Ψ_k Newsletter
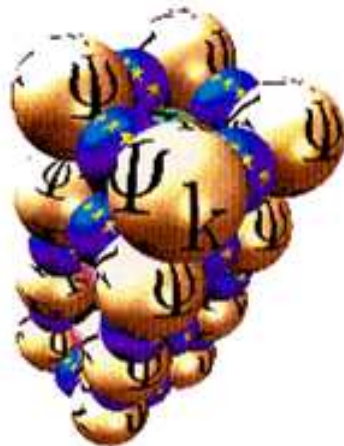
AB INITIO (FROM ELECTRONIC STRUCTURE) CALCULATION OF COMPLEX PROCESSES IN MATERIALS

Number 110   April 2012

Contents

1 Editorial

In this April issue of the Psi-k newsletter we have a number of workshop reports and two reports on collaborative visits. Following those are some announcements of meetings, positions and abstracts of newly submitted/recently published papers.

The scientific highlight article of this issue is by Xavier Andrade (Harvard), Joseba Alberdi-Rodriguez (Donostia),David A. Strubbe (Berkeley), Micael J. T. Oliveira (Coimbra), Fernando Nogueira (Coimbra), Alberto Castro (Zaragoza), Javier Muguerza (Donostia), Agustin Arruabarrena (Donostia), Steven G. Louie (Berkeley), Alán Aspuru-Guzik (Harvard), Angel Rubio (Donostia & Berlin), and Miguel A. L. Marques (Lyon) on "TDDFT in massively parallel computer architectures: the OCTOPUS project".

For details please check the table of content of the newsletter.

The *Uniform Resource Locator* (URL) for the Psi-k webpage is:

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	function
psik-coord@stfc.ac.uk	**messages to the coordinators, editor & newsletter**

Z (Dzidka) Szotek, Martin Lüders, Leon Petit and Walter Temmerman
e-mail: psik-coord@stfc.ac.uk

2 Psi-k Activities

"Towards Atomistic Materials Design"

2.1 Reports on the Workshops supported by Psi-k

2.1.1 Report on 5th International School & Workshop on "Time-dependent density-functional theory: Prospects and Applications"

Benasque, Spain

January 3-17th, 2012

SPONSORED BY:

- CECAM Headquarters (Lausanne)
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O r g a n i z e r s

A. Rubio (Univ. País Vasco and ETSF, Spain),
E. K. U. Gross (MPI Halle, Germany),
M. A. L. Marques (LPMCN, Université Lyon I, France),
F. Nogueira (CFC, Univ. de Coimbra, Portugal),
A. Castro (BIFI, Universidad de Zaragoza, Spain)
http://www.benasque.org/2012tddft/

Report

The fifth School and Workshop on Time-Dependent Density Functional Theory was hosted, as the previous ones, by the Benasque Center for Science. It is the fifth of a very successful series that started in 2004. The purpose has always been to (1) make a very intense introduction to both the theory, the practice, and the numerical implementation of time-dependent density-functional theory (TDDFT) mainly (but not exclusively) oriented to young scientists willing to initiate or strengthen their knowledge and skills on TDDFT, followed by (2) a workshop on the subject in which all the main aspects are to be covered by the leading experts. The physical conditions offered by the new Benasque Center for Science (http://www.benasque.org/) allow for a very fruitful and informal contact between the students and the teachers, that we strongly encouraged.

The results has been a very fruitful School, and an exciting Workshop. Regarding the School, all the pedagogical documents of the school are made available to the everybody through the webpage of the meeting. Also, we provided all the students with the new book "Fundamane-tals of Time-Dependent Density-Functional Theory", edited by M.A.L. Marques, N. Maitra, F. M. S. Nogueira, E. K. U. Gross, and A. Rubio, *Lecture Notes in Physics* **837** (Springer, Berlin/Heidelberg, 2012), as it has all the basic material to be discussed in the lectures. The students could access, throgh the lectures and the discussion with the teachers, to all the relevant aspects of the theory, as well as to the key aspects of the experimental results addressed by the theory (some experimental lecturers were invited).

In order to allow for the students to better discuss their research projects among themselves and with the teachers of the school, we asked them to present posters describing their current work and/or planned research project. Two of these posters were then selected as oral contributions to the international workshop and were granted the "Pedro Pascual Prize" for the best posters of the school. As in previous editions of this event, the number of applications surpassed all expectations and, of course, also the limit of places that we had to satisfy in order for the students to get the maximum benefit from the school, and also due to space and computer resource limitations.

School

The School covered the topics that we believe constitute the core of the theory, and also included a few lectures about the experimental aspects that TDDFT intends to describe.

- Foundations of the theory, cornerstone theorems.

- Foundations of many-electron theory, which permits to study complementary theories for the description of many-electron dynamics: GW, Bethe-Salpether.

- Overview of spectroscopies: description of experiments, by leading experimentalists in the field.

- TDDFT approach to quantum electronic transport.

- Advanced concepts: memory, etc.

- Applications.

The theoretical work was complemented, in the afternoons, by practice work in the computer room of the Benasque Center for Science, where the students could access state-of-the-art software for TDDFT calculations. The practical work was divided in three sections:

- Software development: guided construction of a TDDFT code. The students had the opportunity of analyizing a basic TDDFT code, in order to develop skills in software engineering of this kind of programs.

- Tutorial on the `octopus` code.

- Tutorial on the `yambo` code.

Workshop

The aim of the Workshop was to assess the present status of TDDFT approaches to the study of spectroscopic properties of real materials, and explore their capability for applications in further systems with technological and biological interest. The recent developments of TDDFT covered during the workshop include TDDFT versus current-DFT, van der Waals interactions, applications to biological systems, new functionals, transport phenomena, optical spectra of solids, etc. Due to the different methods used to tackle this problem (Many-Body Theory, Density Functional Theory, Configuration Interaction, semi-empirical approaches), this Workshop was intended as a way to promote links among scientists coming from different communities working or interested in electron excited states. Also it was intended as a follow-up event for the students attending the school as it was a good opportunity for them to see the real implications of the school lectures and get the new theoretical advances in the the development of exchange-correlation functionals as well as applications to complex systems (nanostructures, bio-molecules, interstellar molecular analysis, solids, etc.) Our goal was to bring together scientists working on foundations and different applications of TDDFT and many-body theory, trying to assess the capability of current approximations to be applied to real systems of increasing complexity. The invited and contributed talks covered:

- Fundamental topics on TDDFT, essential theorems.

- Many-Body Theory, and electron transport theory.

- New approximations to the exchange and correlation potential and kernel.

- Quantum optimal control.

- New experimental results, mainly in the areas of high intensity fields and atto-second Science.

Two poster sessions were scheduled; the posters were hanged at the beginning of the event, and were kept at all times, so that students and participants could discuss at any time. All the posters were photographed and can be consulted at:

http://www.benasque.org/2012tddft/cgi-bin/pictures.pl

In order to incentivate the creation of good posters, we organized a contest in which the two best posters (as qualified by a poll in which all School teachers could vote) got the "Pedro Pascual Prize". These two awards were given to Federica Agostini ("Exact factorization of the time-dependent electron-nuclear wave-function: A mixed quantum-classical study", and Daniel Whitenack ("Derivative discontinuities, orbital energies and lifetimes in density functional resonance theory"). Both of them, who received equal number of votes, were invited to present their work at the Workshop.

The work presented at the Workshop has demonstrated the outstanding capabilities of TDDFT to describe the various forms of spectroscopy that probe the electron dynamics of matter. Spectroscopies, in general, are the tools used to study the microscopic structure of matter. The experimental results obtained with these tools can only be interpreted correctly with the help of accurate theoretical methods, capable of simulating the microscopic behavior of matter subject to external perturbations. A number of spectroscopic methods address electronic excited states (e.g. optical absorption spectroscopy, photo-electron emission spectroscopy, etc), and hence the need of first principles theoretical methods capable of addressing the excited state many-electron problem. Time-dependent density-functional theory (TDDFT) is one of such methods.

It is not, however, the only approach to the excitations of many-electron systems. In fact, more accurate (yet more expensive) techniques (based on many-body perturbation theory, for example) exist, and therefore these alternatives have also been covered in both the workshop and the school in particular their relation and comparison to TDDFT. However, TDDFT achieves a good balance between accuracy and computational cost. This workshop has clearly shown how its use is increasing, and it is fast becoming one of the tools of choice to get accurate and reliable predictions for excited-state properties in solid state physics, chemistry and biophysics, both in the linear and non-linear regimes.

We have witnessed in this Workshop the most recent developments of TDDFT (and time-dependent current functional theory): the description of photo-absorption cross section of molecules and nanostructures, electron-ion dynamics in the excited state triggered by either a small or high intense laser fields, van der Waals interactions, development of new functionals coping with memory and non-locality effects, applications to biological systems (chromophores), transport phenomena, optical spectra of solids and low- dimensional structures (as nanotubes, polymers, surfaces...).

Programme

School

Wednesday, January 4th

- 0920 0930 Opening Remarks

- 0930 1030 TDDFT I (E. K. U. Gross).

- 1030 1130 TDDFT II (E. K. U. Gross).

- 1130 1230 Introduction to Green's function (S. Kurth).

- 1230 1330 Many-Body: GW I (M. Gatti).

- 1530 1830 Introduction to the practical classes and codes.

  - Introduction to octopus (D. Strubbe).
  - Introduction to yambo (D. Sangalli).
  - Introduction to the coding exerciese (A. Castro).

Thursday, January 5th

- 0930 1030 Overview of spectroscopies I (F. Lepine).

- 1030 1130 Overview of spectroscopies II (F. Lepine).

- 1130 1230 TDDFT III (E. K. U. Gross).

- 1230 1330 Many-Body: GW II (M. Gatti).

- 1530 1830 Basic TDDFT code I.

Friday, January 6th

- 1600 1700 TDDFT IV (E. K. U. Gross).

- 1700 1800 The link between experiment and theory (S. Botti).

- 1800 1900 Many-Body: BSE I (I. Tokatly).

- 1900 2000 Many-Body: BSE II (I. Tokatly).

Saturday, January 7th

- 0930 1030 TDDFT as a tool in chemistry I (I. Tavernelli).

- 1030 1130 Models in time-dependent phenomena I (M. Lein).

- 1130 1230 Quantum transport (S. Kurth).

- 1230 1330 TDDFT as a tool in chemistry (I. Tavernelli).

- 1530 1830 Basic TDDFT code II.

Sunday, January 8th

- 1630 1730 Basic TDDFT code III.

Monday, January 9th

- 0930 1030 Advanced TDDFT I (N. Maitra).

- 1030 1130 Models for time-dependent phenomena II (M. Lein).

- 1130 1230 Advanced TDDFT II (N. Maitra).

- 1230 1330 Current TDDFT I (C. Ullrich).

- 1530 1830 octopus I.

- 1830 2000 POSTER SESSION.

Tuesday, January 10th

- 0930 1030 TDDFT as a tool in chemistry III (I. Tavernelli).

- 1030 1130 Non-linear material response properties with TDDFT (D. Strubbe).

- 1130 1230 Current TDDFT II (C. Ullrich).

- 1230 1330 Models for time-dependent phenomena III (M. Lein).

- 1530 1830 octopus II.

Wednesday, January 11th

- 0930 1030 TDDFT vs. Many-Body I (R. van Leeuwen).

- 1030 1130 TDDFT as a tool in chemistry IV (I. Tavernelli).

- 1130 1230 Overview of spectroscopies III (S. Huotari).

- 1230 1330 Advanced TDDFT III (N. Maitra).

- 1530 1830 yambo I.

- 1830 2000 POSTER SESSION.

Thursday, January 12th

- 0930 1030 TDDFT vs. Many-Body II (R. van Leeuwen).

- 1030 1130 Models for time-dependent phenomena IV (M. Lein).

- 1130 1230 Overview of spectroscopies IV (S. Huotari).

- 1230 1330 Current TDDFT III (C. Ullrich).

- 1530 1830 yambo II.

- 1830 1900 Final remarks.

Workshop

Friday, January 13th

Chairperson: C. Ullrich

- 0920 0930 Opening remarks (A. Rubio).

- 0930 1015 Time-dependent current density functional theory: Rigorous lattice formulation (I. Tokatly).

- 1010 1050 Semiclassical Correlation in Density-Matrix Propagation (N. Maitra).

- 1050 1130 Exact Factorization of the Time-Dependent Electron-Nuclear Wave Function (A. Abedi).

- 1130 1200 COFFEE BREAK.

Chairperson: I. Tokatly

- 1200 1240 (TD)DFT: fundamentals and a new functional (M. Ruggenthaler).

- 1240 1320 Accurate density-functional methods based on correlation energy functionals within the exact- exchange random phase approximation (A. Görling).

- 1320 1400 Optics of semiconductors from meta-GGA-based time-dependent density-functional theory (V. Nazarov).

Chairperon: E. K. U. Gross.

- 1600 1640 The derivative discontinuity in transport (S. Kurth).

- 1640 1720 Electron-ion entanglement and decoherence dynamics in trans-polyacetylene oligomers (H. Appel)

- 1720 1750 COFFEE BREAK

- 1750 1830 Intersubband spin plasmons in quantum wells: spin-orbit coupling and many-body effects (C. Ullrich).

- 1830 1910 Excitation gaps of finite-sized systems from Optimally-Tuned Range-Separated Hybrid Functionals (L. Kronik).

Friday, January 13th

Chairperson: A. Castro.

- 1600 1640 How to get what you want from optimal control (C. Koch).

- 1640 1720 Optimal control of many-electron systems and quantum revival (E. Räsänen).

- 1720 1800 Show me the light: Optimal Control with Strong Lasers (D. Kammerlander).

- 1800 1830 COFFEE BREAK

Chairperson: A. Rubio.

- 1830 1910 Attosecond time-resolved molecular dynamics (M. Vrakking).

- 1910 1950 Multichannel Tunneling in Strong Field Ionization (O. Smirnova).

- 1950 2030 Probing electron excitations with inelastic x-ray scattering spectroscopies (S. Huotari).

Sunday, January 15th

Chairperson: R. van Leeuwen.

- 0930 1010 Ab initio colors (and a few thoughts on functionals for spectroscopy) (S. Baroni).

- 1010 1050 Interaction effects in time-dependent quantum transport: TDDFT versus MBPT (G. Stefanucci).

- 1050 1130 TDDFT for Electron-Phonon Dynamics with Applications to Quantum Dots (O. Prezhdo).

- 1130 1200 COFFEE BREAK

Chairperson: F. Nogueira.

- 1200 1240 Nonadiabatic molecular dynamics coupled to time dependent external potentials: towards TDDFT-based local control (I. Tavernelli).

- 1240 1320 Progress at the interface of wave function and density functional theories (N. Gidopoulos).

Chairperson: S. Kurth

- 1530 1610 Towards inclusion of dissipation in TDDFT (E. Suraud).

- 1610 1650 Beyond the Hubbard model: a realistic description of metal-insulator transitions in transition-metal oxides (M. Gatti).

- 1650 1730 Plasmonics and non-local interactions from TDDFT: graphene and metal surfaces (T. Olsen).

- 1730 1750 COFFEE BREAK.

- 1750 1830 Photoinduced charge separation in light-harvesting supramolecular systems (C. A. Rozzi).

- 1830 1850 Exact factorization of the time-dependent electron-nuclear wave-function: A mixed quantum- classical studio (F. Agostini, poster prize award).

- 1850 1910 Derivative Discontinuities, Orbital Energies and Lifetimes in Density Functional Resonance Theory (D. Whitenack, poster prize award).

Monday, January 16th

Chairperson: N. Maitra

- 0900 0940 The interaction of ultrashort high-intensity x-ray pulses with dense atomic gases (N. Rohringer).

- 0940 1020 Studies of dipole-forbidden electronic excitations (A. Sakko).

- 1020 1050 COFFEE BREAK.

- 1130 1210 Bootstrap approximation for exchange-correlation kernel of TD-DFT (S. Sharma).

- 1130 1210 Role of discontinuities in linear response TDDFT (M. Hellgren).

- 1210 1220 FINAL REMARKS.

