Stability, Energetics, and Magnetic States of Cobalt Adatoms on Graphene†

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Research in the adsorption of transition metal adatoms on graphene has grown rapidly because of their possible use in spintronic applications. We investigate the stability and electronic properties of a single Co atom on graphene [1] with near-exact auxiliary-field quantum Monte Carlo (AFQMC) calculations [2]. We performed exact calculations on several model systems to benchmark the accuracy of various density functional theory methods. A frozen-orbital embedding scheme was combined with AFQMC to increase the reach in system sizes. Several energy minima are found as a function of the distance h between Co and graphene. Energetics only permit the Co atom to occupy the top site at $h = 2.2$ Å in a high-spin $3d^8 4s^1$ state, and the van der Waals region at $h = 3.3$ Å in a high-spin $3d^7 4s^2$ state. The findings provide an explanation for recent experimental observations with Co on free-standing graphene [3].

† This research work is supported by DOE, NSF, and ONR. Computing is provided by DOE (INCITE), NCSA Blue Waters, and W&M HPC facility.

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