**All-electron electronic structure theory for new materials for light harvesting and light emission**

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Materials for light harvesting and light emission are immensely fertile ground for electronic structure theory, for the simple reason that key physical phenomena behind a particular materials' function arise at the atomic scale. Key challenges for theory include: Structures can be complex; the computationally desired phase may compete for stability with an *a prio*ri unknown zoo of possible secondary phases; accurate predictions of excited-state phenomena in large structures remain a challenge for the field. In this talk, we describe our progress on these issues using the all-electron approach in the FHI-aims code [1], a high-accuracy, scalable and affordable general-purpose code for molecular and materials simulations. Examples include the nature of polymeric carbon-nitride materials for photocatalytic hydrogen evolution [2], new quaternary semiconductor materials I2-II-IV-VI4 (I=Cu, Ag; II=Sr,Ba; IV=Ge,Sn; VI=S,Se) for photovoltaics [3,4], and identifying the nature of carried levels in layered organic-inorganic perovskites with optically active organic components by large-scale hybrid density-functional theory[5]. We finally describe ongoing work on an open-source infrastructure "ELSI" to bridge between electronic structure user codes and massively parallel solver libraries for the large-scale (generalized) Kohn-Sham electronic structure problem, and we describe progress on distributed-parallel GPU architectures.

This overview talk covers joint work with many colleagues around the world, including the groups of Bettina Lotsch (Stuttgart/Munich), David Mitzi (Duke), Yosuke Kanai (UNC), Matthias Scheffler (FHI Berlin), Xinguo Ren (USTC Hefei), group members at Duke University, and the very large developer community of the FHI-aims code - all these and further interactions, as well as support from the National Science Foundation and the US Department of Energy, are gratefully acknowledged.

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