Ψ_k Newsletter

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

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Contents

1 Editorial

In this Psi-k newsletter, in the **General News** section, we remind you what is the best way of getting your messages to Psi-k mailing list or the Psi-k newsletter processed most efficiently. The current newsletter contains only one workshop report, with abstracts of presented papers, and it can be found in the **RTN2** (f-electron Network) section. Call for workshop proposals for 2005 is in the **ESF** section. We announce future workshops in both the **ESF-** and the **General Workshop/Conference Announcements** section. The announcements of available positions and abstracts of the newly submitted papers are in their usual sections.

In the **UK's CCP9** section we announce the CCP9 Conference which will take place in Daresbury Laboratory, UK, in November 2004. At this conference Professor Matthias Scheffler (FHI Berlin), the winner of the Institute of Physics Max Born Medal and Prize for 2004, will deliver the Max Born lecture. We encourage our readers to register for this conference which promises to be a very stimulating scientific event. Note that the number of places is limited to about 80, so do not leave it too late to register.

In this newsletter we have a short section presenting a new network, which is the NANOQUANTA Network of Excellence. In this section, apart from a short description of the network, there is a full information on this network's job vacancies, and in particular PhD and postdoctoral positions. Please see the section **Presenting New Networks** for details.

The scientific highlight of this Psi-k newsletter is by Takayuki Kitamura, Yoshitaka Umeno and Akihiro Kushima (*Kyoto University*) on **"Ideal strength of nano-structured components"**. Please check the table of contents for further details.

The *Networks* have a home page on World Wide Web (WWW). Its *Uniform Resource Locator* (URL) is: **http://psi-k.dl.ac.uk/**

The above contains information on the Psi-k 2004 workshops and hands-on courses (subject to funding). In these pages you can also find information on how to apply for funding to make collaborative visits.

Please submit all material for the next newsletters to the email address below.

The following email addresses, which remain in operation, are repeated for your convenience, and are the easiest way to contact us.

	function
psik-coord@daresbury.ac.uk	**messages to the coordinators, editor & newsletter**
psik-network@dl.ac.uk	**messages to the whole Ψ_k community**

Dzidka Szotek, Walter Temmerman and Martin Lüders
e-mail: psik-coord@dl.ac.uk

2 General News

2.1 Sending Electronic Messages to Psi-k Mailing List

This is just to remind everybody that in order to make sure that your contributions to the Psi-k newsletter get swiftly processed is to submit them to

psik-coord@dl.ac.uk.

Three persons have access to this mailbox and in case of absence of one or two of them there is always another person to take care of the contributions.

In order to get announcements distributed across the Psi-k mailing list promptly, it is advised to send your e-mails to

psik-network@dl.ac.uk.

The Psi-k mailing list is moderated and every message sent to the network needs to be approved to stop SPAM or other undesired messages arriving to your mailboxes. But still by sending your announcements to the above e-mail address you will have a better chance of getting prompt distribution than when sending them to any of our individual addresses. If any of us is away at conferences then others still can process your messages. Note please that the approval happens via a UNIX workstation, as a result of which many attachements get scrambled. Therefore we advise simply to distribute ASCII files that form the main body of the electronic message, referring to web-pages instead of attaching pdf- or ps-files to the message. We welcome any suggestions for improvements of the Psi-k services. Please note that we have no dedicated secretarial help and all Psi-k services are provided by the scientists.

3 News from the RTN2 Network

"f-electrons"

3.1 Reports on the RTN2 Workshops

3.1.1 Report on the Riksgränsen Workshop

Workshop on the Electronic Structure of the Light Actinides, Riksgränsen, Sweden, May 11-16, 2004

The first workshop on the electronic structure of the light actinides was held at the ski-resort Riksgränsen, Sweden, May 11-16, 2004. The focus of the workshop was both on the latest theoretical developments as well as on the experimental aspects of the light actinides. The scientific program included reviews, invited talks and contributed oral and poster presentations. There were 17, 1 hour long, invited talks. The workshop started with three experimental talks (by Dr.T.Gouder, Dr.J.Joyce and Dr.L.Havela) on the electronic structure of elemental Pu (in the α and δ phase) and Pu based compounds (e.g. $PuCoGa_5$, Pu-chalcogenides, and Pu-pnictides). Different interpretations of the various observed features were discussed, in particular the effect of multiplet structures versus band formation. The initial experimental presentations were followed by theory, both based on self-interaction correction formalism (presented by Dr.A.Svane) and LDA/GGA (in a talk given by Dr.P.M.Oppeneer). Dr.Svane also presented spectroscopic features based on combining atomic multiplett calculations with band structure theory (via the so called Hubbard I approximation) and Dr.Oppeneer presented recent successful calculations of the superconducting properties of light actinides, solving the gap equation.

On the second day, Dr.P.Korzhavyi presented theoretical calculations of the oxidation states of UO_2 and PuO_2 and Dr.A.Ruban described the details of a recently developed first principles electronic structure method (the so called relativistic exact muffin-tin orbital method - R-EMTO). Numerical examples were included, in particular the magneto crystalline anisotropy of selected compounds. Theoretical work using a combination of unrestricted Hartree-Fock theory and density functional theory was presented by Dr.M.S.S.Brooks and the spin and orbital moments of selected 3d, rare-earth and actinide materials was compared favourably to experimental data. A critical evaluation of the ground state properties of the light actinide elements was presented by Dr.M.Richter, who reported that drastically different calculated cohesive properties exist in the literature. Dr.Richter argued that properly performed calculations should not deviate more than a few percent from each other. This talk was followed by two talks on strongly correlated electron physics in nano-devices and the magnetic properties of UNi_2Si_2.

Dr.J.Akella started the third day of the workshop by describing recent advancements in the high pressure diamond anvil cell technique, and its application to the light actinides. In particular the disagreement between the results for the equation of state, obtained from the Karlsruhe group and the Livermore group were discussed. Dr.W.Temmerman followed this work by presenting the recent developments in the single site self-interaction corrected KKR-CPA code, and numerical examples of e.g. Ce were given. Dr.R.Ahuja then presented first principles calculations of structural phase transitions in actinide systems, e.g. actinide pnictides and chalcogenides. Dr.Ahuja also presented elastic constants for selected systems, including materials with a negative C_{12} constant. The work on elasticity was followed by a presentation by Dr.L.Nordström, who presented recent advancements in first principles theory of non-collinear magnetism of rare-earth and actinide systems. The discussion of the formalism was followed by a description of several numerical examples. The electronic structure of super-heavy elements, such as element 114 and 126 was presented by Dr.A.Delin, who showed that the g-shell is starting to become filled for element 126. Recent developments in the dynamical mean field theory was presented by Prof. A.Lichtenstein, and a numerical calculation of the ground state properties, as well as the excitation spectrum, of δ-Pu was presented. The last section of the work shop contained a experimental presentation of non-colinear magnetism (helical anti-ferromagnetism and helical ferromagnetism) of $Ce\ _2(Fe_{1-x}Mn_x)_{17}$, by Dr.A.Kuchin. This presentation was followed by a talk of M. Colarieti-Tosti who described a method to calculate the mixed valency of elements and compounds, and a numerical example was given (Yb). The last talk of the conference was given by Prof.M.Katsnelson, who gave an inspiring overview of unconventional actinide and rare earth materials, including heavy fermion systems, Kondo impurity and Kondo lattice systems, as well as intermediate valence systems.

The location of the workshop, Riksgränsen, is an excellent ski-resort in the most northern part of Sweden where the participants also enjoyed the combination of skiing, both alpine and cross-country, and midnight sun. The workshop ended with a snowmobile outing with fishing for some Nordic Char, and a lunch prepared over an open fire. The full program of the workshop including pdf-files of the talks of the invited speakers can be found at www.fysik.uu.se/theomag/workshop2004.

The workshop was supported by the ESF Psi-k programme in connection with its Working Group 10 'Psi-k f-electrons', the EU Research Training Network 'psi-k f-electron' and FOI, Sweden.

PROGRAMME

Tuesday May 11:
18.00 Welcome reception

Wednesday May 12:
8.30 Opening address **O. Eriksson**
8.45 Photoemission study of actinide thin films, from Th to Am **T.Gouder, Karlsruhe, Germany**
9.45 Coffee
10.15 Photoelectron spectroscopy of U, Np and Pu compounds **J. Joyce, Los Alamos National Lab, USA**
11.15 Am doped δ-Pu; experiment **L. Havela, Charles University, Czech Republic**
12.15 Lunch
Skiing+hiking
16.30 Total energy and electronic structure of actinides compounds **A. Svane, Univ. of Aarhus, Denmark**
17.15 Electronic structure investigations of actinides **P. M. Oppeneer, Uppsala University, Sweden**
18.15 Dinner
Midnight skiing

Thursday May 13:
8.30 Oxidation of plutonium dioxide: theory **P. Korzhavyi, KTH, Sweden**
9.30 Coffee
10.00 Green's function fully relativistic exact muffin-tin orbitals method **A. Ruban, KTH, Sweden**
11.00 Theory of spin and orbital magnetization densities with screened exchange interactions **M. S. S. Brooks, Karlsruhe, Germany**
12.00 Lunch
Skiing and hiking
18.00 Dinner
19.30 Evaluation of heavy element lattice geometries in LDA: state of the art **M.Richter, Dresden, Germany**
20.15 Methods of f-electron physics, applied to nano-materials in non-equilibrium **J.Fransson, Uppsala, Sweden**
21.00 The first-order magnetic phase transition in UNi_2Si_2 compounds triggered by correlation effects of Ising spins. **S. Khmelevskyi, Vienna, Austria**

Friday May 14:

8.30 Uranium revisited and newer static high pressure and temperature DAC work at LLNL. **J.Akella, Lawrence Livermore National Lab., USA**
9.30 Coffee
10.00 Single-Site Self-Interaction Correction in the KKR-CPA **W. M. Temmerman, Daresbury, England**
11.00 High pressure properties of actinide elements: theory **R. Ahuja, Uppsala University, Sweden**
12.00 Lunch
13.00 Non-collinear magnetism in the rare earths **L. Nordström, Uppsala University, Sweden**
13.45 Will there ever be g-electron physics? **A. Delin, KTH, Sweden**
14.30 Efficient LDA+DMFT scheme for f-electron systems **A ...Lichtenstein, Hamburg University, Germany**
15.15 Coffee
15.45 Helimagnets Ce2Fe17-xMnx (Hy) **A. Kuchin, Ekaterinburg, Russia**
16.05 On the pressure induced mixed valency in Yb **M. Colarieti-Tosti, Karlsruhe, Germany**
16.50 Problem of anomalous f-electron systems: Kondo lattices and intermediate valence compounds **M. Katsnelson, Uppsala University, Sweden**
18.00 Workshop banquet
Midnight skiing

Saturday May 15:

9.00 Snowmobile trip with lunch and fishing

ABSTRACTS

Photoemission study of actinide thin films

T. Gouder

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We will compare photoemission studies of various U, Pu and Am thin film systems. U has extended f-states, which in many compounds are itinerant. Am, on the other hand, has localized f-states. Pu is at the localization threshold, and depending on the environment, the f-states may be weakly itinerant or localized. Photoemission has been used to study the electronic structure of actinides, and in particular the localization-delocalization of the f-states. One important question is whether the spectra really reflect the ground-state density of states, or whether final state effects (multiplets, correlation satellites) have to be taken into consideration. We present a comparative photoemission study of different actinide systems with different degree of localization. Examples of hydrides, pnictides and chalcogenides will be discussed. Also the influence of dilution and reduced dimensionality on the electronic structure will be discussed. We will show that in most Pu systems, such final state features are important.

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Photoelectron Spectroscopy of U, Np and Pu Compounds

J. Joyce

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The electronic structure of actinides is investigated using photoelectron spectroscopy from a laser plasma light source as well as synchrotrons. The central role of the 5f electrons in defining the electronic properties of actinides is examined and the extent of hybridization between 5f states and conduction electrons is qualitatively assessed. Photoemission provides a rather direct probe of the electronic structure of materials and affords an opportunity to compare to electronic structure calculations. The photoemission results will be compared against mixed-level model calculations for a range of Pu materials. Experimental data for δ-Pu will be compared against several computational schemes. With tunable photons as the excitation source in photoemission one is able to probe different orbital components via cross-section arguments and thereby investigate hybridization and bonding issues of 5f electron behavior. To identify controlling principles in actinide electronic structure, we extend the scope of the investigation to include compounds as well as the elements, achieving a greater range of hybridization possibilities as the actinide separation distance, crystal structure and available ligands, are varied systematically. Within this framework, a description of the electronic structure of transuranics, and actinides is being developed. For Pu the 5f electrons are found in two configurations, one localized and the other iterant. This dual nature for the 5f electrons occurs in δ-Pu metal, $PuCoGa_5$, $PuIn_3$, and $PuSb_2$ which exhibit a wide variety of crystal structures and correlated electron properties including magnetism, superconductivity and enhanced mass. The controlling conditions which dictates the dual nature of the 5f electrons over a broad crystal structure range but not over all Pu compounds is as yet unknown. Independently, the extension of this 5f electron dual nature model to other systems including uranium compounds and plutonium oxide has recently been published. Synchrotron results using angle-resolved photoemission yield further insight into the behavior of families of actinides extensible to transuranic systems. Sequences of actinide systems with U, Np and Pu components are presented. These sequences indicate localization effects in the 5f actinides which are predictable and quantifiable in binding energy. Finally, the intrinsic nature of the 5f states in some magnetic uranium compounds is identified as narrow band features and the implications for physical properties are discussed.

Work funded by the U.S. Department of Energy, Office of Science.

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Am-doped δ-Pu: experiment

L. Havela

Charles University,
Prague, Czech Republic.

The main part of the lecture is covering the issue of electronic structure of δ-Pu and its variations upon the doping with variable amount of Am (15-40 %), which leads not only to the δ-phase stabilization, but also to a considerable lattice expansion. Photoelectron spectroscopy results (UPS and 4f core-level XPS) suggest that this lattice expansion has surprisingly no apparent effect on the degree of 5f localization up to about 35% Am. This finding might give a hint as to the theoretical description of δ-Pu.

The other part will review results of recent magnetic studies of nanocrystalline U-based itinerant systems (UN and US). It will be demonstrated than any crystallographic disorder tends not only to a suppression of magnetic order, but also of local U moments in this class of materials.

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Total Energy and Electronic Structure of Actinide Compounds.

A. Svane

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DK-8000 Aarhus C, Denmark.

The ground state configuration of AcX compounds (X= N, P, As, Sb, Bi, O, S, Se, Te, Po) is determined from total energy calculations using the self-interaction corrected local-spin-density approximation. Emphasis is put on the PuX compounds, which are compared to their Sm counterparts. The photoemission spectra of SmX and PuX compounds are modelled by including multiplet effects in the atomic limit, as proposed by Lichtenstein and Katsnelson [1]. The analysis supports the proposition of Wachter [2] that PuSe may be considered similar to the high pressure phase of SmSe.

[1]: A. I. Lichtenstein and M. I. Katsnelson, Phys. Rev. B **57**, 6884 (1998).

[2]: P. Wachter, Sol. State Commun. **127** 599 (2003).

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Electronic structure investigations of actinide compounds

P. M. Oppeneer

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Uppsala, Sweden

The electronic structure and electronic properties of several fascinating actinide compounds are investigated. We concentrate here on the heavy-fermion superconductor UPd2Al3, the newly discovered medium-high Tc superconductor PuCoGa5, and the Am monopnictides. For UPd2Al3 the symmetry of the superconducting order parameter is studied selfconsistently by numerical solutions of the gap equation, adopting physically motivated pairing potentials that are either due to small q-phonon processes, or to spin- fluctuations. The same unconventional d-wave order parameter is obtained with both types of pairing potentials. Implications for the pairing mechanism are discussed. Electronic structure calculations for PuCoGa5 and several other actinide 115 compounds reveal that the lattice constants are well-given assuming delocalized 5f's. The electronic states at the Fermi energy are dominated by the Pu 5f's. The superconductivity emerges thus out of states dominated by the Pu 5f electrons. Our investigation in particular reinforces the similarity to the unconventional Ce115 heavy fermion superconductors, yet with a stronger coupling constant due to a stronger 5f hybridization. For the Am compounds we investigate their electronic structure in relation to recent UPS and XPS photoemission experiments.

Work done in collaboration with G. Varelogiannis, I. Opahle, S. Elgazzar, A. Shick, D. Ghosh, S.K. De, T. Gouder and F. Wastin.

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Oxidation of plutonium dioxide: Theory

P. A. Korzhavyi

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Royal Inst. of Technology (KTH), Stockholm, Sweden

The corrosion behaviour of nuclear materials is often unusual and difficult to interpret, both due to the complex electronic structure of the actinide elements and to the difficulties caused by their radioactivity. In recent experiments, most unexpectedly, plutonium dioxide was found to react with water to form higher oxides up to $PuO_{2.27}$, whereas PuO_2 was always thought to be the highest stable oxide of plutonium. Past experimental attempts to oxidize PuO_2 using atomic oxygen or ozone have been negative, so that no direct calorimetric measurements are possible. In this study, we obtain the lacking thermodynamic data from first-principles total energy calculations. On the basis of the obtained theoretical results, combined with well-established thermochemical data, we have performed a thorough analysis of this complicated problem. The reactions of PuO_2 with either O_2 or H_2O to form $PuO_{2+\delta}$ are calculated to be endothermic, that is, in order to occur they require a supply of energy. However, our calculations show that, under

certain conditions, this reaction may be driven by radiolysis of water through the formation of strong oxidants (e.g., hydrogen peroxide, H_2O_2) and their spontaneous reaction with PuO_2.

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Green's function fully relativistic exact muffin-tin orbitals method.

A.V. Ruban

Department of Materials Science and Engineering, Royal Institute of Technology, Stockholm, Sweden.

