

## ***Statement of Need for CCP3***

### **1. Please state the scientific area this CCP focuses on and give a brief description of the scope of its scientific remit.**

CCP3 is the Collaborative Computational Project for Surfaces. The focus of its activities lies in developing tools and methodologies for the computer simulation of the structure and properties of surfaces and interfaces, and the theory and codes used in interpreting associated experimental techniques.

Throughout its history CCP3 has flourished through close collaboration between theoreticians and experimentalists, tackling large-scale scientific software development projects, and maintaining and distributing key software resources. Illustrative of the software development activities of CCP3 are:

- DL-EXCURV, a code for the analysis of EXAFS data for local structure determination; (662 global licenses)
- CRYSTAL, a first-principles all-electron Gaussian-based code for predictive modelling of materials properties; (475 global licenses)
- DLVisualise, a graphical user interface (GUI) providing a common interface to CRYSTAL, GULP (a classical simulation code) and experiment-interpretation codes (DL\_EXCURV, DL\_LEED for the analysis of low-energy electron diffraction, ROD for the analysis of surface X-ray diffraction, ...); (1043 licenses since 2005).
- CASTEP: a first-principles pseudopotential plane-wave-based code for analytic and predictive modelling of materials properties; (140 UK academic licenses; significant industrial user base).

Many CCP3 developments have been collaborative projects with international research partners, exploiting opportunities for leveraged funding and world-wide expertise. Explicitly excluded from CCP3 remit is software for operating an experiment.

Through networking activities, in the UK, in Europe and the USA, CCP3 has provided hands-on training and user support, and fostered interactions within the UK surface science community. In doing so, CCP3 has gained a large user community, greatly enhancing the impact of UK research in the field.

### **2. Is this an existing UK CCP, or a new CCP? If it is an existing CCP, please explain the continued requirement for this CCP.**

CCP3 is an existing CCP, active since 1978. Overseen by a working group comprising of experimentalists, theoreticians, large-facility users, industry representatives, senior and junior scientists, the project has been constantly renewed through continuous engagement with its large and broad community, and through regular reviews of the working group membership.

The need for CCP3 is as strong as ever, with surfaces and interfaces playing a crucial role in determining the properties of materials that are being actively pursued in response to the major challenges of today:

- Environmental (e.g., corrosion, catalysis, ...)
- Security (e.g., chemical sensing, ...)
- Energy (e.g., solar energy harvesting, thermal management, ...)

- Technology (e.g., information processing/storage, catalysis, adhesion, lubrication, ...)

Surface and interface science is also fundamental to the field of nanotechnology, where the small dimensions mean surfaces and interfaces dominate the properties of the new materials and devices.

The science of surfaces and interfaces is extremely complex, even in the controlled lab environment. The need to understand the structure, energetics, electronic and magnetic properties, and chemical processes at surfaces under working conditions for practical applications is highly challenging, and a long-term goal in the field. Progress is achieved through the combined application of multiple experimental techniques, and computational modelling, the latter playing a role both in the interpretation and simulation of experiment and through the application of first-principles methods, capable of answering fundamental “what if” and “how” questions.

CCP3 underpins UK research in this field, by collating, standardising, maintaining and developing the software infrastructure needed by the community, by providing advice, expertise, training and support to users of numerous codes and packages, and as a forum for collaboration and discussion through which future software needs can be identified and met. Within its library of software are codes whose use is **essential** to the meaningful analysis and interpretation of a range of experiments, and unique tools that facilitate their use by an ever broadening user community.

Future challenges demand the continual development of these software resources. For example, a fundamental problem in first-principles modelling is that of electron correlation, and CCP3 core support has enabled continual code development to make available to users the latest theoretical developments. This includes ongoing work on hybrid exchange time-dependent density functional theory, making possible accurate simulation of excited state properties fundamental to research in solar energy harvesting. Furthermore, CCP3 support ensures codes that run on and are optimised for the latest computer architectures, enabling their use in investigating the most challenging of problems.

