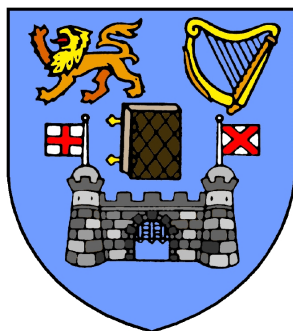


***7th* Irish Atomistic Simulators Meeting**
Trinity College Dublin
16-17 December 2002



The Annual Meeting of Irish Atomistic Simulators brings together Research Scientists from Condensed Matter Physics and Solid State Chemistry Active in Theory and Computation.

The organisers wish to thank Science Foundation Ireland for financial support

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3 Timetable

Short Talks = 15 minutes Presentation + 5 minutes Discussion

Long Talks = 35 minutes Presentation + 5 minutes Discussion

Monday 16 December 2002

Erwin Schrödinger Theatre (Old Physics Building)

14.10-14.50	Edward Timoshenko 4.21	14.50-15.10	Ronan Connolly 4.2
15.10-15.30	Aonghus Lawlor 4.7	15.30-15.50	Gavin McCullagh 4.9
15.50-16.10	Feng-Shou Zhang 4.23	16.10-16.30	Break
16.30-16.50	Lisa Moore 4.10	16.50-17.10	Giuseppe Colizzi 4.1
17.10-17.30	Andreas Larsson 4.6	17.30-17.50	Cristian Sanchez 4.19
17.50-18.10	Dónal Mac Kernan 4.8	18.30	Reception
20.00	Dinner		

Tuesday 17 December 2002

Basement Theatre: Sami Nasr Institute for Advanced Materials (New Physics Building)

09.10-9.30	Michael Nolan 4.14	09.30-09.50	Scott Monaghan 4.12
09.50-10.10	Clive Harris 4.3	10.10-10.30	Henry Pinto 4.17
10.30-10.50	Karen Johnston 4.4	10.50-11.10	Sarah Moore 4.11
11.10-11.30	Break	11.30-11.50	Mauro Pereira 4.16
11.50-12.30	Stefano Sanvito 4.20	12.30-12.50	Alexandre Reily Rocha 4.18
12.50-14.00	Lunch	14.10-14.30	Małgorzata Wierzbowska 4.22
14.30-15.50	Charles Patterson 4.15	14.50-15.10	Svjetlana Mulaomerović 4.13
15.10-15.30	Nikolaos P. Konstantinidis 4.5	15.30-15.50	Closing Remarks

4 Abstracts

4.1 Investigating the disappearance of ferroelectricity in H-bonded crystals under pressure by *ab initio* calculations: The case of KH_2PO_4 (KDP) and DH_2PO_4 (DKDP)

Giuseppe Colizzi(speaker)

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Abstract

A series of recent experiments carried out for several members of the family of H-bonded ferroelectric materials has shown that the ordered phase disappears when sufficient pressure is applied. This fact, which is well-known also for perovskite ferroelectrics, exhibits, nevertheless, two remarkable peculiarities [1]:

1.the critical temperature vanishes when the protons are still located off-center, at a distance $\delta = 0.2\text{\AA}$ from the center of the H-bonds.

2.this distance appears to be universal, in the sense that it does not depend on the material and the dimensionality of the H-bond network.

For this reason it has been postulated [1]that this is a property of individual O-H...O H-bonds, and tentatively ascribed to the zero-point-motion of the O-O vibrations. In our study of ferroelectricity of KH_2PO_4 (KDP) and DH_2PO_4 (DKDP) materials by *ab initio* calculations we aimed to investigate also at this aspect of the phase transition. Our calculations, which disregard the quantum nature of nuclei, do not show the behaviour that seems to emerge from experiments. So, if the interpretations of experimental data is correct, we conclude that this behaviour is intimately connected with the quantum nature of hydrogen atoms. Calculations in which nuclei are also quantum mechanically treated are necessary in order to shed light on this aspect of structural phase transitions in H-bonded materials. Work in this direction is in progress.