List of participants

1. ABEDI, Ali: Max-Planck-Institut for microstructure physics, Halle

2. AGOSTINI, Federica: Max Planck Institute of Microstructure Physics

3. APPEL, Heiko: Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

4. BAISHYA, Kopinjol: University of Illinois at Chicago

5. BARONI, Stefano: SISSA

6. BOTTI, Silvana: LSI, Ecole Polytechnique

7. BRUNETTO, Gustavo: UNICAMP

8. CASTAÑEDA MEDINA, Arcesio: Max-Planck Institute for Microstructure Physics

9. CASTRO, Alberto: BIFI, Universidad de Zaragoza

10. CERQUEIRA, Tiago: Universidade de Coimbra

11. CRAWFORD, Alison: ETSF Nano-Bio Spectroscopy Facility (Donostia-San Sebastin)

12. CUDAZZO, Pierluigi: ETSF San Sebastian

13. DAVIDE, Sangalli: MDM Lab. - IMM - CNR, Agrate Brianza (MI)

14. DAVYDOV, Arkady: Max-Planck-Institut fr Mikrostrukturphysik

15. DE GIOVANNINI, Umberto: University of the Basque Country

16. EICH, Florian: Max-Planck-Institute for Microstructure Physics

17. ESCARTÍN, Jos Mara: Laboratoire de Physique Thorique, U. Paul Sabatier Toulouse / CNRS

18. FARZANEHPOUR, Mehdi: Nano-bio Spectroscopy Group Univesidad del Pais Vasco UPV/EHU

19. FIOLHAIS, Carlos: University of Coimbra

20. FLICK, Johannes: Fritz-Haber-Institut der Max-Planck

21. FUKS, Johanna: Basque Country University

22. GAO, Xianlong: Zhejiang Normal University

23. GARCA RISUEÑO, Pablo: University of Zaragoza

24. GATTI, Matteo: UPV-EHU San Sebastian

25. GIDOPOULOS, Nikitas: ISIS-STFC, Rutherford Appleton Laboratory

26. GIESBERTZ, Klaas: University of Jyväskylä

27. GOERLING, Andreas: University of Erlangen-Nuremberg

28. GOYENOLA, Cecilia: Linkping University

29. GROSS, E.K.U: Max Plank Institute of Microstructure Physics

30. GUEORGUIEV, Gueorgui: Linkping University

31. HELBIG, Nicole: Forschungszentrum Juelich

32. HELLGREN, Maria: Max-Planck Institute for Microstructure Physics

33. HJORTH LARSEN, Ask: Center for Atomic-scale Materials Design. Technical University of Denmark

34. HUNG, Linda: Ecole Polytechnique

35. HUOTARI, Simo: University of Helsinki

36. IACOMINO, Amilcare: CSIC and Nano-Bio Spectroscopy Group Universidad del Pais Vasco UPV/EHU

37. JADIDI, Tayebeh: university of osnabrueck

38. JOKAR, Jeiran: Forschungszentrum Jülich

39. KAMMERLANDER, David: LPMCN Université Lyon 1 et CNRS

40. KARLSSON, Daniel: Lund University Institute of Physics

41. KIM, Youngseok: University of Illinois at Urbana and Champaign

42. KOCH, Christiane: University of Kassel

43. KRONIK, Leeor: Weizmann Institute of Science

44. KURTH, Stefan: Univ. del Pais Vasco, San Sebastian

45. LEIN, Manfred: Leibniz Universität Hannover

46. LEPINE, Franck: Laboratoire de Spectrométrie Ionique et Moléculaire, Universidad de Lyon

47. LEVY, Roi: Ben Gurion University

48. MAITRA, Neepa: Hunter College City University of New York

49. MANRIQUE, David Zsolt: Lancaster University

50. MARINI, Andrea: Phys. Dept. University of Rome "Tor Vergata"

51. MARQUES, Miguel: LPMCN, Université Lyon I

52. MATEO, David: Universitat de Barcelona

53. MAURER, Reinhard J.: Technical University Munich

54. MELO, Pedro: Universidade de Coimbra

55. MICCIARELLI, Marco: Universit'a degli studi di Napoli Federico Secondo

56. MIRTSCHINK, Andre: VU Amsterdam

57. MYRTA, Gruning: Centro de Fisica Computacional, Universidade de Coimbra

58. NAZAROV, Vladimir: Research Center for Applied Sciences, Academia Sinica

59. NOGUEIRA, Fernando: Centro de Fsica Computacional, Universidade de Coimbra

60. OLIVEIRA, Micael: Universidade de Coimbra

61. OLSEN, Thomas: CAMD, Techn. Univ. of Denmark

62. PENG, Degao: Duke University

63. POTOTZKY, Klaus: Max Planck Institute of Microstructure Physics, Halle (Saale)

64. PREZHDO, Oleg: University of Rochester

65. QUARTI, Claudio: Politecnico di Milano

66. RÄSÄNEN, Esa: University of Jyväskylä

67. RIBEIRO, Raphael: University of Minnesota

68. ROHRINGER, Nina: Max Planck Advanced Study Group, Hamburg

69. ROSA, Marta: Università di Modena e Reggio Emilia

70. ROZZI, Carlo Andrea: CNR - Istituto Nanoscienze

71. RUBIO, Angel: Universidad del Pais Vasco (EHU/UPV)

72. RUGGENTHALER, Michael: Nanoscience Center, Physics Department, University of Jyväsylä

73. SAKKO, Arto: Aalto University

74. SANDERS, Jacob: Harvard University

75. SAVARESE, Marika: University of Naples "Federico II"

76. SHARIFZADEH, Sahar: Lawrence Berkeley National Laboratory

77. SHARMA, Sangeeta: MPI Halle

78. SMIRNOVA, Olga: Max-Born Institut Berlin

79. SON, WON-JOON: North Carolina State University

80. SPONZA, Lorenzo: LSI - ETSF : Ecole Polytechnique

81. STEFANUCCI, Gianluca: Universitá di Roma Tor Vergata

82. STELLA, Lorenzo: Universidad del País Vasco,CSIC-UPV/EHU-MPC and DIPC

83. STRUBBE, David: University of California, Berkeley

84. SURAUD, Eric: LPT IRSAMC UNiv. P. Sabatier Toulouse 3

85. TAVERNELLI, Ivano: EPFL

86. TOKATLY, Ilya: Universidad del País Vasco UPV/EHU

87. ULLRICH, Carsten: University of Missouri

88. VAN LEEUWEN, Robert: University of Jyväskylä

89. VARSANO, Daniele: University of Rome

90. VOORA, Vamsee: University of Pittsburgh

91. VOROS, Marton: Budapest University of Technology and Economics

92. VRAKKING, Marc: Max Born Institute in Berlin

93. WALKENHORST, Jessica: Universidad del País Vasco

94. WANG, Lei: Institute for Theoretical Physics, ETH Zurich

95. WANKO, Marius: Universidad del Pais Vasco (EHU/UPV)

96. WHITENACK, Daniel: Purdue University

97. WOBBE, Milena: University College London

98. ZHANG, Qing: Department of Chemistry, the University of Hong Kong

2.1.2   Report on Workshop "Topological Insulators and Non-Perturbative Spin-Orbit Coupling"

CECAM-HQ-EPFL, Lausanne, Switzerland

January 9–11, 2012

CECAM, Psi-k, ESF

Oleg Yazyev (EPFL),
Joel Moore (UC Berkeley),
David Vanderbilt (Rutgers U)

http://www.cecam.org/workshop-564.html

REPORT

Scope of the workshop.– Topological insulators are the recently discovered materials that have a bulk electronic band gap, but also exhibit conducting surface states. These surface states, originating from the combination of strong spin-orbit interactions and time-reversal symmetry, exhibit a number of novel properties such as the chiral spin textures and topological protection from backscattering. Many materials have now joined the list of confirmed topological insulators. Related materials include quantum anomalous Hall insulators, topological superconductors, and Weyl semimentals.

The emerging field of topological insulators and related materials has now become one of the most rapidly developing areas of physics. This workshop is the first attempt to bring together active computational electronic structure researchers pioneering the field of topological insulators. The topics addressed by the speakers included methodological developments, first-principles investigation of novel physical properties of known topological insulators, and in silico discovery of new topological materials. In addition, several leading phenomenological theorists and experimentalists participated in the workshop.

General aspects and methodology.– The introductory talk given by Joel Moore (Berkeley) covered the theoretical basics of the field and highlighted a number of problems which can be solved by the computational physics community. David Vanderbilt (Rutgers) and Ivo Souza (San Sebastian) discussed the calculations of orbital magnetoelectric response. Raffaele Resta (Trieste) presented a local Chern number approach towards mapping topological order in real space. On the purely methodological side of the field, Andrea Dal Corso presented his new PAW methodology which naturally includes spin-orbit interactions. Hubert Ebert (Munich) addressed the problem of modeling ARPES spectra using the so-called one-step-model of photoemission that describes excitation, transport to the surface and the escape to the vacuum in a

coherent way. The problem of modeling electronic transport properties of topological insulators was addressed by Stephan Roche (Barcelona). Finally, the entertaining talk of Pekka Pyykkö (Helsinki) covering the role of spin-orbit interactions in heavy-element chemistry earned special attention.

Experiments.– The first session of the workshop included two experimental presentations. The first talk was given by Zahid Hasan (Princeton) who pioneered the field of 3D topological insulators. Hasan spoke both about the discovery of bulk topological insulators as well as about recent works on the Tl–Bi–S–Se system in which the topological phase transition can be achieved by tuning the chemical composition. This system was proposed as a platform for studying Weyl fermion physics. He also covered their studies of magnetically doped bismuth chalcogenide topological insulators. The second experimental talk was given by Marco Grioni (EPFL) who presented ARPES investigations of surface systems with strong spin-orbit interactions.

Bismuth chalcogenide topological insulators.– Bismuth chalcogenides (Bi_2Se_3, Bi_2Te_3 and Sb_2Te_3) are currently the most extensively investigated bulk topological insulators, both experimentally and theoretically. Several speakers focused specifically on these materials. Oleg Yazyev (EPFL) reported calculations of spin polarization of topologically protected charge carriers in these materials and proposed a spintronic device based on thin films of these materials capable of independent control of spin and charge transport. He also covered the recent advances in *GW* quasiparticle band structure calculations of these materials. Hyoung Joon Choi (Yonsei) reported calculations of the orbital angular momentum of the surface states and an investigation of the topologically protected states at the side surfaces of these materials. Jürgen Henk (Halle) presented the results of their calculation on Mn-doped Bi_2Te_3. Kyungwha Park (Virginia Tech) reported the results of first-principles calculations on interfaces between bismuth chalcogenides and silicon.

Novel topological insulators.– Predicting novel topological insulators *in silico* is currently the major direction of computational first-principles research in this field. Arun Bansil (Boston) and Binghai Yan (Bremen) covered their theoretical prediction of thallium-based topological insulators derived from bismuth chalcogenides, which was followed by experimental confirmation. Eugene Chulkov (San Sebastian) introduced a large family of bismuth-based topological insulators that contain group-IV elements. Stanislav Chadov (Mainz) presented an extensive search for topological insulators in the family of half-Heusler alloys. Many speakers presented even more dramatic departures from known topological insulators. For instance, Hai-Jun Zhang (Stanford) considered strongly correlated actinides while Zhong Fang (Beijing) spoke about Ag_2Te and $HgCr_2Se_4$.

Topological electronic phases in graphene.– An important part of the workshop concerned the 2D topological electronic phases on the honeycomb lattice. Motivated by the models introduced by Haldane and by Kane and Mele, a number of research groups aim at realizing these phases in practice. Jaroslav Fabian (Regensburg) presented their results on the origin of intrinsic spin-orbit gap in graphene and proposed a recipe towards enhancing spin-orbit coupling in graphene by means of hydrogen adatoms. Joaquin Fernandez-Rossier (IINL-Braga) considered the Kane-Mele model in the presence of electron-electron interaction introduced via the Hubbard term. Stefan Blügel (Jülich) discussed the possibility of realizing the quantum anomalous Hall effect

by depositing heavy-element magnetic adatoms on graphene.

Report on selected discussion.– Several speakers identified important issues that have to be addressed by the computational community. In particular, in his introductory talk Joel Moore (Berkeley) highlighted the problem of interfaces between topological insulators and other materials that can be addressed by means of first-principles computations. In particular, he mentioned the interfaces with magnetic materials and superconductors, which are important in the context of realizing Majorana fermion systems. A somewhat related aspect concerns the modeling of thermoelectric properties of topological insulators and their interfaces. The last very important problem mentioned in this talk points at the need to develop an approach for defining the relevant topological invariants in strongly correlated systems. Many participants have also stressed the need in accurate beyond-DFT methodologies adapted to the topological electronic phases. Precise treatment of band gaps and relative band alignments, the properties poorly reproduced by the standard DFT schemes, are critical for modeling the topologically non-trivial properties of these materials. Oleg Yazyev (EPFL) presented the results of GW calculations of bismuth chalcogenide topological insulators, which essentially bridge the known disagreements between experimental data and DFT calculations for these materials. However, developing a consistent methodology involving both spin-orbit interactions and quasiparticle self-energy corrections remains a challenge. Hyoung Joon Choi (Yonsei) has mentioned the need to develop an approach for calculating orbital moments of electronic states in topological materials.

The workshop has certainly attained its two most important objectives identified in the proposal. First, the workshop allowed active researchers who have started working in this rapidly developing field to report their recent works. Second, the workshop allowed for unimpeded interaction between the representatives of the computational physics community on one side and several leading experimentalists and phenomenological theorists on the other. The joint efforts allowed determining a number of important objectives for future research. It is worth mentioning that despite the apparent diversity of the topological insulators community (which is typically the case for any emerging field), all participants of our workshop freely communicated to each other. One of the objectives of our workshop was to develop a strategy for the computational search for new topological insulators. Although several talks dedicated to the search for novel materials were given during the workshop, the speakers reported using different approaches such as the high-throughput investigation of a large number of structurally related compounds or the compositional changes of known topological insulators. It is now clear that only having several different approaches rather than one unified strategy can ensure rapid progress in this direction. Another objective was related to the comparison of accuracy of various schemes for treating spin-orbit interactions in first-principles methodologies. The computations presented by the participants were making use of different DFT codes, which utilize different methods for calculating spin-orbit couplings. It turned out, however, that these methods tend to give very similar results. The main limitation in the predictive power of first-principles computations applied to topological insulators is thus density functional theory itself. Most of the participants have touched to some extent on the problem of determining topological phases from electronic structure calculations, which is the last objective of the workshop. A significant amount of progress is being made in this direction, but any conclusion would be preliminary at this point.

The topic of topological electronic phases enabled by spin-orbit interactions will certainly see a

number of computationally-oriented workshops and conferences in the near future. A specialized workshop on this topic is already scheduled to take place in Bremen (August 13–17, 2012). We recommend that a follow-up CECAM workshop with a scope similar to ours should be held again within two years. For any such workshop, we recommend that it is absolutely necessary to actively involve experimentalists and phenomenological theorists in such computational events. This often-overlooked condition is crucial for the development of any emerging field, including that of topological insulators.

WORKSHOP PROGRAMME

Day 1 - January 9, 2012

8:45 to 9:00 – Welcome

9:00 to 9:40 – Joel Moore
Topological Insulators: Overview and Need for Electronic Structure Theory

9:40 to 10:20 – Zahid Hasan
Surface States in Topological Insulators and Superconductors: Discovery and Recent Results

10:20 to 10:50 – Coffee Break

10:50 to 11:30 – Marco Grioni
Spin-Split States at Surfaces: the ARPES View

11:30 to 12:10 – Andrea Dal Corso
Projector Augmented-Wave Method with Spin-Orbit Coupling

12:10 to 12:30 – Hyungjun Lee
The Role of d-orbitals in the Spin-Orbit Induced Splitting in Noble Metal Surfaces

12:30 to 14:00 – Lunch

14:00 to 14:40 – Oleg Yazyev
Bismuth Chalcogenide Topological Insulators from First Principles: DFT and Beyond

14:40 to 15:20 – Zhong Fang
Topological Insulators and Topological Semimetals

15:20 to 15:50 – Coffee Break

15:50 to 16:30 – Arun Bansil
Electronic Structure and Modeling of Highly Resolved Spectroscopies in Topological Insulators and Other Complex Materials

16:30 to 16:50 – Jürgen Henk
Spin Texture and Magnetism in Pure and Mn-doped Bi_2Te_3

16:50 to 18:00 – Discussion and Poster Highlights

18:00 to 20:00 – Poster Session

Day 2 – January 10, 2012

9:00 to 9:40 – Warren Pickett
Impact of Spin-Orbit Coupling in Transition Metal Pnictides and Oxides

9:40 to 10:20 - Hyoung Joon Choi
Chiral Orbital-Angular-Momentum and Side-Surface Dirac Cone of Topological Insulators

10:20 to 10:50 – Coffee Break

10:50 to 11:30 – Stefan Blügel
Electrically Tunable Quantum Anomalous Hall Effect

11:30 to 12:10 – Joaquin Fernandez-Rossier
Magnetism at the Edges and Surfaces of Topological Insulators

12:10 to 12:30 – Jaroslav Fabian
Spin-Orbit Coupling in Graphene: from Monolayer to Graphite

12:30 to 14:00 – Lunch

14:00 to 14:40 – David Vanderbilt
Orbital Magnetoelectric Coupling and Surface Anomalous Hall Effect

14:40 to 15:20 – Ivo Souza
First-Principles Calculation of the Orbital Magnetoelectric Response

15:20 to 15:50 – Coffee Break

15:50 to 16:30 – Raffaele Resta
A "Topological Marker" in Coordinate Space

16:30 to 16:50 – Pekka Pyykkö
Relativistic Effects in Heavy-Element Chemistry and Physics

16:50 to 17:10 – Hubert Ebert
From Simple Rashba Physics to Topological Insulators: Actual Trends in ARPES

17:10 to 17:30 – Stephan Roche
Exploring Spin-Orbit Coupling Effects in Models of Topological Insulators

19:00 Dinner

Day 3 – January 11, 2012

9:00 to 9:40 – Gustav Bihlmayer

Exploring 2D Topological Insulators under Realistic Conditions by Density Functional Theory

9:40 to 10:20 – Eugene Chulkov
Buried Topological States in a Homological Series of Topological Insulators

10:20 to 10:50 – Coffee Break

10:50 to 11:30 – Stanislav Chadov
Tuning the Topology-Related Properties in Heusler Compounds. First-Principles Study

11:30 to 12:10 – Hai-Jun Zhang
Strongly Interacting Topological Insulators in Actinide Family

12:10 to 12:30 – Binghai Yan
Prediction of Topological Insulators in TlBiSe$_2$ Family of Chalcogenides

12:30 to 14:00 – Lunch

14:00 to 14:20 – George Japaridze
Magnetic Field Induced Quasi Helical Liquid State in a Disordered 1D Electron System with Strong Spin-Orbit Interaction

14:20 to 14:40 – Kyungwha Park
Interface-Induced Magnetism in Topological Insulators Bi$_2$Te$_3$ and Bi$_2$Se$_3$

14:40 to 15:00 – Closing word

LIST OF PARTICIPANTS

Organizers

Joel Moore (University of California at Berkeley, USA)
David Vanderbilt (Rutgers University, New Brunswick, USA)
Oleg Yazyev (Swiss Federal Institute of Technology Lausanne, Switzerland)

Invited speakers

Arun Bansil (Northeastern University, Boston, USA)
Gustav Bihlmayer (Research Center Julich, Germany)
Stefan Blügel (Research Center Julich, Germany)
Stanislav Chadov (University of Mainz, Germany)
Hyoung Joon Choi (Yonsei University, Seoul, South Korea)
Eugene Chulkov (University of the Basque Country, San Sebastián, Spain)
Andrea Dal Corso (SISSA and CNR-IOM-DEMOCRITOS, Trieste, Italy)
Zhong Fang (Chinese Academy of Sciences, Beijing, China)
Joaquin Fernandez-Rossier (IINL, Braga, Portugal)

Marco Grioni (Swiss Federal Institute of Technology Lausanne, Switzerland)

Zahid Hasan (Princeton University, USA)

Warren Pickett (University of California at Davis, USA)

Raffaele Resta (University of Trieste, Italy)

Ivo Souza (University of the Basque Country, San Sebastin, Spain)

Hai-Jun Zhang (Stanford University, USA)

Contributing participants

Amlaki Taher (University of Twente, Enschede, The Netherlands)

Autes Gabriel (Swiss Federal Institute of Technology Lausanne, Switzerland)

Raffaello Bianco (University of Trieste, Italy)

Alberto Crepaldi (Swiss Federal Institute of Technology Lausanne, Switzerland)

Pierre Delplace (University of Geneva, Switzerland)

Hubert Ebert (Ludwig Maximilians University, Munich, Germany)

Jaroslav Fabian (University of Regensburg, Germany)

Daniel Gosalbez Martinez (University of Alicante, Spain)

George Japaridze (Andronikashvili Institute of Physics, Tbilisi, Georgia)

Jad Halimeh (Ludwig Maximilian University, Munich, Germany)

Jinhee Han (Yonsei University, Seoul, South Korea)

Jürgen Henk (Max Planck Institute of Microstructure Physics, Halle, Germany)

Hyungjun Lee (Yonsei University, Seoul, South Korea)

Lukas Müchler (Johannes Gutenberg University, Mainz, Germany)

Masaki Noro (Tokyo Institute of Technology, Japan)

Kyungwha Park (Virginia Tech, Blacksburg, USA)

Pekka Pyykkö (University of Helsinki, Finland)

Stephan Roche (Autonomous University of Barcelona, Spain)

Ryuji Takahashi (Tokyo Institute of Technology, Japan)

Tineke Van den Berg (University of Aix-Marseille, France)

Naunidh Virk (Swiss Federal Institute of Technology Lausanne, Switzerland)

Xiaoping Wang (Chinese Academy of Sciences, Beijing, China)

Binghai Yan (University of Bremen, Germany)

Hongbin Zhang (Research Center Julich, Germany)

2.1.3 Report on Workshop "Perspectives and challenges of simulations of bio-materials interfaces"

University of Bremen, Germany

10-14 October 2011

Organizers: Prof. Dr. Thomas Frauenheim, University Bremen, BCCMS, Germany

Prof. Dr. Lucio Colombi Ciacchi, University of Bremen, Germany

Prof. Dr. Viola Vogel, ETH-Zuerich, Switzerland

Summary

The workshop Perspectives and challenges of simulations of bio-materials interfaces was held at the University of Bremen, Germany from October 10th to October 14th 2011. In total, 65 participants from Bulgaria, Germany, USA, UK, Canada, Denmark, Japan, Poland, Sweden, Italy, Iran, France, China and Switzerland attended the workshop.