The existence of CCP3 has ensured high quality, longevity and exploitation of codes and tools, that are responsive to user needs and which adhere to the high software standards of the computational community. The need for CCP3 is therefore as great now as it ever was.

### **3. If this is an existing CCP, please give a summary of the existing support that this CCP receives, including core support, network activities and flagship projects.**

CCP3 has been able to satisfy the needs of an ever growing and broadening user community through:

- Core support: the service level agreement between EPSRC and STFC currently funds 2.35 FTEs, enabling: the continued development, maintenance and distribution of several codes in the CCP3 software library, including *ab initio* codes (CRYSTAL, CASTEP) and experiment-interpretation codes (e.g. DL\_PHASE, DL\_LEED, DL\_EXCURV, ROD, ...); the on-going development, maintenance and distribution of the DLVisualise GUI, and the development of new interfaces for the analysis of data from additional experimental techniques; porting and optimisation of codes onto HPC platforms; administrative support including maintenance of the CCP3 website and code portal, and organisation and running

of networking activities; teaching at training events; support for the preparation of flagship grants and European networking support grants; and involvement in demonstration projects.

- **Networking activities:** past national networking activities have been supported via network funding requests attached to flagship grant proposals. Support at the level of £12k p/a has enabled: biannual working group/management meetings; participation in CCP Steering panel meetings; scientific workshops and meetings; training courses on computational science (e.g. the annual *Ab Initio* Modelling in Solid State Chemistry (MSSC) schools); outreach activities such as provision of computational sessions in training schools for experimental methods (e.g. the theory day at the annual ISIS neutron training course); an international visitor programme. In addition, CCP3 has attracted EU funding for its international networking activities. It was a node in the EU-FP5 Synchrotron Radiation Research Theory Network (2000-2005), and the follow-up Light Source Theory Network (2006 – 2010).
- **Flagship projects:** CCP3 has a successful history of securing funding for flagship projects through responsive mode funding, and of bringing the investment to long-term fruition to a large community. The most recent flagship project ended on 31 Jan 2010, and enabled the analysis of single-molecule vibrational spectroscopy experiments. In the best tradition of CCP3, the project centred on collaboration between experimentalists, theorists, and STFC staff. Over the long-term flagship project funding has typically enabled one PDRA per annum to be employed on computational development activities.

#### **4. Please explain what types of EPSRC support this CCP would require.**

##### **a) Core support provided through the STFC Computational Science and Engineering Department.**

The ongoing core activities detailed above will continue to require support at the present level (2.35 FTE) beyond March 2011, with no gap in provision if the needs of the CCP3 user base are to be met. We believe a strong case can be made for further core support at the level of 1 FTE, beginning April 2011, required to meet the growing scientific challenges and particularly a **broadening** of the CCP3 user community. This will enable CCP3 to provide core support for the uniquely capable ONETEP code, a need that emerged during consultations with our user base.

##### **b) Support for networking activities.**

Continued operation of CCP3 in support of the surface and interface science community will require funds to support its range of UK network activities, at the level of £12k p/a. This is an immediate need, as the current review exercise has superseded normal submission of a project proposal containing network support provision. CCP3 will continue to participate in international bids to secure funding to support its overseas networking activities, and to this end is currently establishing a working relationship with the European Theoretical Spectroscopy Facility<sup>#</sup> (<http://www.etsf.eu>).

##### **c) Support for software development projects.**

The surface and interface science community has articulated needs for several innovative and challenging software development projects that are required over the medium term. These will need support at the level of 1 PDRA per annum, beginning six months after the announcement of the renewal of CCP3 support to enable appropriate planning.

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<sup>#</sup> ETSF offers a suite of ab initio spectroscopy codes and a mechanism for supporting experimental and industrial users, and opens up the possibility of making CCP3 codes available to European experimental and industrial scientists.

