-called zero-variance property of the energy^{2,1}. We illustrate on diatomic molecules (Li_2 , C_2 , etc.) that accurate potential energy curves and accurate equilibrium geometries are obtained⁴.

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Combined Ab Initio and Density Functional Approach Based on a Partition of the Orbital Space

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The operator $\hat{H}_0$ generates a set of orbitals. $E(S)$ is the ground-state energy of the system S with hamiltonian $\hat{H}$. Let us increment $\hat{H}$ with a projector $\hat{O}$ acting on the virtual orbitals of $\hat{H}_0$. The system S_O with hamiltonian $\hat{H} + \hat{O}$ has the ground-state energy $E(S_O)$. Calculations for $E(S_O)$ were performed when S is an atom (A), and independently when S is the homogeneous electron gas (G). Mainly two classes of operators $\hat{O}$ were confronted: the first shifts the virtual levels of $\hat{H}_0$ with a constant G_0 whereas the second removes the eigenstates of $\hat{H}_0$ lying in an energy band of upper bound c above the Fermi level.

By decreasing G_0 or c , systematic improvement is obtained in connecting the non-interacting system based on $\hat{H}_0$ to the fully interacting system S . Along that path, we studied the possibility of introducing for the quantity $E(A_O)$ local approximations based on the homogeneous electron gas. On the other hand, the remaining part $E(A) - E(A_O)$ was calculated via ab-initio treatments. The He and Be iso-electronic series were investigated in the present, as for these systems exact $E(A_O)$'s can be estimated with high accuracy. Notably, among the two classes of operators $\hat{O}$ mentioned above, with the latter we achieved approximate $E(A_O)$'s very close to their supposed exact values, and for a relatively large domain of c .

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Modelling Ionic Nucleation in Small Neon Clusters

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Experimental and theoretical evidence¹⁻³ suggests that rare gas cluster ions consist of an arrangement of neutral atoms bound by polarization forces to a positively charged central core. Our previous calculations⁴ on Ne_n^+ clusters, up to Ne_{10}^+ , using an all-electron DFT treatment that employed the full Hamiltonian of each n-atom system, showed that for these small clusters the core is the dimer ion in which the distance is quite constant and very close to the one of the isolated Ne_2^+ molecule.

Guided by these results, in the present work for the study of larger clusters we calculate the potential between a Ne_2^+ molecule at its optimized geometry and an atom of Ne in order to model the full potential of a Ne_n^+ cluster as the sum of the interactions between the dimer and the remaining (n-2) neon atoms and the ones between each two neutral neon atoms. We further model the full potential as the sum of atomic potentials and compare the results between possible growth mechanism within the ionized clusters.

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Structures of OCS Molecules in H₂ Clusters and in (H₂, He) Mixed Clusters

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We have calculated the energy of the system at a MP4 level using a AUG-cc-pVTZ base for the three possible couples of θ_2 and ϕ angles (being θ_2 the angle formed by the axis linking the centers of mass of the two molecules (R) and the H-H bond, and ϕ the dihedral angle). Those couples of value were chosen because they well represent the three possible orientations of the H₂ molecules with respect to OCS.

After having interpolated the energy values so calculated in order to obtain an R and θ_1 homogeneous grid (and consequently, the three potential energy surfaces), we averaged the three possible orientations so obtaining a potential function depending only on R and θ_1 which can in turn be used to describe the interaction of OCS with an averaged H₂ molecule. By further using the interaction potential between He and H₂¹, and between OCS and He² which we know already, the next step will be to start a Diffusion Montecarlo^{3,4} study of the larger clusters of OCS in mixtures of He+H₂.

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Atomic Impurities in Small ^4He Clusters: Structures and Energetics From DMC Calculations

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The $^4\text{He}_3$ and $(^4\text{He})_N X$ (with $X = {}^{23}\text{Na}, {}^7\text{Li}, {}^1\text{H}^-, {}^1\text{H}^-$) systems are investigated using an algorithm for Diffusion Monte Carlo (DMC) with importance sampling based on a short-time expansion of the Green's functions for the motion in three dimensions.

The interaction forces are obtained as sums of two-body (2B) interaction potentials and the most interaction functions are selected.