The programme consisted of 29 invited lectures, one poster session and different social events allowing for scientific discussions and exchange. The lectures were scheduled to last 40 minutes, including 10 minutes discussion time. In addition to this extended time for discussion, the chairpersons were instructed to introduce the subject of the session and to actively participate in the discussion. This Gordon-conference-style was essential to guarantee a vivid discussion. The organizers ensured that well-established scientists acted as invited speakers and chairpersons.

Concerning the poster session, we accepted only 22 posters to allow for an intense exchange of ideas at each single poster. Here, we encouraged in particular the young scientists to ask questions. The participation of young researchers were supported by partly covering local accommodation costs.

Due to the compact organization and accommodation in one hotel only all participants had to stay together for the whole time of the conference, which additionally enforced the scientific discussion which was mandatory since scientists from various separated fields, i.e. advanced quantum chemistry, quantum Monte-Carlo, many-body perturbation theory, time-dependent DFT, etc. were attending the meeting to merge ideas and formulate a common goal for future method developments.

Scientific content and discussion

The physical/chemical behavior of hybrid bio-organic/inorganic interfaces in the focus of the workshop results from a delicate interplay between the electronic or mechanical properties of the inorganic phase and the surface bonding of biological molecules, which may undergo a drastic change of their structure and thus of their functionality upon interaction with the solid. Chemical reactions at the phase boundaries and other processes involving the transfer of electrons or the exchange of ions across the interface characterize uniquely the behavior of the composite material. Since such effects are not trivial to be analyzed with high resolution experiments or predicted a priori, computer modeling offers a viable way to investigate them on the basis of fundamental physical principles, thus complementing and expanding the information obtained by means of experimental techniques.

The investigation of phenomena at hybrid biomaterials interfaces poses so far unresolved challenges to accurate, atomistic computational methods, since it involves dealing with mutually interacting phenomena spanning multiple time and length scales and requiring different levels of precision. In the biological community, deciphering the physics of complex units from motor proteins to ribosomes, from membrane channels to DNA packaging in the cell nucleus or DNA-sequencing has become possible by the advent of many new technologies to analyze and manipulate molecular systems at highest precision. Combining high-resolution structural analysis with high-performance computing enabled furthermore to simulate how the intrinsic structural movements of biological nano-scale systems combined with their optical, electrical or mechanical properties control or regulate their functions. Also aided by high-performance computing, new functional hybrid-materials were designed, some of which were inspired by biological systems. Understanding life from its molecular foundation on a qualitative level, learning from it for technical applications and elucidating how the interactions between living structures interact and the technical world may stimulate novel routes for materials design has become a very attractive field of research these days.

To accomplish this goal successfully computational research request different methods from quantum and classical atomistic simulations through coarse grained techniques and further bottom upscale to finite element methods (FEM), and this is done traditionally in quite separate research communities.

Since the subject of the workshop is so interdisciplinary, also the background and scientific communities of the lecturers and participants were quite diverse. It was therefore the aim of the workshop to familiarize the participants with different subjects, to encourage interdisciplinary interactions, and to share experience of different research fields with one another. In this way, we managed to foster the exchange of ideas and methods, to highlight the most recent advances in experiments and computational method developments and applications, and hopefully stimulated new and fruitful collaborations and future projects across subject boundaries.

Assessment of the results and impact on future direction of the field

It became apparent from the presentations and the corresponding discussions that the modelling in each component of bio-materials interfaces is indeed very challenging. In some areas of the research field the methods and approaches have still not matured, so that intrinsic technical and conceptual problems persist. In the workshop the following key objectives have been achieve:

The main advantages and disadvantages of currently available modeling techniques for the specific case of simulations of biological/inorganic interfaces have been discussed and specified. The techniques considered comprise (but not are limited to): (i) QM: essentially DFT-based techniques for ground states configurational sampling, including Order-N techniques; (ii) MM: atomistic force fields and many-body potentials; (iii) QM/MM: hybrid quantum/classical with static QM zones; (iv) D-QM/MM: hybrid quantum classical with dynamically moving and evolving QM zones; (v) CG: coarse-grained; (vi) HCG hybrid coarse-grained coupling different levels of precision.

The workshop impressively demonstrated the already existing intense collaboration between experiment and theory being the basis for improving our understanding of fundamental interactions and the functional interplay at bio-materials interfaces. To foster this process even further invited overview talks by highly recognized experimentalists have been presented.

In the workshop future directions for method developments and improvements of existing techniques to address physical and chemical phenomena at bio-organic/inorganic interfaces have been discussed. This has set the basis for scientific collaborations between the participants in order to foster methodological advances with respect to the state of the art.

Invited Speaker

Professor Aleksei Aksimentiev	Urbana, (US)
Dr. Markus Buehler	Cambridge, (US)
Professor Helmut Cölfen	Konstanz, (DE)
Dr. Stefano Corni	Modena, (IT)
Dr. Gabor Csanyi	Cambridge, (UK)
Professor Gianaurelio Cuniberti	Dresden, (DE)
Professor Alessandro De Vita	London, (UK)
Professor Marcus Elstner	Karlsruhe, (DE)
Dr. Colin Freeman	Sheffield, (UK)
Dr. Frauke Graeter	Heidelberg, (DE)
Professor Hongbin Li	Vancouver, (CA)
Professor Lutz Mädler	Bremen, (DE)
Professor Siewert-Jan Marrink	Groningen, (NL)

Professor Marcus Müller	Göttingen, (DE)
Professor Roland Netz	Berlin, (DE)
Dr. Christine Peter	Mainz, (DE)
Professor Mark Rodger	Coventry, (UK)
Professor Sandro Scandolo	Trieste, (IT)
Mr. Julian Schneider	Bremen, (DE)
Professor Klaus Schulten	Urbana, (US)
Dr. Ralf Seidel	Dresden, (DE)
Dr. Chris-Kriton Skylaris	Southampton, (UK)
Dr. Laura Treccani	Bremen, (DE)
Professor Nico van der Vegt	Darmstadt, (DE)
Dr. Daniele Varsano	Rome, (IT)
Dr. Tiffany Walsh	Coventry, (KH)

Participants

Mrs. Anke Butenuth	Freiburg, (DE)
Dr. Leonardo De Maria	Bagsvaerd, (DK)
Mr. Tuan Do	Stuttgart, (DE)
Mr. Stephen Fox	Southampton, (UK)
Mrs. Wenke Friedrichs	Greifswald, (DE)
Mrs. Meike Gummich	Bremen, (DE)
Mr. Jens Kahlen	Mainz, (DE)
Dr. James Kermode	London, (UK)
Dr. Susan Köppen	Bremen, (DE)
Dr. Tomas Kubar	Karlsruhe, (DE)
Mr. Malte Launspach	Bremen, (DE)
Dr. Chunli Li	Darmstadt, (DE)
Mrs. Susanne Liese	Garching bei München, (DE)
Mrs. Elisa Londero	Göteborg, (SE)
Dr. Michael Maas	Bremen, (DE)
Dr. Faramarz Mehrnejad	Tabriz, (IR)
Mr. Robert Meißner	Bremen, (DE)
Mr. Niraj Modi	Bremen, (DE)
Mr. Letif Mones	Cambridge, (UK)
Dr. Gianpietro Moras	Freiburg, (DE)
Professor Pablo Ordejon	Bellaterra, (ES)
Dr. Amedeo Palma	Monterotondo S. , (IT)
Dr. Lionel Perrin	Toulouse, (FR)
Mrs. Hanna Rademaker	Bremen, (DE)
Dr. Francisco Rodriguez-Ropero	Darmstadt, (DE)
Mr. Andrzej Rzepiela	Freiburg, (DE)

Mrs. Svea Sauer	Bremen, (DE)
Mr. Lennart Schmidt	Garching bei München, (DE)
Dr. Jia-Wei Shen	Mainz, (DE)
Mr. Sineng Sun	Kaiserslautern, (DE)
Mrs. Maria Velinova	Sofia, (BG)
Mr. Yann von Hansen	Garching bei München, (DE)
Mr. First name Senbo Xiao	Heidelberg, (DE)
Mr. hijun Xiao	Heidelberg, (DE)
Mr. Chao Zhang	Juelich, (DE)
Mrs. Beifei Zhou	Heidelberg, (DE)

Program

Monday, October 10th 2011(Best Western Hotel Schaper Siedenburg)
18:00 22:00   Registration

Tuesday, October 11th 2011 (House of Science Downtown)
08:00-08:50   Registration
08:50-09:00   Opening and welcome, Lucio Colombi Ciacchi / Thomas Frauenheim

Session: Surfaces I
Chair: Viola Vogel
09:00-09:40   Klaus Schulten, University of Illinois at Urbana-Champaign, (USA)
                    Computer modeling in biotechnology, a partner in
                    development
09:40-10:20   Elisa Molinari University of Modena (Italy)
                    Protein specific adhesion on the gold surface in water by first
                    principle simulations
10:20-10:50   Coffee Break
10:50-11:30   Kurosch Rezwan, University of Bremen (Germany)
                    Antibacterial surface functionalisation
11:30-12:10   Alessandro de Vita, Kings College London (UK)
                    Supramolecular self-assembly at surfaces: from direct linkage to long range
                    interactions
12:10-14:00   Lunch Break

Session: Surfaces II
Chair: Lucio Colombi Ciacchi
14:00-14:40   Lutz Maedler, University of Bremen (Germany)
                    Controlled solubility of nanoparticles in physicological environment

14:40-15:20	Tiffany Walsh, The University of Warwick (UK)
	Atomistic simulations of the aqueous peptide-inorganic interface
15:20-15:50	**Coffee Break**
15:50-16:30	Stefano Corni, University of Modena (Italy)
	Protein-gold interfaces in water: insights from atomistic simulations
16:30-17:10	Markus Buehler, Massachusetts Institute of Technology, Cambridge (USA)
	Deformation and failure of protein materials in physiologically extreme conditions and disease
19:00-21:30	**Welcome Reception** (Bremen Town Hall)

Wednesday, October 12th 2011 (House of Science Downtown)

Session: Biomechanics
Chair: Frauke Graeter

08:30-09:10	Ralf Seidel, Dresden University of Technology (Germany)
	Nanomechanics of DNA origami
09:10-09:50	Viola Vogel, ETH Zurich (Switzerland)
	Mechanisms by which forces can switch the structure-function relationships of proteins

Session: Biomineralization
Chair: Pablo Ordejón

09:50-10:30	Helmut Coelfen, University of Konstanz (Germany)
	Polymers at mineral interfaces and nonclassical crystallization
10:30-11:00	**Coffee Break**
11:00-11:40	Colin Freeman, Sheffield University (UK)
	What goes on at the interface between molecules and minerals?
11:40-12:20	Barbara Aichmayer, Max-Planck-Institute of Colloids and Interfaces, Potsdam (Germany)
	Biogenic and biomimetic organic-inorganic hybrid particles: intracrystalline interfaces of calcite and inclusions of organic molecules
12:20-14:20	**Lunch Break**

Session: Computational methods and techniques I
Chair: Thomas Frauenheim

14:20-15:00	Chris-Kriton Skylaris, University of Southampton (UK)
	Biomolecular interactions from linear-scaling density functional theory calculations with thousands of atoms
15:00-15:40	Sandro Scandolo,The Abdus Salam, International Centre for theoretical Physics, Trieste (Italy)
	Ab-initio parametrized potentials for oxide interfaces
15:40-16:10	**Coffee Break**

16:10-16:50 P. Mark Rodger, The University of Warwick (UK)
Control and form of mineral nanoparticles

16:50-17:30 Gábor Csányi, University of Cambridge (UK)
Adaptive QM/MM simulations of chemical reactions in solution

19:00-23:00 **Conference Dinner** (Juergenshof Bremen)

Thursday, October 13th 2011 (BCCMS University Campus, IFWTAB-Building)

Session: Computational methods and techniques II
Chair: Fabrizio Cleri

08:30-09:10 Sievert-Jan Marrink, University Groningen (The Netherlands)
Hybrid simulations: combining atomistic and coarse-grained force fields using
virtual sites

09:10-09:50 Christine Peter, Max-Planck-Institute for Polymer Research, Mainz (Germany)
Development of multiscale simulation models for biological hybrid materials

09:50-10:30 Nico van der Vegt, Darmstadt University of Technology (Germany)
The conditional reversible work method for systematic coarse graining of complex
fluids

10:30-11:00 **Coffee Break**

11:00-11:40 Marcus Mueller, University of Goettingen (Germany)
Simulation of soft coarse-grained models for membranes

11:40-12:20 Luigi Delle Site, Max-Planck-Institute for Polymer Research Mainz (Germany)
Multiscale modeling and simulation of Liquids and Liquid-solid interfaces

12:20-14:20 **Lunch Break**

Session: Mechanical properties and folding
Chair: Gianaurelio Cuniberti

14:20-15:00 Frauke Graeter, Heidelberg Institute for Theoretical Studies (HITS) (Germany)
Mechanics of biomaterials: how proteins propagate forces

15:00-15:40 Hongbin Li, University of British Columbia, Vancouver (Canada)
Tuning the mechanical properties of proteins: from single molecule to biomaterials

15:40-16:20 Roland Netz, Technical University Munich (Germany)
Simulation approaches to friction in proteins and polymers

17:00-21:00 **Poster Session** & Buffet (Catering Service)

Friday, October 14th 2011 (BCCMS University Campus, IFWTAB-Building)

Session: Particle and charge transport
Chair: Nico van der Vegt

08:30-09:10 Daniele Varsano, University of Rome La Sapienza (Italy)
Multi - scale modeling of DNA derivatives

09:10-09:50	Giovanni Cuniberti, Dresden University of Technology (Germany)
	Charge migration and heat phenomena in biomolecular systems:
	two sides of the same medal?
09:50-10:20	**Coffee Break**
10:20-11:00	Alexei Aksimentiev, University of Illinois at Urbana-Champaign (USA)
	Modeling transport of biomolecules through synthetic nanochannels
11:00-11:40	Marcus Elstner, University of Karlsruhe (Germany)
	A coarse grained QM/MM approach for the description of hole transfer
	in DNA and proteins
12:00	**Departure**

2.1.4 Report on Workshop "Computational Condensed Matter Physics, Materials Science and Nanoscience from First-Principles"

Barcelona, Spain

January 12-14, 2012

Psi-k, ESF, ZCAM-CECAM, MICINN, Generalitat de Catalunya

P. Ordejón, J. Iniguez, M. Stengel and D. Sánchez-Portal

http://www.icmab.es/mini2012

Report

The workshop Computational Condensed Matter Physics, Materials Science and Nanoscience from First-Principles (mini2012) was held in Barcelona between the 12th and 14th of January 2012. This workshop was part of the series of International Workshops on Computational Physics and Materials Science: Total Energy and Force Methods, which take place at Triestes ICTP in odd years and at a different location in even years, the previous one having been in Shanghai in 2010. The workshop had around 90 participants, including invited speakers and members of the International Scientific Committee. We had very lively sessions covering the latest developments in first-principles methods, featuring quantum Monte Carlo techniques, new DFT functionals, and schemes for large scale simulations. The sessions focused on applications covered problems in magnetism and topological phases, electrochemistry and photocatalisys, electron dynamics, strong correlations, as well as miscellaneous topics in material science. The scientific quality of the talks and discussions was very high, and the same applies to the very crowded poster session that we had in the first evening. All in all, we believe the workshop succeeded in providing a broad view of the current state of the field, combined with some topics that are not so well-known to the community at large or to the regular attendees of the series. The activity was evaluated very positively by the International Scientific Committee and regular participants. This opinion is shared by the local organizers, as the workshop exceeded our expectations regarding both its scientific quality and the number and liveliness of the interactions it fostered.