References

- [1] McMahan, Plitz and Nelmes, *Ferroelectrics* 108, 277 (1990)

4.2 Monte Carlo and theoretical studies of conformations of dendrimers in solutions

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Abstract

The regular and well-defined structures of dendritically-branched polymers (dendrimers) provide potential for their application as new advanced materials, superior catalysts[1], potential drug delivery vehicles[2] amongst other things.

In order to better understand the structures of these polymers, conformations of isolated dendrimers of G=1-7 generations with D=1-6 spacers were studied under various solvent conditions[3], by means of a version of the Gaussian self-consistent (GSC) method[4] and Monte Carlo (MC) simulations in continuous space based on the same coarse-grained model.

Here, the scaling law for the dendrimer size, the radial density and the degrees of bond stretching will be analysed. It will also be confirmed that while smaller dendrimers have a dense core, larger ones develop a hollow domain at intermediate distances from the core in good solvent. Finally, the unimolecular micelle conformations of amphiphilic co-dendrimers will be briefly discussed.

Acknowledgements

The authors are grateful to Professors F. Ganazzoli and G. Allegra, Dr. G. Raos, G. Flanagan, M. O'Brien and C. O'Farrell for relevant discussions and to IRCSET and Enterprise Ireland for funding.

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4.3 Theory of Band Structure of Pentacene Organic Crystals

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Abstract

Very high carrier mobilities were reported in ultra pure pentacene crystals[1], providing many possible device applications. We carried out a systematic investigation of the band structure of such crystals using a density functional theory approach (the VASP simulation package), followed by a much simpler tight-binding (TB) approach. The calculated band dispersions are consistent with the measured mobility values, with a relatively large dispersion implying a high carrier mobility at temperatures close to 0 K. We also studied the variation of band width with respect to direction within the crystal structure and found it to be anisotropic, with the maximum band width along the **b** axis and the minimum along the **a** axis. Our results suggest that the highest mobility should be obtained along the **b** direction, with the lowest mobility in the **ab** plane along the **a** direction. This compares with the reported directions of maximum and minimum mobility being at 30° to the **b** and **a** directions respectively.

References

- [1] J. H. Schön, Ch. Kloc, B. Batlogg, *Phys. Rev. B*, **63**, 245201

4.4 Atomic Scale Modeling of Strontium Titanate Surfaces

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Abstract

The atomic structure of the SrTiO₃ (001) surface has been investigated using the *ab initio* full-potential linear muffin-tin orbital method (FP-LMTO). There are two possible terminations of the (1×1) surface reconstruction: the TiO₂- and the SrO-termination. I have calculated the atomic relaxations of both surface terminations. The relative stabilities of the two surfaces have been calculated as a function of the TiO₂ chemical potential.

A (2×1) reconstruction has been observed by Castell [1] and Erdman *et al.* [2] using scanning tunneling microscopy (STM). Three atomic structure models were proposed. I have used the FP-LMTO method to relax each of these models and calculate the surface energies as a function of the TiO₂ chemical potential and O partial pressure.

Acknowledgements

I would like to thank my supervisor Tony Paxton and the rest of the ASG for their help and advice. I would also like to thank DEL and the ASG for financial support.

References

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- [2] Natasha Erdman, Kenneth R. Poeppelmeier, Mark Asta, Oliver Warschkow, Donald E. Ellis and Laurence D. Marks. The structure and chemistry of the TiO₂-rich surface of SrTiO₃ (001). *Nature*, **419** (2002).

4.5 Diagonalization Of Model Hamiltonians Using Their Spatial Symmetry Properties

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Abstract

Diagonalization of model Hamiltonians is limited to systems of small sizes due to exponential growth of the Hilbert space with the size of the system. The maximum size of systems studied can be increased if the symmetry of the Hamiltonian under the interchange of particles is taken into account. In this way, the Hamiltonian matrix is block factorized in subspaces with the same spatial symmetry. The method is used to study the Heisenberg model as a “toy” model in spin clusters which have the connectivity of fullerene molecules. The effect of frustration in the low energy spectrum is examined.