The Green's function fully relativistic exact muffin-tin orbitals method is based on the Andersen's exact muffin-tin orbitals theory developed for highly accurate and efficient electronic structure calculations. It is based on a simplified muffin-tin geometry and uses a minimal basis set. The full charge density technique provides the accuracy of the full potential method for the total energy calculations. The use of the Green's function formalism in practical implementation of the method, makes it suitable for calculating random alloys, interaction parameters as well as for including correlations effects in the framework of the dynamical mean-filed theory. In this talk talk a general description of the method will be given together with its fully relativistic generalization. The results of the fully relativistic calculations for magnetic anisotropy and exchange interactions will be presented.

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Theory of Spin and Orbital Magnetization Densities with Screened Exchange Interactions

M.S.S. Brooks

ITU Karlsruhe, Germany

An approximate orbital dependent treatment of screened exchange interactions is used to replace the local spin density approximation. Both spin and orbital polarization effects are included and the polarizing density is that part of the density which changes sign under sign reversal symmetry.

We report application of this approximation to Fe, Co, Ni, US, $CeFe_2$ and UFe_2. For Fe, Co, and Ni the calculated spin and orbital moments are similar to those obtained in the local spin density approximation for the spin, but there is a slight increase in the orbital moments. For US the spin and orbital moments are $1.25\mu_B$ and $-2.92\mu_B$, respectively. For $CeFe_2$ a detailed analysis of the spin and orbital magnetization densities shows that the measured magnetic amplitude for Ce is consistent with cerium 4f and 5d spin magnetic moments of about $-0.2\mu_B$ each, a 4f orbital moment of $0.08\mu_B$ and a diffuse spin magnetic moment of $-0.04\mu_B$. For UFe_2 the spin and orbital moments at the U site cancel almost exactly.

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Evaluation of heavy element lattice geometries in LDA: state of the art

M. Richter

Dept. of Theoretical Solid State Physics,
Dresden, Germany

The following questions will be addressed:
- Is LDA outdated ?
- Is the accuracy of 20th century codes sufficient to judge the value of approximations to DFT ?
- Which level of accuracy can be achieved today ?

Further, the following recent results will be shown:
- Atomic volumes and bulk moduli of light actinides
- Hexagonal Uranium films on W(110)
- Comparison of calculations for trivalent d-metals

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Methods of f-electron physics, applied to nano-materials in non-equilibrium

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Mesoscopic quantum systems used for transport, provide excellent tools for studying phenomena related to strong electron correlations. In this talk I will discuss application of a method based on non-equilibrium many-body Green functions in the context of nanotransport. The method, originally designed for f-electron systems, has proven successful for the interpretation of important features recorded in experiments on, for example, double quantum dots. By describing the interacting region (quantum dot, coupled quantum dots, molecule, nanowires etc) exactly in terms of its many-body eigenstates (including the interactions), the contact reservoirs are treated as perturbations to the system. By way of examples, the method will be described along with numerical results.

[1]: J. Fransson, O. Eriksson, and I. Sandalov, Phys. Rev. Lett. **88** 226601 (2002).
[2]: J. Fransson, O. Eriksson, and I. Sandalov, Phys. Rev. B **66** 195310 (2002).
[3]: I. Sandalov, B. Johansson, and O. Eriksson, Int. J. Quantum Chem. **94** 113 (2003).
[4]: J. Fransson, O. Eriksson, and I. Sandalov, Photon. Nanostruct. **1** 89 (2004).
[5]: J. Fransson and O. Eriksson, J. Phys.: Condens. Matter **16** L85 (2004).
[6]: J. Fransson, I. Sandalov, and O. Eriksson, J. Phys.: Condens. Matter **16** L249 (2004).
[7]: J. Fransson, Phys. Rev. B **69** 2013XX(R) (2004).

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The first-order magnetic phase transition in UNi_2Si_2 compounds triggered by correlation effects of Ising spins.

S. Khmelevskyi and P. Mohn

Center for Computational Material Science, Vienna University of Technology, Vienna, Austria.

We show that the sequence of the magnetic phase transitions observed in the intermetallic compound UNi_2Si_2 can be described within pure axial Ising model with three uniaxial competing interactions. Such description can be achieved only by inclusion of correlation effects beyond the mean-field approximation. To treat the effects of non mean-field fluctuations we use a cumulant expansion of the free energy up to third order (Kirkwood approximation). It appears that the first order transition between the uncompensated antiferromagnetic state (UAF) and the ordinary AF state occurring in UNi_2Si_2 at elevated temperature can be regarded as a macroscopic manifestation of correlation effects in the simple Ising model.

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Uranium revisited and newer static high pressure and temperature DAC work at LLNL.

J. Akella

Lawrence Livermore National Laboratory, USA

The crystallographic anisotropy effects in the compression of the orthorhombic alpha-uranium phase were studied. Akella et.al (1990) first proposed "differential" compression of the orthorhombic axes in uranium. Newer detailed DAC experiments showed interesting variation in both axial ratios b/a and c/a with increasing pressure. Results obtained indicate that the c-axis of the orthorhombic cell is the least compressible while there is a crossover point in the compression of a and b axes at 83 GPa. Contrary to what Bihan et al. (2003) postulated, our observed axial ratio trends are independent of the pressure medium used.

We at LLNL have developed newer " designer diamond-anvil " technology in order to investigate the electrical properties of materials at megabar pressures. Also with tailor made designer anvils more reliable high temperature data could be obtained in the P and T space. Nanosecond ultrasonics to study phase transitions in solid and liquid systems at high P and T is another newer technology. Preliminary data obtained using these newer technologies will be presented.

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Single-Site Self-Interaction Correction in the KKR-CPA

W. M. Temmerman

Daresbury Laboratory, UK

Some problems of the local spin-density (LSD) approximation when applied to d- or f- electron systems are related to the spurious self-interaction of the LSD. It has already been demonstrated that these shortcomings can largely be overcome by removing this spurious self-interaction from the LSD for all occupiued orbitals. However, traditional SIC-LSD calculations are computationally very demanding, and applications to alloys, for instance, are very difficult.

Here we will propose a simplified version of SIC-LSD, based on the multiple scattering formalism, with a local self-interaction correction (L-SIC), and discuss its implementation in the KKR method. The multiple-scattering aspect of this new SIC method allows an generalisation to the coherent potential approximation (CPA) in a straight forward manner. This facilitates applications of the SIC to alloys and pseudo alloys allowing to describe intermediate valences. This approach provides a first step towards a dynamical SIC, which will allow for dynamical valence fluctuations.

As a demonstration of the method, we present results for the well-known α-γ transition of Ce, where we also show how the SIC operates in terms of multiple scattering theory. Finite temperature calculations and the L-SIC-CPA are employed to calculate the phase-diagram and in particular the critical point of the transition. It is discussed how the treatment of the intermediate valence of the α-phase will influence the picture, and a strategy for treating this intermediate valence is outlined.

The method is also employed to study Ce impurities in metallic hosts and in particular to determine if Ce impurity is magnetic or not. Excellent agreement with experiment is obtained.

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High pressure properties of actinide elements: theory

R. Ahuja

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Uppsala, Sweden

Density-functional electronic structure calculations have been used to investigate the high pressure behavior of Am. The phase transition from fcc to the low symmetry structure is shown to originate from a drastic change in the nature of the electronic structure induced by the elevated pressure. For the low density phase, an orbital polarization correction to the local spin density (LSD) theory was applied. Gradient terms of the electron density were included in the calculation of the exchange/correlation energy and potential, according to the generalized gradient approximation (GGA). Theory compares rather well with recent experimental data of S.Heathman et al.(Phys.Rev.Lett. **85**, 2961 (2000)) which implies that electron correlation effects are reasonably modeled in our orbital polarization scheme.

Using the same approach, we have looked in to the elastic constants of AnTe where An = Th, U, Pu, Am and Np. They compare very well with the recent experimental data. In addition, we will also present our results for high pressure structural phase transition in thorium pnictides and chalcogenides. We compare the calculated structural stabilities with experimental data. For most of the compounds LDA gives the correct ground state but it fails in case of ThSe, ThAs and ThSb. If we instead use the GGA, the correct ground state is obtained also for these compunds. At high pressure all the compounds show a NaCl → CsCl phase transition except ThTe and ThBi which are stable in the CsCl structure already at ambient pressure.

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Non-collinear magnetism in the rare earths

L. Nordström

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Uppsala, Sweden

Although the magnetism of the rare earths show a very rich range of interesting phenomena, it was for a long time been an almost untouched problem for first principles density functional based methods. This stems from the fact that the magnetism arises within the partially filled but localized $4f$ shell. It will be discussed how the $4f$ magnetism can be treated, and results will be presented for e.g. the stabilised helical spin density waves, anisotropy, and equilibrium lattice constants. Since the tri-valent rare earths are chemically very similar, they are ideal for multi-layer constructions. We will present calculations for some magnetic rare earths multi-layers.

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Will there ever be g-electron physics?

A. Delin

Department of Materials Science and Engineering, Royal Institute of Technology (KTH),
Stockholm, Sweden

Inspired by the recent experimental results on forming superheavy nuclei (113 and 115, in the beginning of this year), I discuss the possiblity of g-electron physics, that is, if we will ever see any atoms belonging to the 5g-series.

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Efficient LDA+DMFT scheme for f-electron systems

A. Lichtenstein

University of Hamburg, Germany

We analyzed different ways to go beyond the LDA approximation and includes the effects of Coulomb interactions in f-electron systems. Realistic dynamical mean field theory (LDA+DMFT) which takes into account a frequency dependence of the self energy have been combined with the first-principle LMTO method. A simplified version of the Exact-Diagonalization scheme as well as the Quantum Monte-Carlo method are tested for spin-polarized multi-orbital case. The accuracy and efficiency of proposed LDA+DMFT scheme will be discussed.

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Helimagnets; $Ce_2Fe_{17-x}Mn_x(H_y)$

A. Kuchin

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The results of magnetic and neutron diffraction studies of the new system Ce2Fe17-xMnx (x $\leq$ 2) at ambient and high pressure and also after hydrogenation are reviewed. The magnetic ground state of the compounds changes with Mn content from collinear ferromagnet to helical antiferromagnet and then to "helical ferromagnet". The stability of these states to change of lattice parameters owing to external pressure or insertion of hydrogen is quite different.

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On the pressure induced mixed valency in Yb

M. Colarieti-Tosti

ITU Karlsruhe, Germany
Physics Department, Uppsala University Uppsala, Sweden

A first-principle based method for calculating the electronic structure and total energy of solids in an intermediate valence configuration will be presented. The method takes into account correlation effects (d-f Coulomb interaction) and many-body renormalisation of the effective hybridisation parameters. As an example the formation of a pressure induced intermediate valence state in Yb metal is considered and its electronic structure and equation of state are calculated and compared to experimental data.

⋆⋆⋆⋆⋆

Problem of anomalous f-electron systems: Kondo lattices and intermediate valence compounds

M. Katsnelson

Physics Department, Uppsala University
Uppsala, Sweden

A review is done of electronic spectra, optical and magnetic properties of "anomalous" rare - earth and actinide compounds, that is, intermediate valence and heavy fermion systems. An importance of correlation effects in these systems is emphasized. An interplay of "excitonic" effects (attraction between d-electrons and f-holes) and Kondo screening of magnetic moments is considered as well as a competition between the Kondo screening and interatomic magnetic interactions. Possible scenario of non-Fermi-liquid behavior in the anomalous f-systems as a result of this competition is discussed.

List of participants

O. Eriksson	Uppsala University	Sweden
T. Gouder	Karlsrhue	Germany
J. Joyce	Los Alamos National Lab	USA
L. Havela	Charles University	Czech Republic
A. Svane	University Of Aarhus	Denmark
S. Khmelevskyi	Vienna	Austria
P. Korzhavyi	KTH	Sweden
M. S. S. Brooks	Karlsrhue	Germany
M. Richter	Dresden	Germany
A. Delin	KTH	Sweden
A. Ruban	KTH	Sweden
J. Fransson	Uppsala University	Sweden
J. Akella	Lawrence Livermore National Lab	USA
W. Temmerman	Daresbury	England
R. Ahuja	Uppsala University	Sweden
L. Nordström	Uppsala University	Sweden
P. M. Oppeneer	Uppsala University	Sweden
A. Lichtenstein	Hamburg University	Germany
M. Colarieti-Tosti	Karlsrhue	Germany
A. Kuchin	Ekaterinburg	Russia
M. Katsnelson	Uppsala University	Sweden
P.Andersson	FOI	Sweden
A. Grechnev	Uppsala University	Sweden
T. Burkert	Uppsala University	Sweden
T. Bjorkman	Uppsala University	Sweden
S. Simak	Uppsala University	Sweden
F. Bultmark	Uppsala University	Sweden
E. Sjöstedt	Uppsala University	Sweden
R. Lizarraga	Uppsala University	Sweden
E. Holmström	Uppsala University	Sweden
J. Wirstam	FOI	Sweden
M. Goliat	FOI	Sweden
I. Opahle	Dresden	Germany
S.K. Kwon	Uppsala University	Sweden
G. Santi	University of Aarhus	Denmark
S. Lebègue	Uppsala University	Sweden
L. Pourovskii	University of Nijmeguen	Netherlands

4 News from the ESF Programme

”Towards Atomistic Materials Design”

4.1 Call for Workshop Proposals for 2005

There will be no support for workshops in 2005 because of our own Psi-k2005 Conference on 17-21 September 2005, but instead people are encouraged to submit ideas for invited speakers grouped into a symposium at the conference: see call for suggestions in August or October Newsletter.

However support for hands-on tutorials and perhaps one or two other activities not conflicting with the conference may be considered.

A proposal should be submitted by email (in the form of a single simple email letter WITHOUT ATTACHMENTS OR LATEX, just plain ASCII) to the Psi-k Programme secretary, Walter Temmerman, at

psik-coord@dl.ac.uk

by 15 September 2004 as absolutely final deadline.

The proposal should include the following points (preferably numbered in this order).

1. Title and purpose of the event, with names and addresses (including email) of the organisers.
2. The scientific content and why this event would be useful at this time.
3. A tentative list of speakers whom it is hoped to have.
4. The number of participants it is planned to invite or attract, and their scientific involvement, e.g. as simulators, related experimentalists, code developers, etc., and young scientists in the subject.
5. Plans for a tutorial element and for attracting new researchers into the subject of the workshop. (If this is thought inappropriate for this workshop, please explain why.) The purpose of the ESF Psi-k Programme is to help everyone in our community to do good quality research, and in our expanding field this implies some outreach and tutorial activity.
6. Budget, expressed in Euro, and how much is being applied for from the ESF Psi-k Programme. In the past, this has been limited to about 9k Euro.

7. A statement about other organisations which will be applied to for co-sponsorship and additional funding, e.g. any EU RTN Network, CECAM, CCP9 in the UK, etc..
8. Where it is hoped the event would be held.
9. Plans about inviting and financing any participants from countries outside Europe including America and Japan.

Peter Dederichs
Chairman of ESF Psi-k Programme

4.2 ESF Workshop/Conference Announcements

4.2.1 ESPA-2004 Workshop

ELECTRONIC STRUCTURE: PRINCIPLES AND APPLICATIONS
(ESPA2004)
UNIVERSITY OF VALLADOLID, SPAIN
SEPTEMBER 15-17, 2004
http://www.uva.es/ESPA2004
The workshop is funded by the Spanish Ministerial Department of Science and the Psi-k ESF programme

SCOPE OF THE WORKSHOP

It is intented to consolidate an existent forum of spanish theorist on computational chemical-physics in a more international context by increasing the participation of scientist from other countries. Aside from this general purpose, the present edition of the conference will dedicate special attention to the methods and theoretical applications dealing with chemical reactivity studies, as well as to recent advances in DFT, its applications in chemistry and how comparison with experiments behaves. Additionally, other techniques applied in the study of the molecular structure, heterogeneous catalysis, biomolecules, chemistry of transition metal systems, etc... will be addressed.

Speakers include:

M. Aschi (L'Aquila, Italy),
A. D. Becke (Queens, Canada),
H. F. Schaefer (Georgia, USA),
G. Borstel (Osnabrueck, Germany),
J. M. Soler (Madrid, Spain),
M. Persico (Pisa, Italy),
E. Kotomin (Stuttgart, Germany),
P. Pyykoo (Helsinki, Finland),
R. J. Bartlett (Florida, USA),
D. Clary (Oxford, UK),
M. Ruiz-López (Nancy, France),
G. E. Scusseria (Austin, USA),
P. Hobza (Prague, Czech Republic),
J. A. Sordo (Oviedo, Spain),
M. Solá (Gerona, Spain),
V. Moliner (Castellón, Spain),

J. M. Ugalde (San Sebastian, Spain),
A. Sanchez de Merás (Valencia, Spain).

The titles of the invited talks are now shown on the workshop website

http://www.uva.es/ESPA2004

Final reminder - Deadlines:

For early registration : June 15

For abstract submission: June 30

For registration: July 15

Organizing committee: Antonio Largo, Luis C. Balbás, Inmaculada Martin, Carmen Barrientos, Carmen Lavin, Pilar Redondo

Contact:

Pilar Redondo
Departamento de Química-Física
Universidad de Valladolid
E-47005 Valladolid, Spain
4-983-423013
espa2004@uva.es

Workshop on Ab Initio Methods for Correlated Spintronics Materials

Mont St. Odile, Strasbourg, 23-26 September 2004

Organizers: M. Alouani, S. Biermann and A. Lichtenstein
Web page: http://www-ipcms.u-strasbg.fr/odile2004/

This workshop in condensed matter physics is motivated by recent advances in the treatment of electronic correlation effects, particularly in the context of realistic electronic structure calculations. The workshop will attempt to balance two important components: the development of new methods, and applications of these methods to experimentally relevant systems. The main objective is to gain new physical understanding into the working of complex materials.

