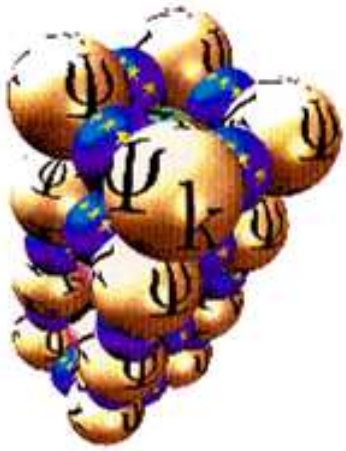
Ψ_k Newsletter

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

Number 115 February 2013



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Sponsored by: UK's CCP's and Psi-k

Contents

1 Editorial

Welcome to the first 2013 issue of Psi-k Newsletter. As usual at this time of year it does not have many contributions. Straight after the editorial we comment on problems with the Psi-k Portal and possible ways of avoiding at least some of them, before a robust fix is found. In addition, the present issue contains one workshop report, a job announcement and a few abstracts of newly submitted and/or recently published papers.

The scientific highlight of this issue is replaced by a memorial article, where some of the collaborators of Professor Balazs L. Gyorffy (University of Bristol, UK), who died last October, celebrate his physics, giving account of his many scientific accomplishments and their personal reminiscences on working with him. The title of the article is "Reflections on the Life and Science of Balazs L. Gyorffy (1938-2012) "Life without U"". The contributors to the article are: James F. Annett (University of Bristol, UK), Paul J. Durham (Daresbury Laboratory, UK), Ingrid Mertig (University of Halle and MPIfM Halle, Germany), Julie B. Staunton (Warwick University, UK), G. Malcolm Stocks (Oak Ridge National Laboratory, USA), Walter M. Temmerman (Daresbury Laboratory, UK), and Peter Weinberger (Formerly, Technical University of Vienna, Austria).

Please check the table of content for details.

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

http://www.psi-k.org.uk/

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

The email address for contacting us and for submitting contributions to the Psi-k newsletters is

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

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2 General News

2.1 Problems with Psi-k Announcements and Portal

A number of users have complained about problems with the Psi-k Portal due to the server being down, the window for uploading information being too small or scrambled (note that the latter does not occur when using the Internet Explorer), information being distributed multiple times or not at all, etc..

Some of the problems (e.g. the outages or the small size of the editor window) are related to the portal software we use. We tried updating the software on a test machine and found that the new version fixed those problems, but unfortunately created some new problems. We are currently investigating this with the portal developers.

Other problems can easily be avoided by the users. Here are the two main issues:

- **Multiple submissions:**
  When submitting an announcement, it can take several minutes to complete. Furthermore, a considerable time may pass until the announcement gets distributed. Therefore, please be patient and do not press the **Add Announcement** button several times.

- **Announcement is not visible:**
  A point which often causes confusion is the **Availability** option. Please, leave this option as **Show**. The dates, which one may specify, should *not* be the dates of the event or of the deadline for a job application. They are meant to define the time span that one wants an announcement to be visible on the portal.

As a reminder, below we repeat steps to be followed when uploading announcements to the portal:

To login to the Psi-k Portal one has to go to the Psi-k webpage

`http://www.psi-k.org`

and when there click on the option **'Announcements and Psi-k Portal'**. This will give one a choice to either login to the Psi-k Portal and be able to view and upload announcements, workshop reports and proposals, etc., or just only view the announcements present on the Psi-k Portal, for which there is no need to login.

Since uploading information to the Psi-k Portal seems to give problems to some users, below we repeat the steps one needs to follow to successfuly upload e. g. an announcement to the Psi-k Announcements folder and have it distributed by e-mail to all the Portal members.

1. **'Login to Psi-k Portal'**;

2. While already on the Psi-k Portal click on **'Psi-k Announcements'**;

3. Click on **'Add'** to get a window for uploading an announcement and then fill in all the required fields;

4. Hyperlinks have to be added to any URLs of the web pages ocurring in the body of the announcement by clicking on the option above the main text window, which looks like **'binnacles on a blue ball'** and typing the URL into the subsequently opened field;

5. **Add Attachements**, if any, by pressing this option;

6. When all is typed and filled, then press **'Preview'** to see if all looks right;

7. If not, then press **Edit**;

8. If yes, then for **'Email Notification'** choose **'High - All Participants'**;

9. **Don't do** anything about 'Access' or 'Availability' please, and leave them as they are by default;

10. Press **'Add Announcement'**, only once (!), and all is done.

3 Psi-k Activities

"Towards Atomistic Materials Design"

3.1 Reports on the Workshops supported by Psi-k

3.1.1 Report on CECAM workshop: Vibrational coupling: most important, often ignored, and a challenge for ab-initio theory

Lausanne, November 6 - 9, 2012

Sponsors: CECAM, Psi-k, MM1P.de

Organizers: Heiko Appel, Christian Carbogno, Claudia Draxl, Matthias Scheffler

Web page: `http://th.fhi-berlin.mpg.de/th/meetings/vc2012/`

Summary

We have organized and implemented a successful workshop on the ab-initio description of vibronic and vibrational couplings, held from November 6 - 9 at the CECAM headquarters in Lausanne. With the workshop we brought together researchers from different backgrounds but common interest in vibrational and vibronic couplings to identify and address open problems and challenges which are shared among the different fields. In each of these research areas there is a large body of specialized expertise available. We therefore were aiming to establish within the workshop a common forum to allow for an exchange of methodologies and to stimulate new developments which help to tackle the challenges posed by applications. The main goal of the workshop was to enhance the quality of theoretical and computational research on the various phenomena that are affected by electron-vibrational interactions.

The workshop was organized in a Gordon conference type format. Each topic was introduced by a discussion leader which also then moderated the invited talks of the corresponding session. Invited speakers lectured for 40 minutes, followed by 20 minutes of guided discussion. In total, the workshop encompassed 19 invited speakers and 20 poster contributions out of 42 participants and 4 organizers. Almost every participant was therefore actively contributing to the scientific discussions. We also reserved two evenings for poster sessions, which in particular gave younger participants a much broader stage to showcase their own research work. When we prepared the program of the workshop, we reserved ample time for discussion after the talks. This turned out to be essential to stimulate many discussions and often the critical points were only revealed in the discussion periods.

Description of the scientific content of and discussion at the event

Following the timeline of the workshop program, we briefly summarize below the main outcomes of the presentations given in the workshop.

The first day of the workshop was concerned with non-adiabatic effects that arise when the coupling of electrons and nuclei is treated beyond a standard ground-state Born-Oppenheimer description. After an introduction of H. Appel on experimental observables that are not properly captured in an adiabatic Born-Oppenheimer picture, E.K.U. Gross illustrated in his contribution how to make the Born-Oppenheimer approximation exact and thus gave a new perspective on potential energy surfaces and the non-adiabatic coupling of electrons and nuclei. In the second talk of the session, I. Tavernelli outlined how to compute non-adiabatic coupling matrix elements in first and second order response in a time-dependent DFT framework and exemplified the approach for protonated formaldimine and graphene. In the last talk of the first day, A. Horsfield illustrated the shortcomings of Ehrenfest dynamics and introduced the correlated electron-ion dynamics (CEID) approach, highlighting also the problems associated with the electron correlation in the CEID formalism. In the evening of the first day the first poster session took place. The session stimulated many lively discussions that extended way beyond the official end of the session.

