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Editorial

First-Principles Approaches to Metals, Alloys, and Metallic Compounds

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1. Introduction and Scope

At the beginning of the 21st century, electronic-structure theory has matured to a degree that allows for accurate phase prediction and computational characterization of various kinds of materials; in particular, elemental metals adopting whatever allotropic structure, various intermetallic compounds, and other complex metal-rich phases. Hence, fundamental theoretical progress has been made and is rapidly continuing in both physics and chemistry. From a more applied, engineering-like perspective, there is an urgent need for novel metallic structural materials, such as advanced steels, to address future challenges arising in both mechanical and civil engineering as well as energy production and conversion. While it is clear that different microstructural features influence the macroscopic behavior, modern techniques for simulation and modeling of metals and intermetallic phases at the atomic scale may enormously accelerate and guide the entire development process. In particular, atomistic understanding is a key issue because it allows for the generation of (spin-dependent) structural models of crystalline phases and the calculation of enthalpies and other free energies as a function of pressure and temperature. In combination with evolutionary algorithms and advanced thermochemical and phase-field approaches, these methods provide a solid ground for a novel methodological approach to the physics, chemistry, and engineering of metals and metal-rich materials. Furthermore, fundamental insights obtained in this manner may be incorporated, either as input parameters or key assumptions, into larger-scale models, whether purely theoretical or computational, rendering atomistic simulations essential for the development of multiscale approaches. Thus, this Special Issue focusing on first-principles approaches to metals, alloys, and metallic compounds tries to follow that train of thought, and it also aims at allowing for a wider perspective on metallic materials, to be studied by physicists, chemists and materials scientists, as well as engineers.

2. Content

To begin with, and as a timely object, high-strength high-manganese steels are at the very core of modern metal engineering, so Sevsek and Bleck [1] demonstrate an ab initio-based modelling of high-manganese steels depending on first-principles calculations of short-range ordering energies, a question of paramount importance for the Collaborative Research Centre 761 ("Steel ab initio") funded by the German Research Foundation (DFG). Both configurational structures and the impact of alloying elements are analyzed, finally providing good agreement with experimental data. In a somewhat similar manner, Song et al. [2] provide a combined small-angle neutron scattering and ab initio investigation on the Mn–C short-range ordering in an X60Mn18 steel. Not only does the experiment prove the presence of such ordering upon recrystallization, theory provides evidence for cluster formation and its evolution, which also translates into a stress-strain curve. The role of carbon, in particular carbon precipitates, is covered in the contribution by Sawada et al. [3] using the examples

of titanium and niobium carbide. While the interface energy between carbide and iron is obtained via large-scale first-principles theory, the estimated coherent-semi-coherent TiC transition diameter agrees with experiment.

The aforementioned three papers already allude to bridging the gap between atomistic and continuum levels, directly covered in the contribution by Korbmacher et al. [4] who utilize Ni–H as a reasonable system to model phase equilibria. By considering various effects, they arrive at a fully quantitative agreement for the chemical potential without adjustable parameters. Likewise, Weikamp et al. [5] present a selection of scale-transfer approaches from the electronic to the continuum regime for topics relevant to hydrogen embrittlement. Eventually, they develop an approximative scheme to estimate grain-boundary energies for varying C and H contents, and they consider the dependence of hydride formation on the grain-boundary stiffness. When it comes to time evolution and dynamical phenomena, the paper by Zhang and Jiang [6] deals with molecular-dynamics simulations of crack propagation in nanoscale polycrystalline nickel. The strain rate has an important effect on the mechanism of crack propagation, and for higher strain rates local, non-3D-crystalline atoms show up, and Lomer–Cottrell locks are formed.

If we ignore nonmetallic elements for the moment and focus on intermetallic binaries, Herrig et al. [7] show how to perform low-temperature syntheses of smooth face-centered FeMn thin films provided proper guidance by ab initio theory. The latter indicates very strong interfacial bonding of the Cu nucleation layer to an alumina substrate and between fcc FeMn and Cu, hence local epitaxial growth is enabled. With respect to binary phases such as HfOs, HfIr, and HfPt, Li et al. [8] study their structural, electronic, and elastic properties using first-principles theory and confirm the order of thermodynamic stability as HfPt > HfIr > HfOs. On the other side, the calculated bulk moduli follow the order HfOs > HfIr > HfPt, and the anisotropy of acoustic velocities, Debye temperatures, and thermal conductivities are obtained.

Coming back to the critical role of hydrogen, Hüter et al. [9] present a multiscale modelling of H transport and segregation in polycrystalline steels from a chemo-mechanical model taking into account stress gradients as well as microstructural trapping sites; the energetic parameters are determined from ab initio calculations. A scale-bridging description of dislocation-induced H aggregation is accessible, but there are limitations hindering a quantitative comparison to experimental data. Likewise, Timmerscheidt et al. [10] investigate possible H-trapping effects connected to the presence of Al in the grain interior by employing density-functional theory, and they aim at understanding the relevance of short-range ordering effects because of the occurrence of Fe_3AlC κ-carbides. The individual H–H/C–H interactions are repulsive, but Mn enhances H trapping. All that can be expressed mathematically, such as to numerically describe hydrogen embrittlement. And yet, full hydrogen content bridges the gap to inorganic chemistry as shown by Gong and Shao [11] who model stability, electronic structure, and dehydrogenation of pristine and doped 2D MgH_2 from first principles. The study has implications regarding dynamical stability and dehydrogenation, and it shows that the Mn-doped system exhibits good performance for hydrogen storage and dehydrogenation kinetics.

That being said, this Special Volume includes 11 original contributions, and 7 of them deal with high-manganese steels which have come to light within CRC 761 (“Steel ab initio”). In particular, the research deals with short-range ordering from experiment and theory, and the contributions also highlight carbide-like precipitates. In addition, the authors of this volume bridge the gap between atomistic and continuum levels, in the spirit of scale-transfer approaches, in particular for hydrogen embrittlement. Then, molecular-dynamics simulations play their role in terms of crack propagation. First-principles theory is helpful for growing better intermetallic thin films, and such approaches predict structural and elastic properties of metallic binaries, too. Also, multiscale modelling of hydrogen transport is provided, and the chemical reasons for H-trapping κ-carbides are highlighted. Eventually, stability and dehydrogenation of metal hydrides are looked at. Indeed, first-principles theory has acquired a firm and supportive role in the fundamental and applied research of metals, alloys, and metallic compounds. What a wonderful evolution to witness and also to be part of!

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