4.6 The nature of endohedrally doped fullerenes

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National Microelectronics Research Centre and
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Abstract

It was postulated and experimentally verified shortly after the discovery of C_{60} , that the hollow carbon cage could encapsulate individual atoms to form the so-called endohedrally doped fullerenes, opening up the possibility to use fullerenes with engineered properties as building blocks for a range of technical applications. In this report, we present results on the electronic properties of the group V atoms nitrogen and phosphorus encapsulated by C_{60} .

It has been found that when nitrogen or phosphorus is encapsulated within C_{60} they retain their 'atomic' properties. This has been shown experimentally[1] and by calculation[2, 3]. It appears as if these atoms are shielded from their environment by the C_{60} cage, in that the endohedral atoms do not form bonds to the cage, there is no charge transfer between the atom and the fullerene, and the group V atoms maintain a quartet spin state. The encapsulating C_{60} cage appears to be little disturbed by the endohedral dopants. We will show that these group V atoms are suspended in the centre of the cage due to repulsive interaction with the cage. These properties of the endohedral species NC_{60} and PC_{60} suggest their use for applications (e.g. quantum computing) where it is desirable to have isolated quantum systems, with controllable interactions to their external environment.

References

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- [2] *Chem. Phys. Lett.* **326** (2000) 567.
- [3] J.A. Larsson, J.C. Greer, W. Harneit, and A. Weidinger *J. Chem. Phys.* **116** 7849 (2002).

4.7 Are there universal laws present when systems “arrest”

Aonghus Lawlor^{1,a,c}(**speaker**), Dan Reagan², Gavin D. McCullagh^{1,c}, Paolo De Gregorio¹, Piero Tartaglia³, and Kenneth A. Dawson^{1,b,c}

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Abstract

One of the ubiquitous observations of nature is that many systems solidify without crystallizing. Such processes are termed variously ‘gellation’, vitrification, dynamic arrest, ‘solidification’, jamming, ergodic-non-ergodic transition.

We introduce and discuss a new order parameter of dynamic arrest[1] and study its behaviour in some examples of simple models of dynamic arrest. We find that, when the (vanishing) diffusion constant is plotted against the new order parameter (‘dynamically accessible volume’) a high degree of universality emerges.

This, along with some connection to ‘gellation’ suggest that many varied processes of solidification (where the system does not crystallize) may in fact be quite ‘Universal’ phenomena of nature.

References

- [1] Aonghus Lawlor, Dan Reagan, Gavin D. McCullagh, Paolo De Gregorio, Piero Tartaglia, and Kenneth A. Dawson, *Phy. Rev. Lett.*, **89**, 245503, (2002).

4.8 A Trotter Type Integrator for Quantum-Classical Non-Adiabatic Dynamics

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Abstract

Quantum-classical dynamics describes systems composed of a quantum subsystem coupled to a classical environment and provides a means to study the dynamics of many-body systems that are not amenable to investigation using full quantum dynamics. A non-adiabatic description is necessary when the Born-Oppenheimer approximation breaks down, as is the case for many electron and proton transfer processes in chemistry and biology.

In the Wigner approach we use[1], the isolated quantum subsystem and bath obey quantum mechanics and classical mechanics, respectively, but their coupled evolution is given by quantum-classical equations of motion where a simple Newtonian description of the environmental degrees of freedom no longer exists.

In recent work, we showed that this approach reproduces essentially exact results for the spin-boson model[2, 3] but the demonstration was limited to short and medium times. Currently we are developing an integrator based on a Trotter break-up of the propagator which unlike Dyson equation based integrators is bounded about unity. It should, therefore, be better suited to generating long time dynamics. In my talk, I will explain how this propagator is obtained and implemented numerically.