The second day was devoted almost exclusively to thermoelectrics, an emerging hot topic in material science. A theoretical description of thermoelectric effects, i.e., the accurate assessment of (electronic) charge and (electronic and vibrational) heat transport at arbitrary low and high temperatures still constitutes a considerable challenge, as discussed and reviewed in the introductory talk by J. Sofo. On the same topic, but from quite a different perspective, J. Grin illustrated the challenges that experimentalist face in the search and engineering of novel and optimized themoelectric materials and how theory can aid and guide this process, for instance by disentangling the contributions that arise from changes in the character of the interatomic bonding. The following presentations discussed different techniques that have recently been developed and implemented in this context: B. Xu and N. Bonini critically examined how perturbational techniques based on state-of-the-art density functional theory allow to compute the transport coefficients at low temperatures and how these approaches can be extended to enable an assessment of the Seebeck coefficient and/or contributions arising from impurities, defects and alloying. Conversely, C. Carbogno and I. Savic focused on the higher (classical) temperature regime and discussed how molecular dynamics (MD) based approaches allow to tackle the problem of vibrational heat conduction. While C. Carbogno discussed the specific adaptions that MD techniques require when applied in first-principles frameworks, I. Savic presented the influence of meso- and nano-scopic structuring, as investigated by extended MD simulations on the basis of semi-empirical potentials. Last but not least, K. Reuter framed the previously discussed methodologies in a completely different light by presenting their potential application for problems in the field of heterogeneous catalysis and surface science.

The third day started with an introductory talk given by C. Draxl about the importance of electron-phonon coupling for accurate ab-initio electronic structure calculations, e.g., in the computation of band gaps at finite temperatures. In the following, F. Giustino discussed the perturbative treatment of the electron-phonon coupling and the role of various Feynman di-

agrams for different material classes (metal, insulators, superconductors). Along these lines, A. Eiguren illustrated the application of such concepts for the computation of quasi-particle spectra. Eventually, X. Gonze introduced the nuts & bolts of density functional perturbation theory, the underlying assumptions and the influence of the resulting Feynman diagrams (Fan term, diagonal and non-diagonal Debye-Waller-term) on the band-gap of diamond. B. Batlogg opened the afternoon session of the third day by introducing the challenges that experimentalists face in the assessment, interpretation and description of vibrational and vibronic coupling in semiconductor materials and nanostructures. K. Hannewald then presented his theoretical approach to calculate mobilities in organic molecular crystals, going beyond narrow bands and small polarons. In the last talk of the day, M. Gl?ssl introduced a real-time path integral approach to describe the laser-driven carrier-phonon dynamics in semiconductor quantum dots. In the evening of the third day also the second poster session took place, which gave young participants again the opportunity to present their research work.

On the fourth and last day of the workshop the focus was then shifted to excitations in strong fields and time-resolved excitations. I. Franco opened the stage by addressing the role of vibronic couplings in non-equilibrium situations, decoherence and dephasing effects and related timescales. These aspects were then also taken up in the talk of O. Prezhdo who exemplified the role of dephasing, electron-phonon relaxation, and exciton formation in Silicon and PbSe quantum dots by means of of non-adiabatic molecular dynamics. E. Malic discussed in his talk the thermalization and cooling of graphene after optical excitation with a particular emphasis on orientational relaxation induced by phonons. In the last talk of the workshop R. van Leeuwen focused on the role of electron-phonon coupling in time-dependent quantum transport and non-equilibrium many-body theory and discussed the influence of vibronic effects on I-V curves, spectral functions and transient dynamics.

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

One recurring key point of the talks and of the discussions was the accuracy and correctness of density functional perturbation theory for the calculation of electron-phonon matrix elements. To date, there is no formal derivation or justification for this approach available in the literature. The presentations of C. Draxl, F. Giustino, A. Eiguren and X. Gonze impressively showed that a series of contributions beyond the current state-of-the-art perturbational treatment of the electronic structure are required to achieve a consistent description of vibrational-vibronic effects. Obviously, this fact calls for future collaborations and benchmarking of these approaches. Similarly, the presented electronic and vibrational transport calculations that are based on density functional perturbation theory might require revisitation under this light. As a matter or fact, this holds true for all transport methodologies presented on the third day of the workshop: Each of the discussed approaches (B. Xu, C. Carbogno, N. Bonini, I. Savic) has its own strengths and weaknesses; the underlying approximations limit the applicability of these techniques to certain fields, structures and thermodynamic regions. Naturally, a validation of the individual approaches requires a careful comparison of the different techniques with each other on the

same footing, i.e., at the same level of electronic structure theory. Collaborations in this field can help establish a sound and unified theory of thermal and electronic transport, as required for further advancement in this area of research (Y. Grin). Another recurring aspect in the discussion sessions was the search for a reliable and scalable quantum-classical or quantum-semiclassical method to study non-equilibrium dynamics beyond Ehrenfest. Current approaches fail e.g. to describe proper thermalization between electronic and nuclear degrees of freedom (Ehrenfest dynamics and related approaches), or are difficult to apply for metallic or small gap systems (surface hopping). Most prominent were also the repeated discussions on the expansion of Hamiltonians to higher order in the nuclear displacements, which introduces new vertices for Green's function approaches.

In summary, we think the workshop has been successful in identifing many open questions in the field of vibrational and vibronic coupling. As outlined above, the discussions revealed also several interesting directions for future research and developments which could help to tackle open problems. There were two main lines of research that emerged throughout the workshop: (i) vibronic pertubation theories centered around the Born-Oppenheimer approximation and (ii) non-adiabatic real-time dynamics of coupled electron-ion systems. We feel that it will be beneficial in future to pursue both directions in more detail. This should help researchers in the field to find tailored solutions for remaining open problems.

Program of the CECAM workshop

	Tuesday, November 6: Session I - Non-adiabaticity
14:40-15:00	Heiko Appel - Introduction to the session and discussion moderator
15:00-15:40	Eberhard K.U. Gross - How to make the Born-Oppenheimer approximation exact: A fresh look at potential energy surfaces and Berry phases in the time domain
15:40-16:00	Discussion
16:00-16:20	Coffee Break
16:20-17:00	Ivano Tavernelli - Nonadiabatic couplings and nonadiabatic dynamics within TDDFT
17:00-17:20	Discussion
17:20-18:00	Andrew Horsfield - How do you build a good Hamiltonian for CEID?
18:00-18:20	Discussion
18:20-19:30	Poster session

Wednesday, November 7: Session II - Thermoelectrics and heat transport

09:00-09:40 Jorge Sofo - Introduction to the session and discussion moderator
09:40-10:00 Discussion
10:00-10:40 Bin Xu - Ab initio thermoelectric properties
10:40-11:00 Discussion
11:00-11:20 Coffee Break
11:20-12:00 Christian Carbogno - Thermal Conductivity at High Temperatures from First Principles
12:00-12:20 Discussion
12:20-12:40 Peter Kratzer - Nanostructured semiconductors as thermoelectrics
12:40-14:00 Lunch Break

Wednesday, November 7: Session III - Thermoelectrics and catalysis

14:00-14:40 Juri Grin - Introduction to the session and discussion moderator
14:40-15:00 Discussion
15:00-15:40 Karsten Reuter - Watching phonons getting all excited during oxygen dissociation at Pd(100)
15:40-16:00 Discussion
16:00-16:20 Coffee Break
16:20-17:00 Nicola Bonini - Electrical and thermal transport from first-principles
17:00-17:20 Discussion
17:20-18:00 Ivana Savic - Towards realistic description of thermal transport processes in nanostructured materials
18:00-18:20 Discussion

Thursday, November 8: Session IV - Electronic excitations

09:00-09:20 Claudia Draxl - Introduction to the session and discussion moderator
09:20-10:00 Feliciano Giustino - Electron-phonon calculations across the board: from kinks to band gaps
10:00-10:20 Discussion
10:20-10:40 Coffee Break
10:40-11:20 Asier Eiguren - Self-consistent renormalization of quasi-particles; extending the range of applicability of the quasi-particle theory.
11:20-11:40 Discussion
11:40-12:20 Xavier Gonze - Temperature and zero-point motion effects on the electronic band structure
12:20-12:40 Discussion
12:40-14:00 Lunch Break

