Advanced Electronic Structure Methods for Solids and Surfaces

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Abstract: This Special Topic Issue on Advanced Electronic Structure Methods for Solids and Surfaces contains a collection of research papers that showcase recent advances in the high accuracy prediction of materials and surface properties. It provides a timely snapshot of a growing field that is of broad importance to chemistry, physics, and materials science.

The Journal of Chemical Physics has recently had a successful Special Topic Issue on "Advances in Density Functional Theory". The current Special Topic on "Advanced Electronic Structure Methods for Solids and Surfaces" includes a number of significant Density Functional Theory (DFT) papers, however, the focus is on complementary methods using approaches such as periodic wavefunction methods, embedded quantum chemistry, and quantum Monte Carlo.

The accurate determination of the energetics of solids and surfaces and how molecules interact with them is of critical importance to a long list of phenomena, ranging from gas storage, to catalysis, to the behavior of materials at extreme pressures. Over the last twenty years or so DFT has emerged as the method of choice to make predictions about materials properties based on their electronic structures. Indeed DFT has been spectacularly successful and is one of the great success stories of physics and chemistry. However, DFT has many limitations in its widely applied forms and for many problems in materials science is severely lacking (see e.g. Kieron Burke's Perspective article in this Journal [1]).

There are, of course, many other electronic structure theories that can be applied to the condensed phases of materials, including various flavors of quantum Monte Carlo, periodic-wavefunction methods and embedding techniques that now make highly accurate predictions of the energetics of solids and surfaces possible (see e.g. [2-6]). Although many theoretical and technical challenges remain, it is clear that the use of many-body and quantum chemical methodologies has enormous potential in bringing about a systematically improvable *ab initio* methodology for condensed matter systems.

This Special Topic Issue showcases some recently developed and new approaches as well as their application to a range of important problems in materials science, surface science and catalysis. It includes several examples where periodic second-order Møller-Plesset (MP2) perturbation theory has been applied to condensed phase systems [7,8,9]. Amongst these is a particularly noteworthy contribution from VandeVondele and colleagues in which geometry optimization, full cell relaxation, and energy conserving MP2-based molecular dynamics simulations have been performed for a variety of molecular crystals, with demonstration calculations on a unit cell containing as many as 160 water molecules. Also included in the Special

Topic are a number of interesting and important adsorption studies using so-called incremental approaches [10], a form of DFT in which many body dispersion is taken into account [11], as well as a hybrid MP2:DFT scheme [12]. The use of explicitly correlated (R12/F12) methodology in condensed matter systems, which greatly accelerates basis set convergence and thereby dramatically reduces the cost of the calculations, is also demonstrated by Grüneis [9].

We have only highlighted a few of the many excellent contributions that can be found in this Special Topic Edition. This set of papers makes it clear that the development and application of advanced electronic structure techniques to the condensed phase is a thriving area of research. We hope you enjoy reading the papers in this collection as much as we enjoyed putting it together!

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