The $^4\text{He}_3$ and $^4\text{He}_2 X$ radial and angular distributions produced by the DMC calculations are analysed in detail to extract information on the most probable shapes exhibited by this very floppy trimeric cluster.

The $(^4\text{He})_N X$ clusters total energy, with $N \geq 3$, and radial and angular distributions are investigated to extract information on the most probable exhibited by this doped clusters.

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Ab Initio Three-Body Nonadditivities in Simulations of Liquids and Phase Equilibria

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High-accuracy ab initio pair potentials and three-body nonadditivities obtained recently using the symmetry-adapted perturbation theory (SAPT) have been employed in Monte Carlo simulations of water, argon, and carbon dioxide. The three-body effects turn out to be essential for the proper description of the hydrogen-bonded structure of ambient water, while the role of the higher many-body effects is much smaller. For argon, the complete ab initio three-body nonadditive potential (including the short-range terms) has been shown to faithfully reproduce the experimental phase diagram in the vapor-liquid coexistence region and the parameters of the phase transition. Peculiar cancellations between various physical components of the three-body nonadditivity lead to the Axilrod-Teller-Muto potential being an excellent approximation to the total three-body effect. Importance of the nonadditive forces for vapor-liquid equilibria of carbon dioxide will be discussed based on preliminary results of simulations using accurate ab initio pair interaction potentials and a simple model of three-body nonadditivity including dispersion and induction effects.

***NPT* Simulations of Water Using Ab Initio Potentials**

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One of the known distinctive properties of water is its so-called anomalous density behaviour.¹ The density of water is known to have a maximum value at a temperature of about 4°C. At present there is no known potential which reproduces this density peak except for potentials fitted to do so.² In this work, the most accurate *ab initio* two body potential available³ is used in an *NPT* Monte Carlo simulation⁴ with 512 water molecules to investigate this behavior. Preliminary calculations using an *ab initio* three body potential⁵ are also presented together with results obtained from some empirical potential.

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A Combined Exact Diagonalization - *Ab Initio* Study of the Metallicity and Electron Localization in Nanoscopic Systems

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We present our new method of approach to the interacting electron systems combining the *exact diagonalization* of the Hamiltonian in the Fock space (including *all* hopping integrals and *all* two-site interactions) with an *ab initio* optimization of the single particle (Wannier) functions in the resultant ground state. Such a procedure is applied to the analysis of the electron localization in nanoscopic chains containing up to $N = 12$ atoms. Electrons are essentially localized for the interatomic distance $R > 2.2 \text{ \AA}$. The statistical distribution evolves from the Fermi-liquid limit (at $R \sim 1 - 2 \text{ \AA}$) to continuous momentum distribution for $R > 2.2 \text{ \AA}$. The Mott and the Hubbard criteria of localization are explicitly evaluated and the importance of the former is stressed. Further applications of our method are listed at the end.

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Plane-Wave Density-Functional Theoretic Study of Formation of Clay-Polymer Nanocomposite Materials by Self-Catalysed In Situ Intercalative Polymerisation

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In recent years there has been considerable interest in clay-polymer nanocomposites due to the novel material qualities that they exhibit^{1,2}. They offer enhanced mechanical and thermal properties, which have led to a wide range of applications. A new method of fabricating these materials involves treating raw clay with an aqueous solution of two monomers which are thought to spontaneously polymerize between the structural layers.

We present the results of ab initio calculations looking into the possible ways in which the clay sodium montmorillonite may catalyse the reaction of methanal with ethylenediamine. Catalysis is shown to occur at the clay mineral lattice-edge where hydroxyl groups and exposed aluminium ions act as strong Bronsted and Lewis acid sites, respectively³.

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Electronic Behavior of Carbazole and its Derivatives

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Carbazole is known as an important component of crude oil[1]. It has been mentioned that this species participates in the formation of resins when polimerisation takes place or acts as a mediator in reactions that produce asphaltene flocculation [2]. The detailed role of this compound or its derivatives in such a processes is not yet clear. However an important degree of participation in the stabilization of asphaltene sediment may be present from these compounds due to the intrinsic aromaticity of these molecule that can account as a driving factor for attachment with other organic species. We have carried out a study on the aromaticity and electronic structure behavior of carbazole and some of its substituted derivatives. The study was performed taking advantage of the NICS (Nuclear Independent Chemical Shifts) technique. The changes in aromaticity of each compound were evaluated on the basis of inductive and resonant effects. A correlation of aromaticities for the compounds that varies depending on the different substituents (electron-donor or electron-acceptor) with respect to carbazole molecule is considered and a study of the possible reactivity for each case was performed. Furthermore a proposition of a possible mechanism via a free radical reaction to generate an asphaltene-like molecule is explored.