Scientific Contents and Discussions

The scientific content of the workshop revolved around the latest developments in first-principles electronic structure theory, and more generally on the application of cutting-edge numerical methods in nanoscience, condensed matter physics and materials science. The workshop was organized in a number of invited oral sessions, where internationally recognized experts were called to present what we judged to be the most significant advances of the past few years. While the specific topics of each session were diversified to offer a reliable snapshot of the current status

of the field, there were some recurrent themes that characterized most of the oral presentations, namely: I) the reliable description of electron correlations, II) strategies to overcome the stringent time- and length-scale limitations of DFT, III) electron excitations, ultrafast dynamics and non-adiabatic processes; IV) magnetism and novel materials characterized by non-trivial band topologies. With the partial exception maybe of the last point (topological insulators are a class of materials where the most peculiar aspects of the physics can often be captured already at the single-particle level) these are the most outstanding and difficult challenges that the field is facing, and all the speakers did an excellent job at portraying the serious efforts that are being made worldwide, in order to make significant progress along these crucial methodological issues. Some of the sessions were more oriented towards applications, but nonetheless followed the traditional style of this series of conferences and workshops: dealing with systems or phenomena that are difficult to describe within standard density functional theory, and that require special care to cope, e.g. with quantum nuclear fluctuations in ice (M. Fernandez-Serra), or with the complexity of transition-metal oxides (J. Junquera, J. Kunes, T. Saha-Dasgupta). Most of the oral presentations generated lively discussions, demonstrating once more the novelty of the topics, which are not fully established within the community yet. Overall, we have the feeling that the theory sessions (e.g. on Quantum Monte Carlo, DFT Functionals, Electron Dynamics) were characterized by especially insightful debates, that we believe have produced a constructive and fruitful exchange of ideas between the participants. Alongside the oral sessions, there was also a poster session where all participants were encouraged to present their results. At the end of the first day, after so many oral talks, we were pleasantly surprised to see that the participants weren't tired or intimidated by the sheer amount of information they were exposed to, and got involved into many discussions, thus contributing to a lively and successful poster session. In the poster session we were also pleased to see a large participation of young scientists working locally in Barcelona, and more generally in Spain. One of the motivations behind the MINI series of workshops is to give a marked priority to participants coming from the Country where the workshop is hosted. Also from this point of view, we can say that the workshop fulfilled our aims, and more generally the guidelines of the International Steering committee. All in all, the scientific contents of the workshop were in line with the tradition of high quality and attention to the cutting-edge developments that has characterized the MINI series throughout the years. At the same time, we succeeded at keeping a balance between formal theory and applications, so that the sessions could appeal to a broader audience. The encouragingly warm response, both from the scientific committee and by the regular participants, clearly confirmed this success.

Programme

Thursday, January 12th, 2012

Morning

08:30 REGISTRATION

08:50 OPENING REMARKS

09:00-11:00 SESSION: Quantum Monte Carlo - Chair: Claudia Filippi (U. Twente)

Ali Alavi (University of Cambridge, UK)

Quantum Monte Carlo approach to the full CI problem: recent progress

Sandro Sorella (SISSA, Italy)

Efficient Resonance Valence Bond approach for electronic structure

Shiwei Zhang (College of William and Mary, USA)

Recent progress in electronic structure calculations by auxiliary-field quantum Monte Carlo

11:00-11:30 COFFEE BREAK

11:30-12:50 SESSION: Materials I - Chair: Nicola Marzari (EPFL)

Mariví Fernandez-Serra (Stony Brook University, USA)

Anomalous Nuclear Quantum Effects in Ice

Javier Junquera (Universidad de Cantabria, Spain)

Highly-confined spin-polarized two-dimensional electron gas in SrTiO3/SrRuO3

13:00-15:00 LUNCH BREAK

Afternoon

15:00-17:00 SESSION: Magnetism and Topological Phases - Chair: David Vanderbilt (Rutgers)

Stefan Blügel (Jülich Forschungszentrum, Germany)

Spin relaxation mechanisms in metallic systems: Resonance and anisotropy effects

Ivo Souza (Universidad del País Vasco, Spain)

Wannier-based description of orbital magnetic effects in ferromagnets

Oleg Yazyev (EPFL, Switzerland)

Bismuth chalcogenide topological insulators from first principles

17:00-19:00 COFFEE BREAK AND POSTER SESSION

Friday, January 13th, 2012

Morning

09:00-11:00 SESSION: New DFT Functionals - Chair: Lucia Reining (Ecole Polytechnique)

Sangeeta Sharma (Max Planck Halle, Germany)

Treatment of strongly correlated systems within the framework of reduced density matrix functional theory

Gustavo E. Scuseria (Rice University, USA)

Symmetry breaking and restoration in electronic structure theory

Andreas Savin (Universite Pierre et Marie Curie, France)

Coupling wave function with density functional calculations

11:00-11:30 COFFEE BREAK

11:30-12:50 SESSION: Electrochemistry and Photocatalysis - Chair: Shobhana Narasimhan (JNCASR)

Annabella Selloni (Princeton University, USA)

First principles simulations of materials and processes in photo- and electro-catalysis

Michiel Sprik (University of Cambridge, UK)

Reactivity of holes at the TiO2/water interface

13:00-15:00 LUNCH BREAK

Afternoon

15:00-17:00 SESSION: Electron Dynamics - Chair: Steven G. Louie (UC Berkeley)

Matteo Gatti (Universidad del País Vasco, Spain)

Design of effective kernels for spectroscopy: time-dependent current-density-functional theory

Fernando Martín (Universidad Autónoma de Madrid, Spain)

XUV/X-ray femto- and attosecond laser pulses for ultrafast electronic control in simple molecules: towards attochemistry?

Marco Schiró (Princeton University, USA)

Dynamics of Strongly Correlated Electrons out of Equilibrium

17:00-17:30 COFFEE BREAK

17:30-18:50 SESSION: Large Scale Simulations - Chair: Richard M. Martin (U. of Illinois)

Johannes Neugebauer (TU Braunschweig, Germany)

Subsystem-Based Time-Dependent Density-Functional Theory for Biomolecular Spectroscopy

Joost VandeVondele (University of Zürich, Switzerland)

Simulating large condensed phase systems with GGA and hybrid density functionals

Evening

20:30 CONFERENCE DINNER

Saturday, January 14th, 2012

Morning

09:00-11:00 SESSION: Materials II - Chair: Xingao Gong (Fudan U.)

Jörg Neugebauer (Max Planck Düsseldorf, Germany)
Fully ab initio determination of free energies: Application to modern high-strength steels

Cheol-Hwan Park (Massachusetts Institute of Technology, USA)
Many-body effects on the carrier dynamics of graphene

Ruben Perez (Universidad Autónoma de Madrid, Spain)
Probing nanostructures with forces and currents

11:00-11:30 COFFEE BREAK
11:30-12:50 SESSION: Strong Correlations - Chair: Eric Koch (Jlich)

Jan Kunes (Academy of Sciences, Czech Republic)
Thermal and doping effects in materials with competing multiplets

Tanusri Saha Dasgupta (SN Bose Centre for Basic Sciences, India)
A Realistic Theory of Correlated Oxides

12:50 CLOSING REMARKS
13:00-15:00 LUNCH

List of Participants

<u>Invited Speakers</u>

Andreas Savin, Universite Pierre et Marie Curie, France
Gustavo E. Scuseria, Rice University, USA
Sangeeta Sharma, Max Planck Halle, Germany
Ali Alavi, University of Cambridge, UK
Sandro Sorella, SISSA, Italy
Shiwei Zhang, College of William and Mary, USA
Stefan Blügel, Jülich Forschungszentrum, Germany
Ivo Souza, Universidad del Pas Vasco, Spain

Oleg Yazyev, EPFL, Switzerland

Jan Kunes, Academy of Sciences, Czech Republic

Tanusri Saha Dasgupta, SN Bose National Centre for Basic Sciences, India

Matteo Gatti, Universidad del Pais Vasco, Spain

Fernando Martin, Universidad Autonoma de Madrid, Spain

Marco Schiro, Princeton University, USA

Annabella Selloni, Princeton University, USA

Michiel Sprik, University of Cambridge, UK

Marivi Fernandez-Serra, Stony Brook University, USA

Javier Junquera, Universidad de Cantabria, Spain

Jorg Neugebauer, Max Planck Dusseldorf, Germany

Cheol Hwan Park, Massachusetts Institute of Technology, USA

Ruben Perez, Universidad Autonoma de Madrid, Spain

Johaness Neugebauer, Technical University Braunschweig, Germany

Joost VandeVondele, University of Zurich, Switzerland

Sientific Committee (attending the workshop)

Stefano Baroni, SISSA - Trieste, Italy

Claudia Filippi, Univ. Twente, Netherlands

Xingao Gong, Fudan University, China

Francesco Mauri, Univ. Pierre et Marie Curie,France

Erik Koch, Forschungszentrum Jülich, Germany

Steven G. Louie, Univ. California - Berkeley, USA

Richard M. Martin, University of Illinois, USA

Nicola Marzari, EPFL, Switzerland

Shobhana Narasimhan, JNCASR - Bangalore, India

Pablo Ordejón, CIN2, Spain

Lucia Reining, Ecole Polytechnique - Paris, France

David Vanderbilt, Rutgers University, USA

Organizing Committee (attending the workshop)

Pablo Ordejón, CIN2, Spain

Jorge Iñiguez, ICMAB, Spain

Massimilano Stengel, ICMAB, Spain

Daniel Sanchez-Portal, Centro de Física de Materiales, Spain

Other participants

Hideaki Sawada, Nippon Steel Corporation, JAPAN

Josep María Oliva, Instituto Rocasolano - CSIC, Spain

Alberto Marmodoro, Max-Planck-Institut fur Mikrostrukturphysik, Germany

Jose María Soler,Universidad Autnoma de Madrid, Spain

Peter Koval, Centro de Fisica de Materiales - Donostia, Spain

Nicolai A. Bogdanov, Inst. Theoretical Solid State Physics - Dresden, Germany

David Soriano, ICN, Spain

P. Andre Clayborne, University of Jyväskylä, Finland

Perla Wahnón, UPM, Spain

Diego Carrascal, Univ. Oviedo, Spain

Gian-Giacomo Asara, U. Rovira i Virgili, Spain

Christina Ebensperger, University Erlangen-Nrnberg, Germany

Michelle Fritz, U. Autonoma de Madrid, Spain

Julen Ibañez, Donostia Int. Physics Center, Spain

Jon Mikel Azpiroz, UPV/EHU, Spain

Elisa Jimenez, UPV/EHU, Spain

Andres Vega, Universidad De Valladolid, Spain

Lauro O. Paz Borbon, C. C. Catalysis - Chalmers, Sweden

Emilio Artacho, Nanogune, Spain

Yohanna Seminovki, UPM, Spain

Federico Iori, Universita Di Modena, Italy

Silvia Gallego, CSIC, Spain

Jae Kyung Chang, Sugkyunkwan University, Korea

Changhyun Yi, Sugkyunkwan University , Korea

Kjartan Thor Wikfeldt, University College London, UK

Nadia Binggeli, ICTP, Italy

Ju-Yong Kim, Sugkyunkwan University, Korea

Guillermo Roman-Perez, UAM, Spain

Kerstin Hummer, University Of Vienna, Austria

Gabriel Bester, Max Planck Inst., Germany

Idoia G. De Gurtubay, UPV/EHU, Spain

Jose Javier Plata, Universidad De Sevilla, Spain

Alberto García, ICMAB, Spain

Otto González, ICMAB, Spain

Mathias Ljungberg, ICMAB, Spain

Claudio Cazorla,ICMAB, Spain

Desanka Boskovic, CIN2, Spain

Jose Angel Silva, CIN2, Spain

Rafael Martínez, CIN2, Spain

Jacek Wojdel, ICMAB, Spain

Oswaldo Dieguez, ICMAB, Spain

Ricardo Rurali, ICMAB Spain

Alvaro, Miranda, ICMAB, Spain

Miguel Pruneda, CIN2, Spain

Carlos F. Sanz-Navarro, CIN2, Spain

Oscar Paz, ICMAB, Spain

2.1.5   Report on Workshop "Towards Reality in Nanoscale Materials V"

20th — 22nd February 2012
Levi, Lapland, Finland

`http://trnm.aalto.fi`

Organizers

Prof. Adam Foster
Department of Physics, Tampere University of Technology
Department of Applied Physics, Helsinki University of Technology

Dr. Teemu Hynninen
Department of Physics, Tampere University of Technology

Dr. Arkady Krasheninnikov
Department of Applied Physics, Helsinki University of Technology
Department of Physics, University of Helsinki

Prof. Risto Nieminen
Department of Applied Physics, Helsinki University of Technology

Prof. Kai Nordlund
Department of Physics, University of Helsinki

Report

The "Towards Reality in Nanoscale Materials V" workshop was the fifth in this annual series, although its origins can be traced to a meeting on wet chemical etching in Levi in 2006. Each year the focus changes, with one key topic supported by the regular themes of defects and multiscale modelling, all under the umbrella of nanoscale materials. This year's key topic was graphene interfaces, and we had many contributions on this including invited titles such as 'Graphene on metal surfaces', and 'Quantum confined electronic states in atomically well-defined graphene nanostructures'. Graphene, as expected, was a dominant topic at the meeting in general, but all the studies presented results in keeping with overall theme of *towards reality*.

In total we had over 80 participants at the meeting, continuing the trend of increased numbers every year. The programme was split into 9 invited talks, 25 contributed talks and 48 posters. Unlike the chaos caused last year by a Finnair strike, we lost only four participants at the last minute - two to the snow in Sweden, one to thieves in Italy and one mysteriously in Turkey. In general the meeting was very lively, with good discussion during the presentations, coffee breaks and posters. The switch of the meeting from the dark of December to February was a major bonus, as we actually had daytime rather than just twilight/night this year. As always the traditional Lappish dinner makes a good impression and those braving the cold afterwards were rewarded with the Northern lights (we are at 50% observation chance during the history of TRNM). Plans are already being made for TRNM VI in February 2013.

Programme

8:45	intro		
chair	*Rinke*	*Roche*	*Busse*
9:00	Artacho	Busse	Roche
9:40	Bøggild	Lehtinen	Ahlskog
10:00	Sob	Pietrucci	Makarova
10:20	Borlenghi	Booth	Carva
10:40	*coffee*	*coffee*	*coffee*
chair	*Pantelides*	*Perez*	*Artacho*
11:20	Watkins	Liljeroth	Pantelides
12:00	Barth	Ewels	Minato
12:20	Laasonen	Amara	Girard
12:40	Rayson	Kurasch	Uppstu
13:00	*lunch*	*lunch*	*lunch*
chair	*Liljeroth*		*Bøggild*
16:00	Rinke	*posters*	Sinnott
16:40	Susi	*16:00 – 18:00*	Mittendorfer
17:00	Standop		Perez
17:20	*coffee*		
chair	*Sinnott*		closing
18:00	Chamberlain	*dinner*	
18:20	Björk	*18:00 – 21:00*	
18:40	Philipp		

- Monday

  - Emilio Artacho: Origin of the 2DEG at oxide interfaces, relation with topology and with redox defects, and possible 1DEG

  - Peter Boggild: On chip synthesis and characterisation of nanostructures in-and outside TEM

  - Mojmir Sób: Ab initio study of mechanical and magnetic properties of Mn Pt compounds and nanocomposites

  - Simone Borlenghi: Electronic transport and magnetization dynamics in realistic devices:a multiscale approach

  - Matthew Watkins: Developing realistic models of interfaces from simulation

  - Clemens Barth: Two-dimensional growth of nanoclusters and molecules on Suzuki surfaces

  - Kari Laasonen: Reaction studies of Al-O clusters in water

  - Mark Rayson: Towards large-scale accurate Kohn-Sham DFT for the cost of tight-binding

  - Patrick Rinke: Towards a unified description of ground and excited state properties:RPA vs GW

  - Toma Susi: Core level binding energies of defected and functionalized graphene

  - Sebastian Standop: Spatial Analysis of Ion Beam induced Defects in Graphene

  - Thomas Chamberlain: Utilizing carbon nanotubes as nanoreactors

  - Jonas Björk: Molecular self-assembly of covalent nanostructures: Unraveling their formation mechanisms

  - Patrick Philipp: Simulation of defect formation and sputtering of Si(100) surface under low-energy oxygen bombardment using a reactive forcefield

- Tuesday

  - Carsten Busse:Graphene on metal surfaces

  - Ossi Lehtinen: Detailed structure and transformations of grain boundaries in graphene

  - Fabio Pietrucci: Graph theory meets ab-initio molecular dynamics: atomic structures and transformations at the nanoscale

  - Tim Booth: Discrete removal of carbon atoms by silver nanoparticles in suspended graphene

  - Peter Liljeroth: Quantum confined electronic states in atomically well-defined graphene nanostructures

  - Chris Ewels: Distorting graphene through mechanics and edge chemistry

  - Hakim Amara: Healing Mechanisms During The Growth of Carbon Nanotubes

  - Simon Kurasch: Graphene: An ideal substrate for TEM imaging and spectroscopy

- Wednesday

  - Stephan Roche: Transport Properties in Disordered Graphene: Effects of Atomic Hydrogen and Structural Defects

  - Markus Ahlskog: Size dependence of electronic transport in multiwalled carbonnanotubes

  - Tatiana Makarova: Edge states versus in-plane defects in graphite magnetism

  - Karel Carva: Defect controlled conductivity of graphene with vacancies and N impurities

  - Sokrates Pantelides:Topics in graphene

  - Taketoshi Minato: Mechanism of organic chemical reactions on nanoporous gold

  - Sylvain Girard: Coupling experiments and simulations for the radiation hardening of fiber optics: (I) Experimental results on canonical samples of pure and doped silica

  - Andreas Uppstu: Electronic transport in graphene-based structures: An effective cross section approach

  - Susan Sinnott: Charge Optimized Many-Body (COMB) Potentials for Interfacial Studies

  - Florian Mittendorfer: Graphene on Ni(111): Strong Interaction and Weak Adsorption

  - Ruben Perez: Probing nanostructures with forces and currents: From atomic-scale contrast on graphene and carbon nanotubes to heterofullerene synthesis with planar aromatic precursors

- Posters

  - Harriet Ahlgren: Ion irradiation induced defects in freestanding graphene: an atomistic simulation study

  - Hadi Arefi: A density functional theory study of functionalised silicon surfaces

  - Joseph Bamidele: The role of dispersion forces in the surface stability of oxidised Cu(110)

  - Vladimir Baturin: On heat capacity of superconducting nanoclusters

  - Carla Bittencourt: Imaging the carbon K near-edge polarization dependence of a few layer graphene by NEXAFS-TXM

  - Alexandra Carvalho: Alternative methods for doping nanocrystals and designing junction offsets

  - Vladimir Chashchikhin: DFT modeling of the interaction of small analyte molecules with a dye/silica receptor center