Significant progress in the field of strongly correlated electrons has been achieved recently with the developments of dynamical mean-field theory (DMFT), and on the GW method both in studies of model systems and in combining it with realistic band-theory methods. There are already many indications that realistic implementations of DMFT and GW constitute a scientific opportunity to solve in a quantitative manner problems which have so far defied analysis with more traditional techniques, for example the Mott metal to insulator transition, itinerant ferromagnetism and the behavior of heavy fermions. Taking advantage of this opportunity requires bringing together experts in electronic structure, and many-body theory. The DMFT and GW will be the central methods discussed in the workshop, because they have been extensively developed over the last ten years, and continue to be extended and improved.

Invited Speakers:

O.K. Andersen (MPI Stuttgart, Germany)
B. Arnaud (U. of Rennes, France)
F. Aryasetiawan (JRCAT, Japan)
F. Bruneval (Ecole polytechnique Paris, France)
S. Bluegel (Juelich, Germany)
T. Das-Gupta (Bose Institute, India)
P. Dederichs (Juelich, Germany)
V. Drchal (Prague, Czech Republic)
A. Ernst (MPI Halle, Germany)
O. Eriksson (Upsalla, Sweeden)
A. Georges (Ecole Polytechnique Paris, France)
O. Gunnarson (MPI Stuttgart, Germany)
K. Held (Stuttgart, Germany)
M. Katsnelson (Nijmegen, The Netherland)
J. Kortus (Strasbourg, France)
G. Kotliar (Rutgers, USA)

W. Metzner (MPI, Stutgart, Germany)
E. Pavarini (Pavia, Italy)
M. Rohlfing (Augsburg, Germany)
A. Poteryaev (Humburg, Germany)
A. Robio (San Sebastien, Spain)
M. Rozenberg (Buenos Aires, Argentina)
S. Savrasov (Newark, USA)
A. Schindlmayr (Juelich, Germany)
Z. Szotek (Daresbury, England)
W. Temmerman (Daresbury, England)
R. Zeller (Juelich, Germany)

The workshop will bring together researchers who are using different methodologies for strongly correlated materials along with people from the band theory community who are interested in correlated materials. This workshop will then set a new start for further development in the theory of correlated materials. Furthermore, the workshop will be strongly oriented towards the discussion of a large number of specific materials of great current experimental interest. Sharing insights from diverse materials and different calculation methods should allow to distinguish between common aspects and non universal (material-specific) aspects, hence shedding light on what are the new physical principles that govern the behavior of strongly correlated materials. The basic issues raised (evolution from band behavior to atomic behavior in the presence of orbital degeneracy and multiplet structure, interaction of local moments and charge carriers) are very basic and general. Rather than search for abstract unifying principles, we propose to investigate the specific physical properties of many interesting materials, and compare them to what can be understood using the quantitative tools now under development. This should allow to paint a broader picture of the strong correlation problem, and to evaluate more accurately the current status of theoretical understanding, a key step for further progress.

4.2.2 CMS2004 Workshop

2nd Announcement

The Physics Department, Universita‘ di Cagliari,

and the

Sardinian Laboratory for Computational Materials Science
(SLACS: http://www.slacs.it)
—- organize the —-

XIV WORKSHOP ON
COMPUTATIONAL MATERIALS SCIENCE

”An international workshop on computational techniques and applications to materials science”

18 - 23 September 2004

Calaserena Village, Geremeas (CA), Sardinia, Italy
web page: http://www.dsf.unica.it/ workshop e-mail: workshop@dsf.unica.it
with the sponsorship of
European Science Foundation Programme ”Toward Atomistic Materials Design”
and
Democritos Modeling Center for Research
in Atomistic Simulation
under the patronage of UNESCO

`=====>>>> See TENTATIVE INVITED SPEAKERS below <<<<=====`

FORMAT: About 16 invited lectures plus TALK (∼ 10) and POSTER contributed session.

VENUE: Calaserena Village at Geremeas (CA), an attractive sea resort on the south-eastern coast of Sardinia, providing comforts and an informal atmosphere.

REGISTRATION: The REGISTRATION form can be sent directly from the Workshop

homepage `http://www.dsf.unica.it/~workshop`.

The deadline is June 1st, 2004.

To confirm your registration please send the receipt of your bank transfer by FAX to +39-070-510171 (addressed to F. Bernardini). The fee includes bus transportation, full-board lodging at Calaserena Village (4 days / 5 nights), coffee breaks, refreshments, and amounts to:

Single room: 650,00 Euro/person
Accompanying person: 350,00 Euro/person

Triple/Quadruple room: 550,00 Euro/person

A very limited number of single rooms is available, we will process the Hotel Reservations on the first-come first-serve basis.

PAYMENT: Payment should be effected by bank money transfer, preferably using the IBAN code below, in EURO NET OF ANY BANK CHARGES to the bank account indicated below:

```
Comitato Organizzatore di Attivita
di Fisica Computazionale

BANK NAME: Banco di Sardegna

BANK ADDRESS: Viale Bonaria 33, Cagliari (IT-09125)

IBAN:  IT85 D010 1504 8000 0000 0022 698

ACCOUNT NUMBER: 22698

SWIFT / BIC: SARDIT3S100

BBAN: D 01015 04800 0000000 22698

ABI: 1015

CAB: 04800
```

No fee is requested from the invited speakers.

BUS TRANSFER: We will organize a bus transfer for those arriving at Cagliari-Elmas airport on 18/09 AFTERNOON and leaving on 23/09 MORNING. If you want to use this service please provide your flight infos using the FLIGHT INFO FORM on our web-page.

CONTRIBUTIONS: Contributions in poster format are invited. Deadline for abstract submission: July 15, 2004. LaTeX template for abstract can be downloaded from the Workshop homepage: http://www.dsf.unica.it/ workshop

CONTACT: For scientific and logistic queries: workshop@dsf.unica.it

PS: Please feel free to forward this message to those you deem interested in this event.

INVITED SPEAKERS

A. Filippetti (Cagliari, Italy)
R. Gebauer (Trieste, Italy)
E.K.U. Gross (Berlin, Germany)
G. Lulli (Bologna, Italy)
A. Magistrato (Trieste, Italy)
P. Milani (Milano, Italy)

R. Podloucky (Wien, Austria)
S. Sanvito (Dublin, Ireland)
G. Schmidt (Jena, Germany)
F. Seno (Padova, Italy)
A. Selloni (Princeton, USA)

Series Chairman : Prof. A. Baldereschi - EPFL Lausanne
Workshop Chairman : Prof. F. Meloni - INFM and Universita‘ di Cagliari

Scientific Secretariat:

Dr. F. Bernardini
Dr. V. Fiorentini
Dr. P. Ruggerone
(SLACS and Universita‘ di Cagliari)

5 News from UK's CCP9 Programme

UK's Collaborative Computational Project 9 on "Computational Studies of the Electronic Structure of Solids"

5.1 CCP9 Conference Announcements

5.1.1 CCP9 Conference

Daresbury Laboratory, UK

29-30 November 2004

Organizers:

James Annett and Walter Temmerman

Scope: All ab-initio and materials specific calculations of the electronic properties of condensed matter systems such as metals, semiconductors, magnets, superconductors, biological systems, minerals, surfaces etc.

Topical areas: Magneto-electronics, catalysis, nanotechnology, high temperature superconductors, novel wide band gap semiconductors etc.

Techniques: DFT, GW, TD-DFT, DMFT, QMC, SIC-LSDA, LSDA+U.

All contributions dealing with the solution of the Schrdinger/Dirac equation in condensed matter systems are welcome.

Register on http://www.ccp9.ac.uk

Plenary Speakers

Matthias Scheffler (Berlin) (IOP Max Born Prize Lecture)
Rex Godby (York)
Mikhail Katsnelson (Nijmegen)
Takao Kotani (Osaka)
Richard Martin (Illinois)
Mike Payne (Cambridge)

Local Organisers

Shirley Miller: s.miller@dl.ac.uk, +44 1925 603240
Damian Jones: d.r.jones@dl.ac.uk, +44 1925 603805

6 General Workshop/Conference Announcements

6.1 MSSC2004 Summer School

Ab initio Modeling in Solid State Chemistry

September 20-24, 2004 - London, UK

http://www.cse.clrc.ac.uk/events/MSSC2004/

Directors:

N.M. Harrison, Imperial College and CCLRC Daresbury Laboratory
R. Dovesi, University of Torino (Italy)
F. Corá, The Royal Institution of Great Britain

The Computational Science Department of the Daresbury Laboratory, the Department of Chemistry of Imperial College, and the Davy Faraday Laboratory of the Royal Institution organize a Summer School on the ab initio simulation of crystalline solids. The school is addressed to PhD students, Post-Docs and researchers with interests in solid state chemistry, physics, materials science, surface science and catalysis, and will provide an overview of the possibilities offered by current ab initio quantum mechanical techniques when applied to characterize crystalline solids. The capabilities of CRYSTAL03 will be illustrated, with hands-on tutorials organised in the afternoon sessions.

Outline of the Subjects

+ Introductory lectures:

- Space groups and point symmetry
- Reciprocal space and Bloch functions
- Models, Hamiltonians and Basis sets

+ The structure of the CRYSTAL code: basis set, Hamiltonians, accuracy.
+ Total energy and related quantities: equations of state, solid state reactions, phase transitions, elastic constants and phonon spectra.
+ Geometry optimization: strategies and techniques.
+ One electron properties: DOS, band structure, charge and spin density maps, electrostatic potential.
+ Localized crystalline orbitals - Wannier Functions.

+ Dielectric properties.
+ Spin polarized solutions: magnetism and EPR parameters Surface.
+ Chemistry and heterogeneous catalysis: models, accuracy and limitations.
+ Local defects in solids: the observables of interest; the possible strategies.
+ Post-HF ab initio techniques for crystals.

Program and Location

The school will last five days (20-24 September 2004) and will be held at the Department of Chemistry of Imperial College, in central London (UK). The morning sessions will be devoted to lectures by experts. The afternoons will be dedicated to practical sessions. In the tutorials it will be shown how to obtain information concerning various observables using the CRYSTAL code.

There will be the possibility to present posters, that will be discussed during a special session.

Participation is limited to 40 people. A small number of bursaries will be available, to cover the participation fee of researchers from less favourite countries.

Application forms should be downloaded from

http://www.cse.clrc.ac.uk/events/MSSC2004/

and faxed to Damian Jones at +44 (0)1925 603634.

Teachers

Teachers include the main authors of the code CRYSTAL, supported by qualified UK scientists, including:

M. Alfredsson - Dept. of Earth Science, University College London (UK)
B. Civalleri - Theoretical Chemistry Group, University of Torino (Italy)
F. Corá - Davy Faraday Research Laboratory, The Royal Institution (UK)
R. Dovesi - Theoretical Chemistry Group, University of Torino (Italy)
N. M. Harrison - CLRC and Imperial College (UK)
W. C. Mackrodt - Dept. of Chemistry, University of St. Andrews (UK)
G. Mallia - Imperial College and The Royal Institution (UK)
B. Montanari - Imperial College and Rutherford Appleton Laboratory (UK)
R. Orlando - Theoretical Chemistry Group, University of Torino (Italy)
C. Pisani - Theoretical Chemistry Group, University of Torino (Italy)
C. Roetti - Theoretical Chemistry Group, University of Torino (Italy)
V. R. Saunders - CLRC Daresbury Laboratory (UK)
B. Searle - CLRC Daresbury Laboratory (UK)
A. Wander - CLRC Daresbury Laboratory (UK)

Deadlines and Contact Information:

Submission of application form: May 31st Notification of acceptance: June 15th
Submission of poster abstracts: July 15th

E-mail: D.R.Jones@dl.ac.uk
Telephone: +44 (0)1925 603805
Fax: +44 (0)1925 603634

Postal address:
MSSC2004
c/o Mr Damian Jones
CCLRC Daresbury Laboratory
Keckwick Lane
Daresbury
Warrington
WA4 4AD
UK

6.2 EPSRC/IOP Theory of Condensed Matter Summer School

Physics by the Lake

Ambleside, Cumbria, UK 29 August-10 September 2004

The aim of the School is to teach material underpinning research in modern condensed matter theoretical physics, introducing concepts, techniques and problems that are the subject of current research in the field, illustrated with a wide range of applications. The School is directed towards postgraduate students in condensed matter theory, and theory-inclined students in experimental condensed matter physics, typically in their first or second year of research. The lecture courses will be presented by active members of the community, and accompanied by example classes with full tutorial support. A series of specialist seminars will highlight current research and applications.

This is the eighth in a successful series, following previous schools held in Ambleside (1997-1998 and 2000-2003) and Harlech (1999). The latest EPSRC/IOP Theory of Condensed Matter Summer School will again be held at The University College of St Martin, Ambleside, between 29 August and 10 September 2004. Ambleside is in Cumbria, on the shores of Lake Windermere, in the heart of the beautiful Lake District National Park. The School is organized by the UK Institute of Physics Theory of Condensed Matter Group, and sponsored by the UK EPSRC.

Lecturers:

Prof Graeme Ackland (Edinburgh) Computational Modelling;
Prof James Annett (Bristol) Superconductivity;
Dr.Nick D'Ambrumenheil (Warwick) Correlated Quantum Systems;
Dr Martin Evans (Edinburgh) Statistical Physics;
Prof. Peter King (Imperial) Applications of Statistical Physics;
Prof. Angus Mackinnon (Imperial) Mesoscopics;
Dr Ian Morrison (Salford) Electronic Structure;
Prof. Philip Russell (Bath) Photonics;
Prof. Julia Yeomans (Oxford) Soft Condensed Matter;

Further details available at

http://www.bath.ac.uk/physics/summer/

or contact:

Dr Simon Crampin
Hon. Secretary, IOP Theory of Condensed Matter Group,
Department of Physics, University of Bath, Bath BA2 7AY, UK
(email: s.crampin@bath.ac.uk)

7 General Job Announcements

The Theory Department of the Fritz-Haber-Institut offers a

Head of an Independent Junior Research Group on "Ab initio Statistical Mechanics"

position (W2).

The position is available beginning October 1, 2004.

An excellent knowledge of the electronic structure theory (density functional theory), statistical mechanics (Monte Carlo methods), and of surface physics and the physics of nanostructured systems is required. The candidate should have a track record of scientific achievement (the German habilitation or equivalent standard of scientific activities). The research group leader is expected to possess the necessary scientific skills and leadership qualities to initiate new projects and supervise training and work of students and post-docs.

The appointment will be for 5 years; an extension is possible. Remuneration will be paid at the associate professor level (W2) with all fringe benefits of the German public sector. The position will include an independent budget for personnel, running costs, and investment, office space for students and postdocs, and use of the research infrastructure of the Department of Theory and the institute.

The Max Planck Society seeks to increase the number of women in those areas where they are underrepresented and therefore explicitly encourages women to apply. The Max Planck Society is committed to employing more handicapped individuals and especially encourages them to apply.

Please send your application by June 30, 2004 to:

Fritz-Haber-Institut der MPG
Executive Director
Faradayweg 4-6
D-14195 Berlin
Germany

Postdoctoral position in Condensed Matter Theory

INFM-S3 and Physics Department

University of Modena and Reggio Emilia (Italy)

Nanoscience Group

Field(s): computational physics, condensed matter, theoretical physics

Application deadline: August 31, 2004

Contact: Prof. Elisa Molinari, Prof. Franca Manghi

E-mail: molinari@unimo.it, manghi@unimo.it

Address: INFM-S3 c/o Dipartimento di Fisica, Universita' di Modena e Reggio E.

Job description:

Applications are invited for a postdoctoral research position in solid-state physics at the INFM-S3 research center (c/o Physics Department, University of Modena and Reggio Emilia), within the RTN EU Contract “EXCITING” No. HPRN-CT-2002-00317. The focus of the work will be the microscopic description of electronic excitations in low-dimensional systems (nanostructured and/or (bio)molecular systems). The candidate should have a Ph.D. in computational/theoretical physics with an excellent academic background and good computational skills. Applicants with research experience on ab-initio (Density-Functional Theory) calculations are strongly encouraged to apply.

Additional information:

http://www.s3.infm.it

http://www.nanoscience.unimo.it

http://www.exciting.physics.at

The appointment will be for 18 months, starting at the latest in October 2004. Owing to EU regulations, only candidates from EU countries and associate states can be considered. Interested applicants should submit a curriculum vitae and arrange for letters of reference to be sent to the address below:

Prof. Elisa Molinari

INFM-S3 and Dipartimento di Fisica,

Universita' di Modena e Reggio E.

via Campi 213/A

41100 Moden, Italy

Ph.D. studentship: "Structural Modelling of the Biological Interface with Materials"

Department of Physics and Astronomy University College London

Condensed Matter and Materials Physics Group and London Centre for Nanotechnology

A large collaborative project has just been announced by a consortium of materials simulation groups at University College London (UCL), Cambridge University and Warwick University. The aim of the project is to develop and apply techniques for modelling the interaction of biomolecules with material surfaces in water. The project has wide-ranging relevance to contemporary issues in medicine, nanotechnology and materials modelling, including:

* Biocompatibility: the acceptance of medical implants by the human body depends on the interaction of proteins with the surface of the implant material.

* Biomineralization: the growth of mineral structures in the living world (bones, sea-shells...) depends on the self-assembly of these structures on an organic scaffold. A deeper understanding of these processes will help us to develop completely new ways of fabricating materials by imitating the natural world - 'biomimetics'.

* Protein-directed assembly of nano-particles, which is one of the possible routes towards the controlled large-scale fabrication of nano-structures.

Progress in these and other areas will be greatly aided by the ability to model the interaction between organic molecules and the surfaces of ceramic materials.

In the framework of this collaborative project, we wish to appoint as soon as possible a Ph.D. student to work with Professor Mike Gillan and Dr. Dorothy Duffy in the Condensed Matter and Materials Physics group at UCL. The aim of the Ph.D. project will be to use computer simulation to study the interface between water and various minerals, including calcite (calcium carbonate) and barite (barium sulphate), and the adsorption of simple organic molecules at this interface. We will use state-of-the-art methods based on first-principles quantum mechanics, in order to develop systematic ways of modelling the interactions. The methods we develop will feed into the other parts of the collaborative project being carried out in Cambridge and Warwick.

