Improved STLS approach to the correlation energy of the spin polarized electron gas

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The exchange-correlation energy functional $E_{XC}[n]$ is the key quantity in the Kohn-Sham density functional theory, but it must be supplied from some external source. So far, by use of the Monte-Carlo (MC) data for the exchange-correlation energy of the homogeneous electron gas for a wide range of the density parameter r_s, LDA and its improved version, GGA, are proposed and found to provide a fairly good description of cohesive properties of molecules and solids. As for the dependence on spin polarization ζ, however, it must be noted that the fundamental MC data are obtained only at both unpolarized (ζ=0) and fully-polarized (ζ=1) situations; some interpolation scheme is employed to generate the correlation energy of the homogeneous electron gas at arbitrary r_s and ζ, $\varepsilon_c(r_s,\zeta)$, based on either the ζ-dependence in the exchange energy or that in $\varepsilon_c(r_s,\zeta)$ in RPA [1]. Since RPA is not reliable for r_s larger than unity, we need to calculate $\varepsilon_c(r_s,\zeta)$ at arbitrary ζ with using a better theoretical tool.

As an attempt in this direction, we have revisited the Singwi-Tosi-Land-Sjölander (STLS) scheme [2], in which $\varepsilon_c(r_s,\zeta)$ can be obtained in a self-contained manner by self-consistently determining the local-field factor $G(q)$ and the static structure factor $S(q)$. The scheme is known to provide a very accurate $\varepsilon_c(r_s,\zeta)$ at ζ=0, but not so accurate at ζ=1. We have explored the physical reason why the STLS scheme does not work well at ζ=1 and come to the observation that it is due to the violation of the Pauli principle, as manifestly shown by the negative parallel-spin radial distribution function at zero separation, $g_{\uparrow\uparrow}(0)<0$. With the recognition of the major problem of the original STLS in this way, we have invented a framework for adding a procedure to enforce the Pauli principle in the STLS scheme and succeeded in reproducing the MC data for $\varepsilon_c(r_s,\zeta)$ at ζ=1 as shown in Fig. 1. In the near future, our new improved version of the STLS scheme will be used to give more reliable data for $\varepsilon_c(r_s,\zeta)$ at arbitrary ζ, based on which we shall reconsider a suitable functional form for $E_{XC}[n]$.

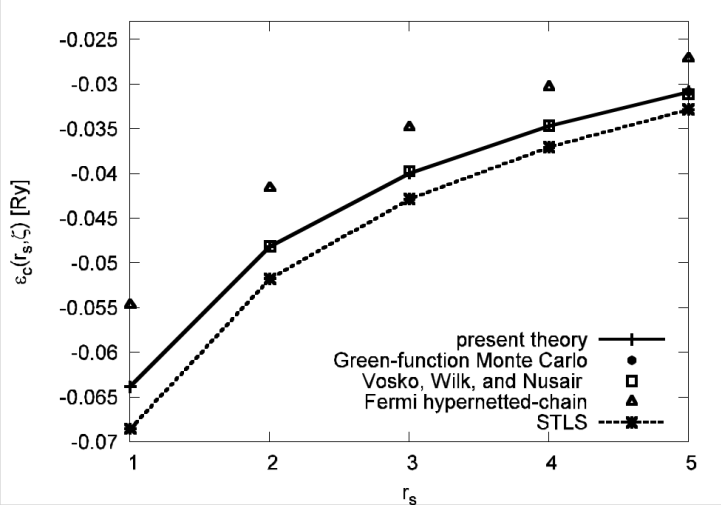

Fig. 1: correlation energy $\varepsilon_c(r_s,\zeta)$ in fully polarized systems (ζ=1) in various methods

[1] J. P. Perdew and Y. Wang, Phys. Rev. B **45**, 13244 (1992).
[2] K. S. Singwi, M. P. Tosi, R. H. Land, and A. Sjölander, Phys. Rev. **176** (1968).