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The Catalytic Cracking of Hydrocarbons: Paraffins in the HY Zeolite

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The catalytic conversion of hydrocarbons represents a strategic point in the refining oil industry¹. We discuss the physical bases and fundamental aspects that give support to the catalytic cracking of paraffins in a cavity of the HY zeolite. We provide new insights on the dynamical process that occurs during the precise moment of the cracking, frequently inaccessible to the experiment, as we did in the case of the HZSM-5 zeolite^{2,3}. With the use of a host model for the HY zeolite and a representative hydrocarbon from the paraffinic family, we quantify the role that the crystal field, Brønsted acidity, temperature and high coverage effects have in the catalysis. The time evolution of structural and electronic variables is elucidated and the use of new variables for particular analysis is highlighted. Our investigation substantiates current views with possible implications in the design of future catalysts.

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Computer Simulation of Proton Rearrangements in 5^{12} and $5^{12}6^2$ Buckyball Water Clusters

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Water clusters have been extensively studied in a large number of experimental and theoretical studies. A special case of the $(\text{H}_2\text{O})_n$ clusters are buckyball water clusters (BWC), where the O atoms have the same topologies as carbon atoms in fullerene clusters. The cavities may be empty, but the stability of these systems increases if the cavities are filled with atoms or small molecules. BWC appear as stable systems in gas phase and as building-units in the clathrate hydrates, in the solvation shell formed around the gas atoms/molecules that penetrate in the liquid water or in the quasi-liquid layers.

The most stable small BWC appear to be $(\text{H}_2\text{O})_{21}^+$ clusters, which consist of a H_3O^+ surrounded by a $(\text{H}_2\text{O})_{20}$ cage, consisting of 20 water molecules arranged in a pentagonal dodecahedral structure (noted 5^{12}). A related structure is the $(\text{H}_2\text{O})_{24}$ cage which has tetrakaidecahedral structure (noted $5^{12}6^2$) with two additional hexagonal faces. Both BWC are structural units of the clathrate hydrates. The properties of various isomers of these BWC are determined by the topology of the hydrogen bond network.

In the present work we investigate the hydrogen rearrangement and its mobility in empty and Ar-filled neutral 5^{12} and $5^{12}6^2$ BWC. The structures are fully optimized at abinitio RHF level using the triple split valence basis sets 6-311G(d) for H and O. In the case of Ar, the core electrons are treated by ECP-SBKJC and the valence shell at the 3G(d) level. We have analyzed 5^{12} and $5^{12}6^2$ types of structures obtained by arranging randomly water hydrogens in agreement with the ice-rules into 10^4 different conformers for each type. Their total dipole moments cover a large range. We studied for both types of structures, the isomers with lowest, middle and higher total dipole moments. We found no direct connection between the total dipole moment and the total energy. The H flipping and the stability of the BWC is, however, determined by interactions of the local dipoles.

The water molecules are 3-coordinated and can be classified into hydrogen mono donors (1d), where one H is oriented towards one of the neighboring O and the other off the cage, and hydrogen two donors (2d), for which both H are oriented towards two neighboring O. The geometrical, electronic and dynamics properties depend on the 1d or 2d type of the interacting water molecules, but this classification is not satisfactory, as a large scatter of geometries is observed in 1d-2d pairs. A very significant improvement is achieved by introducing subclasses according to the relative dipole configurations of the water molecules. Qualitatively, the total energy of an undistorted cluster can be rationalized as a sum of dimer dipole interactions.

Spectral Properties of the 1D t-J Model at Finite Doping

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We developed a new Monte Carlo technique (Hybrid-loop MC) that is able to treat the t-J model at finite doping and incorporates the advantages of the Projector MC (no limitation in the measurable observable) and of the Loop MC (absence of critical slowing down). Using this new technique we are able to study static and dynamical properties of the t-J model in 1D at finite doping in the whole range of parameters (J/t) without sign problem. Here we present the study of the phase diagram (density vs J)¹ and of the spectral functions (photoemission and inverse photoemission) of the model in the different phases (repulsive and attractive Luttinger liquid, and phase separation) at low doping.