## 5. Please explain why each type of support is required and the potential impact of each type of support.

### a) Core support

This is essential for long-term evolution of codes, and the development and dissemination of expertise.

- Core support for *ab initio* codes: Codes used in modern materials modelling result from many man-years of investment, and if they are to continue to reflect the latest advances in electronic structure theory it is essential that they receive long-term, uninterrupted development support. This was strongly articulated to us by the community during the consultation exercise on which this document is based. Specific developments for which the surface and interface community expressed a need for core support include: i) efficient inclusion of finite temperature effects in computations, essential to draw consistent and meaningful parallels between experiment and computations; ii) the inclusion of spin-orbit interactions, to enable accurate modelling of spin structures, e.g. in candidate materials for spintronics applications; iii) tools for the interpretation of experimental spectroscopy and microscopy (e.g., optical, photo-emission, fluorescence, and core level spectroscopy) to maximise scientific insight extracted from experiment in a wide range of applications; and iv) development of the description of excited states and transport properties, to address topical science problems including solar-harvesting research.

The CRYSTAL, CASTEP and ONETEP codes have been identified by the community as those that are collectively required to satisfy these needs. Both CRYSTAL and CASTEP are well established, have large user bases, and incorporate ever expanding sets of functionalities. They are complementary due to their different foundations, with CASTEP best suited for studying metallic surfaces and interfaces whereas CRYSTAL has advantages in the treatment of magnetic and transition metal oxide surfaces and interfaces, important for catalysis. Both perform well on medium-scale problems (up to a few thousand atoms) typical of surfaces, interfaces and nanoparticles. ONETEP is a relatively new order-N code (linear scaling with problem size compared to cubic scaling of conventional algorithms) which has reached the development stage where core CCP support will assist in realising its enormous potential for predictive/analytical surface and interface science. The ability to tackle significantly larger systems will enable the community to study new science problems and undertake increasing realistic modelling, such as providing structural details of complex and defective surfaces, nanostructures, grain boundaries etc that are of technological importance but have hitherto been beyond the reach of first-principles approaches due to prohibitively large unit cells.

The core support will also enable the dedicated engagement of code developers in continued collaboration with experimentalists, who have flagged up the need for: (i) immediate access to theoretical collaborators for fast completion of break-through research; (ii) ongoing support to those experimentalists wanting to perform their own calculations under specialist supervision; (iii) fast access to advice and opinion on published theoretical research. This mechanism impacts very strongly in enhancing the effectiveness of the combination of theory and experiment.

The user community also recognises that the efficient exploitation of the latest HPC capabilities is essential to being able to tackle leading problems on the scale typical of modern surface and interface science. Core support enables code developers to work alongside the computer scientists expert of HPC architectures to ensure long-term development and maintenance of CCP3-supported codes on HPC platforms.

- Core support for analytical tools: The challenges of modern surface science where a full understanding of complex systems is achieved by applying an increasing number of experimental and modelling techniques continues to drive the need for effective analysis of experiment. This enhances productivity and reliability, and is essential for widespread uptake of modelling resources. Continuing support will enable CCP3 to further enhance the DLVisualise GUI in response to user demands (e.g., provide interfaces for the ONETEP code, and photoelectron diffraction experiment-interpretation codes), and in so doing provide a unified interface for experimentalists and modellers wishing to employ a wide range of tools in their investigations. Core support will also enable CCP3 to continue to maintain the existing library of analytic codes based on multiple-scattering theory (e.g., DL\_EXCURV, DL\_LEED) – the consultation exercise has underlined the tremendous on-going demand that exists for these resources.

Core support also plays a key role in administration of the project, such as maintaining the CCP3 web site and assisting in the preparation of proposals for flagship projects and EU networking support. It underpins the management and steering, and networking activities where CCP3-supported staff provide lectures and “hands-on” training sessions, and give presentations at meetings and conferences on the computational developments emerging from CCP3-supported activities. These activities are an essential part of maximising the impact of the work undertaken under the CCP3 banner, and ensure that the user community is made fully aware of new developments and tools and instructed in their use. This is especially beneficial to young scientists and new users.