Thursday, November 8: Session V - Semiconductors and nanostructures

14:00-14:40	Bertram Batlogg - Introduction to the session and discussion moderator
14:40-15:00	Discussion
15:00-15:40	Karsten Hannewald - Polaronic signatures in the static and dynamical conductivities of organic crystals
15:40-16:00	Discussion
16:00-16:20	Coffee Break
16:20-17:00	Martin Gläßl - Real-time path integrals for laser driven carrier-phonon dynamics in quantum dots
17:00-17:20	Discussion
17:20-19:00	Poster Session
19:30-22:00	Social Dinner

Thursday, November 9: Session VI - Excitations in strong fields and time-resolved excitations

09:00-09:20	Ignacio Franco - Introduction to the session and discussion moderator
09:20-10:00	Oleg Prezhdo - Elastic and Inelastic Electron-Phonon Scattering in Nanoscale Materials
10:00-10:20	Discussion
10:20-10:40	Coffee Break
10:40-11:20	Ermin Malic - Ultrafast relaxation dynamics in graphene - Impact of carrier-phonon and carrier-carrier scattering
11:20-11:40	Discussion
11:40-12:20	Robert van Leeuwen - Electron-electron and electron-phonon interactions in time-dependent quantum transport using non-equilibrium many-body theory
12:20-12:40	Discussion
12:40	End of Workshop

4 General Job Announcements

PhD Position in Development of Electronic Structure Codes at Materials Center Leoben, Austria

A PhD position is available at Materials Center Leoben (MCL, Austria) under the supervision of Prof. Peter Puschnig (University of Graz, Austria). The successful candidate will join the PhD program at the University of Graz (http://physik.uni-graz.at/doktoratsschule/index.php), and will perform his/her research activities at the atomistic modelling group at MCL. The MCL is a research centre for materials engineering and development with strong ties to both industry and academia. At the atomistic modelling group, we strive for the accurate description of the thermodynamic and mechanical properties of steels using first-principles methods (http://www.mcl.at/en/forschung/atomistic-modeling/properties-of-steels.html). The principal focus of the PhD project will be the development of state-of-the-art density-functional codes which will open new possibilities in modelling complex materials. Such development will be done in close collaboration with Prof. Andrei Ruban (Royal Institute of Technology, Stockholm, Sweden) and Dr. Oleg Peil (University of Geneva, Switzerland).

Proficient skills in scientific programming (including Fortran) and strong background in solid-state physics are needed. A starting date around July 2013 is anticipated. Please send Curriculum Vitae and two reference letters to peter.puschnig@uni-graz.at (Cc: a.huamantinco@mcl.at). Consideration of candidates will begin immediately and will continue until the position is filled.

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5 Abstracts

Orbital-dependent electron tunneling within the atom superposition approach: Theory and application to W(110)

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Abstract

We introduce an orbital dependent electron tunneling model and implement it within the atom superposition approach for simulating scanning tunneling microscopy (STM) and spectroscopy (STS). Applying our method, we analyze the convergence and the orbital contributions to the tunneling current and the corrugation of constant current STM images above the W(110) surface. In accordance with a previous study [Heinze et al., Phys. Rev. B 58, 16432 (1998)], we find atomic contrast reversal depending on the bias voltage. Additionally, we analyze this effect depending on the tip-sample distance using different tip models, and find two qualitatively different behaviors based on the tip orbital composition. As an explanation, we highlight the role of the real space shape of the orbitals involved in the tunneling. STM images calculated by our model agree well with Tersoff-Hamann and Bardeen results. The computational efficiency of our model is remarkable as the k-point samplings of the surface and tip Brillouin zones do not affect the computation time, in contrast to the Bardeen method.

(Physical Review B 86, 235415 (2012))
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Analytic evaluation of the electronic self-energy in the GW approximation for two electrons on a sphere

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Abstract

The GW approximation for the electronic self-energy is an important tool for the quantitative prediction of excited states in solids, but its mathematical exploration is hampered by the fact that it must, in general, be evaluated numerically even for very simple systems. In this paper I describe a nontrivial model consisting of two electrons on the surface of a sphere, interacting with the normal long-range Coulomb potential, and show that the GW self-energy, in the absence of self-consistency, can in fact be derived completely analytically in this case. The resulting expression is subsequently used to analyze the convergence of the energy gap between the highest occupied and the lowest unoccupied quasiparticle orbital with respect to the total number of states included in the spectral summations. The asymptotic formula for the truncation error obtained in this way, whose dominant contribution is proportional to the cutoff energy to the power $-3/2$, may be adapted to extrapolate energy gaps in other systems.

(Submitted to Phys. Rev. B)
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Comparison of stress and total energy methods for calculation of elastic properties of semiconductors

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Abstract

We explore the calculation of the elastic properties of zinc-blende and wurtzite semiconductors using two different approaches: one based on stress and the other on total energy as a function of strain. The calculations are carried out within the framework of density functional theory in the local density approximation, with the plane wave-based package VASP. We use AlN as a test system, with some results also shown for selected other materials (C, Si, GaAs and GaN). Differences are found in convergence rate between the two methods, especially in low symmetry cases, where there is a much slower convergence for total energy calculations with respect to the number of plane waves and **k** points used. The stress method is observed to be more robust than the total energy method with respect to the residual error in the elastic constants calculated for different strain branches in the systems studied.

(Published in J. Phys.: Condens. Matter **25**, 025803 (2013))
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Thermal conductivity of bulk and nanowire $Mg_2Si_xSn_{1-x}$ alloys from first principles

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Abstract

The lattice thermal conductivity (κ) of the thermoelectric materials, Mg_2Si, Mg_2Sn, and their alloys, are calculated for bulk and nanowires, without adjustable parameters. We find good agreement with bulk experimental results. For large nanowire diameters, size effects are stronger for the alloy than for the pure compounds. For example, in 200 nm diameter nanowires κ is lower than its bulk value by 30%, 20%, and 20% for $Mg_2Si_{0.6}Sn_{0.4}$, Mg_2Si, and Mg_2Sn respectively. For nanowires less than 20 nm thick, the relative decrease surpasses 50%, and it becomes larger in the pure compounds than in the alloy. At room temperature, κ of $Mg_2Si_xSn_{1-x}$ is less sensitive to nanostructuring size effects than Si_xGe_{1-x}, but more sensitive than $PbTe_xSe_{1-x}$. This suggests that further improvement of $Mg_2Si_xSn_{1-x}$ as a non-toxic thermoelectric may be possible.

(Physical Review B, **86**, 174307 (2012))
`http://dx.doi.org/10.1103/PhysRevB.86.174307`
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Rare-Earth vs. Heavy Metal Pigments and their Colors from First Principles

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Abstract

Many inorganic pigments contain heavy metals hazardous to health and environment. Much attention has been devoted to the quest for non-toxic alternatives based on rare-earth elements. The computation of colors from first principles is a challenge to electronic structure methods however, especially for materials with localized f-orbitals. Here, starting from atomic positions only, we compute the color of the red pigment cerium fluorosulfide CeSF, as well as of mercury sulfide HgS (classic "vermilion"). Our methodology employs many-body theories to compute the optical absorption, combined with an intermediate length-scale modelization to assess how coloration depends on film thickness, pigment concentration and granularity. We introduce a quantitative criterion for the performance of a pigment. While for HgS this criterion is satisfied due to large transition matrix elements between wide bands, CeSF presents an alternative paradigm: the bright red color is shown to stem from the combined effect of the quasi two-dimensionality and the localized nature of $4f$ states. Our work demonstrates the power of modern computational methods, with implications for the theoretical design of materials with specific optical properties.

(Proc. Natl. Acad. Sci. USA 110(3) 904-907 (2013) [preprint: arXiv:1301.0630])
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Combined GW and dynamical mean field theory: Dynamical screening effects in transition metal oxides

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Abstract

We present the first dynamical implementation of the combined GW and dynamical mean field scheme ("GW+DMFT") for first principles calculations of the electronic properties of correlated materials. The application to the ternary transition metal oxide $SrVO_3$ demonstrates that this schemes inherits the virtues of its two parent theories: a good description of the local low energy correlation physics encoded in a renormalized quasi-particle band structure, spectral weight transfer to Hubbard bands, and the physics of screening driven by long-range Coulomb interactions. Our data is in good agreement with available photoemission and inverse photoemission spectra; our analysis leads to a reinterpretation of the commonly accepted "three-peak structure" as originating from orbital effects rather than from the electron addition peak within the t_{2g} manifold.

