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Coupled-cluster theory for condensed-phase spectroscopy

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Coupled-cluster theory has recently gained interest as an accurate and systematically-improvable approach for ground-state properties of extended systems. Through the equation-of-motion formalism, coupled-cluster theory can be extended to describe excited states. In this talk, I will first present the use of equation-of-motion coupled-cluster theory (EOM-CC) for the calculation of the one-particle spectral function of the 3D uniform electron gas [1]; importantly, EOM-CC with single and double excitations is able to predict satellite (shake-up) peaks associated with coupling to plasmons and yields a quasiparticle bandwidth in good agreement with experimental photoemission data. I will next describe the development of a Gaussian-based periodic coupled-cluster code, available in the open-source PySCF software package [2]. I will present the application of this approach to the ground-state properties and quasiparticle band structure of simple, atomistic semiconductors such as diamond and silicon [3]. Finally, I will discuss approximate variants of EOM-CC as well as their formal relation to the Green's function based GW approximation.

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[2] Q. Sun, T. C. Berkelbach, N. S. Blunt, G. H. Booth, S. Guo, Z. Li, J. Liu, J. McClain, E. R. Sayfutyarova, S. Sharma, S. Wouters, and G. K.-L. Chan, arXiv:1701.08223
[3] J. McClain, Q. Sun, G. K.-L. Chan, and T. C. Berkelbach, J. Chem. Theory Comput. 13, 1209 (2017).