  - Filippo Federici: NC-AFM energy dissipation mechanisms

  - Filippo Federici: Atomic scale dynamics of frictional processes

  - Filippo Federici: Extreme sensitivity in potential characterization of an insulating step edge

- Yulia Filicheva: Theoretical investigation of the diamond films with implanted oxygen atoms

- Andris Gulans:From short-to long-range correlations in layered materials

- Eero Holmström: Atomic-scale processes set the critical limit for conventional ion beam thinning of Silamellae

- Semran Ipek: Structural and electronic properties of GaAs nanotubes

- Elisa Jimenez: Porous Solids Based on Endohedrally Doped CdS Nanoclusters

- Wonjae Kim: Nonlinear behavior of three terminal graphene junctions at room temperature

- Manana Koberidze: Molecular Dynamics Simulation of Aluminum Oxidation via Reactive Force Field

- Hannu-Pekka Komsa: Defects in electron irradiated transition metal dichalcogenides

- Jaap Kroes: Atomic oxygen chemisorption on a zig-zag carbon nanotube

- Sampo Kulju: Atomic scale characterization of alumina films grown on the MgAl2O4(100) surface

- Alexander Kvashnin: Ultrathin diamond nanofilms as possible two- dimensional insulator: electronic and elastic properties

- Dmitry Kvashnin: Transport properties of graphene-based branched nanoribbons.Theoretical study

- Katri Laaksonen: Optical properties of thermocromic VO2 nanoparticles

- Markus Langer: Imaging and energy dissipation mechanisms on metallic and insulating surfaces studied with AFM in pendulum geometry

- Nikita Marchenkov: Lattice parameter local determination for trigonal crystal systems using several coplanar X-ray reflections

- Taketoshi Minato: Atomic defects on titanium dioxide surface -Electronic structure and manipulation

- Riku Oja: d0 ferromagnetic interface between non-magnetic perovskites

- Ari Ojanpera: Ehrenfest molecular dynamics within the projector augmented- wave method: an ab initio insight on electronic stopping in nanostructures

- Alexander Pyymaki Perros: Interfacial characteristics of in situ CVD- seeded ALD oxide layers on graphene

- Filippo Pizzocchero: Direct density functional theory simulation of single sequential transmission electron microscope images

- Nicolas Richard: Coupling experiments and simulations for the radiation hardening of fiber optics: (II) Ab initio characterization of electronic and optical properties of defects in amorphous silica

- Juho Roukala: Magnetic resonance properties of Sc3C2@C80 in finite temperatures

- Arto Sakko: Time-dependent DFT approach for the dielectric response of nanoscale systems

- Karri Saloriutta: Resonant scattering in graphene: adsorbate fingerprints from ab initio calculations
- Nino Samadashvili:Nanoscale friction of ice
- Dmitri Schebarchov: Interplay of geometric and electronic structure in metalloid gallium clusters
- Martha Scheffler: Local tunneling spectroscopy of the Hydrogen-induced impurity state in quasi-freestanding graphene
- Gabriele Sclauzero: Corrugation-enhanced graphene reactivity boosting the graphene/SiC(0001) interface stability
- Anton Targonsky: New Method for Measuring Rocking Curves in X-Ray Diffractometry for time resolving observation of the crystal defects by Ultrasonic Modulation of the Lattice Parameter
- Stefan Taubert: Molecular dynamics simulations of initial stages of growth of nitrogen-doped carbon nanotubes
- Dmitry Terentyev: Modelling of radiation damage recovery in concentrated Fe-Cr alloys: a multi-scale modelling approach
- Lisa Katharina Tschammer: Nanomanipulation of Graphene using E- beam and Block Copolymers
- Jarkko Vahakangas: NMR parameter of chemically modified graphanes
- Ville Virkkala: Hybrid-Functional Study of Band-Structures of GaAsxNx and GaSbxNx Alloys
- Monika Vsianska: The effect of non-magnetic impurities on magnetic and cohesive properties of grain boundaries in nickel
- Valeria Zagaynova: Influence of boron doping on the defect-induced magnetism of oxygen-eroded graphite
- Martin Zeleny: First-principles investigation of the martensitic transformation in Co-doped Ni2MnGa shape memory alloys

Participants

Åhlgren	Harriet	harriet.ahlgren@helsinki.fi
Ahlskog	Markus	ahlskog@jyu.fi
Amara	Hakim	hakim.amara@onera.fr
Arefi	Hadi	hadi.arefi@tyndall.ie
Artacho	Emilio	e.artacho@nanogune.eu
Bamidele	Joseph	joseph.bamidele@kcl.ac.uk
Barth	Clemens	barth@cinam.univ-mrs.fr
Baturin	Vladimir	baturin@lpi.ru
Bittencourt	Carla	carla.bittencourt@ua.ac.be

Björk	Jonas	jonbj@ifm.liu.se
Bøggild	Peter	Peter.Boggild@nanotech.dtu.dk
Booth	Timothy	tim.booth@nanotech.dtu.dk
Borlenghi	Simone	simonebg@kth.se
Busse	Carsten	busse@ph2.uni-koeln.de
Carva	Karel	karel.carva@mff.cuni.cz
Carvalho	Alexandra	aicarvalho@ua.pt
Chamberlain	Thomas	thomas.chamberlain@nottingham.ac.uk
Chashchikhin	Vladimir	chashchikhin@gmail.com
Ewels	Chris	chris.ewels@cnrs-imn.fr
Federici	Filippo	filippo.federici@tut.fi
Filicheva	Yulia	jfilicheva@gmail.com
Foster	Adam	adam.foster@tut.fi
Girard	Sylvain	sylvain.girard@cea.fr
Gulans	Andris	agl@aalto.fi
Holmström	Eero	eero.holmstrom@helsinki.fi
Ipek	Semran	seipek@gyte.edu.tr
Kim	Wonjae	wonjae.kim@aalto.fi
Komsa	Hannu-Pekka	hannu-pekka.komsa@helsinki.fi
Krasheninnikov	Arkady	akrashen@acclab.helsinki.fi
Kroes	Jaap	jaap.kroes@epfl.ch
Kulju	Sampo	sampo.kulju@tut.fi
Kurasch	Simon	simon.kurasch@uni-ulm.de
Kvashnin	Alexander	AGKvashnin@gmail.com
Kvashnin	Dmitry	cvashnini@gmail.com
Laaksonen	Katri	katri.m.laaksonen@aalto.fi
Laasonen	Kari	kari.laasonen@aalto.fi
Langer	Markus	markus.langer@unibas.ch
Lehtinen	Ossi	ossi.lehtinen@gmail.com
Liljeroth	Peter	peter.liljeroth@aalto.fi
Makarova	Tatiana	tatiana.makarova@physics.umu.se
Marchenkov	Nikita	1414-88@mail.ru,
Minato	Taketoshi	minato@m.tohoku.ac.jp
Mittendorfer	Florian	Florian.Mittendorfer@tuwien.ac.at
Nieminen	Risto	risto.nieminen@aalto.fi
Öberg	Sven	sven.oberg@ltu.se
Oja	Riku	riku.oja@aalto.fi
Ojanper	Ari	ari.ojanpera@aalto.fi
Pantelides	Sokrates	pantelides@vanderbilt.edu
Perez	Ruben	ruben.perez@uam.es
Philipp	Patrick	philipp@lippmann.lu
Pietrucci	Fabio	fabio.pietrucci@epfl.ch
Pizzocchero	Filippo	filippo.pizzocchero@nanotech.dtu.dk

Pyymaki	Alexander	alexander.pyymaki.perros@aalto.fi
Rayson	Markus	mark.rayson@ltu.se
Richard	Nicolas	nicolas.richard@cea.fr
Rinke	Patrick	rinke@fhi-berlin.mpg.de
Roche	Stephan	stephan.roche@icn.cat
Roukala	Juho	juhorouk@paju.oulu.fi
Sakko	Arto	arto.sakko@aalto.fi
Saloriutta	Karri	ksaloriu@cc.hut.fi
Samadashvili	Nino	nino.samadashvili@tut.fi
Schebarchov	Dmitri	Dmitri.Schebarchov@gmail.com
Scheffler	Martha	m.scheffler@ifw-dresden.de
Sclauzero	Gabriele	gabriele.sclauzero@epfl.ch
Sinnott	Susan	ssinn@mse.ufl.edu
Sob	Mojmir	mojmir@ipm.cz
Standop	Sebastian	standop@ph2.uni-koeln.de
Susi	Toma	toma.susi@aalto.fi
Targonsky	Anton	tantva@ya.ru
Taubert	Stefan	stefan.taubert@aalto.fi
Terentyev	Dmitry	dterenty@sckcen.be
Tschammer	Lisa	lkats@nanotech.dtu.dk
Uppstu	Andreas	christer.uppstu@aalto.fi
Vähäkangas	Jarkko	jarkko.vahakangas@oulu.fi
Virkkala	Ville	ville.virkkala@aalto.fi
Vsianska	Monika	230038@mail.muni.cz
Watkins	Matt	matthew.watkins@ucl.ac.uk
Zagaynova	Valeria	valeria.zagainova@physics.umu.se
Zeleny	Martin	martin.zeleny@aalto.fi

2.2 Reports on Collaborative Visits

2.2.1 Report on a Short Collaborative Visit of Viktor Ivády (Hungary) to Sweden

Accurate electronic structure calculation on transition metal defects in silicon carbide polytypes

Viktor Ivády

Wigner Research Centre for Physics,

Hungarian Academy of Sciences,

PO Box 49, H-1525 Budapest, Hungary

Abstract

Relatively little is known about the complexty of point defects that may appear in different polytypes of a given crystal, which significantly influence their electrical and optical properties. Silicon carbide (SiC) is a prototype material for polytypism and many forms can be relatively easily fabricated.

SiC polytypes have a wide band gap where the electronic and optical properties can be significantly altered by introducing impurities. As a PhD student I have started to conduct research on characterizing transition metal defects in different polytypes of SiC by means of *ab initio* supercell calculations with the help of my supervisor. We collaborate tightly with the experimental group led by Professor Erik Janzén at Linköping University in Sweden. We run the *ab initio* codes at the supercomputer center installed at that campus.

The key issue in characterization of the metal impurities in SiC is to accurately calculate the electronic structure of these point defects which is a challenging task because of the strongly correlated d electrons together with the ordinary sp^3 bonds present in the system. So far I have applied a recently developed hybrid density functional in order to circumvent the well-known band gap error of (semi)local density functionals. My key results about the relative stabilities of different transition metal defects have been recently published in a Physical Review Letters paper. In order to calculate the charge transition levels and photoluminescence signals found by Janzén's group I would like to test different functionals beyond the applied hybrid density functional in order to be able to accurately predict the charge transition levels of selected transition metal defects. In addition, the optical signals would be determined by many-body perturbation theory based on the accurate ground state calculation.

The purpose of this short visit is to have an intense personal discussion with the experimental

group at Linköping University in order to select the appropriate detected centers for these important case studies and to optimize the codes installed on the supercomputers at Linköping University mediated by the experts at the supercomputer center for efficient large scale calculations.

Report

The visit was successful and especially beneficial for me. Through the personal discussion with both experimental and theoretical researchers I could see different aspects of the issues of transitional metal related defects in SiC, and they exposed some very important questions. During some visit in the laboratories I could acquire profitable knowledge about experimental techniques. Thanks for this trip I could make progress in finding the appropriate theoretical description of these very important defects.

During the two weeks I was invited for two workshops where I could see the operation of a well organized research group was optimizing their present studies to achieve long-term purposes. Furthermore I could see the tight cooperation between experimental group and theoretical group at these meetings. I was invited for laboratory visits three times when the experimental issues of sample growing and measuring were introduced to me. I was an attendee at the seminar of an theoretical group is leaded by prof. Igor Abrikosov and I had a presentation about my previous work which was published in Physical Review Letters. The most profitable activities were the numerous discussions with experimental as wall as theoretical researchers. I had meetings with researchers from prof. Erik Janzén's group like Nguyen Tien Son, Ivan Ivanov and Andreas Gällström. We tried to find out the explanations of their recent experimental observations based on the results from our calculations. About theoretical questions I had a discussion with prof. Igor Abrikosov which was very useful for my present study.

The projected publications are related to the visit in Sweden: I carried out some calculations for Andreas Gällström about vanadium related defects in SiC to justify his assumptions which may appear in a joint publication. I had an accepted presentation at DPG conference in Berlin with the title: Accurate electronic structure calculation on transition metal defects in SiC by HSE06+U functional. With the same topic we are planning to write an article which we would like to publish in Physical Review Letters.

We want to continue this fruitful collaboration between the Hungarian group and the Swedish groups.

2.2.2 Report on a Short Collaborative Visit of G.Stefanucci (Rome) to Stefan Kurth (San Sebastian)

Scientific Report

Dr. Gianluca Stefanucci

Prof. Stefan Kurth and myself recently published a paper [Phys. Rev. Lett. 107, 216401 (2011)] on the possibility of describing the conductance of the Anderson model in the Kondo regime using Time-Dependent (TD) Density Functional Theory (DFT). We showed that static DFT combined with the Landauer formula exactly reproduces the Kondo plateau at zero temperature provided that the exchange-correlation (xc) potential exhibits a derivative discontinuity. We further showed that at temperatures above the Kondo temperature the very same approach fails dramatically and we traced back the problem to the absence of dynamical xc corrections. The purpose of the visit was to understand how to estimate these corrections so as to suppress the Kondo peak without suppressing the Coulomb blockade side peaks. Even though the research is, at present, limited to rather simple model systems we believe that the qualitative features of the TDDFT xc potential are present also in more realistic situations and can help to shed some light on the current discrepancies between theory and experiments.

In order to estimate the dynamical xc corrections we calculated the TDDFT and the exact density response function by exact diagonalization. These response functions are indeed the basic quantities according to the results derived in Phys. Rev. Lett. 107, 216401 (2011). We then inserted the xc correction into the exact TDDFT formula and study the dependence on temperature, coupling to the leads, Coulomb repulsion U and on site energy at the impurity. The results revealed a strong connection between the TDDFT kernel with space arguments on the leads and the TDDFT kernel with arguments on the impurity. We derived some preliminary results which justify this finding. We also observed that dynamical xc corrections calculated as described above do indeed suppress the Kondo peak. The basic ideas developed during the visit need, however, to be further studied and checked.

We believe that the progress we made during this week holds promise for an important result in the TDDFT approach to quantum transport. It is therefore our intention to continue the collaboration possibly with other short visits.

3 General Workshop/Conference Announcements

3.1 International Workshop on Quantum Monte Carlo in the Apuan Alps VII

Apuan Alps Centre for Physics @ TTI
Vallico Sotto, Tuscany, Italy

Sat 28th July - Sat 4th August 2012

`www.vallico.net/tti/tti.html`

A4 POSTER:
`www.tcm.phy.cam.ac.uk/~mdt26/tti_c_poster_2012.png`

Continuing the series of alternative and very informal meetings at this venue, the Cambridge University Theory of Condensed Matter group is organizing a seventh International Workshop to discuss the development and application of the continuum quantum Monte Carlo method and related topics in condensed matter physics and quantum chemistry. The conference will take place in our 15th Century monastery in the mediaeval high mountain village of Vallico Sotto (in the Tuscan Apuan Alps near the beautiful Italian city of Lucca).

For many types of problem the accuracy of quantum Monte Carlo is much better than that of the more widely-used density functional theory, and its scaling with number of atoms is much more favourable than that of high-level quantum chemistry. Unlike most other methods, it is also fully capable of exploiting the full power of the largest computers in the world (e.g. the Cambridge CASINO code has been shown to have almost perfect parallel scaling to over 100,000 CPU cores). All topics related to applications of QMC and/or theory and algorithm development will be welcome, though it is expected that broader topics in the general area will also be discussed.

The normal format for these events involves formal presentations being restricted to the mornings, with the afternoons left free for relaxed discussion and participation in activities. For the young and vigorous, we organize mountain walks, caving and other healthy outdoor exercise, whilst the unfit and elderly might enjoy artistic tours, city visits, and gentle country strolls, with all participants reuniting in the evening for excellent Tuscan dinners in local restaurants. The monastery is a unique venue where the community spirit and magnificent location have inspired memorable meetings in the past.

This year's workshop will involve up to 50 people, all accommodated on site and in the village. Many speakers will be specifically invited, but anyone who feels that they have something to contribute and who wishes to attend the event is most welcome to contact the organizer (Mike

Towler: mdt26 at cam.ac.uk) for further details. There is no charge either for attendance at the conference or accommodation. A provisional programmme is available at the above website.

FURTHER DETAILS/PHOTOGRAPHS/MATERIAL FROM PREVIOUS WORKSHOPS ACCESSIBLE ON TTI WEB PAGE - CLICK THE 'PUBLIC EVENTS' LINK.

3.2 Summer School on Quantum Monte Carlo and the CASINO program VII

Apuan Alps Centre for Physics @ TTI
Vallico Sotto, Tuscany, Italy

Sun 5th - Sun 12th August 2012

`www.vallico.net/tti/tti.html`

A4 POSTER:
`www.tcm.phy.cam.ac.uk/~mdt26/poster.png`

The seventh international summer school in the series 'Quantum Monte Carlo and the CASINO program' will take place during August 2012 at the TTI monastery in the Tuscan Apuan Alps in Italy, organized and hosted by members (and ex-members) of Cambridge University physics department's Theory of Condensed Matter Group. The aim of the school is to give students a thorough introduction to quantum Monte Carlo as a method for performing high-quality calculations of the electronic structure of atoms, molecules, and materials. The course is designed for young quantum chemists or theoretical physicists who have no previous experience with this technique, though people at any stage of their career who are interested are welcome to take part.

The monastery is a unique venue where the community spirit and magnificent location have inspired memorable workshops in the past. It is a delightful 15th century building incorporating an ancient church, and is situated in the isolated but spectacular setting of the Tuscan mountain village of Vallico Sotto. The church is fully equipped with relevant presentation and computer technology, and all accommodation is on-site. As with all events at the Institute, formal lectures are restricted to the mornings, and participants are given the freedom and space to think and to contemplate and discuss the issues at hand. In addition to hands-on exercises, a programme of healthy recreational activities such as mountain walks will be organized in the afternoons, and it is hoped that by following this strict regime, together with breathing the clean air of the Apuan Alps and by preparing and sampling fine Tuscan cuisine, the participant will be able to return home mentally and physically refreshed as well as better informed.