(Europhys. Lett. (EPL) 100(6) 67001 (2012) [preprint: arXiv:1210.6580])
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Prediction of the bias voltage dependent magnetic contrast in spin-polarized scanning tunneling microscopy

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Abstract

This work is concerned with the theoretical description of the contrast, i.e., the apparent height difference between two lateral surface positions on constant current spin-polarized scanning tunneling microscopy (SP-STM) images. We propose a method to predict the bias voltage dependent magnetic contrast from single point tunneling current or differential conductance measurements, without the need of scanning large areas of the surface. Depending on the number of single point measurements, the bias positions of magnetic contrast reversals and of the maximally achievable magnetic contrast can be determined. We validate this proposal by simulating SP-STM images on a complex magnetic surface employing a recently developed approach based on atomic superposition. Furthermore, we show evidence that the tip electronic structure and magnetic orientation have a major effect on the magnetic contrast. Our theoretical prediction is expected to inspire experimentalists to considerably reduce measurement efforts for determining the bias dependent magnetic contrast on magnetic surfaces.

(Physical Review B 87, 024417 (2013))

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Understanding Structure and Bonding of Multilayered Metal-Organic Nanostructures

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Abstract

For organic and hybrid electronic devices, the physicochemical properties of the contained interfaces play a dominant role. To disentangle the various interactions occurring at such heterointerfaces, we here model a complex, yet prototypical, three-component system consisting of a Cu-phthalocyanine (CuPc) film on a 3,4,9,10-perylene-tetracarboxylicdianhydride (PTCDA) monolayer adsorbed on Ag(111). The two encountered interfaces are similar, as in both cases there would be no bonding without van der Waals interactions. Still they are also distinctly different, as only at the Ag(111)-PTCDA interface massive chargerearrangements occur. Using recently developed theoretical tools, we show that it has become possible to provide atomistic insight into the physical and chemical processes in this comparatively complex nanostructure being able to distinguish between interactions involving local rearrangements of the charge density and long-range van der Waals attraction.

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Renormalized Second-order Perturbation Theory for The Electron Correlation Energy: Concept, Implementation, and Benchmarks

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Abstract

We present a renormalized second-order perturbation theory (rPT2), based on a Kohn-Sham (KS) reference state, for the electron correlation energy that includes the random-phase approximation (RPA), second-order screened exchange (SOSEX), and renormalized single excitations (rSE). These three terms all involve a summation of certain types of diagrams to infinite order, and can be viewed as "renormalization" of the 2nd-order direct, exchange, and single excitation (SE) terms of Rayleigh-Schrödinger perturbation theory based on an KS reference. In this work we establish the concept of rPT2 and present the numerical details of our SOSEX and rSE implementations. A preliminary version of rPT2, in which the renormalized SE (rSE) contribution was treated approximately, has already been benchmarked for molecular atomization energies and chemical reaction barrier heights and shows a well balanced performance [Paier *et al.*, New J. Phys. 14, 043002 (2012)]. In this work, we present a refined version of rPT2, in which we evaluate the rSE series of diagrams rigorously. We then extend the benchmark studies to non-covalent interactions, including the rare-gas dimers, and the S22 and S66 test sets. Despite some remaining shortcomings, we conclude that rPT2 gives an overall satisfactory performance across different chemical environments, and is a promising step towards a generally applicable electronic structure approach.

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6 SCIENTIFIC HIGHLIGHT OF THE MONTH

Reflections on the Life and Science of Balazs L. Gyorffy (1938-2012) "Life without U"

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Introduction

The December issue of the Psi-k Newsletter carried the sad news of the death on October 25, 2012 of Professor Balazs L. Gyorffy, together with a reprint of a formal obituary that was written by Professor Robert Evans, FRS. As part of a broader tribute to Professor Gyorffy, this issue carries a compilation of contributions from some of Balazs' collaborators, friends and colleagues from within Psi-k and outside, reflecting on their experiences of working with him. Balazs' enthusiasm for science and life was irrepressible and a source of pleasure for those that shared in his scientific genius and knew him as a loyal and considerate friend. From the beginning, Balazs focused on what we now call "first principles" electronic approaches and eschewed the introduction of adjustable parameters whenever possible, often humorously positing that in condensed matter physics there is only one parameter - the electronic charge and that is known pretty well! At the center of Balazs' attention was the "electron glue" (possibly his coinage) [1] and how the interactions between the 10^{24} electrons that comprise it give rise to the richness of materials properties and functionalities.

In his long and highly productive career, Balazs made seminal contributions to many areas of condensed matter and materials physics - from the fundamentals of electronic structure theory, multiple scattering theory, alloy theory, superconductivity, magnetism, relativistic electron theory, strongly correlated electron theory, symmetry and symmetry breaking in alloy and magnetic phase transformations, to the subtle manifestations and consequences of Berry curvature in materials physics - a subject on which he published a number of papers in the years after his "official" retirement. Throughout his career, Balazs worked closely with experimentalists,

[1] While the etymology of this evocative term cannot for certain be established it is one that Balazs used early and much loved.

often pioneering the theoretical developments necessary for the theorist to "calculate what is actually measured" without the need of intervening models and unnecessary approximations. While obtaining the most direct contact possible with experiment was a cardinal principle, simply getting agreement with experiment was never sufficient - one had to understand how this agreement arose in terms of the underlying electronic structure from which one can carry the insights and intuition gained to more complex situations and new phenomena.

Although Balazs' scientific life was centered in Bristol, his influence on the progression of condensed matter and materials theory was international and inclusive. During his long career at Bristol, Balazs mentored many students and postdocs that since have gone on to illustrious careers of their own. Indeed many of these are now professors and senior researchers at major universities and laboratories in the UK, continental Europe and North America. In addition Balazs was a valued collaborator and advisor to innumerable scientists and research groups around the world. His ability to see the big picture, to make connections across apparently disparate fields of physics, and his deep insights into electronic structure theory and materials phenomena were an endless source of inspiration to many.

Beginning in the mid-1970's, Balazs took up the challenge of maintaining UK leadership in the development of first principles electronic techniques with his involvement (along with Volker Heine and others) in founding and sponsoring the Collaborative Computational Project-9 (CCP9) - a role that would be extended to the whole of Europe through his efforts in helping shape the Psi-k network. With this in mind it is therefore appropriate that the Psi-k newsletter has agreed to carry a series of articles of remembrance by a few of the many colleagues that had the privilege and pleasure of working with him. The spirit of these is not to chronicle the full scope of Balazs' contributions to science but to highlight some of the areas of greatest impact and provide insights into what it was like to be with him during these scientific adventures.

Malcolm Stocks: "If you have the opportunity, it is a good idea to work with the best"

I believe that I first met Balazs in late 1968 at the University of Sheffield when I was assigned to assist him in supervising an undergraduate Physics "problem" class. What is perhaps most remarkable about this is that I am no longer completely sure of these circumstances; remarkable because it is now so hard to imagine any interaction with Balazs without it being forever imprinted on one's memory. My second point of interaction was centered in Oak Ridge and would lay the foundation for much of my scientific career and greatly influence that of Balazs. The catalyst for this was Sam Faulkner who also met Balazs at Sheffield. Sam had been awarded a Fulbright Fellowship and was on leave from Oak Ridge National Laboratory (ORNL) in the USA. On returning to Oak Ridge, Sam invited Balazs to act as a consultant to the Theory Group in the Metals and Ceramics Division. This 1969 visit began a lifelong relationship with Oak Ridge. It is likely during this visit that Balazs first became interested in the electronic structure theory of disordered systems and multiple scattering theory - according to Sam "out of politeness" for being invited.

