Exploring the promise of diffusion Monte Carlo for Materials Research

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Electronic structure calculations based on Density Functional Theory (DFT) play an increasingly important role in the development and characterization of materials. However, efforts to calculate properties of materials of interest commonly expose the accuracy limitations of current DFT approximations. This situation has led to a renewed interest in beyond-DFT approximations. Since Ceperley and Alder's pioneering calculations on the homogeneous electron gas provided the benchmarks that underpin most current DFT approximations, diffusion Monte Carlo (DMC) has been recognized as one of the most accurate techniques for electronic structure calculations. Unfortunately, despite its modest computational complexity, the large expense of ab initio calculations rendered the method impractical for all but benchmark and model calculations. Within the last decade this situation has changed. The availability of capability class computers and well-designed algorithms has allowed DMC to become feasible for general application. Because the resources required are still quite large, it is essential to understand the size of the approximations in DMC calculations of real materials and how to minimize them. I will present a series of benchmarks showing the current state of the art in this promising method[1] as well as paths towards improving its accuracy.[2] I will also present several recent applications that take advantage of the unique strengths of DMC, including calculating the properties of two dimensional black phosphorus,[3] determining the interaction of large closed shell molecules,[4] and probing the magnetic state of strongly correlated transition metal oxides. I will close by comparing DMC to other modern beyond-DFT methods in the context of materials research.

[1]Shulenburger and Mattsson, PRB **88**, 245117 (2013)
[2]Shulenburger, Mattsson and Desjarlais, arXiv:1501.03850
[3]Shulenburger, Baczewski, Zhu, Guan and Tomanek, arXiv:1508.04788
[4]Benali, Shulenburger, Romero, Kim and von Lilienfeld, JCTC 10, 3417 (2014)

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