Klüner *et al.* Reply: The preceding Comment [1] is concerned with whether we, in our Letter [2], adequately justified our hybrid wave function/density-functional (DFT) embedding theory and the choice of kinetic energy density functionals (KEDFs) used to construct the embedding operator v_{emb}. The Comment has two problems: (i) a misunderstanding of our theory and (ii) an example of questionable relevance.

First, length restrictions did not permit a complete rederivation of the formalism; a thorough, formal justification is given *in detail* in [3,4]. The first criticism leveled is that since $T_s^{nad}[\rho_{\mathrm{I}}, \rho_{\mathrm{II}}]$ is a nonlinear functional of ρ_{I}, use of v_{emb} as an operator must be postulated. This is simply not so. Our embedding potential, derived variationally within (formally exact) DFT as the operator that influences the density in the region of interest due to the presence of the surroundings, is the same as the one written by Wesołowski in his Eq. (1). Our v_{emb} is used *only* as an additional one-electron potential in the Fock operator to obtain the wave function in the embedded region, but *never* to obtain the total energy E_{tot}; i.e., our E_{tot} does *not* contain $\langle\phi_i|\,(\delta T_S^{nad}[\rho_{\mathrm{I}}, \rho_{\mathrm{II}}]/\delta\rho_{\mathrm{I}})\,|\phi_i\rangle$, and hence we do not need to postulate its use and its nonlinearity is not an issue. To clarify, our E_{tot} is calculated as given in Eq. (16) of [4], where the subsystem interaction terms in E_{tot} appear only within the entire system's DFT total energy, $E_{\mathrm{tot}}^{\mathrm{DFT}}[\rho_{\mathrm{tot}}]$. The embedded region total energies in Eq. (16), $E_{\mathrm{I}}^{ab,\mathrm{DFT}}$, involve only terms within that region; i.e., v_{emb} does not appear explicitly (to avoid double counting interactions). v_{emb} does, however, have an implicit effect on $E_{\mathrm{I}}^{ab,\mathrm{DFT}}$, as the wave functions in region I are affected by v_{emb}.

Wesołowski's second concern is the choice of KEDF and its corresponding potential. As stated in [2], details on the construction of the KEDF potential (KEFP)—again we do not use the KEDF itself, only its potential—are published elsewhere [5]. In the Letter we provided a number of tests for the validity and justification of the choice of KEFP: (i) a DFT in DFT embedding, in which the electron density was virtually identical to that of a periodic DFT calculation (verifying the accuracy of the KEFP and the embedding scheme; see Fig. 1) and (ii) the sensitivity of adsorption energies to the choice of KEFP was reported in Table II, Ref. [2]. These latter tests show the importance of including gradient corrections in both the exchange-correlation and KEFP to achieve high accuracy.

As evidence disputing the validity of our KEDF choice, Wesołowski offers references and the Comment's figure, all of which assess the ability of various KEDFs to describe weak intermolecular forces (e.g., dispersion). Surely it is clear that KEDFs appropriate for van der Waals interactions are not necessarily the ones appropriate for metals (the territory of our "claim"); it is well known that in the case of exchange-correlation functionals, those that work well for metals fall short in their description of weakly interacting molecules (the purview of Wesołowski's cases) [6]. We proved in our Letter that our choice of KEDF works properly for metals and whether it works for van der Waals or hydrogen-bonded systems was not claimed by us. (N.B., we did not apply the conventional gradient expansion up to second order as implied by Wesołowski; rather, we introduced a local truncation criterion based on the ratio of the Thomas-Fermi and von Weizsäcker potentials [5]; he does not provide in his papers or this figure results for such a functional.) Last, note that the Comment's figure showing the inaccuracy of the conventional gradient expansion for a molecular dimer is not relevant to our method, since, as explained above, we do not use the KEDF to calculate total energies as he did to generate the Comment's figure; we use only the kinetic energy *potential* in the embedding potential as an additive term to the Fock matrix. This is a second order effect and thus so are the approximations introduced. Therefore, the accuracy of the KEDFs used in our theory is not at all as crucial as in his orbital-free DFT method, where he must use it to obtain a total energy.

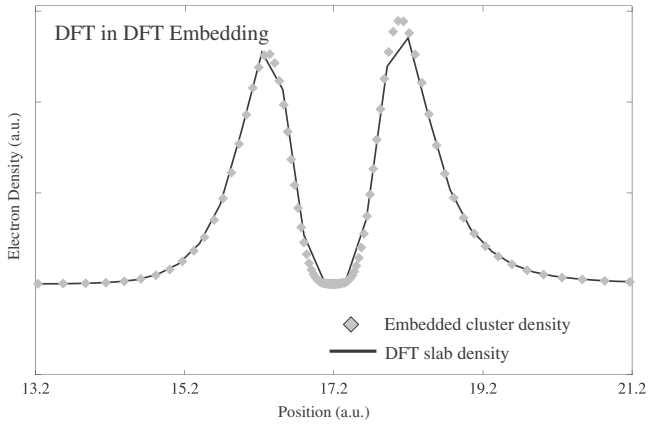

FIG. 1. DFT [local density approximation (LDA)] in DFT (LDA) embedding calculation for a Pd_3 cluster embedded in a three-layer Pd slab.

This work is supported by the NSF, the ARO, the AFOSR, and the DFG.

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Received 15 August 2001; published 3 May 2002
DOI: 10.1103/PhysRevLett.88.209702
PACS numbers: 71.15.–m, 73.20.Hb

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[6] See, for example, S. Kristyan and P. Pulay, Chem. Phys. Lett. **229**, 175 (1994); E. Ruiz *et al.,* J. Phys. Chem. **100**, 12 265 (1996).