By the late 1960's it was becoming increasingly clear that the "coherent potential approximation"

(CPA) held a special place in the theory of disordered systems in that it was the first self-consistent effective medium theory of the effects of disorder on the electronic structure. In addition the CPA was shown to sit at the top of a hierarchy of such theories that included the virtual crystal approximation (VCA) and averaged t-matrix approximation (ATA). Unlike the ansätze of the VCA and ATA, the CPA provided a way to self-consistently determine the "best" effective medium whose properties would accurately represent the configurationally averaged properties of the real disordered alloy. In a nutshell, CPA enforces the condition that replacement of the effective or coherent potential by true electron-ion potential at a site (in modern parlance the Kohn-Sham potential at that site) produce no additional scattering when averaged over all possible occupancies of that site. Not surprisingly the "coherent potential" turns out to be a complex (literally) energy dependent quantity that has to be found iteratively.

Sam Faulkner, who had been a student of Jan Korringa at Ohio State University, was very aware of the power of the CPA. Indeed, his thesis had been on disordered systems theory and it was Jan Korringa who first proposed the ATA. In addition, it was Sam Faulkner (together with Harold Davis and Hugh Joy) who, after coming to Oak Ridge in the early 1960's, had developed the first Korringa-Kohn-Rostoker (KKR) electronic structure code - a code made possible by the power of the then leadership CDC and IBM computers at Oak Ridge. It was into this environment that I came as a postdoc and Balazs came as a visitor. During his first visit to Oak Ridge, Balazs wrote a paper on a generalization of the CPA to liquid metals with short-range order. I, on the other hand, was working on implementing the CPA within the context of generalized tight binding models to systems such as copper-nickel and silver-palladium. In addition I had also been reading the papers of Paul Soven who had proposed a method by which one could implement the CPA within a KKR-based "band structure" formalism. Try as I might, I could not see a way in which Soven's ideas could be converted into a practical computational approach. All that changed during a visit of Balazs to Oak Ridge in 1971 when he left me with a set of notes that he would subsequently publish in Physical Review B 5, 2382 (1972). These notes were a model of clarity, insight, and compactness that I found quite wonderful. The notes laid out a formalism in which the path to implementation of the CPA within the language of KKR was clear. It was this set of notes, together with a little bit of thinking, that persuaded me to move to Bristol to work with Balazs on what became KKR-CPA.

The essential insight of Balazs' formulation was to write the equations of multiple scattering theory in terms of what is now known as the scattering path operator. This formalism then allowed the KKR-CPA equations to be written down in a compact form and furthermore provided a method for calculating the scattering path operator in terms of the inverse of the KKR matrix which turned out to only depend on the t-matrix corresponding to the "coherent potential" at a site and the KKR structure constants that describe propagation between the sites and which in turn depend only on the lattice structure.

Despite the clarity of exposition and the clear pathway to implementation, actually developing the KKR-CPA was extremely difficult, not least because the computer power available at the time was woefully inadequate. Fortunately, we were soon to be joined in our endeavor by Walter Temmerman who was to take up the task of writing a code that used k-space integration to obtain the scattering path matrix while Sandro Guiliano and Roberto Ruggeri, who were visiting Bristol from the University of Messina in Sicily investigated obtaining them in real

space. It is interesting that Sandro refused to fly and so ended up driving the length of Italy and across the rest of Europe to Bristol, dragging Roberto along with him. From these small beginnings but with the endless optimism that 'it would all work out' that collaborating with Balazs engendered (sometimes in spite of considerable evidence to the contrary) the basis of the KKR-CPA methods and codes that we know today were slowly put together. To say that conversations with Balazs about how things were progressing could be animated is to point only to the shadow - charged with espresso, puffing his pipe, armed with a piece of chalk and access to the enormous blackboard that was in my office, "discussions" with Balazs were events to be savored.

For me, the scientific directions that were initiated in Bristol during the early 1970's set the stage for my career and allowed me to lead the privileged life of a research scientist - for the most part at Oak Ridge National Laboratory. In this I was not alone! To follow for a moment but one thread, charge self-consistency (SCF) was added to the basic KKR-CPA in collaboration with Hermann Winter in the early 1980's, in part while we were both spending a year as Walter's guests in Daresbury - Hermann was from the then Kernforschungszentrum Karlsruhe. The theory and implementation of KKR-CPA within DFT was the thesis topic of Duane Johnson while a student of Frank Pinski at Cincinnati. The implementation of the relativistic (Rel) KKR-CPA based on the Dirac equation was first taken up by Julie Staunton as graduate student with Balazs and would set the stage for Paul Strange and Hubert Ebert who followed on as Balazs' postdocs. The early 1980's saw development of the concentration functional approach, which made it possible to predict short-range ordering and order-disorder transformation temperatures entirely from first principles and to identify the features of the electron glue responsible for the specific clustering or ordering tendencies in particular alloy systems. These ideas were first implemented by John Wadsworth, another of Balazs graduate students, and were subsequently much further developed by Julie, Frank and Duane. In short, to work with Balazs was to be constantly assailed by new ideas and possibilities. So, it should be no surprise that so many of those mentioned have gone on to be distinguished scientists in their own right. Following additional threads would serve to multiply this many-fold.

There are many fables about Balazs that, over the years, have spread through the legions of his friends and collaborators. Sometimes Balazs elevated the "bit part" of the absent minded Professor to an Academy award winning performance - perhaps some of his wife Carole's acting and playwright theatrical training had rubbed off on him, but more likely it was intrinsic. For example, I am fairly sure that the number of ways that one can put a transparency on a overhead projector the "wrong way round" is finite - but not to Balazs. Sadly, modern technology and PowerPoint have largely removed the opportunity for young scientists to explore the possibilities.

Above all, Balazs was a kind and loyal friend who would go to great lengths to make sure that his colleagues were taken care of when visiting Bristol. For example, when my wife Barbara and young son Ian first arrived in Bristol, Balazs and Carole not only welcomed us but made sure that we could "inherit" the flat that they were vacating as they moved to their new home in Cotham. Sometimes, however, Balazs' kindnesses would also add to his mythology! During our time in Bristol I had to make a trip to the USA leaving my family behind. At some point Balazs, concerned for how Barbara and Ian were getting along in my absence, knocked on the door of the flat and was duly invited in. Out of politeness Balazs put the pipe that he had

been smoking on his arrival into the pocket of his overcoat. After some conversation, Barbara noticed that smoke was emanating from Balazs' pocket of which she promptly informed him. Unfazed, Balazs simply turned his pocked inside out emptying the burning embers onto the carpet, with quite predictable results - an encounter that remains forever burned on Barbara's memory. Balazs, we shall miss you.

Walter Temmerman: "My recollections of working with Balazs"

Balazs influenced my life profoundly. As a young Ph.D. student in theoretical physics in Prof. Ziman's group at the University of Bristol it was the custom to find one's own thesis adviser from the senior staff. In my days this involved Michael Berry, Bob Evans and Balazs Gyorffy. The project of Balazs Gyorffy, in collaboration with G. Malcolm Stocks, entailed writing a computer programme to solve the KKR-CPA equations for disordered alloys, making use of Ole Andersen's tricks of linearization approximation in band theory. What attracted me to this project was that it was very specific and its success rate could be measured. Also I liked the style in which Balazs and Malcolm where performing physics. A lasting memory of a discussion with Balazs and Malcolm is Balazs happily puffing his pipe, sitting on a pile of computer outputs which Malcolm stored underneath the blackboard.

Life with Balazs was never boring. My Ph.D. was funded by the British Council and after this I was fortunate enough to become Balazs' post-doc on a Science Research Council grant to study the k-dependent band structure of Cu-Ni alloys [Phys. Rev. Lett. 41, 339 (1978)]. But in my first year of post-doc, Balazs was gone on a sabbatical at Stony Brook. He had done nothing with respect to my appointment and his farewell message to me was to go to Senate House and present myself. That was to guarantee my inclusion on the university pay roll. As incredible as this was, it has worked.