The Ph.D. project will provide excellent training in a rapidly expanding field of fundamental importance for future technologies. The CMMP group is one of the major condensed-matter groups in the UK. Members of CMMP are also members of the new London Centre for Nanotechnology (LCN), a joint enterprise between UCL and Imperial College. Details of CMMP and the LCN can be found at their web-sites

http://www.cmmp.ucl.ac.uk

and

http://www.london-nano.ucl.ac.uk.

The Ph.D. position is support by an EPSRC project studentship. To be eligible for this position and the associated studentship you should be a national of one of the countries in the European Union. You should have, or expect to obtain, a degree (at least class 2(i) or 2(ii) with MSc, or equivalent) in a relevant discipline of the physical sciences. Unless there are exceptional circumstances, the degree should have been awarded for a four-year course.

Enquiries should be made in the first instance to Professor Mike Gillan (m.gillan@ucl.ac.uk), attaching a c.v. and the names and e-mail addresses of at least two academic referees.

PhD Studentship: "First-principles studies of nanostructured materials"

With Philip Lindan, University of Kent, Canterbury CT2 7NR, UK

Starting September 2004

I am looking for a PhD student to start in September 2004. You will learn about first-principles computer simulation and apply it to challenging problems at the nanoscale. First-principles methods solve the quantum mechanics of an assembly of atoms using no adjustable parameters or experimental fitting data. Thanks to this they have the capacity to deliver predictions about the properties of matter. These predictions are pivotal in striving to understand the radically-different properties of materials whose dimensions are of the order of a few atomic spacings.

The two main areas from which to choose your topic are: filled carbon nanotubes, and nanocrystalline oxides. In both cases you will benefit from strong collaborative links with experimentalists in the UK, and from being a part of European networks of excellence, offering wide scope for enriching your work. We have top-grade access to the best high-performance computing facilities, and support from the two major UK simulation consortia. This is a very well-supported position, and a great chance to train in a high-impact, rapidly-growing and exciting field.

I am happy to answer informal enquiries via email at p.lindan@kent.ac.uk , and to advise you of the application procedure.

Closing date: Friday 9th. July

Postdoctoral and PhD Positions: Proton Transfer in Biosystems Theoretical Chemistry, Ruhr-Universitaet Bochum, Germany

Applications are invited for research associate and PhD positions. The preferred starting date would be summer/fall 2004.

The ideal candiate would have significant experience in electronic structure theory, ab initio MD, mixed quantum/classical simulation, proton transfer and biomolecular systems. She or he would work primarily with dynamical QM/MM (Car-Parrinello) simulation techniques, including code development, in order to understand proton transfer along hydrogen bonded networks in complex biomatrices. In addition, he/she can also be involved in other challenging research projects including both method development and large-scale applications.

Information on the techniques used and developed are accessible via

http://www.theochem.rub.de/go/cprev.html.

The Theoretical Chemistry Center at RUB offers an exciting interdisciplinary environment with excellent working conditions (including several parallel platforms and a 500 GFlop/s SCI machine in house).

Candidates should send a detailed resume including an outline of their research achievements and interests as well as contact information for academic references as one pdf file to

office@theochem.rub.de.

Consideration of candidates will begin immediately and continue until the position is filled. The University particularly welcomes applications from women.

Dominik Marx
Lehrstuhl fuer Theoretische Chemie
Ruhr-Universitaet Bochum
44780 Bochum, Germany
Email: office@theochem.rub.de
Fax: ++49 234 32 14045
URL: http://www.theochem.rub.de/

Postdoctoral Position: Surface Science at Extreme Conditions
Theoretical Chemistry, Ruhr-Universitaet Bochum, Germany

Applications are invited for a research associate position. The preferred starting date would be fall/winter 2004.

The ideal candiate would have significant experience in electronic structure theory, ab initio MD, surface science, and heterogeneous catalysis. She or he would work primarily on using dynamical (Car-Parrinello) electronic structure methods in order to understand the complex processes involved in chemical reactions occurring at surfaces in contact with dense/hot water. In addition, he/she can also be involved in other challenging research projects including both method development and large-scale applications.

Information on the techniques used and developed are accessible via

http://www.theochem.rub.de/go/cprev.html.

The Theoretical Chemistry Center at RUB offers an exciting interdisciplinary environment with excellent working conditions (including several parallel platforms and a 500 GFlop/s SCI machine in house).

Candidates should send a detailed resume including an outline of their research achievements and interests as well as contact information for academic references as one pdf file to

office@theochem.rub.de.

Consideration of candidates will begin immediately and continue until the position is filled. The University particularly welcomes applications from women.

Dominik Marx
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PhD POSITION IN THEORETICAL SOLID STATE PHYSICS

Institute of Theoretical Physics,
University of Frankfurt am Main, Germany
Condensed Matter Theory Group

Field(s): theoretical solid state physics

Job Description:

Applications are invited for a Ph.D. research position in theoretical solid state physics at the Institute of Physics of the University of Frankfurt, Germany. The candidate should have a Diplom/M.S. in theoretical Physics with an excellent academic background and good computational skills.

The focus of the work will be the microscopic description of the behavior of strongly-correlated electron systems by a combination of Density Functional Theory calculations and Many-body methods. For details please see our webpage. The appointment will be for three years starting in Summer 2004.

Interested applicants should submit a curriculum vitae and arrange for three letters of reference to be sent to the address below.

Prof. Dr. Roser Valenti
Institut für Theoretische Physik,
Universität Frankfurt,
Robert-Mayer-Strasse 8,
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Post-doctoral Positions

Lawrence Livermore National Laboratory (LLNL), California, USA

Applications are invited for post-doctoral positions in the

Quantum Simulations Group (QSG) at the Lawrence Livermore National Laboratory (LLNL) in California, USA,

in the area of quantum simulations of nanostructures and quantum simulations of fluids and solids under extreme conditions.

The candidate should have a PhD in Physics, Chemistry or Materials Science and preferably some experience in electronic structure calculations and quantum simulations, e.g. ab-initio molecular dynamics and quantum monte carlo techniques.

Salary ranges from 60K to 65K per year (before taxes).

Information about the QSG at LLNL can be found at:

http://www-pat.llnl.gov/Research/qsg/

If interested to apply or to nominate candidates, please contact Giulia Galli (galli@llnl.gov) or apply at

`http://jobs.llnl.gov/prod_index.html` .

(Posting number is 001946).

8 Abstracts

Cu L_3 x-ray absorption spectroscopy and the electronic structure of minerals: Spectral variations in non-stoichiometric bornites, Cu_5FeS_4

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Abstract

Synthetic bornites with compositional variations within 2 at% of the stoichiometric formula of Cu_5FeS_4 show dramatic changes in their Cu $L_{2,3}$ absorption spectra, determined using synchrotron radiation. Many of the bornites show L_3 spectra with a strong peak at 932.6 eV and a broader region of intensity in the region 933 eV to 940 eV with specific features at 935.6 eV and 938 eV. However, the intensity of the peak at 932.6 eV varies greatly, and there is a clear correlation between the loss of intensity of this peak and the gain of intensity of the peak at 935.6 eV. EPMA reveals that this change in relative intensities is directly correlated to the excess of valence electrons present in the bornites, calculated the using formal valences of Cu^+, Fe^{3+} and S^{2-} [The Cu $L_{3,2}$ spectra are typical of Cu(I) sulfides and Fe L_3 edge spectra reveal the Fe to be present as Fe^{3+}]. A simple cluster model can explain this transfer in intensity among the three different peaks. The three peaks in the bornite XAS $2p$ spectrum can be assigned (in order of increasing energy) to the final states $cd^{10}Ls^2$, $cd^{10}s$, and cd^9s^2; the transition to an empty s state is possible for all three configurations, $d^{10}Ls \rightarrow cd^{10}Ls^2$, $d^{10} \rightarrow cd^{10}s$, and $d^9s \rightarrow cd^9s^2$. When the amount of excess electrons increases, the holes are filled and the weight of the $d^{10}Ls$ and d^9s configurations will decrease, whereas that of the d^{10} configuration will increase, exactly as observed in the spectra. This study demonstrates the value of metal L-edge spectroscopy, and Cu $L_{2,3}$ spectra in particular, in investigations of minerals with variations in stoichiometry and chemistry; the information derived on electronic structure can be used to explain bonding and other variations in physical and chemical properties of the minerals.

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Reprints available from g.van_der_laan@dl.ac.uk

Soft x-ray resonant magnetic scattering from a Ni layer with modulated magnetic anisotropy in a chemically homogeneous Cu/Ni/Cu system

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European Synchrotron Radiation Facility, Grenoble, France

Abstract

We report soft x-ray resonant magnetic scattering studies on a magnetically modulated, but nominally chemically homogenous, 5 nm Ni layer in a Cu/Ni/Cu/Co/NiO/GaAs(110) system. We were able to estimate the main chemical structure of the sample on the basis of the results from specular reflectivity and rocking scans, probing the scattering vector components in the perpendicular and in-plane direction to the Ni wires, respectively. The magnetic scattering using polarized x-rays demonstrates the magnetic modulation of the Ni layer.

Published in J. Synchrotron Rad. **11**, 254–260 (2004).
Manuscript available from g.van_der_laan@dl.ac.uk

The Electronic Structure of Europium Chalcogenides and Pnictides

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Abstract

The electronic structure of some europium chalcogenides and pnictides is calculated using the *ab-initio* self-interaction corrected local-spin-density approximation (SIC-LSD). This approach allows both a localised description of the rare earth $f-$electrons and an itinerant description of s, p and d-electrons. Localising different numbers of f-electrons on the rare earth atom corresponds to different nominal valencies, and the total energies can be compared, providing a first-principles description of valency. All the chalcogenides are found to be insulators in the ferromagnetic state and to have a divalent configuration. For the pnictides we find that EuN is half-metallic and the rest are normal metals. However a valence change occurs as we go down the pnictide column of the Periodic Table. EuN and EuP are trivalent, EuAs is only just trivalent and EuSb is found to be divalent. Our results suggest that these materials may find application in spintronic and spin filtering devices.

(Submitted to J. Phys. CM; cond-mat/0403763)

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Self-interaction corrected relativistic theory of magnetic scattering of x rays: Application to praseodymium

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Abstract

Resonant magnetic scattering spectra of x-rays from elemental praseodymium with the face-centered cubic crystal structure are computed and analyzed. The scattering amplitudes are calculated using a standard time-dependent perturbation theory to second order in the electron-photon interaction vertex. In order to calculate the cross section reliably an accurate description of the electronic states in the material under investigation is required and this is provided by the density functional theory (DFT) employing the Local Spin Density Approximation combined with the self-interaction corrections (SIC-LSD). The magnetic x-ray resonant scattering (MXRS) theory has been implemented in the framework of the relativistic spin-polarized LMTO-ASA band structure calculation method. It is shown that the theory quantitatively reproduces the dependence on the spin and orbital magnetic moments originally predicted qualitatively (Blume, J. Appl. Phys, **57**, 3615, (1985)) and yields results that can be compared directly with experiment.

(Submitted to Phys. Rev. B; cond-mat/0404543)
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Changes in the electronic structure of cerium due to variations in close packing

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Abstract

Here we use electron energy-loss spectroscopy in a monochromated transmission electron microscope with 100 meV energy resolution and 2 Å spatial resolution to show that the electronic structure of face centered cubic γ and double hexagonal close-packed β cerium are considerably different, contrary to previous assumptions in literature. These results are supported by synchrotron-radiation-based x-ray absorption, multielectronic atomic spectral simulations, and local density approximation calculations, illustrating that changes in {111} stacking sequences can drive substantial electronic changes in close-packed phases of cerium that have a similar atomic volume.

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Surface effects in Mn $L_{3,2}$ x-ray absorption spectra from (Ga,Mn)As

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Abstract

We have identified a Mn-rich layer on the surface on (Ga,Mn)As thin films which significantly influences soft x-ray absorption measurements. The Mn $L_{3,2}$ x-ray absorption spectra of the untreated films show a strong multiplet structure, consistent with earlier observations and characteristic of MnO. After removal of the surface layer, the multiplet structure is less pronounced and the spectrum is shifted to ~0.5 eV lower photon energy. Comparison with calculated spectra imply a localized Mn ground state for the untreated sample and a hybridized ground state after etching. In addition, a large x-ray magnetic circular dichroism is observed at the Mn $L_{3,2}$ edge in the etched film. These results may explain several peculiarities of previously reported x-ray absorption studies from (Ga,Mn)As.

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Magnetic anisotropy of aligned magnetic stripe domains in FePd studied by soft x-ray resonant magnetic scattering, magnetic force microscopy and micromagnetic modeling

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Abstract

FePd thin film samples with different perpendicular magnetic anisotro-pies have been studied with magnetic force microscopy (MFM), micromagnetic calculations and soft x-ray resonant magnetic scattering (SXRMS). The competition between perpendicular magnetic anisotropy (PMA) and shape anisotropy leads to the formation of highly ordered stripe domain patterns with a magnetization component perpendicular to the filmplane. The magnetic stripes with a period of $sim100$ nm, which are seen in the MFM images and can be modeled by micromagnetic calculations, give rise to magnetic peaks in the diffraction pattern. Closure domains occur in samples with a low to medium PMA, while a high PMA inhibits their formation. The in-plane magnetization component of the closure domains is not observable with MFM. In the presence of closure domains the interference between the scattering amplitudes in SXRMS from perpendicular and in-plane magnetized domains gives rise to a circular dichroism in the transverse geometry, where the scattering plane is along the stripes.

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Manuscript available from g.van_der_laan@dl.ac.uk

Mn $L_{3,2}$ x-ray absorption from (Ga,Mn)As and (Ga,Mn)N

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Abstract

We present Mn $L_{3,2}$ x-ray absorption measurements from two dilute magnetic semiconductor systems: *p*-type (Ga,Mn)As, which is ferromagnetic with $T_C = 140$ K; and n-type (Ga,Mn)N, in which the magnetic impurities are predominantly paramagnetic. After removing a Mn-rich oxide surface layer by chemical etching, the Mn $L_{3,2}$ spectra from (Ga,Mn)As appear less localized than in previous reports, which is ascribed to screening due to *p*-*d* hybridization. Our results suggest that previous studies may have been influenced by the quality of the surface. In contrast, in the (Ga,Mn)N film the Mn ground state is closer to pure d^5.

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June 1, 2004 – Volume **95**, Issue 11, pp. 7166-7168.
Reprints available from g.van_der_laan@dl.ac.uk

Comment on “Field-Induced Magnetic Circular Dichroism in Paramagnetic Solids: A New Magneto-Optical Effect”

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Abstract

Recently, Ebert and Man’kovsky [Phys. Rev. Lett. **90**, 077404 (2003)] showed theoretically that x-ray magnetic circular dichroism (XMCD) occurs also in paramagnetic solids in the presence of an external field and they proposed modified sum rules for the spin and orbital susceptibility. However, not only are these modified sum rules a natural extension of the old ones, the “new magneto-optical effect” for paramagnets has already been well established experimentally. Moreover, from the experimental verification of the sum rules it is clear that the theoretical analysis by Ebert and Man’kovsky contains a major inconsistency in the way the spin-orbit interaction is evoked to account for the different magnetic contributions in paramagnets.

Accepted for publication in Physical Review Letters.
Preprints available from g.van_der_laan@dl.ac.uk

The hallmark of perfect graphene

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Abstract

Using first-principles calculations we show that the adsorption of atomic hydrogen on graphene opens a substantial gap in the electronic density of states is which lies a spin-polarized gap state. This spin is quenched by the presence of a rotated C-C bond (a Stone-Wales defect) adjacent to or distant from the H atom. We explain these findings and discuss the implications for nanotubes and magnetic nanographene. Furthermore, we demonstrate that the combined effect of high curvature and a Stone-Wales defect makes H_2 chemisorption close to being thermodynamically favorable.

(accepted by: Physical Review Letters)
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Ab initio study of the half-metal to metal transition in magnetite

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Abstract

A first-principles theoretical study of the stability of the half-metallic ground state of magnetite with respect to strain and substitutional doping is presented. In response to uniaxial, epitaxial, or hydrostatic strain, magnetite undergoes a half-metal to metal transition due to a shift of the Fermi level from inside the band gap of the majority-spin electrons to below the top of the valence band, so that both spin channels become metallic. A qualitatively similar metallic character is also found for Mn substitutionally doped magnetite with the stoichiometry $Fe_{2.5}Mn_{0.5}O_4$.

(submitted to Phys. Rev. Lett.)
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Structure determination of isolated metal clusters via far-infrared spectroscopy

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Abstract

We present a new method for the size selective structure determination of small isolated metal clusters in the gas-phase. The technique is applied to cationic vanadium clusters containing 6 to 23 atoms, whose far infrared (FIR) absorption spectra are measured in the 140–450 cm^{-1} spectral range. The spectra are unique for each cluster size and are true fingerprints of the cluster's structure. By comparing the experimental spectra to spectra obtained from density-functional theory, the geometric cluster structure can be identified.

(accepted by: Physical Review Letters)
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On the accuracy of DFT for describing hydrogen bonds: dependence on the bond directionality

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Abstract

A set of representative hydrogen bonded dimers has been studied employing density functional theory (DFT) in the Perdew, Burke and Ernzerhof generalized gradient approximation (PBE-GGA). Our results for hb strengths and hb geometry parameters show a good agreement with those obtained by Moller-Plesset (MP2) or Coupled-Clusters (CC) methods. We observe that the reliability of DFT for the description of hbs is closely connected to the bond directionality: With increasing deviation from linearity DFT errors for the association energies and geometry parameters increase.