References

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4.9 "Interpreting particle gellation as a glass transition - a Lattice Model Approach"

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Abstract

Dense colloidal systems with mainly repulsive interactions have, for some time, been described successfully by glass theory. More recently however, the discovery of a second glass-type transition driven by extremely short ranged attractive interactions has greatly fuelled interest in this field[1, 2]. We have proposed that the phenomenon of particle gellation may be viewed precisely as a glass transition, and used theory and molecular dynamics to develop this idea. However, research into this area by simulation is in general problematic, due to the long time-scales involved. We therefore seek to develop a very simple lattice model that exhibits such phenomena that can be studied in detail. The basic model we are using, is a development from the original work by Mezard & Biroli[3] that displays the repulsive glass transition on a cubic lattice and gives much useful information about it[4]. We report extensions made to this model representing short ranged attractions in order to investigate the attractive glass transition and show some early results from the extended model.

Acknowledgements

The project is funded by COST P1. I also receive funding from Enterprise Ireland.

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- [2] Giuseppe Foffi, Gavin D. McCullagh, Aonghus Lawlor, Emanuela Zaccarelli and Kenneth A. Dawson, Francesco Sciortino and Piero Tartaglia, Davide Pini and George Stell. Phase equilibria and glass transition in colloidal systems with short-ranged attractive interactions. Application to protein crystallization. *Phys. Rev. E*, **65**, 031407 (2002).
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4.10 How big is an atom? Solution and diffusion of Ga in Al

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The addition of gallium to aluminium is known to enhance grain boundary mobility and in high concentrations leads to intergranular embrittlement. Thomson *et al* [1] performed calculations on a $\Sigma=11$ boundary to study the segregation of Ga at the boundary. They found that despite Ga being a larger atom than Al, when substituted in Al, Ga had an atomic volume similar to that of Al and that Ga was attracted to tight sites at a boundary, where its presence relieved local stress. In the present study we investigate the effect of the addition of Ga to bulk Al through *ab initio* calculations. We present results for the heat of solution and mobility of Ga in Al. We compare these results with those for pure Al in the same configurations and explain our results with the use of the '2-radii' as derived from pseudopotential theory (See Heine and Weaire) [2].

Acknowledgements

I would like to thank my supervisor Mike Finnis and the members of ASG for their support and advice. I would also like to acknowledge DEL for the financial support.

References

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- [2] V. Heine and D. Weaire. Pseudopotential theory of cohesion and structure *Solid State Phys.*, **24**, 249 (1970).

4.11 Tight-binding calculations of electron scattering in dilute nitride semiconductors

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Abstract

We will show how the tight-binding matrix elements can be altered in the vicinity of a nitrogen substitutional defect in GaAs to reproduce the appropriate scattering cross-section for electrons near the conduction band edge. The approach used in reproducing the scattering cross-section is analogous to the atomic pseudo-potential approach used in first-principles, plane-wave calculations. We investigate two schemes for fitting the scattering obtained in a two-level model of the conduction band interaction with the defect, which has been devised to fit the experimental variation of the band-gap with nitrogen composition, and find that varying the matrix elements on the first-shell Ga atoms is more effective than varying the Ga-N matrix elements or the N on-site energies.

4.12 Structure and Stability of the Zirconium and Hafnium Pseudo-Binary Oxides for High-K Dielectric Applications

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Abstract

As electronic devices progressively improve in performance, increase in functionality, and decrease in size, research and development in the semiconductor device industry is expected to advance in accord with Moore's law. However, the silicon dioxide gate insulator—a crucial component of the most fundamental active device of all circuitry: the Metal–Oxide–Semiconductor Field Effect Transistor (MOSFET), is approaching obsolescence as the gate oxide dielectric.

