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Large two-dimensional electronic systems: Self-consistent energies and densities at low cost

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We derive a self-consistent local variant of the Thomas-Fermi approximation for (quasi-) two-dimensional (2D) systems by localizing the Hartree term. The scheme results in an explicit orbital-free representation of the electron density and energy in terms of the external potential, the number of electrons, and the chemical potential determined upon normalization. We test the method over a variety 2D nanostructures by comparing to the Kohn-Sham 2D local-density approximation (LDA) calculations up to 600 electrons. Accurate results are obtained in view of the negligible computational cost. We also assess a local upper bound for the Hartree energy.

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I. INTRODUCTION

Orbital-free density-functional theory (OF-DFT) is a computationally appealing method to deal with large systems beyond the reach of conventional DFT. At present, OF-DFT methods can handle systems up to a million atoms.[1] These methods await to be fully explored in the context of low-dimensional systems and nanoelectronic devices. In two-dimensional (2D) physics one of the main challenges of DFT is to deal with regions of the 2D electron gas[2] comprising hundreds or thousands of interacting electrons, e.g., in the quantum Hall regime.[3]

As the name suggests, OF-DFT is free from the use of the Kohn-Sham orbitals needed in the calculation of the Kohn-Sham kinetic energy, and thus the only explicitly needed variable is the electron density $\rho(\mathbf{r})$. The earliest OF-DFT method dates back to the Thomas-Fermi (TF) theory employing the exact result of the homogeneous electron gas for the kinetic energy, and the Hartree approximation for the *e-e* interaction. In fact, most orbital-free schemes can be regarded as modifications or improvements to the TF method.[4]

The crux of the problem in OF-DFT is to find an approximation for the (noninteracting) kinetic-energy functional that may be generally applicable. For this ambitious goal, a promising approach is an orbital-free formulation exploiting the potential rather than the the density as the basic variable.[5] In 2D the TF approximation for the kinetic energy works remarkably well—in fact, the gradient corrections to it vanish to *all* orders.[6–10] Moreover, for the 2D Fermi gas in harmonic trap the TF kinetic energy yields the exact noninteracting kinetic energy when the exact density is used as the input.[6] In this work we replace the Hartree term with a much simpler *local* expression that significantly speeds up the calculations.

Recently, a 2D orbital-free expression for the energy was shown to lead to a major improvement over the TF results when applied to quantum dots and slabs up to 200 interacting electrons.[11] The energies were not obtained self-consistently, but by using the electron densities from the 2D local-density approximation (2D-LDA). The expression of the functional is

$$E_{\rm tot}[\rho(\mathbf{r})] = T_{\rm TF}[\rho(\mathbf{r})] + W[\rho(\mathbf{r})] + \int d\mathbf{r}\, \rho(\mathbf{r})\, v_{\rm ext}(\mathbf{r}), \quad (1)$$

where

$$T_{\rm TF}[\rho(\mathbf{r})] = \frac{\pi}{2}\int d\mathbf{r}\, \rho^2(\mathbf{r}) \quad (2)$$

is the TF kinetic energy in 2D and the last term is the energy contribution due to the external scalar (confining) potential $v_{\rm ext}(\mathbf{r})$. The total electron-electron interaction energy is given by

$$W[\rho(\mathbf{r})] = \frac{\pi}{2}\sqrt{\frac{N-1}{2}}\int d\mathbf{r}\, \rho^{3/2}(\mathbf{r}), \quad (3)$$

which was obtained from a crude approximation by using a Gaussian ansatz for the *cylindrical* average of the pair density, and enforcing an overall linear behavior under uniform coordinate scaling. This was also partially inspired by the fact that an analogous Gaussian ansatz for the one-body reduced-density matrix eventually leads to highly accurate exchange energies.[12]

In Eqs. (1) and (3) it is apparent that for $N = 1$ the functional reduces to the *noninteracting* TF approximation. For $N \neq 1$ the interaction contribution is similar to the form of the exchange energy in the 2D-LDA, but with a different prefactor $\sqrt{N-1}$. This approximation completely eliminates the computational burden of the Hartree term in the TF approximation. We point out that Eq. (1) has been employed by others to compute the total energy of a realistic semiconductor quantum dot formed in gate- and etching-defined devices.[13] The results confirm the good balance between accuracy and efficiency of the functional.