(submitted to: J. Phys. Chem. A (in press))
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Phonon-spectra and thermodynamic properties of the infinite polyalanine α-helix: A DFT based harmonic vibrational analysis

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Abstract

We have performed a density-functional theory harmonic vibrational analysis of the infinite polyalanine α-helix. The calculated phonon dispersion spectrum shows excellent agreement to available experimental data, except for the high frequency hydrogen stretching modes which show characteristic shifts due to anharmonic effects. A major advantage compared to previously performed empirical force field studies is that long range effects such as electrostatic interaction and polarization are fully taken into account. Our results indicate that these effects are crucial to accurately describe the low frequency acoustical branches and lead to a significantly better agreement with experiment for the specific heat in the low temperature range.

(submitted to: Physical Review Letters)

Contact person: Lars Ismer (ismer@fhi-berlin.mpg.de)

Ab initio atomistic thermodynamics and statistical mechanics of surface properties and functions

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Abstract

Previous and present "academic" research aiming at atomic scale understanding is mainly concerned with the study of individual molecular processes possibly underlying materials science applications. In investigations of crystal growth one would for example study the diffusion of adsorbed atoms at surfaces, and in the field of heterogeneous catalysis it is the reaction paths of adsorbed species that is analyzed. Appealing properties of an individual process are then frequently discussed in terms of their direct importance for the envisioned material function, or reciprocally, the function of materials is somehow believed to be understandable by essentially one prominent elementary process only. What is often overlooked in this approach is that in macroscopic systems of technological relevance typically a large number of distinct atomic scale processes take place. Which of them are decisive for observable system properties and functions is then not only determined by the detailed individual properties of each process alone, but in many, if not most cases also the interplay of all processes, i.e. how they act together, plays a crucial role. For a *predictive materials science modeling with microscopic understanding*, a description that treats the statistical interplay of a large number of microscopically well-described elementary processes must therefore be applied. Modern electronic structure theory methods such as density-functional theory (DFT) have become a standard tool for the accurate description of the individual atomic and molecular processes. In what follows we discuss the present status of emerging methodologies which attempt to achieve a (hopefully seamless) match of DFT with concepts from statistical mechanics or thermodynamics, in order to also address the interplay of the various molecular processes. The new quality of, and the novel insights that can be gained by, such techniques is illustrated by how they allow the description of crystal surfaces in contact with realistic gas-phase environments, which is of critical importance for the manufacture and performance of advanced materials such as electronic, magnetic and optical devices, sensors, lubricants, catalysts and hard coatings.

(to appear in: Handbook of Materials Modeling Vol. **1**, Fundamental Models and Methods, (Ed.) Sidney Yip, Kluwer Publishing)

Contact person: Karsten Reuter (reuter@fhi-berlin.mpg.de)

The C 1s NEXAFS spectrum of benzene below threshold: Rydberg or valence character of the unoccupied σ orbitals

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Abstract

High-resolution C 1s NEXAFS spectra of benzene and benzene-d_6 were measured in total ion yield, and compared with the results of state-of-the-art density functional theory (DFT) calculations. Excellent agreement between experiment and theory is obtained in many details. The spectrum below threshold is dominated by four resonances, where two can be assigned to excitations into π-orbitals, while the other two originate from excitations into σ-orbitals. The π-type resonances have been ascribed in previous work unanimously to antibonding valence orbitals, while the character of the σ-type resonances is still the subject of discussion. Based on the present theoretical results and the observed vibrational fine structures both σ-type final-state orbitals are assigned to be of valence character with minor admixtures of Rydberg character.

(submitted to:) Chemical Physics Letters
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Representing high-dimensional potential-energy surfaces for reactions at surfaces by neural networks

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Abstract

The determination of dissociative adsorption probabilities based on first-principles total-energy calculations requires a numerically efficient and accurate interpolation scheme in order to be able to run a sufficient number of trajectories. Here we present a neural network scheme for the construction of a continuous potential-energy surface (PES). We illustrate the accuracy and efficiency of our method for H_2 interacting with the (2×2) potassium covered Pd(100) surface. The sticking probability of H_2/K(2×2)/Pd(100) is determined by molecular dynamics simulations on the neural network PES and compared to results using an independent analytical interpolation.

(submitted to:) Chem. Phys. Lett.

Contact person: Matthias Scheffler (scheffler@fhi-berlin.mpg.de)

Design of Potential Hydrogen-storage Material using First-principle Density-functional Calculations

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Abstract

The crystal and electronic structures of the entire series of alkali aluminium and alkali gallium tetrahydrides ($AB\mathrm{H}_4$; $A =$ Li, Na, K, Rb or Cs; $B =$ Al or Ga) are systematically investigated using an *ab initio* projected augmented plane-wave method. For structural stability studies we have considered several possible structural modifications, and reproduced successfully the equilibrium structures for the known phases $\mathrm{LiAlH_4}$, $\mathrm{NaAlH_4}$, $\mathrm{KAlH_4}$, $\mathrm{NaGaH_4}$, and $\mathrm{KGaH_4}$. Moreover, we predict the equilibrium structures of the other unknown members of this series. $\mathrm{RbAlH_4}$, $\mathrm{CsAlH_4}$, $\mathrm{RbGaH_4}$, and $\mathrm{CsGaH_4}$ should crystallize with the $\mathrm{KGaH_4}$-type structure, and $\mathrm{LiGaH_4}$ with the $\mathrm{NaGaH_4}$-type structure. According to the density of states all these compounds have non-metallic character with a finite band gap of around 5 eV. Charge-density plot and electron-localization-function analyses show that the $[B\mathrm{H}_4]$ sub-units almost look like a separate molecular species spread over the A matrix. An ionic type of interaction is present between the A and the $[B\mathrm{H}_4]$ units. Crystal-orbital Hamilton-population analyses reveal that the interaction between the B and H atoms is stronger than the other possible interactions present in these compounds.

(Cry. Growth & Design 4 , 471-477 (2004).)
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Structural stability and electronic structure for Li_3AlH_6

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Abstract

The structural stability and electronic structure of the potential hydrogen-storage material Li_3AlH_6 have been investigated up to 90 GPa using density-functional total-energy calculations. At ambient condition Li_3AlH_6 stabilizes in space group $R\bar{3}$. The structure consists of isolated, close-to-regular $[AlH_6]^{3-}$ octahedra, which are connected via six-coordinated Li. We predict that this α phase of Li_3AlH_6 undergoes three successive structural phase transitions on application of pressure: α to β at 18.64 GPa, β to γ at 28.85 GPa, and γ to δ at 68.79 GPa. All modifications of Li_3AlH_6 should have non-metallic character with estimated bandgaps varying between 2.72 and 4.12 eV.

(Phys. Rew. B 69 , 020104(R) (2004)).

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Spin, charge, and orbital orderings in oxides with dual-valent transition-metal ions

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Abstract

Using generalized-gradient-corrected full-potential density-functional calculations we have studied the electronic structure and magnetic properties of $YBaMn_2O_5$, $Sr_4Fe_4O_{11}$, and $Ca_3Co_2O_6$. In these phases the $3d$ transition-metal ions have dual valence. We have studied the electronic structure using site-, angular momentum-, and orbital-projected density of states. The charge and orbital ordering are analyzed in terms of the calculated electron-density distribution, charge density, and electron localization function. The oxygen vacancy, cation radii, and crystal field effects are found to play an important role for the various ordering phenomena in these compounds.

(J. Ceramics International (in press 2004)).

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Chemical-bonding and High-pressure Studies on Hydrogen-storage Material

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Abstract

From gradient-corrected, all-electron, full-potential, density-functional calculations, including structural relaxation, it is shown that the metal-hydride series $RT\mathrm{InH}_{1.333}$ (R = La, Ce, Pr, or Nd; T = Ni, Pd, or Pt) violate the "2-Å" rule as well as the hole-size requirement. These hydrides possess unusually short H−H separations which in the most extreme case for $\mathrm{LaPtInH}_{1.333}$ is as short as 1.454 Å. These findings have been analyzed in terms of charge density, charge transfer, electron-localization function, crystal-orbital Hamilton population, and density of states analyses. From high-pressure studies it is predicted several successive pressure-induced structural transitions in MgH_2 within the 20 GPa range. Calculations have also shown several pressure-induced structural transitions in alkali aluminum tetrahydrides with large volume reductions at the phase-transition points and small energy differences between the ambient-pressure and subsequent high-pressure phases.

(Comput. Mat. Sci. (in press 2004)).

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Electronic Structure and Excited-state Properties of Perovskite-like Oxides

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Abstract

The role of structural distortion, magnetic ordering, and Coulomb-correlation effect on the electronic structure of perovskite-like oxides is analyzed. The density-functional theory (DFT) is originally devised to describe the ground-state properties of materials. However, during recent years it has been shown that the DFT also with success can be used to study excited state properties. We have recently calculated the electronic structure and linear optical properties for the series LaXO$_3$ (X = Sc$-$Cu) and found that the gradient-corrected DFT describes correctly the insulating behavior of the ionic insulator LaScO$_3$ and the charge-transfer insulators LaCrO$_3$, LaFeO$_3$, and LaMnO$_3$ although the band gaps are systematically underestimated. For example, the good agreement between experimental and theoretical reflectivity spectra for LaCrO$_3$ clearly demonstrates that accurate full-potential DFT calculations not only describe the occupied and unoccupied states of the bands well, but also reproduce their characters. We have also calculated XPS, XANES, and magneto-optical spectra for perovskite-like oxides. For Mott-Hubbard insulators such as LaTiO$_3$ and LaVO$_3$ the DFT failed to predict insulating behavior and here the LDA+U method is applied to describe the electronic structure correctly.

(J. Crys. Growth (in press 2004)).

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Effect of oxygen stoichiometry on spin, charge, and orbital ordering in manganites

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Abstract

Using full-potential density-functional calculations we show that oxygen stoichiometry plays an important role on spin, charge, and orbital ordering in manganites. The electronic structure and magnetic properties of $\mathrm{LaBaMn_2O_{5+\delta}}$ have been studied for $\delta = 0$, 0.5, and 1; For $\delta = 0$ and 0.5 the system exhibits charge, orbital, and antiferromagnetic spin ordering, whereas at $\delta = 1$, the charge and orbital orderings disappear but the spin ordering remains. We also bring out an insulator-to-metal transition upon going from $\delta = 0$ to 1. The study suggests that one can manipulate the charge and orbital orderings in certain perovskite-like oxides by merely varying the oxygen stoichiometry and hence design oxides with desired electrical and magnetic properties.

(Phys. Rev. B 69 , 092405 (2004)).
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Search for metal hydrides with short hydrogen-hydrogen separation by *ab-initio* calculations

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Abstract

The present investigation is a part of a series on metal hydrides with extraordinary short H-H separations. The electronic structure, chemical bonding, and ground state properties of RTIn (R = La, Ce, Pr, Nd; T = Ni, Pd, Pt) and their saturated hydrides $R_3T_3\mathrm{In}_3\mathrm{H}_4$ (= $3RT\mathrm{InH}_{1.333}$) are systematically studied using the full-potential linear muffin-tin-orbital method. The effect of the metal matrix on the H-H separation in $RT\mathrm{InH}_{1.333}$ is analyzed in terms of chemical bonding, and bond strength is quantitatively analyzed using the crystal-orbital-Hamilton population. Force and volume optimizations reveal that all these hydrides violate the "2-Å rule". The insertion of hydrogen in the metal matrix causes highly anisotropic lattice changes; a large expansion along c and a small contraction in the a direction. Among the 12 studied hydrides the hypothetical $\mathrm{LaPtInH}_{1.333}$ phase exhibits the shortest H-H separation (1.454 Å). The optimized unit-cell parameters and atomic coordinates fit very well with the experimental findings for $R\mathrm{NiInH}_{1.333}$, R = La, Ce, and Nd. Examination of the effect of the metal matrix on the H-H separation in $RT\mathrm{InH}_{1.333}$ suggests that on a proper choice of alloying element one may be able to reduce the H-H separation below 1.45 Å.

(Phys. Rev. B in press (2004)).

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First-principles study of adsorption of methanethiol on Co(0001)

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Abstract

Investigation of the resident site and the adsorption phase structure of thiolates is of fundamental importance for understanding the formation of self-assembled organic monolayers on metal substrate surfaces. In the present study, we have investigated adsorption of methanethiol, CH_3SH, on the ferromagnetic Co(0001) surface using density functional theory calculations. We find that the dissociative adsorption of CH_3SH forming an adsorbed methylthiolate (CH_3S) and an adsorbed H atom is energetically favorable, and that the CH_3S molecule adsorbed at the three-fold fcc and hcp hollow sites is most stable. The adsorption energy at the bridge site is only ~0.2 eV smaller than that at the three-fold hollow site, and the adsorption of CH_3S at the atop site is unstable. For the $(\sqrt{3}\times\sqrt{3})$R30°, (2×2) and (2×3) adsorptions, we find that the S-C bond tends to be normal to the surface, whereas for the (2×1) adsorption it tilts away from the surface normal direction by ~40°. The (2×1) adsorption phase is much less stable. The reduction of the adsorption energy with the increasing coverage is attributed to the repulsive interaction between the adsorbates. Our calculations show that the $(\sqrt{3}\times\sqrt{3})$R30° structure may form in the process of methylthiolate adsorption on Co(0001) due to its adsorption energy being only 0.1 eV lower than that for the (2×2) and (2×3) structures. We find that there is a charge transfer from the substrate surface atoms to the S atoms, and that the S-Co bond is strongly polar. The surface Co atoms bound to S have a magnetic moment of $\sim 1.66\mu_B$, while the surface Co atoms unbound to S have a larger magnetic moment of $\sim 1.85\mu_B$. The S atom in the adsorbed CH_3S acquires a magnetic moment of $\sim 0.08\mu_B$.

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Magnetism of Fe-Co nanostructures on flat and stepped W(110) surfaces: Effects of dimensionality and substrate

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Abstract

We present an ab-initio local-spin-density investigation of the magnetism in nanostructures (ultrathin films, stripes, wires) of Fe, Co, and Fe-Co alloys on flat and stepped W(110) surfaces. Particular attention is devoted to the influence of the reduced dimensionality and of adsorbate-substrate hybridization on the magnetic properties. In pseudomorphic Fe-Co films on flat W(110) interface, hybridization is shown to strongly reduce the tendency towards ferromagnetic ordering with increasing Co content. Reconstructed close-packed Co films remain ferromagnetic. In biatomic Fe-Co stripes on W(970) surface, hybridization effects enhanced by relaxation lead to a rapid decrease of the magnetic energy differences with Co concentration. Monoatomic Fe-Co wires are predicted to order antiferromagnetically at all compositions. We demonstrate that antiferromagnetism results from the combined influence of the reduced dimensionality, the strain in the wires, and of the hybridization with the substrate.

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Coordination dependence of hyperfine interactions at impurities on fcc metal surfaces
Part I: electric-field gradient

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Abstract

We present a comparison between accurate *ab initio* calculations and a high-quality experimental data set (1986-2002) of electric-field gradients of Cd at different sites on Ni, Cu, Pd and Ag surfaces. Experiments found a systematic rule to assign surface sites on (100) and (111) surfaces based on the main component of the electric-field gradient, a rule that does not work for (110) surfaces. Our calculations show that this particular rule is a manifestation of a more general underlying systematic behavior. When looked upon from this point of view, (100), (111) *and* (110) surfaces behave in precisely the same way. The physical mechanism behind the systematics of the EFG for other 5sp impurities (Cd-Ba) at different fcc surfaces sites is revealed, showing in a natural way why the first half of the 5p elements shows a coordination dependence that is opposite with respect to the second half.

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Coordination dependence of hyperfine interactions at impurities on fcc metal surfaces

Part I: magnetic hyperfine field

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Abstract

We present a comparison between accurate *ab initio* calculations and a high-quality experimental data set (1990-2002) of magnetic hyperfine fields of Cd at different sites on Ni surfaces. The experimentally observed parabolic coordination number dependence of this hyperfine field is verified as a general trend, but we demonstrate that individual cases can significantly deviate from it. It is shown that the hyperfine fields of other 5sp impurities at Ni surfaces have their own, typical coordination number dependence. A microscopic explanation for the different dependencies is given in terms of the details of the s-DOS near the Fermi level.

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Theoretical and experimental evidence for a post-perovskite phase of MgSiO3 in Earth's D" layer

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Abstract

The Earth's lower mantle is believed to be composed mainly of (Mg,Fe)SiO3 perovskite, with lesser amounts of (Mg,Fe)O and CaSiO3. But it has not been possible to explain many unusual properties of the lowermost ~150 km of the mantle (the D" layer) with this mineralogy. Here, using ab initio simulations and high-pressure experiments, we show that at pressures and temperatures of the D" layer, MgSiO3 transforms from perovskite into a layered CaIrO3-type 'post-perovskite' phase. The elastic properties of the post-perovskite phase and its stability field explain several observed puzzling properties of the D" layer: its seismic anisotropy, the strongly undulating shear-wave discontinuity at its top and possibly the anticorrelation between shear and bulk sound velocities. This new phase is therefore likely to be a major Earth-forming mineral, and its discovery will change our understanding of the deep Earth's interior.

(Accepted for Nature, in press, 2004)
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9 Presenting New Networks

9.1 NANOQUANTA Network of Excellence

Nanoscale Quantum Simulations for Nanostructures and Advanced Materials

Nineteen Administrative, Postdoctoral and PhD vacancies

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NANOQUANTA is a Network of Excellence, encompassing 10 nodes and over 100 researchers, approved for funding under the European Commission's 6th Framework Programme. The Network is expected to run from 1 June 2004 to 31 May 2008.

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Laboratoire des Solides Irradiés (Paris) (Dr. Lucia Reining)
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Rome: Many-body effects on electronic states and optical properties of nanostructures
Rome: *Ab initio* calculation of optical properties of surfaces
San Sebastián: Response functions, excitons and transport properties of low-dimensional structures
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Please see the network's home page, http://www-users.york.ac.uk/∼rwg3/nanoquanta.html, for links to the full advertisements of each post.