**b) Support for networking activities.**

The networking activities of CCP3 are a *sine qua non* element in its efforts to support the UK surface and interface science community. Working Group meetings steer the project by providing a forum for discussion and collaboration, and a mechanism for identifying future software needs. Young scientists are encouraged to join, promoting their integration into the community. The Working Group includes members also active within other complementary CCPs, enabling effective cross-CCP collaboration. Regular schools and workshops address the demands for training and induction in the latest simulation and modelling tools, by a continually broadening and growing community. The visitor programme enables CCP3 to bring to the UK international experts for strategic collaboration and ensure the project remains at the forefront of the latest developments in the field. Science meetings provide essential opportunities to disseminate new computational and theoretical developments to a wide audience, to bring together experimentalists, theoreticians and experts from different disciplines to tackle increasingly interdisciplinary science problems, to foster discussion and collaboration, and to help shape the future path of the project.

**c) Support for flagship projects**

The CCP3 community has expressed a need for ongoing support of collaborative software development projects. These are essential to catalyse a cohort of researchers, often of mixed theoretical and experimental expertise, around step-change projects in computational development at the leading edge of surface and interface science. Flagship support ensures projects can focus on maximising the utility to the community of the resulting codes, which can be in conflict with the need to obtain novel science and publications within conventional research projects, as well as ensuring software developments feed through to as many as possible of CCP3's codes, rather than just one. The potential impact of such projects is best exemplified through examples of specific needs expressed by the CCP3 community in this area, given below. As in the past, we

envisage the Working Group playing a fundamental role in prioritising and shaping flagship project activities.

- The development of a spectral function solver to interface to first-principles codes within the CCP3 stable, enabling the interpretation of experimental angle-resolved photoemission spectroscopy (ARPES). This cutting-edge experimental method, for example under development at the DIAMOND Light Source, is present at synchrotron facilities world-wide. This software development would enable scientists using ARPES to maximise the output of their experiments in terms of scientific insight, and would provide standardisation of ARPES data interpretation enabling meaningful comparison between different ARPES experiments.

- The development of refinement techniques in DLVisualise for the interpretation of combined experimental techniques. Current methods for quantitatively determining surface atomic structure surfaces are based upon refining agreement between data obtained from a given experiment (e.g. LEED, SXRD, photoelectron diffraction, optical or X-ray spectroscopies) and simulation based upon structural models. An ability to refine structural models by simultaneously interpreting multiple experimental techniques combined with first-principles modelling will significantly increase the accuracy and reliability of structural determinations.

- Step-change projects for the development of an efficient description of excited states, electron transfer and chemical dynamics in a complex surface environment, such as the development of efficient methods for the treatment of excited states including real time nuclear propagation. The scientific opportunity realised by such projects is an understanding of the role of excited states in the functioning and design of photoinduced electronic and chemical processes at surfaces, for example to optimize transduction of solar energy to chemical or electronic forms.

- Multi-scale modelling projects (e.g. large catalysis problems, fracture etc.). The aim here is to obtain information about properties at the macroscopic scale, based on the fundamental mechanisms identifiable at the atomistic scale which is the more traditional focus of CCP3 activity. With such problems typically needing tailored solutions requiring expertise and codes spanning two or more CCPs, CCP3 members see cross-CCP flagship projects as playing a vital role in tackling them.

**6. Please describe who will benefit from the existence of this CCP, including the number and type of researchers in the UK who are likely to benefit and the research disciplines that it will benefit.**

The ubiquity of surfaces and interfaces in nature and their prominent role in determining the properties and function of systems mean that beneficiaries of CCP3 span a very wide range of research disciplines including: physics, chemistry, materials science, biochemistry, nanotechnology, chemical engineering, earth and environmental science.