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The Interaction of Lead with Iron Clusters

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Lead is going to be considered as a liquid coolant in future Accelerator Driven Systems (ADS) nuclear plants. The understanding of the effects of liquid lead on the mechanical and structural properties of the steels involved in the cooling system is thus of primary importance. Significant insights can be achieved by the interpretation of experimental results¹ made on the bases of molecular dynamics simulations of microscopic-scale models. Fe-based steels (i.e. T91) are actually considered to contain the ADS cooling systems. With the aim of producing molecular dynamics simulation of Fe-Pb systems, a force-field coupling these two species is needed which is currently unavailable due to the lack of experimental data (these two species are immiscible at all temperatures). As a preliminary step to define a Fe-Pb force-field, we have attempted to study the behavior of iron and lead in some simple structures (Pb on Fe_n clusters) using density functional calculations and Car-Parrinello molecular dynamics simulations. Resulting data will be cast into an empirical form.

Our results of the pure Fe_n clusters (with $n = 2, \dots, 13$) are in good agreement with previous experimental data² and all-electron³ and pseudopotential calculations⁴.

In all the considered Fe_nPb complexes (with $n = 7, 13$), we find that the binding energy (3.4 eV) is three-four times larger than that evaluated by Legris et al.^{1b} in the case of low index Fe surfaces. A similar behavior is reported by Jackson et al.⁵ for Fe_n(NH₃)_m complexes. The Fe-Pb distance ranges from 2.249 Å in FePb to 2.789 Å in Fe₇Pb and to 2.791 Å in Fe₁₃Pb. The main effect of the Fe-Pb interaction is the increase of the Fe-Fe distances of those atoms belonging to the surface containing the Pb atom.

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CMSPortal: A Web User Interface to Computational Materials Science Applications

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CMSPortal (**Computational Materials Science Portal**) is a *web user interface* to atomistic simulation applications.

The key features of CMSPortal can be divided in two categories: user side and administrator side features. Here we give a short list of both of them.

User side features

CMSPortal:

- **organizes** the run performed under the interface
- **drives** the user through the configuration of the simulation
- **checks** values assigned to variables
- **helps** the user to compile
- **manages** the run, enabling submission, control of the status and deletion
- enable **control** of the progress of the simulation through either files reading or plotting
- enable both **upload** and **download** of input and output files

In the forthcoming release file sharing mechanism, based on UNIX access permissions, will be added to enable and encourage knowledge condivision.

Administrator side features

CMSPortal:

- **limits** the **access** to computer facility
- minimizes effort spent in teaching **computer facility access** mechanism and policy
- widely and easily **configurability**

To date we imported two of our own codes under CMSPortal (TBPack and CMPTool, our own tight binding and classical molecular dynamics codes). CMSPortal can support any application which have the input of type **KEY = VALUE**. On the other hand, also applications with a different type of input can be used under CMSPortal, in this latter case you only need to write a proper PHP filter (or ask us to do it!). We are planing to import CPMD¹ and ABINIT², two well known DFT-planewaves codes, under CMSPortal.

Last but not least, CMSPortal is based on open source software: PHP, Smarty and JpGraph.

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2. Copyright ©1998-2001 ABINIT group (XG).

Theoretical Investigation of Magnesium-Tris(8-hydroxyquinolino)Aluminum (Alq₃) Interface

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Modern organic light emitting (OLED) and thin film transistor (OTFT) devices are prepared by vapor deposition of a low work function metal cathode onto an organic electron carrier/photoemitter. It is of great interest to study the formation of chemical bonds at the Alq₃/Me interface, since these may affect the performance of the devices in an important way. Recent XPS and UPS experiments on Alq₃/Mg^{1,2} system suggest the formation of organometallic bonds in these systems. Accurate first principle calculation could help to unravel the chemistry of this compound.