The vacancies shown are funded by the Nanoquanta Network of Excellence grant from the European Commission; such appointments are *not* subject to the age, nationality and residence restrictions that apply, for example, to Marie Curie appointments, and details of eligibility are set by the policy of the employing institution. Each position involves participation in the development of the collaborative infrastructure within the network, in addition to the research topic shown.

For general enquiries about the network, please contact the Coordinator, Rex Godby (rwg3@york.ac.uk).

10 SCIENTIFIC HIGHLIGHT OF THE MONTH

Ideal strength of nano-structured components

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Abstract

The ideal (theoretical) strength was originally defined as the stress or strain at which perfect crystal lattice became mechanically unstable with respect to arbitrary homogeneous infinitesimal deformation. This has been intensely investigated because the ultimate strength without defects is a fundamental mechanical characteristic of materials. In the analyses, the instability criteria have been studied on the basis of elastic constants. Recent developments in computational technology make it possible to analyze the ideal strength on the basis of quantum mechanics. On the other hand, it is well known that the mechanical strength of components is dependent not only on (1) material (atom species), but also on (2) loading condition and (3) structure. Because most studies on the strength in terms of atomic mechanics have focused on the factor (1) (materials), analysis has mainly been conducted on simple crystal consisting of perfect lattices (*e.g.* fcc and bcc) under simple loading conditions (*e.g.* tension), though some have explored the properties of bulk materials with defects (*e.g.* vacancy and grain boundary). Small atomic components (nano-structured components) such as nano-films, nano-wires (tubes) and nano-dots (clusters) possess their own beautiful, defect-free structure, namely ideal structures. Thus, they show characteristic high strength. Moreover, utilizing the structure at the nanometer or micron level is a key technology in the development of electronic devices and elements of micro (nano) electro-mechanical systems (MEMS/NEMS). Therefore, it is important to understand the mechanical properties not only for the sake of scientific interest, but also for engineering usefulness such as design of fabrication/assembly processes and reliability in service. In the other words, the effects of structure (factor (3); *e.g.* film/wire/dot) have to be understood as the basic properties of atomic components. Thus, the definition of ideal strength should be expanded to include the strength at instability of components with ideal structures under various external loads (factor (2)), which provides fundamental knowledge of nano-structured materials. In this paper, we review works on the strength of ideal nano-structured components in terms of factor (3), mainly under tension and discuss the strength criterion of arbitrary atomic structure/material under arbitrary loading.

1 Introduction

The ideal strength (theoretical strength) was originally defined as the stress or strain at which perfect crystal lattice became mechanically unstable with respect to arbitrary homogeneous infinitesimal strain (*e.g.* [1]). It has been intensely investigated because the ultimate strength without defects is a fundamental mechanical characteristic of materials. However, it is well known that there is an eminent difference between the ideal strength and the actual one observed experimentally, and this fact led researchers to the discovery of dislocation which plays important role in crystal plasticity. Because the ideal strength physically means the maximum resistance against the external stress (strain) that the material possesses, such high strength was experimentally observed in only whisker that included few defects. Recently, the strength obtained by nano-indentation is attracting researcher's attention because the extremely localized evaluation reveals properties in a defect-free region (*e.g.* [2,3]).

On the other hand, investigation of the strength of bulk materials and macroscopic structures has long history (*e.g.* [4]), and the number of related articles published runs into astronomical numbers. The key point derived from the experiences is that the mechanical strength of components depends not only on (1) materials (atom species), but also on (2) loading conditions (mechanical boundary conditions) and (3) structure. Although the environmental effect is also prominent, it lies outside the scope in this article. The interactions among those factors complicate the fracture phenomenon, and this is one of the main reasons why it is difficult to understand the "strength" of materials.

(1) Material The ideal strength gives a fundamental insight into factor (1). In early analyses, the focus was on the simple structure of perfect crystal (*e.g.* fcc and bcc) under simple loading conditions (*e.g.* tension) because it was difficult to correctly calculate the interactions among the atoms. In other words, the inter-atomic potential was not reliable for the crystal lattice with complex structures under high strain conditions. The precise analysis became possible in this decade due to the progress in computational resources and the technique for simulating quantum mechanics.

(2) Loading The homogeneous deformation of a crystal is represented by the change in shape of a unit cell, namely the strain, which is the symmetric second order tensor, ε_{xx}, ε_{xy}, ε_{xz}, ε_{yy}, ε_{yz}, and ε_{zz}. Of course, it can be related to the homogeneous stress tensor through proper constitutive equations. At first, researchers were interested in the strength in uni-axial tension; however, the ultimate strength should be understood as a function of the combination of strain components in general. For example, the strength under the shear along the slip direction of the crystal gives us fundamental knowledge of the dislocation nucleation and glide.

(3) Structure The strain tensor can not fully describe the deformation of a crystal when the unit cell includes many atoms, *e.g.* a perovskite structure. That is, the inner displacement plays important role in the deformation [5], signifying that the structure has a strong influence on

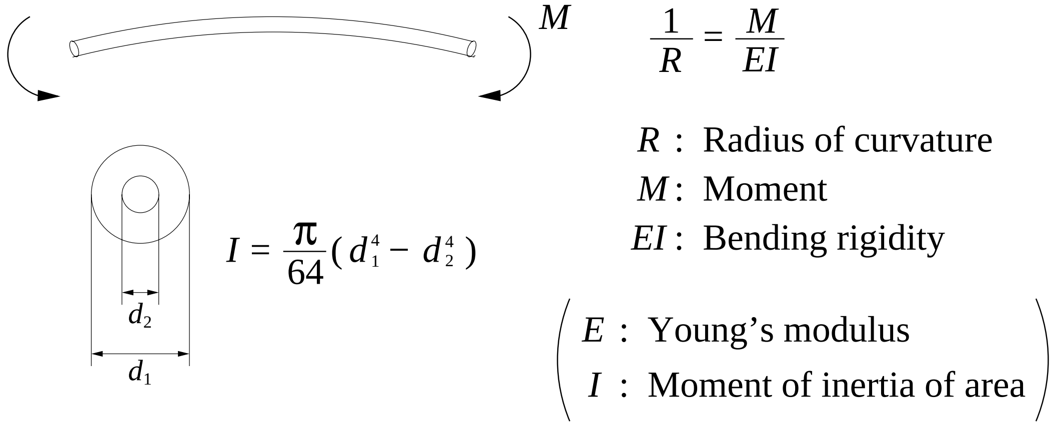


Figure 1: Bending of a tube. The rigidity is dependent on the configuration of tube (the moment of inertia of area).

the strength at the atomic scale. Since we intend to discuss the strength of nano-materials that usually possess characteristic atomic structures, the instability of complex structures becomes a crucial factor in the analysis. In terms of factor (3), we can gain valuable suggestions from the conventional cognition in the mechanical engineering approach on the basis of the continuum mechanics concept. For example, it is well known that the shape of component strongly influences its rigidity and structural strength. The rigidity of the tube illustrated in Fig.1 is determined by the shape of the cross-section (diameter and wall thickness) as well as its elastic coefficient. This, of course, basically represents the carbon nano-tube, which is a typical nano-structured component. Thus, it implies that the structural factor becomes essential, although factor (3) has not been discussed well in past analysis of ideal strength.

In the context of structural analysis, over last decade multi-scale simulation is spotlighted as an *ad hoc* method connecting molecular dynamics to continuum mechanics. However, it is necessary to identify the representative indicator for characterizing the strength of nano-structured materials as fundamental knowledge. Small atomic components such as nano-films, nano-wires (tubes) and nano-dots (clusters) possess their own beautiful, defect-free structures, namely the ideal structure. Consequently, they display characteristic high strength. Moreover, utilizing the structure at the nanometer or micron level is a key technology in the development of electronic devices and elements of micro or nano electro-mechanical systems (MEMS or NEMS). Their complex systems can be decomposed into elements with simple configuration such as the film, wire and dot. For this reason, it is important to understand the mechanical properties not only for the sake of scientific interest but also for engineering applicability such as design of fabrication/assembly processes and reliability in service. As the basic property of component, the effects of structure (factor (3); *e.g.* film/wire/dot) and the loading condition (factor (2); *e.g.* combined load of tension/shear) on the ultimate strength have to be understood. In particular, analysis of instability of component with an ideal defect-free structure gives fundamental insight into the strength of atomic structure. In other words, the definition of ideal strength should be expanded to include the strength at point where nano-structured components with ideal structure (factor (3)) become unstable under various boundary conditions (factor (2)).

Many attractive materials with the ideal structure have been reported not only in scientific journals [6] but also in books (*e.g.* [7-11]; in this series of highlights, there are excellent reviews as well [12, 13]). However, it is not our aim here to review them. We do, however, select some examples investigated by the auth's group, and present characteristic behavior of their strength in Section 2. In terms of the strength, it is clear that mechanical instability under external load or displacement is essential for the analysis of nano-structured materials. Thus, in section 3 we explore the instability criterion.

2 Strength of material with ideal structure

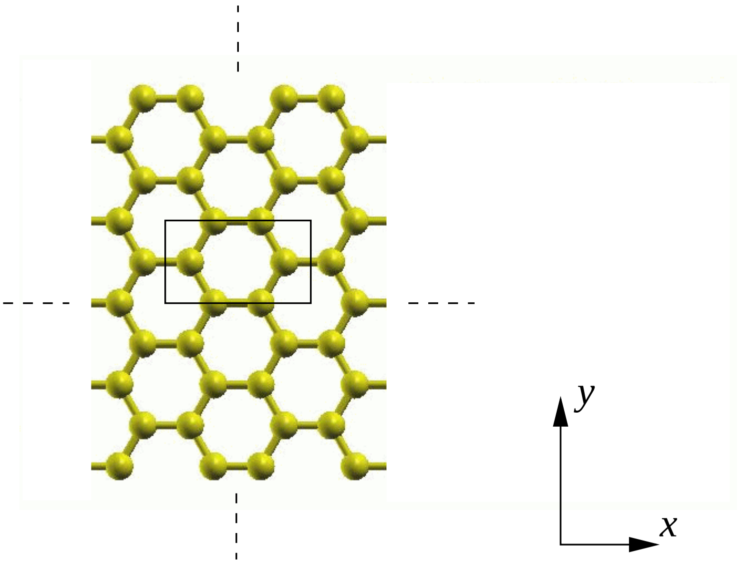
To analyze an "ideal" bulk crystal, the three-dimensional periodic boundary condition was applied to the unit cell. Neglecting one- or two-dimensional periodicity, we can easily obtain typical nano-structured components without defect inside. The periodicity is held in the clean structure, though it is low-dimensional. Thus, there are two-dimensional and one-dimensional materials with ideal structure referred to as "ideal" film and "ideal" wire (or tube), respectively. In this context, the cluster (nano-particle) and the fullerene (cage-like structure) should be classified as zero-dimensional ideal structures.

2.1 Two-dimensional structure

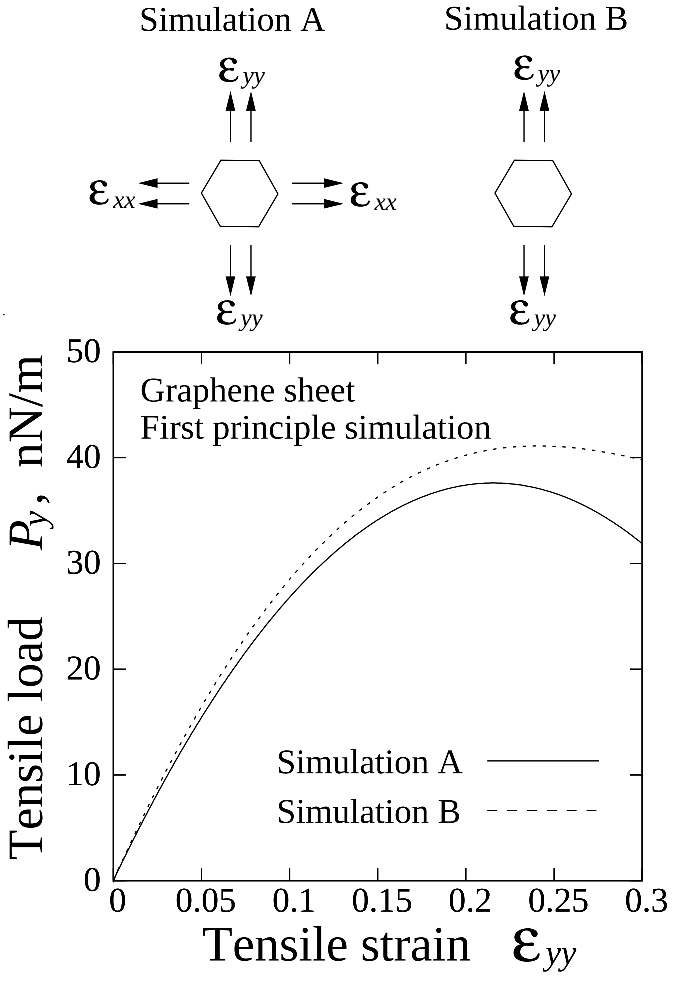
Graphene sheet When the "ideal" film (film with ideal understructure) is defined as the structure with the perfect two-dimensional periodicity, the simplest one is a layer of the graphene sheet which consists of a hexagonal carbon network as illustrated in Fig.2 (a) [14-18]. It is an absolute mono-layer film, for which the unit cell is indicated by the solid square in Fig.2(a)[14]. The equi-tensile strains, ε_{xx} and ε_{yy}, (Simulation A) or the uni-axial tensile strain, ε_{yy}, (Simulation B) are applied to the cell under the constraint condition that the other strain components are kept null [14]. The dotted and solid lines in Fig.2 (b) show the relationships between the load P_y and the strain ε_{yy} along the y-axis, respectively, analyzed by first-principle simulation (LDA, plane-wave basis, norm-conservative pseudopotential). Although it is easy to convert the load into the stress to evaluate the thickness from the distance between layers of stacked graphene sheet, we prefer to represent it here without the conversion in order to avoid ambiguity. This provides fundamental knowledge on the strength of nano-structured carbon such as fullerenes, nano-tubes, hones and so on. Moreover, the ideal strength, which is given by the peak load, in the uni-axial tension is higher than that in the equi-axial tension. Thus, the ideal strength is dependent on the combination of external strains (loading condition).

By comparing the load-strain curves, we can examine the validity of classical potential (*e.g.* the Brenner potential [19]) and the tight-binding (TB) method [20] under the high-strain condition. The result obtained by TB showed excellent correspondence with that in the above first-principle analysis, whereas the Brenner potential does not.

Thin film with reconstructed surface The development of high-vacuum technology enables us to procure well-defined surfaces, and extensive investigations have been devoted to exploring the structure at the surface as well as exotic properties stemming from it. The lattices near

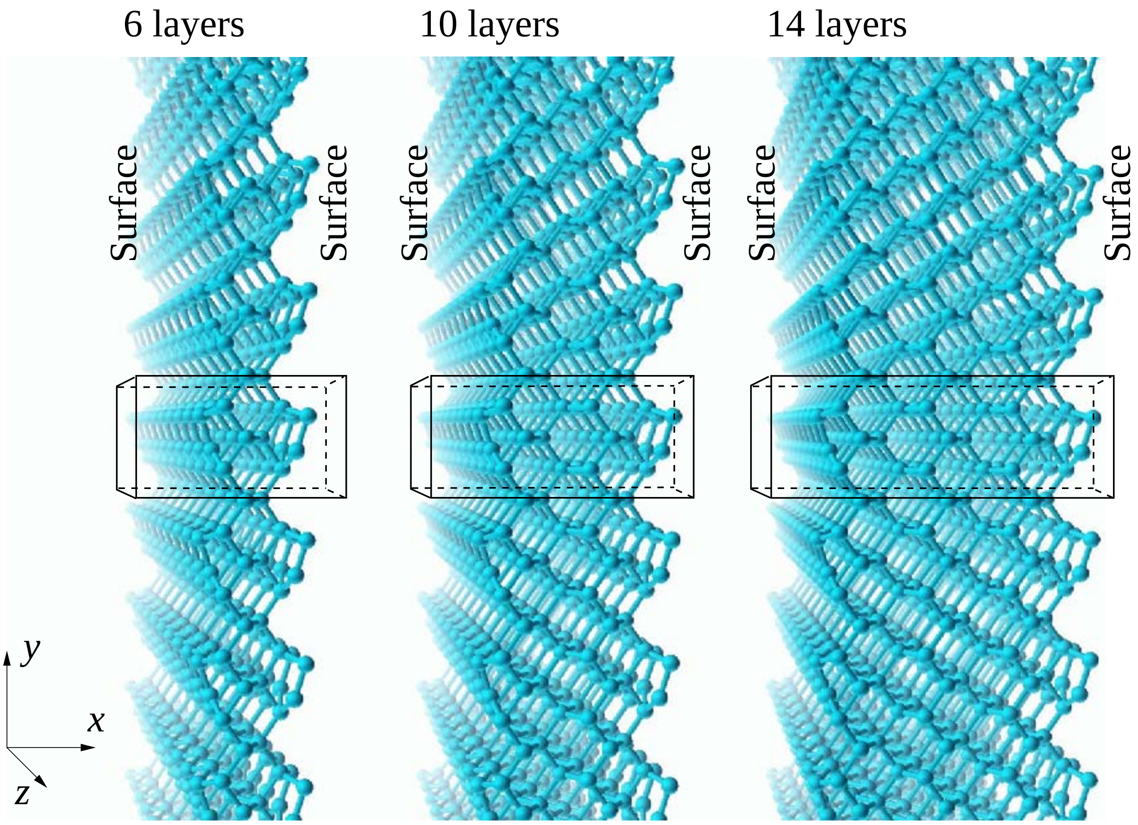


(a) Ideal structure of carbon layer in graphene sheet and the unit cell for calculation.

