

**Erratum: Towards an Exact Treatment of Exchange and Correlation in Materials:  
Application to the “CO Adsorption Puzzle” and Other Systems  
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The novel aspect in our Letter and Ref. [1], as well as in the work by Tuma and Sauer [2], is that for clusters the differences  $E^{XC_{\text{corr}}} = E(\text{XC-better}) - E(\text{XC-approximate-DFT})$  converge well with cluster size. The exchange-correlation (XC) XC-better treatment has been the Møller-Plesset perturbation theory (MP2) in our Letter and Ref. [2] or quantum Monte Carlo (QMC) calculations in Ref. [1]. The smooth convergence of  $E^{XC_{\text{corr}}}$  contrasts the known behavior of total energies calculated with the local-density approximation (LDA), generalized gradient approximation, hybrid-functionals, MP2, coupled-cluster, or QMC calculations. All these total energies change in some irregular manner with cluster size; i.e., their behavior is dominated by quantum size effects, actuated by the kinetic energy operator.

To the best of our knowledge, this short-range nature of the correction of the XC energy had not been discussed, analyzed, or emphasized before. Some earlier publications are, however, related to our findings: (1) Obviously, the employed concept of using three calculations and taking a sum and a difference is not new [3]. It is underlying more or less all embedding or perturbation-theory approaches, e.g., QM:MM and QM:QM. References [4–6] give some examples. (2) Additional support for our approach can be found in the results on jellium spheres by Almeida, Perdew, and Fiolhais [7]. These authors studied the dependence of the XC energy as a function of the sphere radius (i.e., cluster size) for various XC functionals. They address that empirical XC functionals, e.g., B(3)LYP (constructed to describe certain atoms and small molecules), exhibit their shortcomings when applied to metallic systems because they fail to describe the correct limit for the uniform electron density. We also note that Williamson, Hood, and Grossman [8] reported results for  $C_N$  fullerenes (with  $N = 20, 36, 60, 80, 180$ ) showing that the difference in the total energy per C atom between the LDA and QMC results is essentially constant for all clusters:  $0.80 \pm 0.03$  eV as estimated from their Fig. 3.

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