Describing the complex behaviour of materials at the atomic level requires a sophisticated de-

scription of the correlated motion of the electrons. Quantum Monte Carlo (QMC) is an increasingly popular and explicitly many-body method with the unusual capability of yielding highly accurate results whilst also exhibiting a very favourable scaling of computational cost with system size, and perfect parallel scaling up to (at least) hundreds of thousands of computer cores. Over the last twenty years, the Cambridge group have been researching QMC methods and we have created a powerful, general computer program - CASINO - to carry out the calculations. The school will focus both on the basic theory of QMC and on more advanced practical techniques, and will include a thorough introduction to the CASINO program. A background in density functional theory or similar - though not essential - is normally thought to be useful.

Quantum Monte Carlo and the scaling behaviour of CASINO on the fastest computers in the world were the subject of one of last year's Psi-k 'Scientific Highlight of the Month' review articles. See '*Petascale computing opens new vistas for quantum Monte Carlo*' by M.J.Gillan, M.D. Towler and D.Alfè, available for download here:

`www.tcm.phy.cam.ac.uk/∼mdt26/papers/petascale_psik.pdf`

Instructors at the school will include the main authors of the CASINO program, Dr. Mike Towler (Cambridge/UCL), Dr. Neil Drummond (Lancaster) and Dr. Pablo Lopez Rios (Cambridge).

Participants would normally need to book a flight to Pisa airport from where onward transportation will be arranged (though other destinations are possible). Details of previous schools - including photographs - are available under the PUBLIC EVENTS link on the TTI web site.

Those interested should email Mike Towler (mdt26 at cam.ac.uk) for registration and further details.

4 General Job Announcements

Postdoctoral Position on Ab-Initio Thermal Transport
Cornell University, Ithaca, NY, USA

We currently have a post-doc opening available at the Cornell Nanoscale Facility working with Dr. Derek Stewart on ab-initio thermal transport in low thermal conductivity materials with applications for thermoelectrics. The successful candidate will be working on a research project in first principles thermal transport that benefits from a strong international collaboration with research groups at Cornell (Dr. Derek Stewart), Boston College (Prof. David Broido), and CEA-Grenoble (Dr. Natalio Mingo). We have developed a new first principles approach for thermal conductivity that has demonstrated excellent agreement with measured high thermal conductivity materials like silicon, germanium, and diamond. We are now generalizing this approach to study thermoelectric materials. Systems under consideration will include lead chalcogenides, I-V-VI semiconductors, and nanoparticle in alloy-structures. The post-doc will be using both density functional perturbation theory and real space techniques to calculate harmonic and anharmonic interatomic force constants in materials. The post-doc will also gain experience modeling thermal transport using both Greens function approaches and a self-consistent Boltzmann transport approach.

The candidate should have a PhD in Physics, Chemistry, or Material Science with a strong background in theoretical solid-state physics. Prior experience using density functional approaches to model materials and expertise working with common programming languages (Fortran, C/C++, etc) is highly desirable. Experience modeling the phonon properties of materials using ab-initio techniques will be an important asset. The successful candidate should also have strong communication skills. The position is available for one year with a possible extension to two years. Review of the applications will beginning immediately and continue until the position is filled. Informal enquiries on the position are welcome; please send an email to Dr. Derek Stewart (derek.stewart@cornell.edu). For the full application, please send a cover letter, curriculum vitae, including a list of recent publications, and the name, email address and affiliation of three references to derek.stewart@cornell.edu.

The full call can be found at:

`http://sites.google.com/site/dft4nano/positions`

5   Abstracts

Non-adiabatic effects during the dissociative adsorption of O_2 at Ag(111)? A first-principles divide and conquer study

Itziar Goikoetxea[1], Juan Beltrán[2], Jörg Meyer[2,3],
J. Iñaki Juaristi[4,1,5], Maite Alducin[1,5], and Karsten Reuter[2,3]
[1] *Centro de Física de Materiales CFM/MPC (CSIC-UPV/EHU)*
San Sebastián, Spain
[2] *Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4-6, D-14195 Berlin, Germany
[3] *Department Chemie, Technische Universität München,*
Lichtenbergstr. 4, D-85747 Garching, Germany
[4] *Departamento de Física de Materiales, Facultad de Químicas,*
UPV/EHU, San Sebastián, Spain
[5] *Donostia International Physics Center DIPC, San Sebastián, Spain*

Abstract

We study the gas-surface dynamics of O_2 at Ag(111) with the particular objective to unravel whether electronic non-adiabatic effects are contributing to the experimentally established inertness of the surface with respect to oxygen uptake. We employ a first-principles divide and conquer approach based on an extensive density-functional theory mapping of the adiabatic potential energy surface (PES) along the six O_2 molecular degrees of freedom. Neural networks are subsequently used to interpolate this grid data to a continuous representation. The low computational cost with which forces are available from this PES representation allows then for a sufficiently large number of molecular dynamics trajectories to quantitatively determine the very low initial dissociative sticking coefficient at this surface. Already these adiabatic calculations yield dissociation probabilities close to the scattered experimental data. Our analysis shows that this low reactivity is governed by large energy barriers in excess of 1.1 eV very close to the surface. Unfortunately, these adiabatic PES characteristics render the dissociative sticking a rather insensitive quantity with respect to a potential spin or charge non-adiabaticity in the O_2-Ag(111) interaction. We correspondingly attribute the remaining deviations between the computed and measured dissociation probabilities primarily to unresolved experimental issues with respect to surface imperfections.

Magnetic structure of noncentrosymmetric perovskites $PbVO_3$ and $BiCoO_3$: Theoretical analysis

I. V. Solovyev

National Institute for Materials Science,
1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan

Abstract

It is well known that if a crystal structure has no inversion symmetry, it may allow for Dzyaloshinskii-Moriya magnetic interactions, operating between different crystallographic unit cells, which in turn should lead to the formation of long-periodic spin-spiral structures. Such a behavior is anticipated for two simple perovskites $PbVO_3$ and $BiCoO_3$, crystallizing in the noncentrosymmetric tetragonal *P4mm* structure. Nevertheless, we argue that in reality $PbVO_3$ and $BiCoO_3$ should behave very differently. Due to the fundamental Kramers degeneracy for the odd-electron systems, $PbVO_3$ has no single-ion anisotropy. Therefore, the ground state of $PbVO_3$ will be indeed the spin spiral with the period of about one hundred unit cells. However, the even-electron $BiCoO_3$ has a large single-ion anisotropy, which locks this system in the collinear easy-axis C-type antiferromagnetic ground state. Our theoretical analysis is based on the low-energy model, derived from the first-principles electronic structure calculations.

Toward Low-Temperature Dehydrogenation Catalysis: Isophorone Adsorbed on Pd(111)

Wei Liu, Aditya Savara, Xinguo Ren,
Wiebke Ludwig, Karl-Heinz Dostert, Swetlana Schauermann,
Alexandre Tkatchenko, Hans-Joachim Freund, and Matthias Scheffler
Fritz-Haber-Institut der Max-Planck-Gesellschaft,
Faradayweg 4–6, 14195 Berlin, Germany

Abstract

Adsorbate geometry and reaction dynamics play essential roles in catalytic processes at surfaces. Here we present a theoretical and experimental study for a model functional organic/metal interface: isophorone ($C_9H_{14}O$) adsorbed on the Pd(111) surface. Density functional theory calculations with the PerdewBurkeErnzerhoff (PBE) functional including van der Waals (vdW) interactions, in combination with infrared spectroscopy and temperature-programmed desorption (TPD) experiments, reveal the reaction pathway between the weakly chemisorbed reactant ($C_9H_{14}O$) and the strongly chemisorbed product ($C_9H_{10}O$), which occurs by the cleavage of four CH bonds below 250 K. Analysis of the TPD spectrum is consistent with the relatively small magnitude of the activation barrier derived from PBE+vdW calculations, demonstrating the feasibility of low-temperature dehydrogenation.

Random-phase approximation and its applications in computational chemistry and materials science

Xinguo Ren[1], Patrick Rinke[1], Christian Joas[1,2], and Matthias Scheffler[1]

[1]*Fritz-Haber-Institut der Max-Planck-Gesellschaft,*
Faradayweg 4–6, 14195 Berlin, Germany
[2]*Max-Planck-Institut für Wissenschaftsgeschichte,*
Boltzmannstr. 22, 14195, Berlin, Germany

Abstract

The random-phase approximation (RPA) as an approach for computing the electronic correlation energy is reviewed. After a brief account of its basic concept and historical development, the paper is devoted to the theoretical formulations of RPA, and its applications to realistic systems. With several illustrating applications, we discuss the implications of RPA for computational chemistry and materials science. The computational cost of RPA is also addressed which is critical for its widespread use in future applications. In addition, directions of further development and current correction schemes going beyond RPA will be discussed.

Contact person: Xinguo Ren (ren@fhi-berlin.mpg.de)

Molecular-level understanding of WGS and reverse WGS reactions on Rh through hierarchical multiscale approach

Matteo Maestri[1,2] and Karsten Reuter[2]

[1] *Laboratory of Catalysis and Catalytic, Dipartimento di Energia, Politecnico di Milano, Piazza Leonardo da Vinci 32,I-20133 Milano,Italy*

[2] *Department Chemie, Technische Universität München, Lichtenbergstr. 4, D-85747 Garching, Germany*

Abstract

Hierarchically combining semi-empirical methods and first-principles calculations we gain a novel and noteworthy picture of the molecular-level mechanisms that govern the waterCgas-shift(WGS)and reverse waterCgas-shift (r-WGS) reactions on Rh catalysts. Central to this picture is that the WGS and r-WGS follow two different dominant reaction mechanisms: WGS proceeds according to a carboxyl (COOH) mechanism,whereas r-WGS proceeds according to a redox (CO2-COtO) mechanism. The obtained results furthermore underscore the danger of common first-principles analyses that focus on a priori selected dominant-paths. Not restricted to such bias,our herein proposed hierarchical approach thus constitutes a promising avenue to properly transport and incorporate the ab initio predictive-quality to a new level of system complexity.

Contact person: Matteo Maestri, matteo.maestri@polimi.it

TDDFT in massively parallel computer architectures: the OCTOPUS project

Xavier Andrade[1][*], Joseba Alberdi-Rodriguez[2,3], David A. Strubbe[4], Micael J. T. Oliveira[5], Fernando Nogueira[5], Alberto Castro[6], Javier Muguerza[3], Agustin Arruabarrena[3], Steven G. Louie[4], Alán Aspuru-Guzik[1], Angel Rubio[2,7] and Miguel A. L. Marques[8]

[1] Department of Chemistry and Chemical Biology, Harvard University, 12 Oxford Street, Cambridge, MA 02138, USA

[2] Nano-Bio Spectroscopy Group and ETSF Scientific Development Centre, Departamento de Física de Materiales, Centro de Física de Materiales CSIC-UPV/EHU and DIPC, University of the Basque Country UPV/EHU, Av. Tolosa 72, 20018 Donostia/San Sebastián, Spain

[3] Deparment of Computer Architecture and Technology, University of the Basque Country UPV/EHU, M. Lardizabal 1, 20018 Donostia/San Sebastián, Spain

[4] Department of Physics, University of California, 366 LeConte Hall MC 7300, Berkeley, California 94720, USA and Materials Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, California 94720, USA

[5] Center for Computational Physics, University of Coimbra, Rua Larga, 3 004-516 Coimbra, Portugal

[6] Institute for Biocomputation and Physics of Complex Systems (BIFI), Zaragoza Center for Advanced Modelling (ZCAM), University of Zaragoza, Spain

[7] Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

[8] Université de Lyon, F-69000 Lyon, France and LPMCN, CNRS, UMR 5586, Université Lyon 1, F-69622 Villeurbanne, France

Abstract

OCTOPUS is a general-purpose density-functional theory (DFT) code, with a particular emphasis on the time-dependent version of DFT (TDDFT). In this article we present the ongoing efforts for the parallelisation of OCTOPUS. We focus on the real-time variant of TDDFT, where the time-dependent Kohn-Sham equations are directly propagated in time. This approach has a great potential for execution in massively parallel systems such as modern supercomputers with thousands of processors and graphics processing units (GPUs).

For harvesting the potential of conventional supercomputers, the main strategy is a multi-level parallelisation scheme that combines the inherent scalability of real-time TDDFT with a real-space grid domain-partitioning approach. A scalable Poisson solver is critical for the efficiency of this scheme. For GPUs, we show how using blocks of Kohn-Sham states

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[*]xavier@tddft.org

provides the required level of data-parallelism and that this strategy is also applicable for code-optimisation on standard processors. Our results show that real-time TDDFT, as implemented in OCTOPUS, can be the method of choice to study the excited states of large molecular systems in modern parallel architectures.

1   Introduction

One of the main factors that has influenced the wide adoption of first principles electronic-structure methods in Physics and Chemistry was the fast growth of the capabilities of computers dictated by Moore's law [1]. This was combined with the development of new algorithms and software capable of exploiting these capabilities. Thanks to these advances, molecular or solid-state systems containing several thousands of atoms are now accurately modelled using supercomputers.

During the last years, however, we have witnessed a remarkable change in computer architectures. Before, the ever-increasing transistor density was directly translated into an increase of the speed and capabilities of a single processor core. However, problems related to efficiency, power consumption and heat dissipation forced a change of paradigm. So the trend is now to use the extra transistors to provide more processing elements. This is reflected in today's supercomputers, where the number of processor cores is constantly increasing while the capabilities of each processing element are progressing much slower. The same is true for personal computing, the parallelism can be obtained via multi-core central processing units (CPU), but also generally-programmable graphics processing units (GPU). In fact, GPUs effectively convert a desktop computer into a massively parallel computer.

Unfortunately, the task of using efficiently all the available parallel units concurrently is left to the application programmer. This is a very complex task, and presents a real challenge to programmers in general and to scientists in particular. Of course, as new parallel programming paradigms are being introduced, scientific codes also have to evolve. Some of the methods that we can currently use can be adapted to parallel architectures, but many popular scientific computing techniques or algorithms might have to be replaced by others that are more efficient in these new environments. In exchange, this change in computational paradigm offers us the possibility of studying larger systems under more realistic conditions and using more accurate methods than it was possible before.

In this article, we show how time-dependent density functional theory (TDDFT) [2] in real time can be a very competitive approach to study the excited states of electronic systems in massively parallel architectures, especially when combined with a spatial grid representation. In fact, real-time TDDFT is a versatile method to model the response of an electronic system (molecular or crystalline [3]) to different kinds of perturbations. It is useful to calculate properties like optical absorption spectra [4, 5], non-linear optical response [6, 7], circular dichroism [8, 9], van der Waals coefficients [10], Raman intensities [11], etc. The numerical advantage of real-time TDDFT is that the propagator preserves the orthogonality of the states [12]. In practice, this allows us to propagate each one of the states in an independent manner, which is ideal for parallelisation. Since the method does not require expensive orthogonalisation steps, the

numerical cost scales with the square of the size of the system and not cubically as many other methods [13].

Here, we present a summary of the work that has been made during the last years on the optimisation and parallelisation of the code OCTOPUS [14, 15], in order to profit from state-of-the-art high-performance computing platforms, from GPUs to supercomputers. This code is a widely used tool to perform real-time TDDFT simulations. It uses a real-space grid that provides an accurate and controllable discretisation that also allows for an efficient parallelisation by domain decomposition.

Directly following (Section 2), we give a brief description of OCTOPUS and its features. Then (Section 3), we focus our discussion in the parallelisation of OCTOPUS: the various parallelisation modes and how they can be combined depending on the system characteristics and the available processors. We pay special attention to the crucial bottleneck that can be the solution of Poisson's equation. Then we describe how we tackled the GPU parallelisation (Section 4). Finally (Section 5), we present some scalability results to illustrate all the efforts described in the previous sections.

2   Octopus features

2.1   Theory

OCTOPUS was originally written to solve the equations of density functional theory (DFT) in its ground-state [16] and time-dependent [2] forms. In particular, and like the vast majority of DFT applications, we use the Kohn-Sham (KS) [17] formulation of DFT, which leads to a coupled set of single-particle equations whose solution yields the many-body electronic density $n(\boldsymbol{r}, t)$. For example, for the time-dependent case these equations read (atomic units are used throughout this article)

$$i\frac{\partial}{\partial t}\varphi_i(\boldsymbol{r}, t) = \left[-\frac{1}{2}\nabla^2 + v_{\text{ext}}(\boldsymbol{r}, t) + v_{\text{Hartree}}[n](\boldsymbol{r}, t) + v_{\text{xc}}[n](\boldsymbol{r}, t)\right]\varphi_i(\boldsymbol{r}, t) \tag{1}$$

$$n(\boldsymbol{r}, t) = \sum_i^{\text{occ}} |\varphi_i(\boldsymbol{r}, t)|^2 \tag{2}$$

where $\varphi_i(\boldsymbol{r}, t)$ are the single-particle KS states (also called KS orbitals), $v_{\text{ext}}(\boldsymbol{r}, t)$ is the time-dependent external potential that can be the potential generated by the nuclei, a laser field, etc.; $v_{\text{Hartree}}[n](\boldsymbol{r}, t)$ is the Hartree potential that describes the classical mean-field interaction of the electron distribution; and $v_{\text{xc}}[n](\boldsymbol{r}, t)$ is the exchange-correlation (xc) potential that includes all non-trivial many-body contributions.

It is true that the (time-dependent) KS equations are an exact reformulation of (time-dependent) quantum mechanics. However, the exact form of the xc functional is unknown and, therefore, has to be approximated in any practical application of the theory. In OCTOPUS different approximations for this term are implemented, from local and semi-local functionals to more sophisticated orbital dependent functionals, including hybrids [18] and the optimised effective potential approach [19]. Asymptotic correction methods are also implemented [20].

We note that the local and semi-local xc functionals in OCTOPUS were implemented as a separate component, Libxc [21]. This library is now completely independent of OCTOPUS and is used by several projects like APE [22], GPAW [23], and ABINIT [24]. Currently it contains around 180 functionals for the exchange, correlation, and kinetic energies belonging to the local-density approximation (LDA), the generalized-gradient approximation (GGA), the meta-GGA, and hybrid functional families. Functionals for systems of reduced dimensionality (1D and 2D) are also included.