Previous³ DFT based calculations, using an Alq₃ molecule in vacuum as a model of the organic slab, found that addition of a metal atom such as Li, Al and Ca always resulted in the formation of a bond between the metal and an oxygen atom. Following the suggestion of Shen et al.², we systematically tested several chemically different initial configurations, including those proposed in ref.^{1,2}. In all cases the Mg atom resulted bound to oxygen atoms. Similar results were also found when a Mg ion was considered instead of a Mg atom.

Finally we considered a more realistic model in which a Mg atom was introduced in the unit cell of a Alq₃ crystal, which contains four Alq₃ molecules. By performing finite temperature (T=600K) MD simulations we discovered in this case the formation of organometallic structures similar to those suggested in ref.². The significance of these results for the interpretation of the experimental XPS data will be discussed.

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Molecular Dynamics Simulations in the Coalescence of Gold Nanoparticles

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We present a systematic molecular dynamics simulations of the coalescence phenomena in the solid regime between Gold nanoparticles. The symmetry of the particles were icosahedral and fcc cuboctahedral with less than 1000 atoms. The calculations were done within the scheme of the embedded-atom method. Some cases present phase transformations before the two particles coalesced. Two regimes of coalescing times are found as a function of temperature. We found slow surface diffusion of gold atoms, resulting in large coalescing times. We investigate the role of temperature on diffusion, giving insights of the influence of temperature on the coalescing time. We present experimental evidence that supports our simulations and conclusions.

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Chemisorption of Hydrogen on Small Gold Clusters: Theory and Experiment

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We present photodetachment spectra of monohydrogenated gold clusters and calculations based on density functional theory (DFT) that unravel the origin of the vertical detachment energies (VDEs). We investigate the validity of the standard interpretation¹ that associates VDEs to ground-state isomers. We propose a new interpretation that is consistent with both the most probable formation route and the structure of the parent clusters. This gives excellent agreement with experiment. Our results have general implications for the interpretation of VDEs when applied to the study of chemisorption on finite systems.

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Self-Assembling Phenomena and Electronic Structure of Calixhydroquinone Nanotube Arrays: Density Functional Calculations

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Recently, utilizing the computer-aided molecular design approach, we reported arrays of calix[4]hydroquinone (CHQ) nanotubes¹ self-assembled with infinitely long one-dimensional short hydrogen bonds and displaced π - π stacking interactions². Here, we report the predicted assembly phenomena as well as the detailed structure and electronic properties of the CHQ nanotubes investigated after the synthesis and characterization of the nanotubes. Our calculations include both chemical approach based on ab initio theory and density functional theory and physical approach based on first principles calculations using ultrasoft pseudopotential plane wave methods.

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Ab Initio and Molecular Dynamics Studies of Novel Ionophores, Receptors, Enzymes, and Proteins

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We report novel functional molecules including ionophores and receptors based on computer-aided-molecular design approach, and various ab initio, Monte-Carlo, and molecular dynamics results of novel molecular systems. The first amphi-ionophore in aqueous solution is discussed. Cyclohexaananyl should be considered as promising binding agents for both cations and anions in the aqueous phase as well as in the gas phase. We have designed a tripodal system which selectively recognizes acetylcholine, which plays an important role as a neurotransmitter. The selectivity has been verified by experiment. Other novel types of ionophores such as collarenes and beletenes are also studied. In addition, we have studied metabolic enzymes and protein foldings. We report a new type of helix motif in protein folding, which we named as λ -helix¹. This helix is quite different from other types of secondary folding motif known to exist in protein secondary conformation, e.g. α -helix, 3^{10} helix, π -helix, β -sheet, etc. The theoretical background of the unusual helical motif is discussed. This information would be of importance to study secondary stricture in protein folding.

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Coulomb Interactions in Simple Lattice-Models: A Comparison between Counter Ion Model and Random Energy Model

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Many modern applications base on ion conductivity in solid materials. Possible fields for practical application are fuel cells, batteries or sensors. Due to the technical relevance of ion conductivity the research efforts are immense. We focus on the nature of the microscopic dynamics as this knowledge is important for the development of new high tech ion conductors with specified properties. Experiments often analyze macroscopic quantities as the frequency dependent conductivity $\sigma(\omega)$ or relaxation times from NMR-spectroscopy. In contrast, computer simulations can give much important information on the microscopic scale.