Balazs was for life. As the first chairman of the newly created UK's CCP9 (Computational Collaborative Project on "Computational Studies of the Electronic Structure of Solids") in 1980, together with Volker Heine, they employed me as its scientific secretary, placed at Daresbury Laboratory. So, it seemed, I followed my thesis adviser in whatever I did. Balazs was also instrumental in my marriage to Dzidka Szotek, his post-doc, whom he sent from Bristol to Daresbury to get acquainted with the KKR-CPA code. This was the beginning of a very fruitful collaboration and a beautiful life. A few years later, the period of high temperature superconductivity (HTS) arrived. I jumped on the 'bandwagon' and focussed on exploring the materials specific aspects of the parent compounds of high T_c superconductors (HTCS), but in due course joined forces with Balazs. His idea was to solve the Kohn-Sham Bogolubov-de Gennes equations, with a suitably parametrized pairing interaction. Following an earlier study of Nb, involving the first-principles KKR electronic structure and a phenomenological attractive pairing interaction [Phys. Rev. B48, 1202 (1993)], for high temperature superconductors Balazs suggested to combine the so-called eight-band model Hamiltonian, derived from the first-principles electronic structure of $YBa_2Cu_3O_7$ (YBCO) by Ole Andersen, with a phenomenological electron-electron attractive interaction operating between different sites and/or orbitals of the model with a strength defined by a constant K [Phys. Rev. Lett. 76, 307 (1996)]. The eight-band model proved to provide

a good quantitative description of the first-principles local density approximation (LDA) band structure of the superconducting cuprates in the normal state within 2 eV of the Fermi energy ϵ_F. In this 'tight-binding model', orbitals located on non-generic structural elements separating the CuO_2 bilayers, such as the chain in $YBa_2Cu_3O_7$, were deleted. As a consequence, it was strictly speaking a two-dimensional (2D) model. Balazs wanted to focus on establishing where the Cooper force, binding electrons in these materials into pairs, was operating. He suggested to parametrize an electron-electron interaction in terms of the orbitals of the model for the electronic structure of the normal state [Phys. Rev. B58, 1025 (1998)]. Thus one could choose the attraction to occur between two electrons e. g. one at site **R** in the orbital labelled by L and the other at site **R'** and in orbital L', and describe it by the coupling constant $K_{RL;R'L'}$, giving rise to the pairing potential binding the electrons and holes in the appropriate, tight-binding, BdG equations, which would then be solved self-consistently to determine the quasi-particle spectrum and hence the superconducting gap in the ground state. The value of the coupling constant was so chosen that the predicted superconducting transition temperature T_c would agree with the experimentally observed one, namely T_c=92K for YBCO. Among many pairing scenarios the favourite one turned out to be a d-wave pairing, where the members of the Cooper pair resided on the nearest-neighbour Cu atoms in $d_{x^2-y^2}$ orbitals. The calculated gap anisotropy for this d-wave intralayer nearest-neighbour scenario agreed very well with the ARPES experiment of Schabel et al. [Phys. Rev. B 57, 2796 (1997)] for the sample with T_c = 92 K. Similarly, our calculated specific heat compared very well with the measurements [Phys. Rev. Lett. 73, 2744 (1994)] which gave us confidence that our BdG equations could be regarded as capturing the essential physics of the superconducting state. For the low temperature penetration depth our calculations at optimal doping was compared to measurements of A. Carrington et al. [Phys. Rev. B59, R14173 (1999)], revealing paramount importance of the chain in YBCO for zero temperature penetration depth. We have observed a linear dependence of the low temperature penetration depth at all studied doping levels [Phys. Rev B62, 3997 (2000)]. We have also shown that changing the chemical potentials, μ, in the eight-band model, for the favourite d-wave pairing, gave a good quantitative account of the observed rise and fall of T_c with the carrier concentration δ_n, namely doping [Phys. Rev. B58, 522 (1998)]. Moreover, we have obtained very encouraging results for the superconducting gap as a function of doping [Physica C 353, 23-28 (2001)], despite the fact that our approach has been strictly speaking a weak coupling BCS-like theory.

Our studies of the electronic structure of high T_c's have brought us to thinking about the importance of electron correlations in these materials. As suggested by Perdew and Zunger, one way of improving on the LDA description of electron correlations was to introduce the so-called self-interaction correction (SIC) to LDA, which we implemented for solids within our version of LMTO-ASA (SIC-LMTO-ASA) code in the beginning of nineties, and over the years applied to a variety of systems. But Balazs has invented his own "brand" of the self-interaction correction (SIC), the so-called local SIC (LSIC), formulated within the multiple scattering theory, which we have implemented within a KKR-CPA code [Phys. Rev. B71, 205109 (2005)]. Unlike in the SIC implementation within the LMTO-ASA method, where localized states have been described in terms of the exponentially decaying Wannier functions, in the LSIC-KKR-CPA, the localized states have been associated with sharp resonances. Using an 'alloy analogy', we have applied

this local SIC approach to studying the famous $\alpha-\gamma$ phase transition in Ce, and have been able to conclude that the transition is driven by entropy and not the internal energy. Also, we have shown that at high temperatures our first principles LSIC-KKR-CPA approach; where only the thermal fluctuations play a dominant role, has been comparable to the LDA+U+DMFT method. This local SIC contribution typified one of the many issues Balazs has always drawn our attention to. Namely, is it a local effect or does one need to consider the whole crystal lattice? He has been thinking about generalizing this first-principles effective one-electron static approach to a dynamical theory, using the non-local CPA averaging, giving rise to a dynamical mean field-like theory, but without U. Balazs has always referred to such a parameter-free many-body theory as "Life without U", and this has become synonymous with him. As we have already for a while been living in the world without HIM, "Life without U" appears to have acquired an additional complexity.

Julie Staunton: "Balazs"

Those of us lucky enough to have worked with Balazs appreciated his fascination and profound insight for Physics. As condensed matter and materials physicists we know that the essential question for our research concerns the complexity arising from the 10^{24} electrons in a material all interacting with each other and the heavier, slower nuclei as well as external fields. Balazs emphasised the subtleties and beauty of this complex glue and took his characteristically uncompromising approach in describing it. One should start from 'first principles' so that the results can be compared directly with increasingly sophisticated experimental measurements - a standpoint which means that there is no hiding place if the theory does not come up to scratch but enormous satisfaction when it does. This philosophy led him to be such a dominant figure internationally in driving the development of computational electronic structure to which the Psi-k Network owes him so much.

The description of the condensed matter physics of materials gets a little easier if one assumes that the nuclei position themselves on a perfect regular lattice and that thermal effects do not need to be addressed. Balazs impressed on us that Nature is not like this - all materials are disordered in some way - there are lattice defects, the nuclei vibrate about their average positions, the electrons behave differently at different temperatures etc. Balazs pioneered ways to treat the effects of disorder - producing the SCF-KKR-CPA method, as discussed by Malcolm and Walter above, for a quantitative description of disordered alloys and so on. The SCF-KKR-CPA, together with its cousin, the SCF-LMTO-CPA, remains one of the few first principles method for describing electrons in a disordered crystal which can be combined with DFT. With other colleagues I greatly enjoyed working with Balazs to use these ideas on magnetic materials and we devised a theory of metallic magnetism in which these SCF-KKR-CPA techniques deal with thermally induced local moment magnetic fluctuations [J.Phys. F 15,1337, (1984); PRL 69, 371,(1992)]. The resulting disordered local moment (DLM) picture was the first fully first-principles approach to the problem of the temperature dependence of magnetism in metals. In Balazs' words, "at no stage does it map the many-electron problem onto an effective Heisenberg model, and yet it deals, qualitatively, with both the ground state and the demise of magnetic long-range order at the Curie temperature in a material-specific, parameter-free manner". En-

route to developing this theory Balazs proposed a version of a constrained density-functional theory for configurations of non-collinear local moments, an approach now also used in ab-initio spin-dynamics calculations. He used a particularly elegant phrase - "temporarily broken ergodicity" - to describe the time scale separation between the relatively slowly varying local moment orientations and the other faster electronic degrees of freedom. One early success for the theory, which he was quick to realise should emerge from it, was the prediction of a wave-vector and energy-dependent local exchange splitting in the electronic structure of the paramagnetic state of ferromagnetic metals, later observed in PES and IPES experiments on bcc iron.