OCTOPUS can also be used to study model systems of different dimensionalities (currently up to five-dimensional systems are supported). For this, an arbitrary external potential can be given by the user by directly including its formula in the input file. This model is extremely useful, e.g., to study reduced-dimensional systems of interacting electrons in time-dependent fields [25–28]. In fact, the Schrödinger equation describing two electrons interacting in 1D is equivalent to a Schrödinger equation of one independent electron in 2D. In the same way, the problem of two electrons interacting in 2D can be mapped to the problem of one electron in a 4D space.

Another recent incorporation in the code is the possibility of performing multi-scale modelling by combining electronic systems with complex electrostatic environments. For example, this has been used to simulate a molecule placed between two metallic plates at a certain voltage bias [29]. Multiscale QM/MM calculations can be performed as well [30].

Besides ground-state and real-time TDDFT, OCTOPUS can do other types of calculations. It can perform Ehrenfest-TDDFT non-adiabatic molecular dynamics [14, 31] and adiabatic molecular dynamics based on the modified Ehrenfest scheme [32, 33], which inherits the scalability properties of real-time TDDFT, or the standard Born-Oppenheimer and Car-Parrinello [34] schemes. Different response properties in TDDFT [35] can also be obtained using linear-response formalisms like Casida [36], or the density-functional perturbation theory/Sternheimer approach [37–43]. OCTOPUS can also do quantum optimal-control calculations [44–46] and real-time quantum transport calculations [47]. OCTOPUS can generate the DFT data required for GW and Bethe-Salpeter calculations using the BerkeleyGW code [48].

2.2 Grids

OCTOPUS uses a real-space grid discretisation to represent fields such as the Kohn-Sham states and the electronic density. Each function is represented by its value over an array of points distributed in real space. Differential operators are approximated by high-order finite-difference methods [49] while integration is performed by a simple sum over the grid point coefficients.

The real-space grid approach does not impose a particular form for the boundary conditions, so it is possible to model both finite and periodic systems directly. Moreover, in OCTOPUS the grid boundaries can have an arbitrary shape, avoiding unnecessary grid points. For example, for molecular calculations the default box shape corresponds to the union of spheres centred around the atoms (see Fig. 1 for an example).

One of the main advantages of the real-space grid approach is that it is possible to systematically control the quality of the discretisation. By reducing the spacing and increasing the size of the box, the error is systematically decreased, and can be made as small as desired, at the cost of an
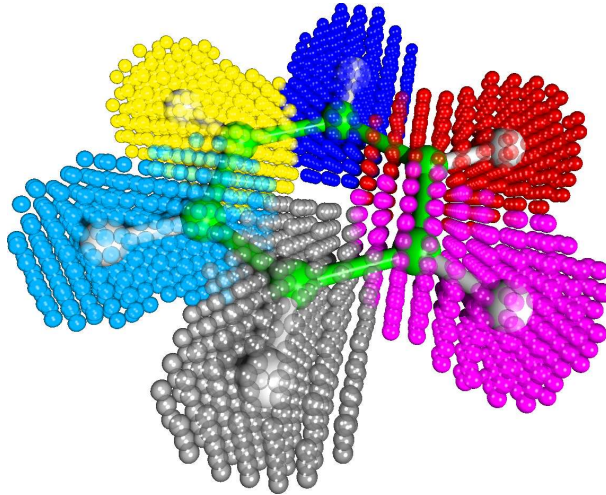
Figure 1: Example of the grid of OCTOPUS for a benzene molecule. The different colours represent different processors in a domain decomposition parallelisation.

increased computational cost. This is of particular significance for response properties [42, 50].

While the real space scheme results in a large number of discretisation coefficients when compared with localised basis set representations, the discretised Hamiltonian is very sparse. In fact, the number of non-zero components depends linearly on the number of coefficients. Moreover, the Hamiltonian only requires information from near neighbours, which is advantageous for parallelisation and optimisation.

Finally, since the description of the core regions is expensive with a uniform-resolution discretisation, in OCTOPUS the ion-electron interaction is usually modelled using norm-conserving pseudo-potentials [51]. At the moment, the code can read pseudo-potentials in several formats: the SIESTA format [52], the Hartwigsen-Goedecker-Hutter format [53], the Fritz-Haber format [54] and its Abinit version [24], and the Quantum Espresso universal pseudo-potential format [55]. Relativistic corrections, like spin-orbit coupling, can also be included by using relativistic pseudopotentials [56, 57].

2.3 The software package

The source code of OCTOPUS is publicly available under the GNU public license (GPL) v.2.0. This allows anyone to use the code, study it, modify it and distribute it, as long as these rights are retained for other users. We believe that this is something of particular importance for scientific work [58]. The code is written mainly in Fortran 95 with some parts in C and OpenCL [59]. Currently it consists of 180,000 lines of code (excluding external libraries).

Since the code is publicly available, it is essential to provide documentation so users can learn how to use it. The OCTOPUS website[1] contains a user manual and several tutorials that teach users how to perform different types of calculations, including some basic examples. Additionally, all input variables have their own documentation that can be accessed through the website or from a command line utility. A mailing list is also available, where users can get help with specific

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[1]http://tddft.org/programs/octopus/

questions about OCTOPUS from the developers or other users.

One of the most important points in developing a scientific code is to ensure the correctness of the results. When a new feature is implemented, the developers validate the results by comparing them with known results from other methods and other codes. To ensure that future changes do not modify the results, we use an automated system (BuildBot [60]) that runs the code periodically and compares the results with reference data. A short set of tests is executed each time a change is made in OCTOPUS while a long one is executed every day. The tests are run on different platforms and with different compilation options, to ensure that the results are consistent for different platforms. Users should also run the testsuite to validate the build on their machine before running real calculations.

To avoid users inadvertently using parts of the code that are being developed or have not being properly validated, they are marked as "Experimental." These experimental features can only be used by explicitly setting a variable in the input file. In any case, users are expected to validate their results in known situations before making scientific predictions based on OCTOPUS results.

3   Parallelisation

In order to take advantages of modern day architectures, OCTOPUS uses a hybrid parallelisation scheme. This scheme is based on a distributed memory approach using the message passing interface (MPI) library for the communication between processes. This is combined with fine-grained parallelism inside each process using either OpenCL or OpenMP.

The MPI parallelisation is mainly based on a tree-based data parallelism approach, even if some steps have already been done in order to take advantage of task parallelism. The main piece of data to be divided among processes are the KS states, an object that depends on three main indices: a combined k-point and spin index, the state index, and the space coordinate. Each one of these indices is associated with a data-parallelisation level, where each process is assigned a section of the total range of the index. Note that since the k-point and spin index are both symmetry related quantum numbers, it is convenient to combine them in a unique index that labels the wave-functions.

This multi-level parallelisation scheme is essential to ensure the scaling of real-time TDDFT. As the size of the system is increased, two factors affect the computational time: first, the region of space that needs to be simulated increases, and second, the number of electrons increases. By dividing each of these degrees of freedom among processors, multi-level parallelisation ensures that the total parallel efficiency remains constant as we increase the system size and the number of processors.

3.1   Parallelisation in K-points and spin

For independent particles, the Schrödinger Hamiltonian can be exactly partitioned according to the k-point and spin labels. This means that each one of the subproblems, *i.e.* for each k-point and spin label, can be solved independently of the others, reducing thereby the dimension of

the Hamiltonian and the computational complexity. Mathematically, in (time-dependent) DFT this is no longer true as the subproblems are mixed by the density. However, a large part of the numerical solution can still be partitioned effectively, making the parallelisation in k-points and spin very efficient as little communication is required. Such a scheme does not always help for scaling, unfortunately. For example, for finite systems only a single k-point is used and in many cases we are interested in spin-unpolarised calculations. In this extreme case, there is absolutely no advantage in this parallelisation level.

3.2   Parallelisation in Kohn-Sham states

The following level of parallelisation regards the distribution of the KS states between processors. The problem is very different for ground-state DFT and for real-time TDDFT, so we discuss these cases separately.

For real-time TDDFT the propagation of each orbital is almost independent of the others, as the only interaction occurs through the time-dependent density. This again leads to a very efficient parallelisation scheme for time propagation that is only limited by the number of available KS states. In fact, when too few states are given to each process (assuming that no other parallelisation level is used), the cost of state-independent calculations starts to dominate (in particular the cost of solving the Poisson equation required to obtain the Hartree potential). As a rule of thumb, one should not have less than 4 states per process in order not to lose efficiency. Note that when ions are allowed to move during the propagation, a complementary parallelisation over atoms is used by OCTOPUS. This ensures that routines that calculate forces and that re-generate the atomic potential at each step do not spoil the very favourable scaling of TDDFT.

For ground-state calculations the parallelisation over states is more complex than for the time-propagation case, as the orthogonality constraint forces the diagonalisation procedure to explicitly mix different states. Regarding parallelisation, the most efficient eigensolver implemented in OCTOPUS turns out to be residual minimisation method – direct inversion in iterative subspace (RMM-DIIS) [61, 62] where the mixing of orbitals is restricted to two procedures: the Gram-Schmidt orthogonalisation [63] and the subspace diagonalisation. To parallelise these operations we use the parallel linear-algebra library Scalapack [64]. State-parallelisation of these procedures is particularly important as they involve matrices whose dimension is given by the number of KS states. Without state-parallelisation a complete copy of these matrices is kept by each process, which can easily lead to memory problems for systems with thousands of atoms.

3.3   Parallelisation in domains

The final level of MPI parallelisation consists in assigning a certain number of grid points to each process. In practice, the space is divided in domains that are assigned to each process. As the finite difference operators, like the Laplacian, only require the values of neighbouring points, the (small) boundary regions between domains need to be communicated between processors. In OCTOPUS this is done asynchronously, which means that the boundary values are copied between processors while the Laplacian calculation is done over the points in the central part

of the domain that do not require this information. This decreases substantially the problems related to latency, thereby improving the scaling with the number of processes.

Note that also the calculation of integrals requires a communication step to add the partial sum over each domain, an operation known as a reduction. The strategy to reduce the cost of this communication step is to group reductions together, as the cost of reductions of small vectors of data is dominated by the latency in the communication.

An important issue in domain parallelisation is selecting which points are assigned to each processor. This task, known as grid partitioning, is not trivial for grids with an arbitrary shape. Not only the number of points must be balanced between processors but also the number of points in the boundary regions must be minimised. OCTOPUS relies on external libraries for this task, with two currently supported: METIS [65] and ZOLTAN [66]. An example of the grid partitioning scheme is shown in Fig. 1.

Certainly the communication cost of the domain parallelisation is considerably higher than for the other schemes. It is also the most complicated to implement. However, once the basic grid operations are implemented it is almost transparent for developers to write parallel code.

3.4 Parallelisation of the Poisson solver

In order to obtain the Hartree potential from the electronic density, we solve the Poisson equation

$$\nabla^2 v_{\text{Hartree}}(\boldsymbol{r}) = -4\pi n(\boldsymbol{r}) \tag{3}$$

When performing real-time propagation, the solution of this equation becomes the main bottleneck when thousands of processors are used [67]. The reason is clear: as there is only one Poisson equation to solve (regardless of the number of KS states), domain partitioning is the only available scheme to parallelise this operation. Unfortunately, the solution of the Poisson equation is highly non-local, mixing information from all points, making it unsuitable for the domain decomposition approach that relies on space locality. This can be easily seen from the integral form of Eq. (3)

$$v_{\text{Hartree}}(\boldsymbol{r}) = \int \mathrm{d}\boldsymbol{r}' \frac{n(\boldsymbol{r}')}{|\boldsymbol{r} - \boldsymbol{r}'|} \tag{4}$$

Fortunately, several solutions to this problem exist, given the wide use of the Poisson equation in different fields. In OCTOPUS this parallelisation level is handled in a special way, and all processes are used independently of the distribution scheme. We found two particularly efficient approaches.

The first one is the use of fast Fourier transforms (FFT) to evaluate (4) in reciprocal space, a very fast method when running on a single processor. However, FFTs are not easy to parallelise. Additionally, the standard FFT approach yields a Poisson potential with periodic boundary conditions, so some modifications have to be made to obtain the free-space solution. Two different FFT-based solvers are available in OCTOPUS: (i) the software package provided by Genovese *et al.* [68] that uses an interpolating scaling functions (ISF) approach and the parallel FFT routine by Gödecker [69]; (ii) the parallel fast Fourier transform (PFFT) library [70] combined with the free-space Coulomb kernel proposed by Rozzi *et al.* [71].
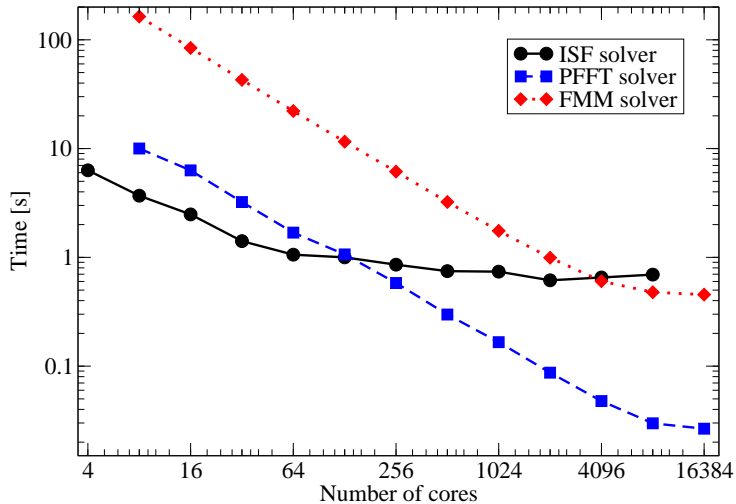
Figure 2: Time required to solve the Poisson equation for different solvers implemented in OCTOPUS. Grid of size 15^3 on a IBM Blue Gene/P system.

The second parallel Poisson solver we tested is the fast multipole method (FMM) [72]. This is an approximate method that reduces the complexity of evaluating Eq. (4) by using a multipole expansion to approximate the far-field effect from several charges into a single term. Our implementation is based on the FMM library [73]. This library is designed to calculate the interaction between point charges, therefore, we have to assume that the density in the grid corresponds to an array of point charges and then calculate a correction for the interaction between neighbouring points [74]. This correction term, essential to obtain the necessary accuracy, has the form of a finite-differences operator, and is therefore simple to evaluate in our framework.

In Fig. 2, we compare the time required to solve the Poisson equation in a parallelepiped box of size 15^3. As we can see, the ISF method is faster for small number of processors, while PFFT and FMM become more competitive as we increase their number. The crossover point is quite low with PFFT method (128 processors) and higher with FMM (4096 processors). More details about the parallel solution of the Poisson equation in OCTOPUS can be found in Ref. [74].

4 GPU parallelisation and code optimisation

Graphical processing units were initially designed for generating graphics in real time. They are massively parallel processors with hundreds or thousands of execution units. Given their particular architecture, GPUs require code written in a explicitly parallel language. The OCTOPUS support for GPUs is based on OpenCL, an open and platform-independent framework for high-performance computing on parallel processors. OpenCL implementations are available for GPUs from different vendors, for multi-core CPUs, and for dedicated accelerator boards. Since the OpenCL standard only defines a C interface, we have developed our own interface to call OpenCL from Fortran. This interface is currently available as an independent library, FortranCL [75].

For optimal performance, GPUs must process several streams of independent data simultaneously. This implies that performance critical routines must receive a considerable amount of data

on each call. To do this, the GPU optimisation strategy of octopus is based on the concept of a block of states, *i.e.*, a small group of KS states. Performance-critical routines are designed to operate over these blocks. This approach provides a larger potential for data parallelism than routines that operate over a single state at a time. It turns out that this strategy also works well for CPUs with vectorial floating units, where parallelisation is based on the OpenMP framework combined with explicit vectorisation using compiler directives.

For both GPUs and CPUs, memory access tends to be the main limitation to the performance of octopus. Working with blocks of orbitals improves memory access, provided that the coefficients are ordered in memory by the state index, so that load and stores are done from/to sequential addresses. However, increasing the KS block size can have a negative effect in memory access as larger data sets are less likely to benefit from cache memory. This is particularly critical for CPUs that depend much more than GPUs on caching for optimal performance.

One routine that can greatly benefit from caching is the application of the finite-difference Laplacian operator required for the kinetic-energy term. This is also an important operation as it represents a considerable part of the execution time of octopus. We devised an approach to improve cache utilisation by controlling how grid points are ordered in memory, *i.e.*, how the three-dimensional grid is enumerated. The standard approach is to use a row-major or column-major order which leads to neighbouring points being often allocated in distant memory locations. Our approach is to enumerate the grid points based on a sequence of small cubic grids, so close spatial regions are stored close in memory, improving memory locality for the Laplacian operator. The effect of this optimisation can be seen in Fig. 3. For the CPU with the standard ordering of points, the performance decreases as the block size is increased, while by optimising the grid order the parallelism exposed by a larger block size allows a performance gain of approximately 40%. For the GPU the effect of the optimisation is less dramatic but still significant.
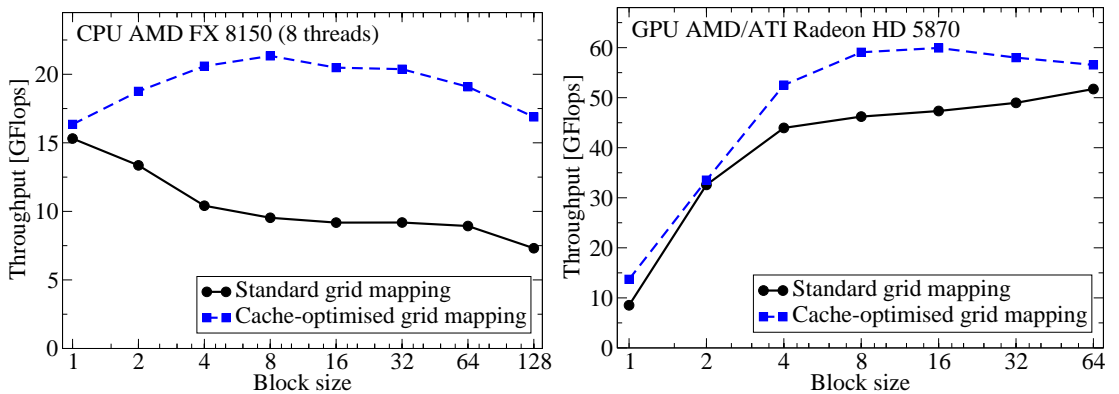
Figure 3: Effect of the optimisation of the grid mapping for data locality in the numerical throughput of the Laplacian operator as a function of the size of the KS states block. *Left* computations with an AMD FX 8150 CPU (8 threads). *Right*: computations with an AMD/ATI Radeon HD 5870 GPU.