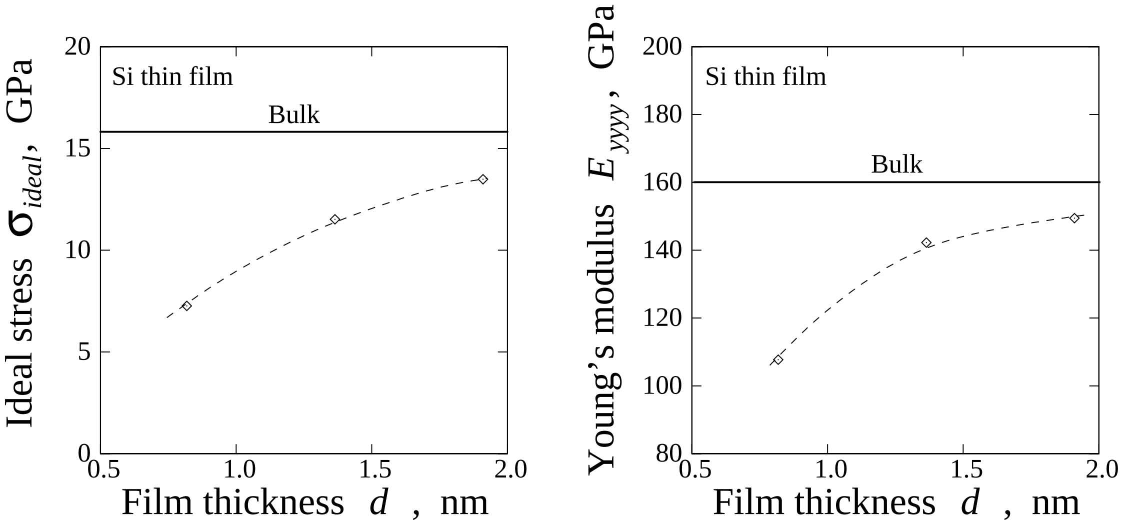


(b) Relationships between the load and the strain under tension.

Figure 2: Strength of graphene sheet [14].



(a) Ideal structures of thin films with 6, 10, 14 silicon layers, and their unit cells.



(b)Dependence of ideal strength and elastic coefficient on the film thickness.

Figure 3: Strength of silicon thin film[26].

the surface are relaxed and they sometimes form notably different structures from those in the bulk. In particular, the first layer shows a unique structure due to the reconstruction when the atoms on the surface have dangling bonds (*e.g.* [21-24]). Since the thin film is interpreted as being a material sandwiched by the surfaces, the mechanical property reflects the nature of the surfaces. In the other words, we may be able to determine the Ideal structure of carbon layer in graphene sheet and the unit cell for calculation.we may be able to determine the ideal surface effect on the strength by comparing the strengths between the bulk and the film [25].

Here, we show the strength of a silicon thin film with the excellent reconstructed surface of (100), of which the simplest structure is 2×1 illustrated in Fig.3 (a), as an example of an ideal film [26]. The freedom of a periodic cell in a film depends not only on the crystal structure but also on the thickness. We prepare a calculation cell of silicon with several thicknesses, and apply the uni-axial tensile strain, ε_{yy}, to the cell under the free transverse-stress condition, $\sigma_{xx} = 0$ by the first-principle simulation (GGA, plane-wave basis, ultrasoft pseudopotential).

Figure 3 (b) shows the dependence of the peak tensile stress and the elastic coefficient, E_{yyyy}, on film thickness. The figure indicates that the peak stress increases as the film thickens [26], and approaches the ideal bulk strength (thick line). It also reveals softening in the thinner film, signifying that the surface inherently possesses a weakening effect. However, the magnitudes of strength and the elastic coefficient, E_{yyyy}, of a 2-nm-thick film reach about 90% of those of the bulk. This implies that the surface property dominates only a region of less than 1 nm deep.

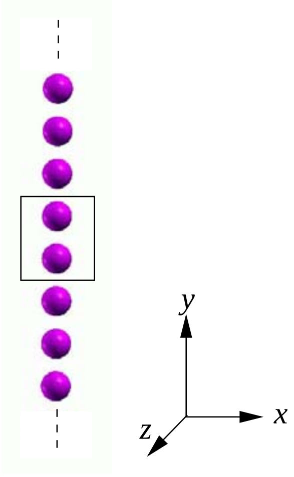
2.2 One-dimensional structure

There are several types of ideal wire with perfect one-dimensional periodicity, such as atomic chains, solid wires and tubes (hollow ones). In this section, we will present a typical strength analysis of them.

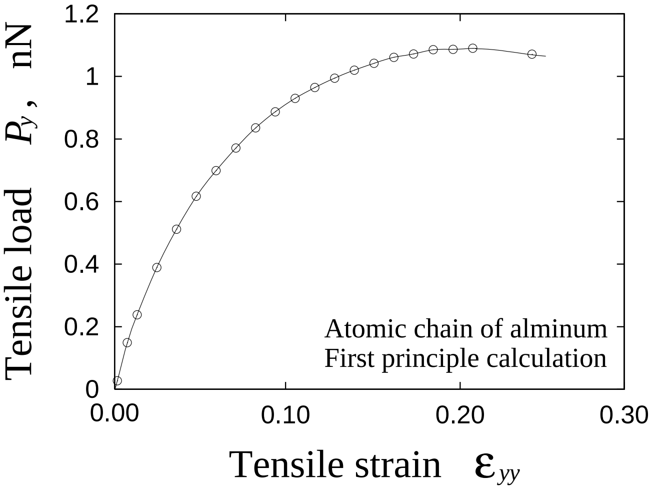
Atomic chain In recent years, it has become possible to arrange atoms in order by employing a sensational manipulation technique. An atomic chain is the simplest wire. It can be experimentally created and be observed *in situ* by an electron microscope (*e.g.* [27]). Figure 4(a) illustrates the atomic chain in which atoms align in a string [28-31].

Numerical simulation (LDA, plane-wave basis set, and norm-conserving pseudopotential) is conducted on the calculation cell of an aluminum atomic chain shown by the dotted square in Fig.4(a)[30]. The result reveals that the equilibrium interval of atoms is much shorter than the closest distance between atoms in the bulk fcc crystal of aluminum. A tensile strain is then imposed on the cell. The load-strain curve shown in Fig.4 (b) indicates a strength of 1N and a critical strain of 0.2. One way of perceiving the chain's strength is to compare it with the bulk providing that the fcc crystal is a bundle of atomic strings as illustrated in Fig.4 (c). Not only is the failure load of the atomic chain much higher than the critical tensile load per string in the bulk, but the elongation is eminently lower [30]. Therefore, we notice that the chain is more brittle than the bulk crystal.

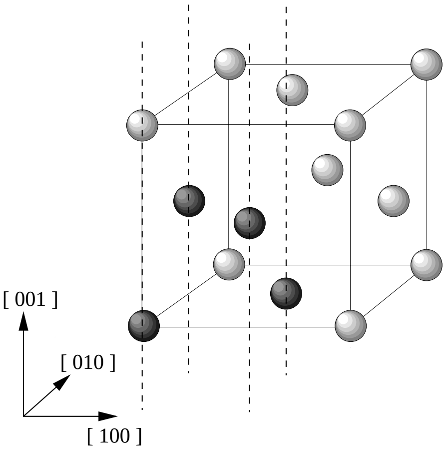
Nano-wire There are two types of structure that form an ideal solid wire. One is a crystal with an ordinary structure surrounded by surfaces, and the other is a very thin wire with an exotic structure, namely a nano-wire. For the former, a similar property as shown in thin film



(a) Illustration of atomic chain and the calculation cell.

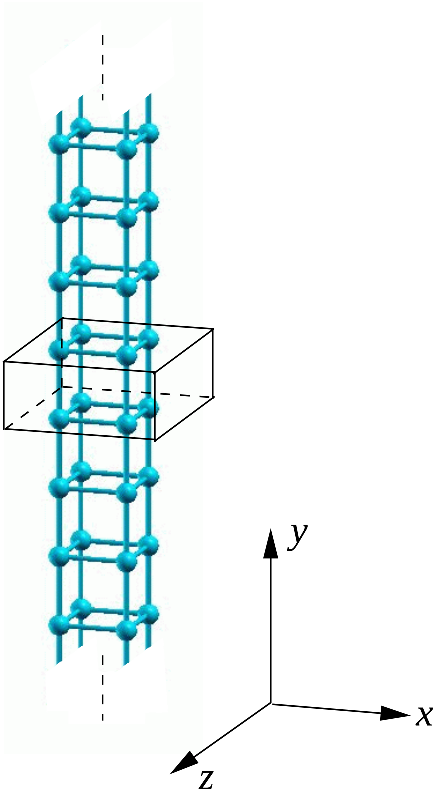


(b) Relationship between the external load and the strain under tension.

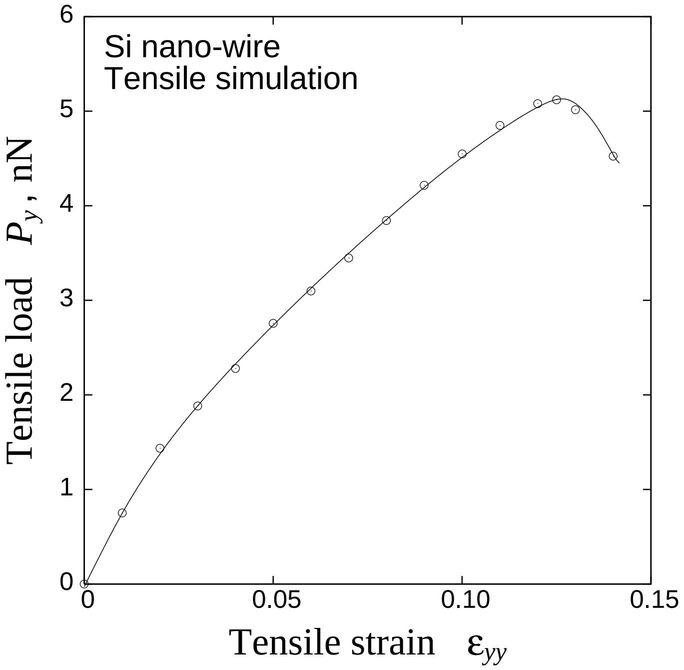


(c) Illustration of virtual strings in the fcc crystal.

Figure 4: Strength of aluminum atomic chain[30].



(a) Ideal structure of silicon nano-wire and the unit cell.



(b) Relationship between the external tensile load and the strain.

Figure 5: Strength of silicon nano-wire[26].

(Fig.3 (b)) is readily conceived. The wire becomes softer and weaker as the diameter shrinks to the extent that it possesses the structure of a crystal lattice.

In the meantime, since the extraordinary structure in the latter [32-38] still holds its periodicity along the wire axis, it should be categorized as an the ideal wire as well. Figure 5 (a) shows an example of the nano-wire structure proposed for silicon [26].

Tensile behavior of the unit cell marked by thick squares in the figure is simulated by the first-principle method (GGA, plane-wave basis set, and ultrasoft pseudopotential) [26]. The relation of load versus strain is shown in Fig. 5 (b), indicating the peak strength of about 5 N. However, care is necessary since the wire possesses various meta-stable structures due to loose constraints imposed by neighboring atoms. This requires strict judgment of instability as discussed in section 3.

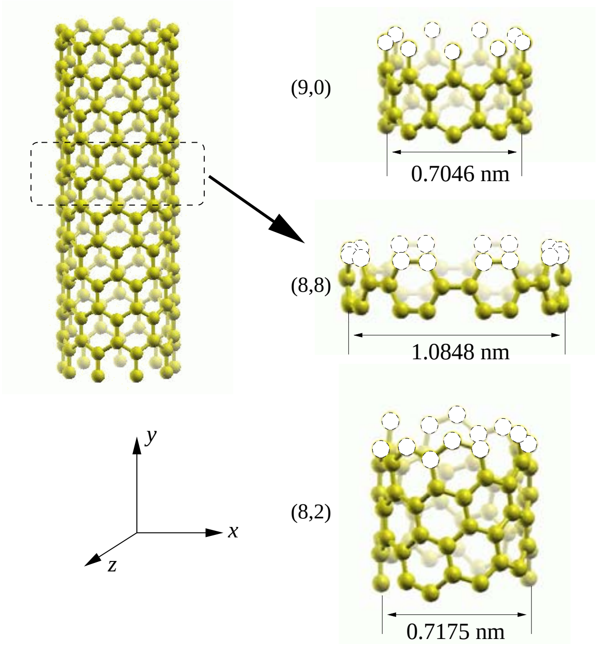
Tube Since the discovery of fullerene, intensive research attention has been directed foward materials with hollow structures at the nano scale. As a wire, the carbon nano-tube (CNT) possesses beautiful tubular structure with rich properties, that have been rigorously investigated both experimentally and theoretically (*e.g.* [39-47]), promising potential use in future miniature devices and machines.

Figure 6 (a) shows the unit cells of CNT with different chirality which represents the understructure of the tube. They have a similar diameter of 0.7-1.0 nm, and tensile simulation is carried out based on the TB method [46]. Here, (9,0) and (8,8) are known as Zigzag and Armchair types, respectively. Referring the graphene layer shown in Fig.2 (a), the strength of CNT (8,8) can be compared with that of raw material. (In the case of Fig.2 (b), the transverse strain is constrainted. For exact comparison, the simulation of the graphene layer should be conducted under the free transverse-stress condition.) This signifies the small effect of curvature on the strength. The effect of chirality can be determined in a similar manner.

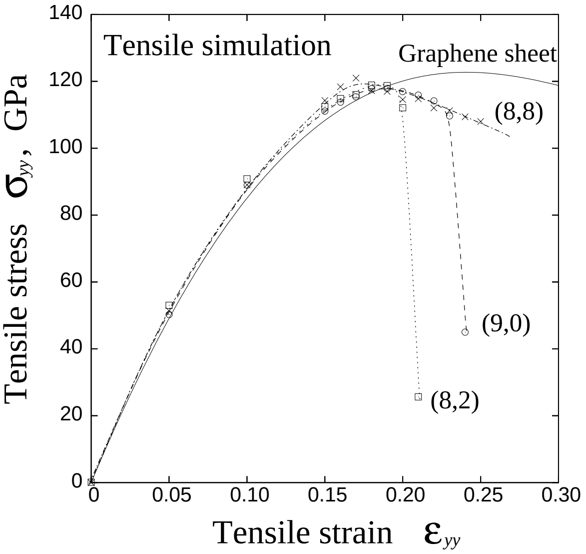
The tensile curves shown in Fig.6 (b) reveal that the Armchair type shows higher critical-stress than the Zigzag type though there are few differences among CNTs with different chirality. The stiffness at the equilibrium (no external load) is also only slight dependent on the understructure. Applying an unbalanced load (strain) to the side wall of CNT as shown in Fig.6 (c), we can analyze the effect of pure bending. Since this provides fundamental knowledge of defect-free nano-structure components under simple loading, it also should be included in ideal strength, though the one-dimensional periodicity along the y-axis is lost. At this point, the instability criterion of an arbitrary structure under an arbitrary external load becomes crucial in the analysis. In general, CNTs inherently possess various shapes including bent tubes, hones, and so on. The strength of typical ones, which provides fundamental comprehension and insight into the strength of the various CNTs, should be categorized as the "ideal strength".

2.3 Zero-dimensional structure

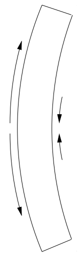
This category includes nano-particles and clusters [48,49] as the solid structure and fullerenes [50-53] as the hollow one. While these do not maintain periodicity, they do possess characteristically beautiful, defect-free structure. Moreover, they are essential members of the family of nano-



(a) Structure of carbon nano-tubes, (9,0), (8,8), (8,2), and their unit cells.



(b) Relationship between the stress and the strain in the axial tension.



(c) Unbalanced load imposed on nano-tube.

Figure 6: Strength of carbon nano-tube[46].

structured materials. Thus, they should also be categorized into the "ideal structures".

In this section, the tensile behavior of the silicon cluster illustrated in Fig.7 (a) [26], which includes six atoms, is examined by the first-principle simulation (GGA, plane-wave basis set, and ultrasoft pseudopotential) as an example. The displacement is applied to the top and bottom of the cluster. Figure 7 (b) depicts the stretched process that discloses the spring-like behavior, and the load-displacement curve is shown in Fig. 7(c).

3 Ideal strength and instability

As described in the Introduction, the strength of an atomic structure is defined as the stress or strain at which unstable deformation takes place. Consequently, it is inevitable to investigate the instability criterion under an external load (or displacement) when we consider in detail the strength of nano-structured components.