Balazs is well-known as one of the pioneers in the use of relativistic quantum mechanics to describe electrons in solids. He played a significant role in establishing the theory of magneto-crystalline anisotropy and analysing relativistic effects on spin-dependent transport, photoemission and magnetic X-ray scattering. As one small example, I recall the fun in revisiting with him the RKKY interaction, expressed of course with multiple scattering theory language, where we found the magnetic anistropy that comes when the jellium, in which the magnetic impurities are embedded, is treated with relativistic quantum mechanics [J.Phys. C 21, 1595, (1988)].

Balazs was exceptionally talented in interpreting the results of often detailed computational results and then producing an elegant, beautiful understanding of an effect. For example a number of us had carried out some calculations of the compositional ordering in NiPt alloys using the first-principles concentration wave theory that Balazs had developed earlier with Malcolm Stocks. We found the $L1_0$ ordering that is observed experimentally. For an isoelectronic transition metal alloy with its almost filled d-bands this was at first a bit of puzzle but Balazs showed how to interpret the electronic mechanism underpinning the size effect produced by the off-diagonal disorder in this alloy. It described electronically why "big" and "small" atoms tend to order into simple structures [PRL, 66, 766, (1991)].

Balazs was a fantastic lateral thinker with an encyclopedic physics knowledge - his ability to make connections and set a new context for apparently disparate topics in theoretical physics is one of the many reasons why physics and his co-workers will miss him so much. Many scientific meetings will be the poorer without his pertinent and thought-provoking points. His dynamism and enthusiasm made him such an inspirational character in Physics and such fun to work with. He showed what it is to be a proper, fully signed up Physicist. Even though Balazs was my Physics father (he supervised my PhD work), as he was to many others, it was because he was a 'Force of Nature' that I had assumed that he would always be around to offer advice and inspire with new ideas. Many of us are massively grateful to his support especially in the early stages of our careers. For my part he played the pivotal role in giving me the confidence to go for jobs, promotions etc. In one instance I am sure he used a technique learnt and adapted from his swimming coaches to build up my confidence. He always made sure that his PhD students broadened and deepened their physics education - e. g. in my case introducing me to Gauge Field Theories and pointing out the connection between the Higgs particle and condensed matter concepts. He was also very receptive and encouraging to ideas from young physicists even if kindly having to point out holes and hitches in them.

Over the last 10 years or so much work in condensed matter physics has been directed at materials whose electrons are particularly difficult to describe accurately owing to their strong correlations

but which have intriguing effects. A useful and productive approach such as LDA+DMFT has been to graft a simpler model of some of the electronic glue attributes onto a more complete, computationally heavier one based on DFT. This model has at least one parameter, a U. Over recent years, as Walter has commented, Balazs had been discussing with a number of us ways to address this physics from 'first principles' and to remove the need for this and other parameters, making his physicist's joke that we should go for 'Life without U'. Unfortunately that phrase has new poignancy which gets it into the title of this article and which we all are trying to come to terms with. Balazs - thank you for everything.

James Annett: "Balazs Gyorffy and Superconductivity"

Balazs made many contributions to the theory of superconductivity, from his early work on estimating electron-phonon coupling to theories of Cooper pair tunnelling and superconducting qubits for quantum computations. Here I just want to highlight some of the themes he was still actively working on in the most recent five to ten years. In particular some of this work had a clear link to ab initio electronic structure, notably in numerical solutions to the Bogoliubov de Gennes equations. While his other work such as on qubits drew on ideas from quantum optics, which went back to his original work on the laser with Willis Lamb.

Balazs took a very active interest in the possibility of spin triplet superconductivity in the material Sr_2RuO_4. He had first suggested that some nearly ferromagnetic metals could become triplet superconductors in a classic paper from 1977 [I. Foukes and B Gyorffy, Phys. Rev. B 15 1395-1398 (1977)]. Together with Karol Wysokinski and Grzegorz Litak (Lublin) we developed a simple phenomenological model based on the known bandstructure [Annett et al., Phys. Rev. B 66, 134514 (2002)]. The approach was very similar to his eight band work on high temperature superconductivity, described above by Walter Temmerman. Balazs was very interested in the additional possibilities caused by the existence of spin-1 Cooper pairs, and he was always looking for a possible "smoking gun" experiment which he could measure the Cooper pair spin directly. This was the motivation for his final project, together with Martin Gradhand, on Berry curvatures and Berry phases in spin-triplet superconductors and his hope was to find a unique spin transport effect which would be a signature of triplet pairing. While this was not completed at the time of his death, we were also able to confirm a pridiction he had made together with Hardy Gross and Klaus Capelle[K. Capelle et al, Phys. Rev. B 58, 473-489 (1998)] for the existence of circular dichroism in spin triplet superdcondutors[K.I. Wysokinski et al, Phys. Rev. Lett. 108, 077004 (2012)]. Again there is a conection to the ideas of Berry curvature for to the Bogoliubov quasiparticles, which he was following up at the time of his death.

Another system where spin triplet pairing can occur is in multilayers of ferromagnets and ordinary singlet superconductors. Again Balazs's interest in this went back to his earlier work, for example his work on ab initio spin-transport and spin-current torques in ferromagnetic multilayers [A. Vernes, B. Gyoerffy and P. Weinberger, Phys. Rev. B 76, (2007)]. Together with experimentlists Mark Blamire and Jason Robinson at Cambridge, Lesley Cohen at Imperial and Victor Petrashov at RHUL Balazs was very interested in developing the theory of spin-transport

in such multilayers, and again looking for a unique transport property which would prove conclusively the existence of spin triplet Cooper pairs. Daniel Fritsch was working together with Balazs and myself on this problem at the time of Balazs's death, and the work is still in progress. He was also exploring with Balazs Ujfalussy an idea to implement the Bogolubov de Gennes equations directly within an ab inito KKR codes.

Finally Balazs was always also looking for applications in superconducting systems of his favourite theories of CPA, non-local CPA (NLCPA) and the disordered local moment (DLM) method. Some years ago we implemented the CPA for disordered superconductors [A.M. Martin et al., Phys. Rev. B 60 7523-7535 (1999)], showing that it is the correct mean-field approach for treating disorder superconducting alloys, just as it is in the normal state. This was later extended to the non-local CPA [R. Moradian et al., Phys. Rev. Lett. 89, 287002 (2002)], extending the normal state work NLCPA done with Julie Staunton and Derwyn Rowlands to the superconducting case. One of Balazs's last PhD students, Garry Batt, then further extended this approach to the problem of 'Fermi arcs' in high temperature superconductivity, combining NLCPA and the DLM to consider a d-wave superconductor with phase fluctuations of the order parameter on neighbouring atoms. This fitted into Balazs's long standing interest in what he called the "pre-formed pair" scenario, by which the anomalous properties of underdoped cuprate superconductors was to be understood as the result of phase fluctuations of the superconducting order parameter[J. Quintanilla et al., J. Pys. Condens. Matter 14 6591-6600 (2002)].

These are only a fraction of the many papers which Balazs contributed to on the theme of superconductivity over his long and productive career. I apologize to the many collaborators, students and colleagues whose work with Balazs I have been unable to mention. Balazs made so many profound contributions and his energy and permanent enthusiasm will be sadly missed very much by all who knew him.

