Comment on "Exact Electron-Gas Response Functions at High Density"

Langreth and Vosko[1] have calculated correlation contributions to the Hohenberg-Kohn energy response function $K_{xc}(\mathbf{q},\mathbf{k})$. They claim that the results support the Langreth-Mehl[2] (LM) treatment of the interacting nonuniform electron gas. We previously pointed out[3] a number of serious deficiencies of LM including (1) restriction of E_{xc} to RPA with cancellation between RPA and gradient corrections from non-RPA (see the discussion of Ref. 2 beginning at the bottom of page 448 and continued to surface applications, in particular) and (2) serious disagreement between LM and jellium surface-energy calculations.[4] Contributions beyond RPA have been considered by Hu and Langreth[5] and are in agreement with Geldart and Rasolt.[6] It follows that nothing is left to account for the large discrepancy between Ref. 2 and Ref. 4 except incorrect summation of higher-order contributions by LM. In addition, (3) the "agreement" of LM for atoms[7] is based on a too restricted sample and (4) $K_{xc}(q)$ has logarithmic terms[3] ignored by LM. Langreth and Vosko[1] state that they provide "an answer to questions raised about this procedure" by us.[3] On the contrary, *not a single one of our objections has been answered.*

A fundamental reason why the LM procedure inevitably fails is that it attempts to force "universal" structure where none exists. The structure factor $S(\mathbf{r},\mathbf{r}')$ or $S(\mathbf{k},\mathbf{k})$ of extended inhomogeneous systems need not vanish as $\mathbf{k}\rightarrow\mathbf{0}$, does not have a universal $\mathbf{k}\rightarrow\mathbf{0}$ limit, and is extremely sensitive (see below) to any external inhomogeneity $V(\mathbf{q})$. Interpolation at small $\mathbf{k}$ is not valid.[2] Furthermore, $S(\mathbf{r},\mathbf{r}')$ develops a long-range power-law behavior in $|\mathbf{r}-\mathbf{r}'|^{-1}$ and is not localized in extended systems.[8] In no way does this violate particle conservation and the sum-rule arguments of LM are inappropriate. The physical basis for this long-range behavior is *imperfect screening* in nonuniform systems.[3,8] This imperfect screening also leads to long-range behavior in the electron-electron interaction[9] $V_{ee}(\mathbf{q}) \sim \sum \mathbf{k} K_{xc}(\mathbf{q},\mathbf{k})$ just as it does for the structure factor[8] $S(\mathbf{k},\mathbf{k}) \sim \sum \mathbf{q} |V(\mathbf{q})|^2 K_{xc}(\mathbf{q},\mathbf{k})$. Note that the small-$\mathbf{k}$ limit of $S(\mathbf{k},\mathbf{k})$ is particularly sensitive to the small-$\mathbf{q}$ ($q<k$) components of $K_{xc}(\mathbf{q},\mathbf{k})$ (Fig. 2 of Langreth and Vosko[1] shows this sensitivity) and therefore to the (nonuniversal) small-$\mathbf{q}$ structure of $V(\mathbf{q})$.

To describe E_{xc} in real systems, two limiting cases can be treated with confidence: (A) arbitrary density variation but slow modulation or (B) small density variation but arbitrary modulation. In principle, one can start from either A or B *provided* that one calculates consistently the necessary correction terms in the expansion parameter which is $\xi^{-1} \sim |\nabla k_F|/k_F, |\nabla^2 k_F|/|\nabla k_F|, \ldots$ in A and $V(\mathbf{q})$ for all $\mathbf{q}$ in B. *The two expansions are not the same.* The crucial importance of consistency is well documented in A for the kinetic energy with realistic density profiles[10] and must apply to E_{xc} also. Finally, the rapid *relative* variation of $K_{xc}(\mathbf{q})$ has little to say about the corresponding expansion in A since a consistent expansion in powers of ξ^{-1} is not provided by linear response alone. This is already clear from the exchange-only $K_x(\mathbf{q})$ which we find to vary rapidly, changing by a factor $\simeq 2.5$ between $q=0$ and $2k_F$ with strong structure near $2k_F$ and even a change of sign (yet giving the results of Rasolt, Wang, and Kahn[11]).[12] The entire range of q (not just q_{TF}) is important for physical systems. It is not true that the Thomas-Fermi screening length is the dominant signature of surface density profiles.

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