Illustrative of the scale and scope of the user community benefiting from CCP3 are:

**DL\_EXCURV**: 122 UK/662 global licenses have been issued, each typically supporting a research team. Users mostly University researchers: students, PDRAs, fellows, through to Professors. **DL\_PHASE**: 179 global licenses; **DL\_LEED**: 7 global licences; **DL\_ROD**: currently under development, expected user base 10-12 UK academic research groups, experimentalists using beamline facilities. **DLVisualise**: Since v2.5 released in 2005, 180 UK licenses/1043 global licenses have been issued, each potentially supporting a research team. Users mostly University researchers: students, PDRAs, fellows, through to Professors. **CASTEP**: 140 UK Academic licenses have been issued. Users include a significant experimental community (e.g. solid state NMR and neutron spectroscopy). Also

used by ... **CONFIDENTIAL CRYSTAL**: Some 50 UK institutions/475 institutions world-wide hold a CRYSTAL license, permitting research group usage. These are mostly Academic or governmental institutions, including Universities and national research centres. We attach letters of support from: a typical experimental user group; the UKCP Consortium; ISIS user community; and the HPC Materials Chemistry Consortium.

This document articulates the community need for future core-support for the **ONETEP** code. This code has a fast-growing user base: ...**CONFIDENTIAL**

Flagship projects have the potential to further extend the CCP3 user base/beneficiaries e.g. development of ARPES-interpretation code is expected to benefit around 20 UK Academic groups involved in both surface science and, beyond the conventional CCP3 community, researchers working on strongly correlated materials.

Networking activities primarily benefit the core surface science community (some 30+ groups in the UK). Individual workshops and meetings attract ~50 delegates. Training activities are designed primarily for new users, and particularly benefit young researchers (and consequently their host groups) – the *Ab Initio* MSSC school attracts 50 students p/a, the annual ISIS neutron training school 30 students p/a. We estimate 100 undergraduates, postgraduates and junior researchers per year are beneficiaries through involvement in projects that employ CCP3-supported codes.

**7. Please make any other comments that you think are relevant to the statement of need.**

From the earliest days when its principle focus was the development of multiple-scattering based codes for experiment-interpretation, through to its more recent embracing of first-principles methods and the synergistic combination of theory, experiment and simulation, CCP3 has been charged with providing the computational tools demanded by the surface and interface science community, and looks forward to a continued future fulfilling this role.

**8. Who was involved in preparing this statement of need? Please list name, institution and research interests.**

This statement of need was prepared through a process of widespread consultation and dialog with various groups including: the current CCP3 Working Group (about 35 members including experimentalists and theorists); the user bases of CRYSTAL, DL\_EXCURV, CASTEP, DLVisualise, ONETEP, DL\_ROD, DL\_LEED (numbers in sec. 6) and their code developers; the Thomas Young Centre, which is an interdisciplinary community of 80 London research groups working to address challenges of society and industry through the theory and simulation of materials; the HPC Materials Chemistry and UKCP consortia, each with nearly 100 registered users; the scientists using and developing the large experimental facilities Diamond Light Source and ISIS neutron facility; in Europe, the ETSF community. A request for statements of need was published on the CCP3 website. In addition to meetings of the CCP3 working group, the opportunity to gather input was also taken at a number of other events, including: the *Ab Initio* MSSC school, the Synchrotron Radiation User Meeting, and the Thomas Young Centre meetings.