Ingrid Mertig: "To Balazs"

A whole community of scientists is indebted to Balazs Györffy for the tools they are using daily in their studies of metals and alloys. In addition, Balazs had a very strong and charismatic personality, an inextinguishable scientific curiosity, and a very broad knowledge in theoretical physics. He definitely stood out as a scientist of fascinating insight and knowledge, with an impressive record of pioneering contributions in theoretical solid-state physics.

My first contact with Balazs was a virtual one via "electron-phonon interaction". I was a PhD student at TU Dresden and calculated electron-phonon coupling constants for Lanthanides and Actinides based on the Gaspari-Györffy theory a method that he developed in 1972 to describe superconductivity in metals.

In real life, I met Balazs at the International symposium on electronic structure of solids in Gaussig (East Germany). He came to Gaussig with his enthusiasm and his charisma and presented his work on the KKR-CPA method for disordered alloys. Everybody was infected by the ideas and all members of the Dresden electronic structure team wanted to do KKR-CPA, meaning that all of us would have liked to collaborate with Balazs.

Since 1990, we frequently met in scientific meetings and have been members in the same European networks, particularly in Psi-k. At one of these meetings he tried to explain to me the Berry phase concept. We ended up with a "twisted belt" in a Bristolian pub. This was the beginning of our collaboration on relativistic effects in semi-classical transport theory based on the Boltzmann equation. Balazs started to visit my group regularly. Having Balazs as a guest for extended stays had a very strong and positive impact for the Martin Luther University and for the Max Planck Institute of Microstructure Physics in Halle.

For me the collaboration was like a "late postdoc" with Balazs. During exciting discussions he opened the door for us to the understanding of the role of Berry curvature in solid-state theory and we developed a method to calculate the Berry curvature within the KKR scheme. We shared the common interest in a relativistic Boltzmann equation and we have been working hard on this subject. This work is not yet finished and we promise to proceed. But we will miss Balazs's brilliant mind.

Peter Weinberger: "From phase shifts to time-dependent theories"

Balazs Gyorffy entered my life for the first time in 1975, when I wrote a letter to the editor on his paper in the Journal of Physics on the electron-phonon interaction making use of phase shifts. The calculation of the latter was how our common experiences started. After the letter to the editor, I got a very friendly collaboration offer from Balazs. Unfortunately I was still busy with my own version of fully relativistic KKR programme, an enterprise for which I was accused to be completely demented. I finally arrived in Bristol exactly on his 40th birthday. Now, anybody who knew about Balazs' commitments to 'Wiener Schnitzel', in particular of the rather large type, would have guessed that 'Schnitzels' were his dearest birthday wish. So there we were, since Carole gave up on this project. Finally we toasted bread together, grinded it in a coffee mill, since bread crumbs - most likely an unknown exotic ingredient at that time in England - were needed, prepared the Schnitzels and fried them in two pans. Balazs was happy! Phase shifts and Schnitzels started our friendship some 35 years ago.

Well, and then came the age of CPA. With the help of the Oak Ridge programme we finally got a relativistic version thereof running. I have to say that in the early eighties Balazs was probably one of the very few around who was convinced that a relativistic approach might turn out to be important. What followed were more visits to Bristol and quite a few summers Balazs, sometimes accompanied by Carole, spent in Vienna.

When after the first success of the TB-LMTO method I posed to Laszlo Szunyogh and Balazs Ujfalussy, in Vienna at the time, the question of why can't we do a similar thing in KKR and a new age in the KKR community started, triggering off quite a few common publications with Balazs, in particular, when a spin-polarized relativistic version thereof started to run. The Screened KKR method became standard since then - essentially thanks to the enthusiasm of Laszlo Szunyogh and Balazs Ujfalussy.

Although within a Greens function approach also electric and optical transport properties became computable on an ab-initio basis, it was Balazs who was a bit sceptical whenever we had current- or magnetic-field induced phenomena in mind. After about a two summer long dis-

cussion, working with Andras Vernes, we finally got it, the ultimate set of equations, namely a time-dependent Dirac equation in the presence of an external electro-magnetic field. Balazs was enthusiastic and insisted to send the paper to PRL, where it had a delay time of about a year, being finally only published as a rapid communication in PRB. This paper I still consider to be our greatest common success, since it is oriented towards the future, to time-dependent magnetic phenomena.

I am pretty sure Balazs loved to spend summers in Vienna. And he had very good reasons:

(1) There were all the Hungarian guys, Laszlo Szunogh, Balazs Ujfalussy, Laszlo Udvardi and a couple of bright PHD students from Budapest. They 'conversed' in Hungarian all day long.
(2) He got a separate swimming lane in the 'Arbeiterschwimmverein', in a 50 m pool, reserved only for him!
(3) He enjoyed tremendously to go down to the Naschmarkt for lunch, an open market nearby, in particular to a very small place serving giant Wiener Schnitzel (and a second one on the house, if the first one was not big enough), and
(4) We, he and I, got on very well as far as physics, politics and cultural engagements were concerned. It was an ideal matching of wave lengths.

From phase shifts to time-dependent theories: it was indeed a long way I had the privilege to accompany Balazs, it was for nearly half of his life-time.

Paul Durham: "Memories of Balazs"

His influence:

For those of us like Walter, Dzidka and I who were lucky enough to work and have daily contact with him when we were young, Balazs's influence was indelible. In those early days in Bristol while we were learning to be researchers we became imprinted with his way of thinking about physics and were all the better for it. Talking with Balazs was a terrific way to understand how physics all fits together. Moreover, you learned how your own work fitted into what everyone else was doing at the time all over the world. Balazs made you feel in the swim of physics. It goes without saying that he had loads of ideas, but the striking thing about him was the generosity with which he shared them with you. A chat with Balazs could give you enough material for a year's research and Balazs's interest in your own ideas gave you a tremendous surge of confidence - you knew you were on to something worthwhile. But most of all, it was such fun to be with him - he was a wonderfully attractive man in every way, and we are so glad to have known him.

The "Uncertinity" Principle:

Balazs spoke and, especially, wrote very good English. If you wrote a paper with him, it was always a good idea for you to do the easy bit - describing what you actually did - and to get him to write the introduction and conclusion, where he could deploy his powers of expression to the full on what he did best - explaining what the work meant. No doubt these powers derived from his very wide reading and he could talk marvellously about European and American literature. I once mentioned that I had just read Joseph Roth's novel - The Radetzky March - and Balazs

immediately launched into a fascinating disquisition on the book as a chronicle of the late Austro-Hungarian Empire. He had some foibles though. I never heard him talking about the uncertainty principle without him pronouncing it "uncertinity" - but then he made this version sound so right, somehow, that I sometimes use it myself.

Car story:

Most conversations with Balazs were very absorbing. A couple of years ago he was visiting Daresbury and I took him to lunch in my new car, of which I was very proud. We talked about physics during lunch, continued on the drive back to the Lab and then on into my office, covering my blackboard, as usual with Balazs. After a few minutes our security guys phoned to say that I had left my car not only unlocked and with the door wide open, but also with the keys in the ignition and the engine running. I guess that talking physics with Balazs was a full-brain activity.

Andrzej's story:

Andrzej Pindor came from Poland to Bristol in the late 1970's and early 1980's and worked with Balazs and Walter mostly on alloys, KKR-CPA and so on. Their discussions, conducted in perfect English voiced in (loud) central European accents, were celebrated in Bristol. Now in Canada, Andrzej writes:

"I certainly have a lot of fond memories of Balazs and our heated and often loud political discussions. It was so telling of Balazs that our quite different political views and the mentioned heated discussions did not affect not only our professional but also our personal relations. One case of such discussions sticks in my mind - we were having one of these discussions in a pub. The pub was closing and we moved to the street in front of the pub (I remember that Sam Faulkner was there too). It must have been well after 11 pm and at certain stage a window in one of the buildings opened and someone shouted [in a Bristol accent]: Be quiet! Go back where you came from and leave us in peace! Balazs, always quick on his feet, shouted back: Hey, I live here! (or something to this effect). This, however, cooled us down and we went our own ways postponing the discussion to a next time.".