In Fig. 4 we show a comparison of the numerical throughput of the GPU and CPU implementations of the Laplacian operator as a function of the size of the KS states block. It can be seen that the use of blocks of KS states represents a significant numerical performance gain with

respect to working with one state at a time. This is particularly important for GPUs where performance with a single state is similar to the CPU one but can be tripled by using a block of size 16 or 32. The same conclusion can be reached by looking at the numerical throughput of the orbital propagation and the total time required for a TDDFT iteration (see Fig. 5 and Table 1). The use of GPUs gives quite spectacular improvements, with a total iteration time being decreased by more than a factor of 6 with respect to the optimised multi-threaded CPU implementation. In fact, the propagation of the KS orbitals is 8 times faster in the GPU, but currently the total speed-up is limited by the Poisson solver that is executed on the CPU. These results mean that using a single GPU OCTOPUS could obtain the absorption spectrum of the C_{60} molecule in about one hour. More details about the GPU implementation in OCTOPUS can be found in Refs. [76, 77].
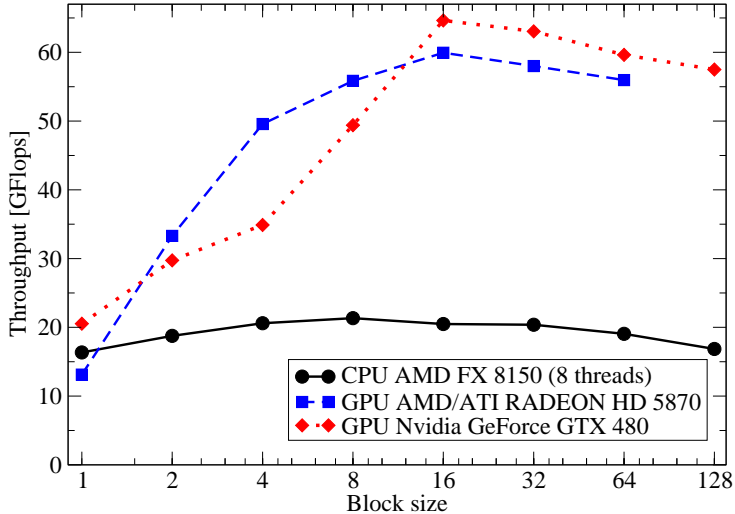
Figure 4: Numerical throughput of the OCTOPUS implementation of the Laplacian operator for different processors as a function of the number of KS states in a block. Spherical grid with 5×10^5 points. For each point the grid ordering has been adjusted for optimal cache usage.

Processor	Time per step [s]
CPU AMD FX 8150 (8 threads)	9.45
GPU AMD/ATI Radeon HD 5870	1.86
GPU NVIDIA GeForce GTX 480	1.72

Table 1: OCTOPUS propagation time per step for different processors. C_{60} molecule with a grid of spheres of radius 10 a.u. around each atom and spacing 0.375 a.u.

5 Scalability in massively parallel systems

In this section we show how all the improvements in algorithms, parallelisation, and optimisation are combined in the simulation of large electronic systems. As a benchmark we used portions of the spinach photosynthetic unit [78] with 180, 441, 650, 1365 and 2676 atoms containing several chlorophyll units. As these are molecular systems, only the state and domain decomposition parallelisation levels are used. We emphasise that these are real-world examples, that were not
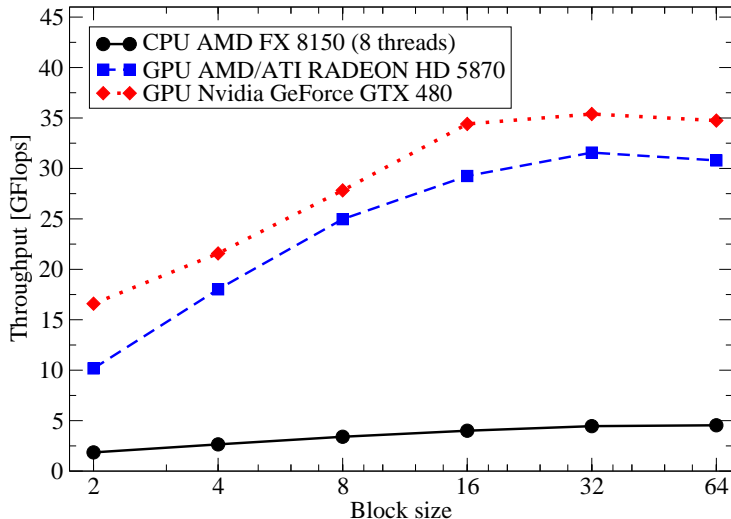
Figure 5: Numerical throughput of the KS state propagation in OCTOPUS for different processors as a function of the size of the KS states blocks. C_{60} molecule with a grid of spheres of radius 10 a.u. around each atom and spacing 0.375 a.u.

tweaked in order to artificially improve throughput or scalability.

We used different supercomputers for our benchmarks, as using more than one machine ensures a better portability of the code and inherent scalability of the algorithm. The first one is Curie, a supercomputer that belongs to the French Commissariat à l'Energie Atomique (CEA) and that consists of Intel Xeon processors interconnected by an Infiniband network. In total it has 11,520 cores available to users. The second platform is the IBM Blue Gene/P, a design based on low power processors (IBM PowerPC 450) interconnected by two networks, one with a toroidal topology and the other with a tree topology. The Blue Gene/P is a challenging architecture as it has a very limited amount of memory per core (512 MiB). Because of this, inside each node OpenMP parallelisation is used as this guarantees a minimal replication of data. Compiler directives are also used to profit from the vectorial floating-point units. The Blue Gene/P calculations presented in this work were performed in the 16,384-core supercomputer of the Rechenzentrum Garching of the Max Planck Society, Germany.

We benchmark separately the two main tasks performed by OCTOPUS. The first concerns the calculation of the ground-state of the system, while the second regards real-time TDDFT runs. We stress again that the parallelisation of the first task is considerably more difficult than of the second. However, the parallelisation of the time-dependent runs is much more important, as this consumes much more computational time than the ground-state runs.

Note that OCTOPUS has to perform several initialisations (create the grid, reserve memory, prepare the MPI environment, etc.) before starting the actual simulations. This time is usually negligible compared to the total simulation time. Our results are therefore measured in terms of elapsed wall-clock time per self-consistent iteration (for the ground-state) or per time step (for the time-dependent runs). The total execution time of a real-world simulation is basically proportional to these numbers.

5.1 Ground-state calculations

We start by showing scalability tests for ground-state calculations for a system of 180 and 441 atoms executed on a Blue Gene/P system. In order to measure the ground-state iteration time, 10 self-consistency cycles were run and the average time is shown in Fig. 6. We also show the parallel speed-up, the relative performance with respect to the run with the smallest number of processors. Excellent scalability is achieved up to 256-512 cores, and up to 4096 cores the time per iteration decreases as the number of processors is increased. As expected, scaling improves for systems with a larger number of atoms.
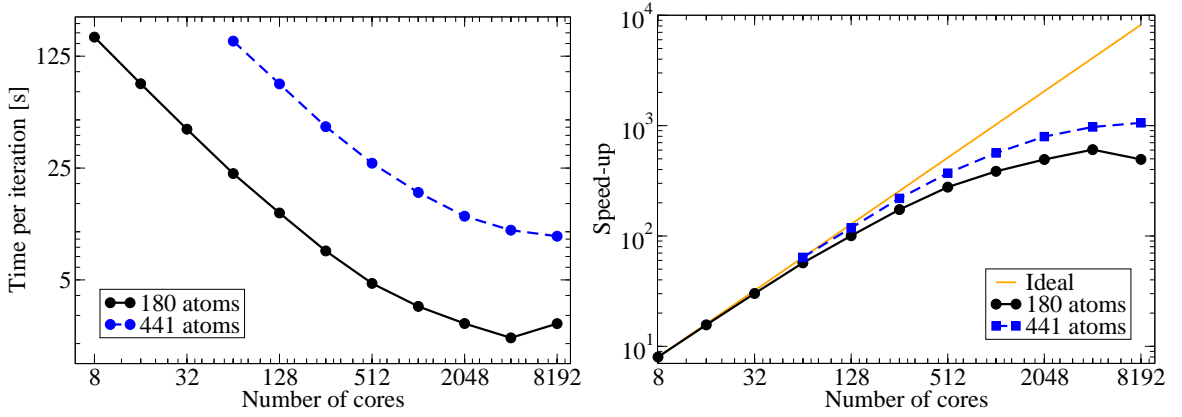
Figure 6: Ground-state calculation on a Blue Gene/P system for different number of atoms. *Left:* Wall-clock time per self-consistency iteration. *Right*: Parallel speed-up compared with the ideal case.

Recall that all runs introduced here were performed in symmetric multiprocessing (SMP) mode, combining inner-loop OpenMP parallelisation with domain MPI parallelisation. This allows us not only to take advantage of the architecture of the CPU, but also to make better use of the (limited) memory available in each of the Blue Gene nodes. For instance, the run with 1024 cores was done launching 256 MPI processes, each of them with 4 OpenMP threads.
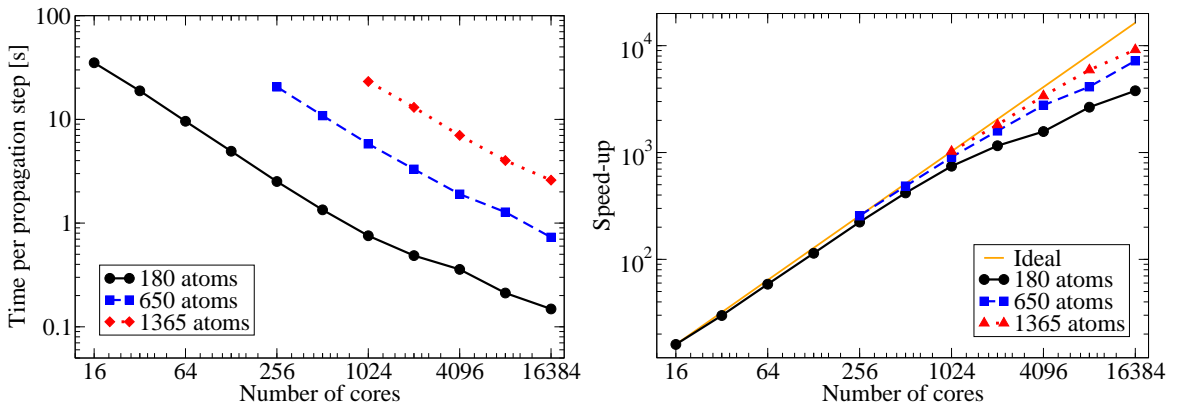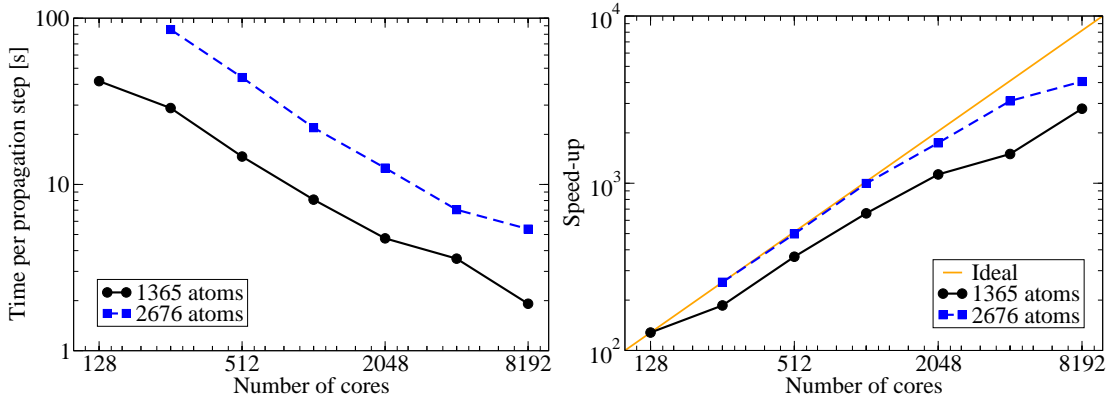

Figure 7: Real-time TDDFT propagation on a Blue Gene/P system for different numbers of atoms. *Left:* Wall-clock time per time step. *Right*: Parallel speed-up compared with the ideal case.

5.2 Real-time TDDFT calculations

In Fig. 7, we show the execution times and scaling for time-propagation runs for the systems of 180, 650 and 1365 atoms executed on a Blue Gene/P system also in SMP mode. We combine different ways to partition the cores between the domain and states parallelisation, selecting only the most efficient combination. We used a minimum of 16 cores with 180 atoms, 256 cores with 650 atoms and 1024 cores with 1365 atoms, as runs with fewer cores were not possible due to memory limitations. Almost perfect scaling was achieved up to 512 cores with the system of 180 atoms, and a remarkable speed-up of 3785 was obtained with 16384 cores, reaching an efficiency of 23% with 91 cores per atom. In the case of the system composed by 650 atoms the ideal speed-up is reached up to 2048 cores, while for 16384 cores a value of 7222 is achieved. Even better performance is obtained with the system of 1365 atoms, with a speed up of 9157 for 16384 cores. Thus, we can maintain an almost ideal speed-up with around 3 cores per atom, and acceptable speed-ups up to 50 cores per atom.

Benchmark tests also were done in the Curie supercomputer. The results are show in Fig. 8. The main advantage of this machine is that it has more powerful processors and more memory per core. However, the drawback is that the network is not as reliable as the Blue Gene one. The network is a shared resource and depends not only in the current run but also in all other running jobs. Consequently, only the minimum iteration time is shown here as an example of the best possible execution. Nevertheless, and confirming the scalability of the algorithm, remarkable speed-ups are achieved up to 8192 cores.

Figure 8: Real-time TDDFT propagation on the Curie system for different number of atoms. *Left:* Wall-clock time per time step. *Right:* Parallel speed-up compared with the ideal case.

We note that these tests were performed with the PFFT Poisson solver, which shows a great improvement when a large number of nodes are used when compared with the default ISF solver. The improvements made to the Poisson solver are reflected in the entire time-dependent iteration time. For large numbers of processes, the total iteration time is improved by up to 58% with respect to the ISF solver.

5.3 Combined MPI-GPU parallelisation

OCTOPUS can also combine the MPI and OpenCL parallelisations in a hybrid approach to use

multiple GPUs. This has created some additional challenges when compared to the multiple-CPU parallelisation. First of all, as GPUs offer higher computational capabilities than CPUs, the time spent in communication becomes a larger fraction of the total execution time. In second place, communication times can become higher as transferring data between GPUs is a three-step procedure: first the data is copied to main memory using OpenCL, then it is sent to the other process using MPI; and finally it is copied to the memory of the second GPU using OpenCL. Still OCTOPUS can scale reasonably well when running on multiple GPUs, as it can be seen in Fig. 9. The speed-up for 8 GPUs is 5.7, an efficiency of 71%. Note that the main limitation to scalability in this case is the lack of a GPU accelerated parallel Poisson solver.
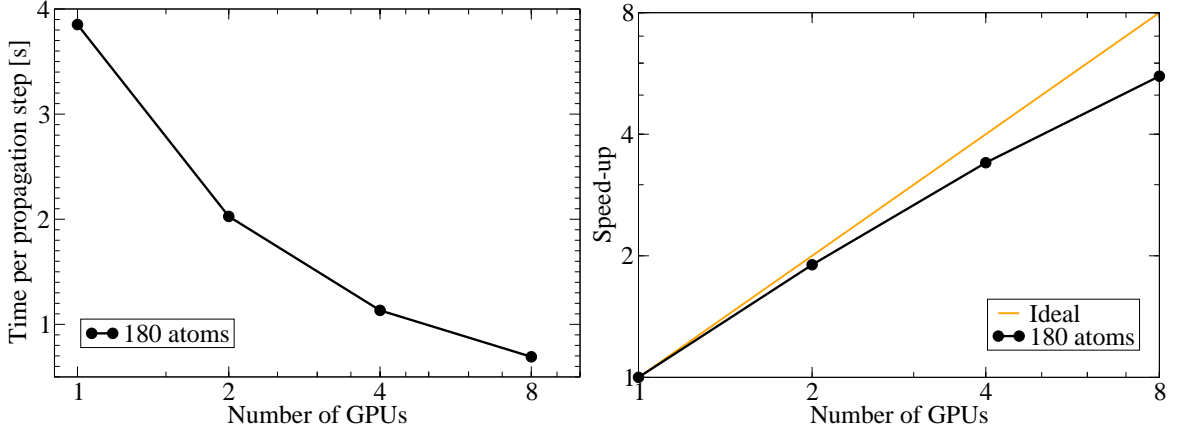
Figure 9: Scaling of time-propagation with multiple Nvidia Tesla M2090 GPUs. *Left:* Wall-clock time per time step. *Right*: Parallel speed-up compared with the ideal case.

6 Conclusions

In summary, our results show that real-time solution of TDDFT equations is a very parallelisable task. In our calculations we can scale with a reasonable efficiency to almost 100 cores per atom, which paves the way for TDDFT real-time simulations of systems with thousands of atoms. We would like to remark that all the calculations in this article were performed using simulation parameters that correspond to actual calculations, and that they were not tweaked to obtain better scalability. This parallelisability can also be exploited for efficient execution in parallel processors, including GPUs and multi-core CPUs with vectorial floating point units.

We also have shown a hybrid parallelisation scheme for execution on clusters of GPUs. This approach combines the high-level MPI parallelisation with low-level parallelisation based on OpenCL. For the moment our tests have been limited to a small number of GPUs, but clusters with thousands of GPUs are already available so certainly this approach will be developed with an eye on exaflop computing.

The current capabilities of OCTOPUS ensure that the excited-states properties of systems with thousands of atoms can be studied in the immediate future. The flexibility of real-time TDDFT method means that not only electronic linear response properties can be studied. In fact, non-linear phenomena or the effect of ionic motion in the response can all be tackled with this formalism. This is of particular interest, for example, in the study of quantum effects in

biological molecules, or in the interaction of matter with strong laser fields.

7 Acknowledgments

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