This work looks at the zirconium and hafnium pseudo-binary oxides as possible replacement gate dielectric materials, and investigates through first principles methods, their structure, composition and thermodynamic stability. Very little is currently known about these materials, but they are reported to be good candidates as replacement gate oxides because of their predicted thermodynamic stability in contact with silicon and the gate electrode of a MOSFET.

Simulations in this work show for the first time that all of these silicate solid solutions (except zircon and hafnon) are thermodynamically unstable, which in many cases results in micro-scale phase segregation.

Phase segregation severely degrades the electrical properties of a dielectric material, and if confirmed by experiment, these silicates—with the exception of zircon and hafnon, will be unsuitable for use as gate dielectrics in MOSFET applications.

4.13 Electron-hole excitations and optical spectra of insulators and semiconductors

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Abstract

The influence of the electron-hole interaction on the dielectric and optical properties has been known at least for about twenty years [1]. However, computational limits had not allowed a complete investigation of that interaction until the present time. The electron-hole attraction leads to bound states (excitons) below the band gap and the effect is especially important in wide gap insulators. We investigate these effects within many-body perturbation theory which leads to the Bethe-Salpeter equation.

The dielectric function and energy loss function of condensed argon are calculated by solving the Bethe-Salpeter equation using a method similar to that reported by Rohlfing and Louie [2], and by conventional RPA technique. It has been shown that inclusion of electron-hole attraction is crucial for an adequate comparison of the theoretical and experimental optical spectra.

References

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4.14 Electron Transport Through Single Molecule Nanowires

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Abstract

The Study of molecular as active components of nanoelectronic devices is growing all the time. Experimental work has stimulated the development of rectifiers, wires, transistors and memory elements [1, 2, 3]. There still remains much that is unknown about the bonding of molecules to metal contacts, the mechanisms of electron transport and the techniques needed to fabricate truly single molecule devices [4, 5, 6, 7].

In this work we consider as metal-molecule-metal assembly resulting from the interaction of thiol molecules, e.g benzenedithiol, with 13 atom gold nanoclusters:

$\text{Au}_{13} - \text{Molecule} - \text{Au}_{13}$. Benzenedithiol has been the prototypical molecule in experimental and theoretical studies of molecular electronic devices [1, 6]. The effect of an applied electric field is the main consideration of this work. We demonstrate how the electronic structure is modified and how some molecular orbitals polarise much faster than other orbitals. The results obtained are rationalised in terms of a simple model, which explains very well the behaviour seen in the DFT calculations.

Acknowledgements

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4.15 Role of oxygen in magnetism of transition metal oxides

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Abstract

Many models for transition metal oxides focus on the metal ions and implicitly assume that oxygen ions play a passive role in the magnetism, by mediating interactions between spins on adjacent sites (superexchange), for example. Recent *ab initio* calculations by our group on manganites and on vanadates [1] show that in some oxides, some oxygen ions have fluctuating magnetic moments that cannot be neglected in models of the magnetism. These particular ions mediate very strong ferromagnetic coupling of adjacent metal spins. These observations help to explain (i) why the double exchange Hamiltonian fails to describe magnetism in manganites and (ii) the magnetic structures observed in the series $\text{La}_{(1-x)}\text{Ca}_x\text{MnO}_3$ by neutron scattering.

Acknowledgements

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4.16 Non-equilibrium Theory for Absorption and Gain in Quantum Cascade Lasers

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Abstract

Quantum Cascade Lasers open a fascinating door for manipulations of a man-made material system for infra red device applications.[1, 2] Only electronic sub bands are involved and consequently, the usual quasi-equilibrium approximations used for electron-hole transitions can not be applied, i.e. non equilibrium distributions must be used to describe gain operation consistently. Here we use Keldysh Non Equilibrium Greens Functions and calculate numerically both gain and absorption in structures that, under an applied voltage can operate as quantum cascade lasers. Wannier or Wannier-Stark states, [3] are used to expand the relevant Green's functions and the interplay between different many particle corrections is illustrated through numerical solutions of our equations, using techniques that extend previous methods successfully used for absorption and gain of conventional semiconductor lasers.[4] Exchange and Depolarization contributions are considered in the numerics and the general theory developed supports high order Coulomb scattering beyond the Screened Hartree Fock level. The theory can be used to study and predict new transport and optical properties of QCL structures and as the starting point for realistic device simulation and design.

