Ab initio many-body study of cobalt adatoms adsorbed on graphene

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Research interest in the adsorption of transition metal adatoms on graphene has grown rapidly because of their possible use in spintronic applications. Single Co atoms on graphene have been extensively studied recently [1–3], and possible Kondo effects have been considered [4]. Calculations of Co/graphene systems have largely been done at the density functional theory (DFT) level with local or semi-local functionals, or with an empirical Hubbard on-site repulsion U (DFT+U). These calculations show significantly varying results on the bonding nature of Co/graphene system. We use auxiliary-field quantum Monte Carlo (AFQMC) and a size-correction embedding scheme to accurately calculate the binding energy of Co/graphene [5]. We find that as a function of the distance h between the Co atom and the six-fold hollow site, there are two states that provide binding and exhibit a double-well feature with nearly equal binding energy of 0.4 eV at $h = 1.51$ and $h = 1.65$ Å, corresponding to low-spin ^{2}Co ($3d^9 4s^0$) and high-spin ^{4}Co ($3d^8 4s^1$), respectively. A recent experimental study [6] reported good agreement with DFT+U in predicting the adsorption site of Co/graphene. We systematically investigate the accuracy of DFT+U calculations for this system with near-exact AFQMC calculations.

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[1] T. O. Wehling, A. V. Balatsky, M. I. Katsnelson, A. I. Lichtenstein, and A. Rosch, Phys. Rev. B **81**, 115427 (2010).

[2] D. Jacob and G. Kotliar, Phys. Rev. B **82**, 085423 (2010).

[3] T. O. Wehling, A. I. Lichtenstein, and M. I. Katsnelson, Phys. Rev. B **84**, 235110 (2011).

[4] V. W. Brar *et al.*, Nat. Phys. **7**, 43 (2011).

[5] Y. Virgus, W. Purwanto, H. Krakauer, and S. Zhang, Phys. Rev. B **86**, 241406(R) (2012).

[6] T. Eelbo *et al.*, Phys. Rev. Lett. **110**, 136804 (2013)