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FOREWORD

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Guest Editor

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It would be fair to say that in the past few decades, theory and computer modeling have played a major role in elucidating the microscopic factors that dictate the properties of functional novel materials. Together with advances in experimental techniques, theoretical methods are becoming increasingly capable of predicting properties of materials at different length scales, thereby bringing in sight the long-sought goal of designing material properties according to need. Advances in computer technology and their availability at a reasonable cost around the world have made tit all the more urgent to disseminate what is now known about these modern computational techniques.

In this special issue on computational methodologies for materials by design we have tried to solicit articles from authors whose works collectively represent the microcosm of developments in the area. This turned out to be a difficult task for a variety of reasons, not the least of which is space limitation in this special issue. Nevertheless, we gathered twenty articles that represent some of the important directions in which theory and modeling are proceeding in the general effort to capture the ability to produce materials by design. The majority of papers presented here focus on technique developments that are expected to uncover further the fundamental processes responsible for material properties, and for their growth modes and morphological evolutions. As for material properties, some of the articles here address the challenges that continue to emerge from attempts at accurate descriptions of magnetic properties, of electronically excited states, and of sparse matter, all of which demand new looks at density functional theory (DFT). I should hasten to add that much of the success in accurate computational modeling of materials emanates from the remarkable predictive power of DFT, without which we would not be able to place the subject on firm theoretical grounds. As we know and will also see from the collection of works here, DFT also provides a platform for testing, improving, and evaluating the feasibility of more approximate methods whose need has become even more urgent. This is understandable since functional materials, given their limited translational symmetry, necessitate the usage of unit cells with a large number of atoms (sometimes in hundreds). Even if DFT codes were efficient enough to handle several hundred atoms in the calculational super-cell, the extraction of equilibrium geometry for such systems requires injection of more efficient methodology, as geometry is the input and not the output of a DFT calculation. Equally important is the need to calculate the temperature dependencies of material properties and for simulations to be carried out at length scales suitable for incorporating kinetic effects from competing processes and cooperative effects from constituting entities. It is true that codes based on DFT are becoming increasingly efficient and that methods such as *ab initio* molecular dynamics simulations are available for simulations of systems at temperatures above 0 K. However, such approaches still have a way to go before they can be readily applied to materials with complex geometries and composition, and for time and length scales that are relevant to realistic environments in the laboratory. Several articles here represent some of the recent advances towards 'multi-scale' modeling of materials.

Among the articles that focus exclusively on DFT, the contribution by Weinert *et al* [1] summarizes some of the advances made to better describe magnetic properties and entropic effects. The article by Kyrychenko and Ullrich [2] discusses recent developments in time dependent DFT to describe transport properties and absorption spectra of solids. Their model allows for a comprehensive treatment of electron–electron interaction, screening and correlation effects which are necessary for proper description of properties of the excited state. The contribution by Langreth and co-workers [3] summarizes their recent efforts at incorporating non-local van der Waals forces into DFT so as to make it suitable for accurate description of the physical and chemical properties of the ground state of sparse/soft matter. Their applications to molecules, layered systems, and hybrid structures are promising and mark the beginning of work in another important set of materials for which insights could be obtained from DFT. The paper here by Tang *et al* [4] focuses on the usage of grid-based methods for calculations of local charge densities. The virtue of the method is that charge densities are not confined to a lattice. Finally, as applications of DFT, the article by Groß [5] is representative of the usage of DFT in tailoring the electronic structures of surface alloys and other nanostructures, while the contribution by Bohnen *et al* [6] is a further example of the applicability of density functional perturbation theory in accurate descriptions of the lattice dynamics of functional nanomaterials such as carbon nanotubes.

For the modeling of amorphous materials, Biswas and co-workers [7] present a review of methods such as the reverse Monte Carlo (RMC) and 'experimentally constrained molecular relaxation' models which impose constraints to ensure that the final model meets *a priori* requirements on structure, topology, chemical ordering, etc. In a similar vein, the papers by Rossi and Ferrando [8], and Rogan *et al* [9], summarize advances in the determination of the equilibrium structure of nanoparticles and nanoalloys through global optimization strategies such as genetic and Basin-hopping approaches, diversity-driven unbiased searches and the conformational space annealing method. Structure determination itself relies on the knowledge of the system energy landscape, the saddle points and the transition states. In this issue the work of Pedersen *et al* [10] is an example of how a saddle point search method can be used to study dislocation mobility in a covalent material, which can be a very challenging task for a complex material. Trushin *el al* [11] present a related procedure for understanding atomistic mechanisms and energetics of strain relaxation in heteroepitaxial systems and transitions from the coherent epitaxial (defect free) state to the state containing an isolated defect (localized or extended).

To facilitate the simulation of rare events, Fichthorn *et al* [12] elaborate on the adoption of the bond-boost method for accelerated molecular dynamics (MD) simulation and its application to kinetic phenomena relevant to thin-film growth. They also present the state-bridging bond-boost method to address the dynamics of systems residing in a group of states connected by small energy barriers and separated from the rest of phase space by large barriers. In the genre of accelerated schemes which also seek to address the issue of completeness in the determination of reaction rates we include here the 'off-lattice' self-learning kinetic Monto Carlo method presented by Kara and co-workers [13] and its application to atomic cluster diffusion on fcc(111) surfaces. Further ramifications of the self-learning kinetic Monte Carlo method are presented in the paper by Nandipati *et al* [14], who apply the recently developed optimistic synchronous relaxation (OSR) algorithm as well as the semi-rigorous synchronous sublattice (SL) algorithm for parallel computation of the coarsening of islands on fcc(111) surfaces.

The above and related methods also lend themselves to the examination of morphological evolution of functional materials. The contribution by Hamouda *et al* [15] summarizes the effect of impurities on epitaxial growth and on shape evolution of systems. Similarly, using an atomistic lattice-gas model Li *et al* [16] describes the key features of the complex mounded morphologies which develop during deposition of Ag films on Ag(111) surfaces. Also, using a combination of a Monte Carlo method and continuum elasticity theory, Uhlík *et al* [17] present an efficient computational method for finding the equilibrium concentration profiles which minimize the free energy of intermixed heteroepitaxial islands of assigned shape and composition.

The contribution by Leuenberger and Sham [18] establishes how the process of Umklapp-assisted recombination can be used to optically detect the spin state of the nucleus of a phosphorus donor. They present two methods to improve the optical detection of the spin state of a single nucleus in Si:P. The work of Ni *et al* [19] is an example of the application of the molecular dynamics technique to determine the thermal conductances across covalently bonded interfaces between oriented single crystal diamond and completely aligned polyethylene chains. Finally, the paper by Yildirim *et al* [20] illustrates the application of standard lattice dynamics and molecular statics methods to identify the novel characteristics of nanoalloys, as a function of composition and geometry.

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