Acknowledgements

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4.17 First-principles study on the structure of the (001) surface of $\gamma - Al_2O_3$.

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Abstract

The controlled growth of alumina films by atomic layer deposition (ALD) is of great interest to the electronics industry, as high-k dielectrics are being sought for the next-generation MOSFETS [1]. Many aspects of the surface structure and reactivity of alumina are still unknown, but are amenable to computation. We present a theoretical study of the alumina polymorph $\gamma - Al_2O_3$. The calculations are based on density functional theory (DFT). In order to predict the bulk crystal structure of $\gamma - Al_2O_3$, we have compared the total energies of 17 non-equivalent possible structures of a 40 atom cell, derived from the perfect spinel by replacing the Mg by Al and removing Al. The lowest energy structure has widely separated Al-vacancy sites, in agreement with previous works [2]. The second part of this work deals with alumina surfaces. For $\gamma - Al_2O_3$, we estimated the energy of several relaxed surfaces, namely (001), (111) and (110). The atomic and electronic structure of the most stable (001) surface is discussed.

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4.18 Non-equilibrium Green function's formalism for transport in magnetic point contacts

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Abstract

The importance of understanding the behavior metallic nanowires (NW) lies on their potential use for technological applications of nanoscaled devices. In particular, we focus our attention on studying the transport properties of magnetic point contacts, in which the current is spin polarized. These systems present a strong non-linearity in the current-voltage profile (I - V) which remains unexplained [1].

In this talk we describe the transport in a magnetic point contact using a non-equilibrium Green function formalism (NEGF) [2] with a tight-binding Hamiltonian [3], which accounts for charging effects in the nanowire. We specifically apply this method to Ni point contacts, for which experimental data are available [1]. Our model system consists of two charge reservoirs formed by a simple cubic semi-infinite arrangement of Ni atoms and of a three-atom-long NW. Each Ni atom is described as a 2-level system (plus spin) with s and d orbitals, and the occupation is fitted self consistently to that of bulk fcc Ni.

When the system is under bias our results show that the asymmetry of the coupling of the nanowire with the electron reservoirs causing charge accumulation in the NW. This results in asymmetric I - V curves, which become more symmetric when the temperature is increased.

Acknowledgments

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4.19 Molecular reorientation of water adsorbed on charged Ag(111) surfaces

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Abstract

Double layer modelling plays a fundamental role in theoretical electro-chemistry since an accurate knowledge of the double layer is needed in any attempt to describe other electrochemical phenomena such as charge transfer processes. The work described in this talk represents a first step toward the fully ab-initio description of the charged electrochemical interface. We have studied the adsorption of a water molecule on charged Ag(111) surfaces using a method recently developed in our group [1]. The orientation relative to the surface for different surface charge densities was obtained. The results agree with experimental findings that water orientation in the electrochemical interface changes from oxygen-up to oxygen-down on going from negative to positive charges. The critical surface charge needed for full orientation with the field has been found to be around $15 \mu\text{C cm}^{-2}$. These results provide new information about the role of the water-metal interaction in determining the properties of the inner layer.

Acknowledgements

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4.20 Building spintronics materials: diluted magnetic semiconductors

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Abstract

Diluted magnetic semiconductors (DMS) are obtained by doping ordinary semiconductors with transition metals. This is usually achieved under non-equilibrium conditions, in order to overcome the low solubility limit of transition metals in semiconductors. However, despite the low concentration of transition metal achievable (up to $\sim 10\%$), ferromagnetism is detected with Curie temperature (T_c) of the order of 100 K for (Ga,Mn)As. This is usually understood in terms of Zener model, where the Mn spins are coupled via itinerant holes.