3.1 Instability of homogeneous crystals under homogeneous external stress (strain)

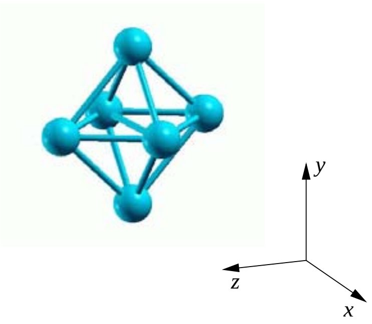
In the 1960s, Milstain [54] made a famous landmark analysis on the stability of cubic lattices under tensile deformation. The homogeneous deformation of cubic lattices is attributed to the components of the strain tensor in order that the distorted lattice shape is represented by the axial lengths, $a_1 - a_3$, and the angles, $a_4 - a_6$, as illustrated in Fig.8, respectively. It is then defined as "stable" when the total energy (free energy) of the system, Π does not decrease for an arbitrary incremental disturbance of strain under the external load. If it decreases, the component deforms without increase in external load, namely unstable deformation. Thus, the instability criterion for the homogeneous crystal under the homogeneous external strain is given by the sign of determinant A including its minor matrices. Here, the matrix A is in the form

$$\mathbf{A} = \left(\mathrm{A}_{ij}\right) = \left(\frac{\partial^2 \Pi}{\partial a_i \partial a_j}\right). \tag{1}$$

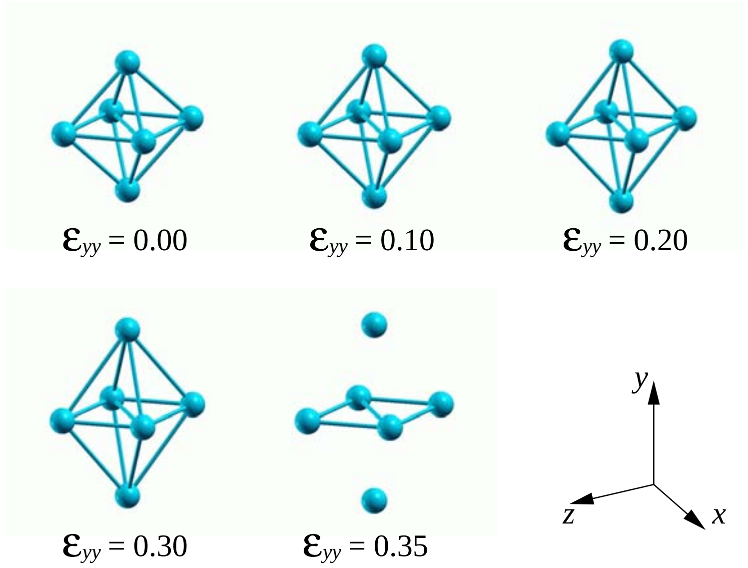
The lattice is stable when all of the determinants are positive. The criterion, of course, can be described by the differentials for strain instead of the variables a_1 to a_6. Since the elastic coefficient is defined as the second differential of the potential energy for the strain component, the instability criterion means the instance that the distorted structure loses its stiffness. Then, by using the elastic coefficient, the criterion for metals with cubic lattice (fcc and bcc) is rewritten as [1]

$$\begin{aligned} E_{xxxx} + 2E_{xxyy} &> 0 \\ E_{xyxy} &> 0 \\ E_{xxxx} - E_{xxyy} &> 0. \end{aligned} \tag{2}$$

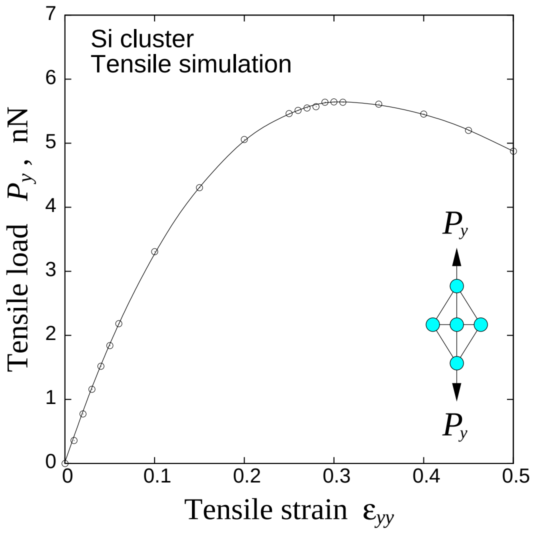
Hill [55, 56] pointed out that the instability criterion is dependent on the coordinates used for the evaluation of strain and stress. In other words, it is strongly dependent upon the loading boundary condition (loading device or external atomic system that yields the load on the



(a) Ideal structure of cluster consisted of six silicon atoms.



(b) Sketches of cluster in the elongated process.



(c) Relationship between the load and the strain under tension.

Figure 7: Strength of silicon cluster[26].

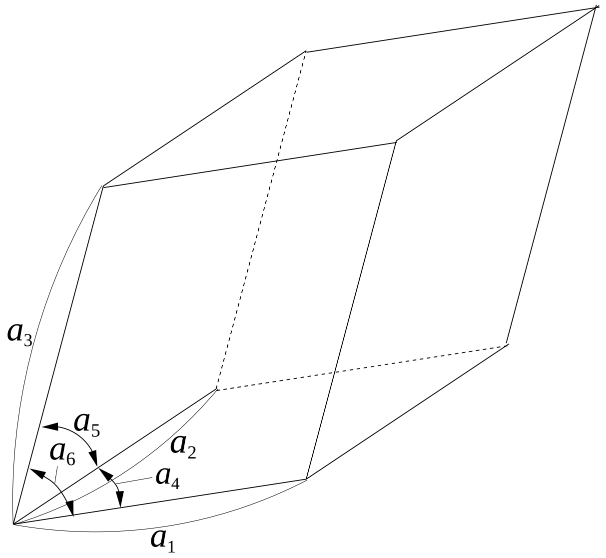


Figure 8: Variables $a_1 - a_6$ that represent the deformation of unit cell.

component in question). This stems from the fact that the freedom of deformation is restricted by only six variables in the analysis. As the nano-material has a larger number of degrees of freedom in general, the effect must be considered in discussions of the instability criterion.

Wang *et al.* [57,58] expanded the discussion on the basis of free energy and corrected the criterion using the stiffness coefficient for the bulk crystal. On the other hand, the criterion can be discussed in the context of the propagation of sound waves. The criterion of the transformation of crystal structures has been explored in terms of the soft mode of phonons by Hill [59]. As the elastic coefficient has a close relation with the long-wave mode of phonons, the ideal strength, which is a kind of structural transformation under an external load, surely can be generalized by the viewpoint of the soft mode, including the influence of the internal displacements. This phonon-based analysis is effective as far as the components hold periodicity, though the effect of external force is not explicitly included in the strictest sense.

Rapid progress in computational technology makes it possible for us to analyze the ideal strength on the basis of quantum mechanics. Investigations are rigorously conducted not only for pure metals with simple structures but also complex crystalline structures, *e.g.* intermetallic compounds. Sob *et al.* [60] produced an excellent review on the ideal strength of metals in the Highlight No. 58. In that report, they pointed out the importance on the coupling of ideal deformation (mechanical property) with electrical or magnetic properties in an ideal crystal. Several researchers (*e.g.* [60]) have successfully extended the analysis on the change in the electrical or magnetic properties due to deformation. For example, the electric property of the CNT is strongly correlated not only to its chirality but also to the strained condition [44-47].

3.2 Local instability in atomic components

In order to generalize the discussion on the "strength" of nano-structured components, it is necessary to explore the instability criterion of an arbitrary structure in the atomic scale.

The criterion for the dislocation emission from a crack tip under an external stress was proposed by Rice [61]. This have an impact on the following works on local instability of atomic components, though his analysis was totally based on the continuum mechanics concept. In his analysis, Rice derived the criterion from an instability of plastic flow under a singular stress-strain field near the crack tip, with the consideration given to the crystallographic slip direction. This stimulated researchers who were working on material strength at the atomic scale, leading to studies using numerical simulations by classical molecular dynamics (*e.g.* [62]). As a result, criterion based on the atomic mechanics concept is required to discuss the above problem precisely. In the nano-structured components, the switching or breaking of bonds between atoms under an external load generally brings about a sudden load-drop or displacement acceleration, namely unstable deformation. In a crystal, the dislocation nucleation was the shear collapse of local lattice. Thus, it was investigated from perspective of lattice instability [63, 64].

The stress, σ_{ij}, and the elastic coefficient, E_{ijkl}, are the quantities in continuum mechanics originally defined for a large region, which includes enough atoms. Those inherently can not be evaluated for each atom or local region. However, under the appropriate assumption (for example: the inter-atomic potential of embedded atom method [65]), σ_{ij} and E_{ijkl} in a local region can be reasonably estimated in consideration of short-range influence [66]. Vliet *et al.*[67] proposed the local instability criterion (Λ-criterion) based on the soft mode of long-wave phonons referring to the relationship between the long-wave property and σ_{ij} and E_{ijkl} in the region. They successfully applied it to dislocation generation under nano-indentation. Moreover, the criterion is generalized to the soft mode of shorter-waves by Dmitriev *et al.* [68].As we will describe in the next section, the exact mechanical criterion for the instability of arbitrary atomic structures under external load requires enormous numerical calculations due to its large number of degrees of freedom for deformation. Thus, it becomes important to make an approximation (local instability criterion) to extract the effective condition based on local information.

Strictly speaking, the analysis of phonons can be applied only on the structure with periodicity. As the low-dimensional structures discussed in section 2 do hold the periodicity in their dimensions, the characteristic instability can be explained by their soft mode. The instability at the surface of a nano-film with strained lattices has recently analyzed using the phonon dispersion property. The results may provide an important clue not only on the dislocation generation from the surface under strain, but also on the ideal strength of low-dimensional components.

3.3 Instability criterion for arbitrary structures under external load [69, 70]

Let us consider an arbitrary body comprising N atoms (N: number of atoms) under a strained state at the temperature of 0 K. Under an external load, the deformation is described by the number of degrees of freedom, $M = 3N - 6$, excluding the translation and rotation of the body. The total energy in the structure under an arbitrary deformation, Π which is the sum of strain energy, U, and the external work (negative), V, is in the function of only coordinates of atoms, $R = (R_1, \cdots, R_M)$. Then, the Taylor's series expansion of Π in terms of infinitesimal deformation, ΔR, gives the following relation

$$\Pi(\boldsymbol{R}+\Delta\boldsymbol{R}) = \Pi(\boldsymbol{R}) + \sum_{m=1}^{M} \frac{\partial \Pi}{\partial R_m}\Delta R_m + \frac{1}{2}\sum_{m=1}^{M}\sum_{n=1}^{M} \frac{\partial^2 \Pi}{\partial R_m \partial R_n}\Delta R_m \Delta R_n + \cdots. \quad (3)$$

The second term on the right-hand side vanishes since the system is at equilibrium. Attention should be paid on the fact that this term does not denote the external load.

Ignoring the higher-order terms, we have

$$\Delta\Pi \;=\; \Pi(\boldsymbol{R}+\Delta\boldsymbol{R})-\Pi(\boldsymbol{R})=\frac{1}{2}\,{}^{t}\Delta\boldsymbol{R}\,\boldsymbol{A}\,\Delta\boldsymbol{R}, \tag{4}$$

where the components of the matrix, A, are given by

$$A_{mn}=\frac{\partial^2\Pi}{\partial R_m\partial R_n}. \tag{5}$$

The structure is stable when $\Delta\Pi$ is positive, but becomes unstable and consequently deformation progresses when $\Delta\Pi$ becomes negative. Thus, the critical magnitude for the structural stability is given by $\Delta\Pi$ =0.

The matrix, A, is diagonalized using the matrix, $\mathrm{P}=(\mathrm{p}_1\ \ \mathrm{p}_2\ \ \cdots\ \ \mathrm{p}_\mathrm{M})$, where p_1, p_2,$\cdots$, p_M are the eigenvectors of A, as

$$\boldsymbol{P}^{-1}\boldsymbol{A}\boldsymbol{P} \;=\; {}^{t}\boldsymbol{P}\boldsymbol{A}\boldsymbol{P}=\begin{pmatrix}\eta_1 & & \boldsymbol{O}\\ & \ddots & \\ \boldsymbol{O} & & \eta_M\end{pmatrix}. \tag{6}$$

Here, η_m ($\eta_1 < \cdots < \eta_M$) is the eigenvalue of A. Introducing the matrix, $\Delta\mathrm{Q}=\mathrm{P}^{-1}\Delta\mathrm{R}$, we get

$$\begin{aligned}\Delta\Pi \;&=\; \frac{1}{2}\,{}^{t}(\boldsymbol{P}\,\Delta\boldsymbol{Q})\,\boldsymbol{A}\,(\boldsymbol{P}\,\Delta\boldsymbol{Q})\\ &=\; \frac{1}{2}\,{}^{t}\Delta\boldsymbol{Q}({}^{t}\boldsymbol{P}\boldsymbol{A}\boldsymbol{P})\Delta\boldsymbol{Q}=\frac{1}{2}\sum_{m=1}^{M}\eta_m\,(\Delta Q_m)^2\,. \end{aligned}\tag{7}$$

Then, the instability criterion ($\Delta\Pi$ =0) is attained by

$$\eta_1=0, \tag{8}$$

under

$$\Delta Q_m \;=\; 0 \quad (m=2,\cdots,M). \tag{9}$$

(Strictly speaking, the third differential term in Eq.(3) must be negative.) Moreover, Eqs.(7)-(9) point out that

$$\Delta\boldsymbol{R} \;=\; \Delta Q_1\cdot\boldsymbol{p}_1. \tag{10}$$

This means that the deformation at the instability is proportional to the eigenvector for the minimum eigenvalue, signifying the deformation mode at the instability. As the scheme is purely dependent on the system's energy, it can be applied to the quantum mechanical analysis without any collection.

Since the criterion is versatile, it is applicable to the instability of materials without periodicity or symmetry, such as the nucleation of dislocations [70], the delamination of thin film from a substrate [71], and so on. Even the plastic deformation of amorphous and metal glass, which stems the bond switching of local atoms, can be analyzed.

3.4 Structural instability

Above, we mainly discussed the instability of a lattice. In the meantime, there is another type of instability caused by the combination of load and global structures. It happens even when the lattice strain is not large. We can classify it into the "global" instability instead of "local" instability because it takes place in a macroscopic elastic body. A typical example of global instability is "buckling" of thin bar.

Figure 9 (a) [26] shows a silicon nano-wire with the same structure as the one shown in Fig.5(a). Axial compression is applied to the wire, of which calculation cell consists of 1 unit cube (Simulation A) or 5 cubes (Simulation B). The periodic boundary condition in the first-principle simulation (GGA, plane-wave basis set, and ultrasoft pseudopotential) fixes both ends of the wire. Simulation A, of course, exhibits extremely high strength, whereas the load-strain curve in Simulation B shown in Fig.9 (b) has low strength due to the buckling. This signifies that the buckling strength can not be predicted by the ideal strength, nor can it be explained in terms of local strain (local lattice instability). On the other hand, an analogy does exist for the relation between the nano-structured component and macroscopic continuum body. In this sense, the conventional continuum mechanics of an elastic body provides an interesting insight to the global collapse of nano-structured components such as devices and MEMS/NEMS. Moreover, it should be noted that the instability criterion discussed in section 3.3 can be applied not only to the local instability, but also to the global one.

4 Conclusion

We discussed the strength of nano-components with the ideal structure, with particular attention focused on the peak stress of low-dimensional material to extend the concept of the conventional ideal (theoretical) strength of a perfect crystal. In the same framework, we could discuss the ideal strength of grain boundaries and interfaces with periodicity, even though they are not nano-components. Materials possessing a super lattice structure, of course, should be such interesting targets. Thus, the "ideal structure" could be defined as the model structure by which simulation provides us with fundamental knowledge.

In the latter half of this paper, we investigated the instability that is closely related to the strength on nano-structured components. Since there are many unknowns in the instability criterion, further research is required.

In this review, we make no mention of not only the influences of temperature and chemical factors, but also of the deformation in the post-unstable criterion, the multiple instabilities (*e.g.* yielding in crystals) and irreversible cyclic deformation (*e.g.* fatigue). It is clear from the history of the strength of macroscopic components that these factors play also crucial roles in the strength of nano-structured components.

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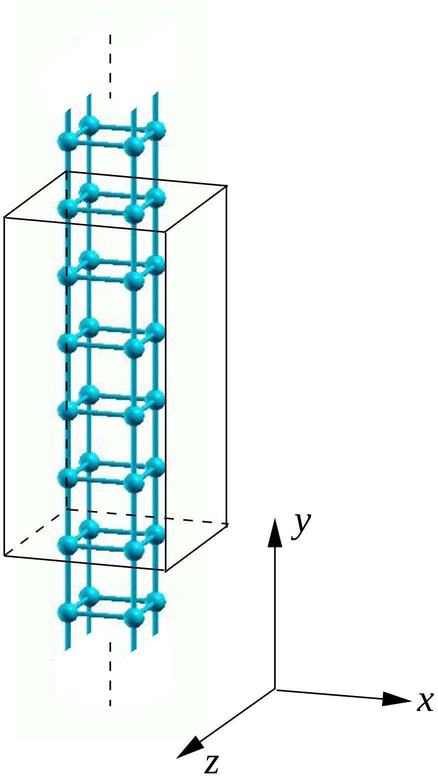
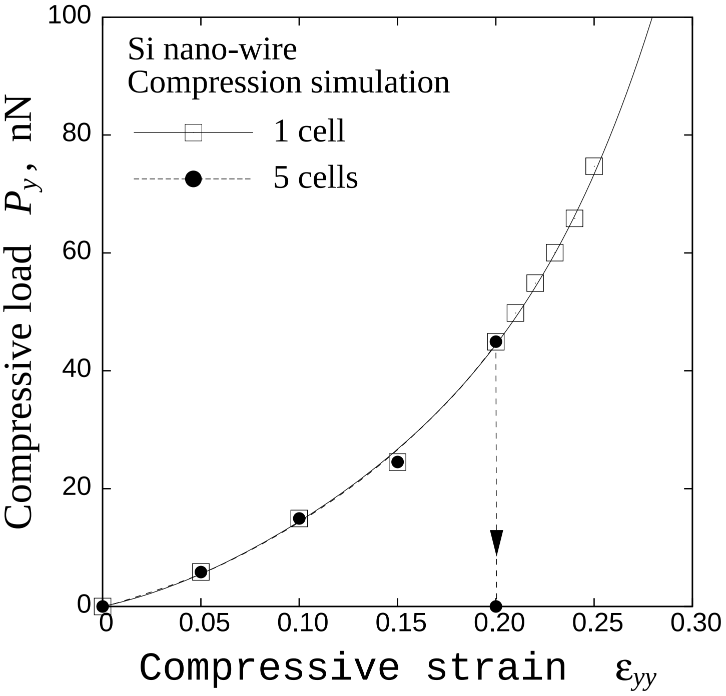


(a) Structure of nano-wire (5 cubes) for analysis of buckling.



(b) Relationships between the compressive load and strain. The sharp load drop in the wire with 5 unit cells point out the occurrence of buckling.

Figure 9: Buckling of silicon nano-wire as an example of global instability[26].

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