Here the focus lies on Coulomb interaction as its role for the mobility of particles in ionic conductors is still under debate. To clarify the effect of Coulomb interaction we perform Monte Carlo simulations on two simple lattice models (Counter Ion Model^{1,2} and Random Energy Model³) which contain Coulomb interaction between the positively charged mobile particles, moving on a static disordered energy landscape. We find that the nature of static disorder plays an important role if one wishes to explore the impact of Coulomb interaction on the dynamics. This Coulomb type interaction impedes the dynamics in the Random Energy Model, but enhances dynamics in the Counter Ion Model in the relevant parameter range. Both models are designed to reflect important aspects of glassy ion conductors, which are a high degree of disorder, mobile ions in a fixed glass network and ionic charges. The models differ in the way how the static disorder is realized. On this poster we present an analysis of the interplay of the Coulomb interaction among ions with this static disorder. In particular we ask whether the Coulomb interaction between the mobile particles increases or decreases their mobility.

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A Mathematical Approach to Conformational Dynamics

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The present approach extends the concepts of dynamical systems put forward by Dellnitz and Junge to conformational dynamics within the framework of statistical physics. It allows to define "conformation" in terms of the dynamical behaviour of the molecular system, and to characterise the dynamical stability of conformations. The proceeding is as follows: As a first step the frequency of conformational changes is expressed in terms of statistical ensembles leading to the definition of a specific Markov operator T , that describes the conformational transition probabilities within the canonical ensemble. Second, a discretisation of T by means of Monte-Carlo techniques yields a stochastic matrix P . The principal idea is to identify the molecular conformations with the almost invariant subsets of the dynamical (Hamiltonian) system. These can be computed via the eigenvectors of the stochastic matrix P corresponding to eigenvalues close to 1.

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Mott-Hubbard Metal-Insulator Transition in Paramagnetic V_2O_3 : An LDA+DMFT(QMC) Study

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The electronic properties of paramagnetic V_2O_3 are investigated by the computational scheme LDA+DMFT(QMC). This approach merges the local density approximation (LDA)¹ with dynamical mean-field theory (DMFT)^{2,3} and uses numerically exact quantum Monte Carlo simulations (QMC) to solve the effective Anderson impurity model of DMFT⁴. Starting with the crystal structure of metallic V_2O_3 and insulating $(V_{0.962}Cr_{0.038})_2O_3$ we find a Mott-Hubbard metal-insulator-like transition at a Coulomb interaction $U \approx 5$ eV⁵. The calculated spectrum is in very good agreement with experiment^{6,7}. Furthermore, the occupation of the $(a_{1g}, e_{g1}^\pi, e_{g2}^\pi)$ orbitals and the spin state $S = 1$ determined by us agree with recent polarization dependent X-ray-absorption experiments⁸.

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Optical Properties of Nanocrystals from Ab Initio Supercell Calculations

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We present theoretical investigations of the optical and electronic properties of Si and Ge nanocrystallites. Embedment in a wide-gap material results in a type-I heterostructure with localization of both electrons and holes, as does a hydrogen passivation of free crystallites. Embedment in a matrix with a somewhat smaller gap produces a type-II system. We show how this translates into trends in optical absorption and electronic structure changes.^{1,2}

In order to describe the properties relevant to luminescence, we calculate the transition probabilities close to the absorption edge. Si and Ge turn out to be very different, with strong lowermost transitions occurring only in Ge. Moreover, the transition probabilities in Ge crystallites prove very sensitive to strain. This can be explained with reference to bulk band structures under strain. We predict germanium nanocrystals, embedded with only little strain in some wide-gap material, to be more promising than silicon crystallites in terms of expected electroluminescence.

In order to translate our results into spectra amenable to measurement, the influence of different environments can be modeled within effective medium theory. Spectra calculated for Ge crystallites in sapphire can be compared quite successfully with measured data².

The nanocrystallites are described within density functional theory (DFT) in local density approximation (LDA) and by means of the projector-augmented wave (PAW) method³. We use the VASP program package⁴.

The drawbacks of the LDA and the independent-particle approximation have to be accounted for. We present self-energy calculations on the GW level, as well as pair excitation energies obtained within the Δ SCF method. The Stokes shift between absorption and emission is also discussed for some systems.

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