In this talk I will present a theoretical study of the structural, magnetic and transport properties of two important DMS: (Ga,Mn)As and (Ga,Mn)N [1]. In (Ga,Mn)As there is a strong hybridization of the Mn- d states at the top of the valence band of GaAs, and density functional theory in the local spin density approximation provides a good description of the ground state. Moreover it is also appropriate to describe several key features as 1) the solubility limit of Mn in GaAs [2], 2) the role of intrinsic defects [3], and 3) the effect of electron confinement [4].

In contrast the Mn- d states lie at mid-gap in GaN and they only weakly hybridize with either the valence or conduction band. In this case the local density approximation fails to describe the localized character of the d shells and the exchange coupling is largely overestimated. I will then present some preliminary results obtained with a self-interaction correction scheme to the local density approximation. This reveals orbital ordering for a single Mn impurity in GaN, and an almost un-split valence band. This does not support an itinerant model of ferromagnetism, since no holes are left in the valence band.

Acknowledgements

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4.21 Intra-chain pair distribution functions of isolated open, ring and star homopolymers in solution

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Abstract

Results of Monte Carlo study [1] of the monomer–monomer radial distribution functions, static structure factor and asphericity characteristics of a single homo-polymer in the coil and globular states will be presented for three distinct architectures of the chain: ring, open and star. In the repulsive regime, where a stretched exponential times a power law form (de Cloizeaux scaling [2]) can be applied, the scaling exponents δ and θ have been obtained. These are found to be in a good agreement with the theoretical prediction from the recent improved higher–order Borel–resummed normalization group calculations [3, 4].

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4.22 LDA+U method for diluted magnetic semiconductors: implementation in the localised atomic orbital basis set code SIESTA

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Abstract

One of the most popular methods of computational condensed matter is the density functional theory (DFT), within its local density (LDA) or gradient corrected (GGA) flavour. Although its usefulness is unquestionable, some failures appears whenever strongly correlated system are studied. This is the case of the partially filled d-shells of transition metals or the f-shells of rare earths, which are strongly localised. In these situations an additional term needs to be added to the standard LDA exchange-correlation potential, namely an on-site repulsive Coulomb interaction U acting on the aforementioned bands. The method, known as LDA+U [1], is successful in properly describing the band picture of Mott-Hubbard insulators and of the magnetic phase of intermetallic compounds. Despite its success, the determination of 'magic' parameters U and J is not straightforward and it may employ machineries from computational quantum chemistry. While leaving the ab-initio determination of the U parameter for a later time, we start our work by implementing the LDA+U method into the fast order-N DFT code SIESTA [2]. The systems we want to study are wide-gap diluted magnetic semiconductors [3]. These are ordinary III-V wide gap materials with embedded magnetic impurities, which form a long-range ferromagnetic order with remarkably high Curie temperatures. These materials, which preserves both semiconducting and magnetic properties are very promising for future magneto-opto and electronic devices.

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4.23 Vibrational Relaxation of I_3^- in liquids and glasses

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Abstract

Vibrational dephasing rates $1/T_1$, $1/T_2$, $1/T_2^*$ of I_3^- in xenon, ethanol, and methyltetrahydrofuran solutions at temperatures ranging from room temperature to 100 K are studied using equilibrium molecular dynamics simulations. The triiodide ion is described by a valence bond model which responds to the field of the classical solvent molecules. One finds the pure vibrational dephasing rate decreases slowly as the temperature is lowered and does not show a discontinuity when the fluid solidifies in agreement with experiment. To interpret the results, a perturbation theory expression for the pure vibrational dephasing rate is evaluated for simulation of a rigid model of the ion and found to agree well with the direct observations. It shows that, unusually, both the solvent force and the solvent potential curvature, contribute to the pure dephasing of the symmetric mode. The correlation time is very short, insensitive to the mobility of surroundings, which may explain the results.

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