Theoretical, computational, and experimental scientists providing key input were:

- Theoretical and computational scientists:
  - Dr S. Crampin (Department of Physics, University of Bath) current CCP3 Chairman: electronic structure and dynamics.
  - Dr B. Montanari (Computational Science and Engineering Department, STFC Rutherford Appleton Laboratory), current CCP3 science officer: organic materials and nanostructures, metal oxides and materials for photovoltaics.
  - Prof. A. Fisher (Physics and Astronomy Department, University College London): spin dynamics, quantum information processing, conduction in nanostructures, scanning tunnelling microscopy.
  - Dr. P. Haynes (Department of Materials and Physics and Thomas Young Centre, Imperial College London): development of linear-scaling methods (ONETEP) and their application to nanostructures.
  - Dr. A. Mostofi (Department of Materials and Physics and Thomas Young Centre, Imperial College London): development of methods for large-scale electronic structure calculations and their application to the study of defects in semiconductors, transport in nanostructures, strongly-correlated systems and weakly-bonded systems.
  - Prof. M. Finnis (Department of Materials and Physics and Thomas Young Centre, Imperial College London): exploring the links between electronic structure of materials, the behaviour of their atoms, thermodynamic functions and the evolution of microstructure.
  - Prof. R. Godby (Department of Physics, York University): *ab initio* theory of electronic structure and dynamics in matter.
  - Prof. G. P. Srivastava (Department of Physics, Exeter University): surface physics, nanophononics, nanostructures and graphene-based systems.
  - Prof. Werner Hofer (Surface Science Research Centre, University of Liverpool): tunnelling spectroscopy, surface adsorption, supramolecular structure, transport.
  - Dr. L. Bernasconi (Computational Science and Engineering Department, STFC Rutherford Appleton Laboratory): theoretical electronic spectroscopy, *ab initio* molecular

dynamics, quantum chemistry.

- Dr. C. Bailey (Computational Science and Engineering Department, STFC Daresbury Laboratory): electronic structure methods, metal fluorides, materials for photovoltaics and graphene-based systems.
- Dr. B. Searle (Computational Science and Engineering Department, STFC Daresbury Laboratory): graphical user interfaces, core level spectroscopy, transition metal oxides.
- Dr. K. Refson (Computational Science and Engineering Department, STFC Rutherford Appleton Laboratory): lattice dynamics, metal hydrides, materials under extreme pressure.
- Prof. Nicholas Harrison (Computational Science and Engineering Department, STFC Daresbury Laboratory and Thomas Young Centre, Imperial College London): predictive modelling of advanced materials.
- Experimental scientists:
  - Dr. C. Nicklin (Diamond Light Source Ltd.): Surface X-ray diffraction, ultrathin oxide layers, dynamics of phase changes, self assembly on templated substrates
  - Prof. P. Woodruff FRS (Department of Physics, Warwick University): experimental quantitative determination of the structure of atomic and molecular adsorbates on surfaces of metals and oxides using electron, photoelectron and X-ray diffraction, and ion scattering
  - Dr R. Lindsay (Department of Materials, Manchester University): surface structure and chemistry of metal oxides; surface chemistry of corrosion and its inhibition; oxide based solar cell devices
  - Prof. P. Weightman (Department of Physics, Liverpool University): mechanisms of biological organisation using high power terahertz sources and studies of molecular interactions at solid-liquid interfaces.
  - Dr Moritz Hoesch (Diamond Light Source Ltd.): lattice dynamics from inelastic X-ray scattering and angled-resolved photoemission (ARPES).
  - Prof. A. F. Lee (Department of Chemistry, Cardiff University): metal and metal oxide nanoparticles for catalytic clean technologies including biofuel synthesis, surface adsorption and reactivity, X-ray synchrotron methods, magnetic thin films and bimetallic nanostructures.
  - Dr. Mervyn Roy (Department of Physics and Astronomy, University of Leicester): quantum dots, electronic structure, metallic nanoclusters, X-ray polarimetry.
  - Prof. Geoff Thornton (Department of Chemistry, University College London) nanoscience and surface science of metal oxides.
  - Dr Adrian Finch (Department of Earth Sciences, University of St Andrews): climate change.
  - Prof. Steven P. Brown (Department of Physics, University of Warwick): Experimental Solid-State NMR, notably investigations of hydrogen bonding and  $\pi$ - $\pi$  interactions that drive self-assembly of organic molecules in the solid-state, e.g. applications to pharmaceuticals, supramolecular chemistry.