In this work we verify that the same approach works well in a fully self-consistent framework. In addition, an important modification improves its performance. As remarked in Ref. 11, the derivation of Eq. (3) employs, among others, a Hartree-Fock expression for the pair density that leads to a

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particular choice for the overall coefficient. Here we propose a different coefficient that has a nonempirical justification as explained below. Our proposal also leads to an estimate for a *local* upper bound of the Hartree energy in 2D systems. Finally, we test the derived approximation self-consistently for an extensive set of 2D systems. Remarkably accurate results for the total energy and density are obtained in view of the simplicity of the scheme and the negligible computational cost.

II. THEORY

A. Upper bound for the Hartree term

Lieb and co-workers have proved that the TF theory is asymptotically exact for large quantum dots.[14] In addition, Burke and co-workers have pointed to the fact that successful DFT approximations are those that become *asymptotically* exact for the class of systems under investigation.[15] It is natural to follow the same principle in 2D, where semiconductor quantum dots play the role of "artificial atoms." The fundamental question is whether the form in Eq. (1) can be made, in some sense, asymptotically correct. We provide an affirmative answer by exploiting the existence of a *local* upper bound for the Hartree energy:

$$\frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\leqslant\frac{1}{2}C\sqrt{N}\int d\mathbf{r}\rho^{3/2}(\mathbf{r}), \qquad (4)$$

where C is a constant to be estimated.[16] The latter expression suggests that, for large N, we may be able to energetically approach an exact (TF) theory "from above." Of course sole energy bounds do not allow us to directly control the behavior of the functional derivatives of the obtained approximations. This is expected to affect the accuracy of self-consistent densities. Moreover, the considered bound does not allow size consistency, which may have severe effects on the chemical potentials as well as on "multicenter" systems with separated confining potentials.

In order to find an "optimal" constant C in Eq. (4), we consider a harmonically confined quantum dot, where the confinement potential is kept fixed while adding more electrons. Eventually the density becomes relatively flat, resembling a disk with radius R. In the large-N regime the Hartree energy dominates over the exchange and correlation energy. For example, for $N = 6$, 30, and 600 we have $|E_{\rm xc}|/E_H \approx 0.33$, 0.15, and 0.03, respectively, with the oscillator strength $\omega = 0.5$. For a completely flat 2D density, the Hartree energy is given exactly by[17]

$$E_H^{\rm disk}=\frac{8}{3\pi}\frac{N^2}{R}. \qquad (5)$$

Combining Eqs. (4) and (5) we see that asymptotically Eq. (4) tends to an *equality* with $C^{\rm disk}=\frac{16}{3\sqrt{\pi}}$. Previously, taking as reference a Gaussian-like density,[16] it was suggested that $C=\frac{3\pi}{\sqrt{2}}$, i.e., a value significantly greater than $C^{\rm disk}$. Therefore, we could expect that our result may work as a good estimation for the upper bound of E_H.

Figure 1 shows the Hartree energy E_H from a self-consistent 2D-LDA calculation with respect to our upper bound estimate $E_{\rm bound}$. Three different quantum-dot systems are considered: (i) Harmonic dots defined by $v_{\rm ext}(r)=\omega^2 r^2/2$ with $\omega = 0.5$, (ii) circular dots defined by a hard-wall potential [$v_{\rm ext}(r) = 0$ at $r \leqslant R$ with $R = 10$, $v_{\rm ext}(r)\to\infty$ at $r > R$], and (iii) rectangular hard-wall dots[18,19] with side lengths L and $2L$, where $L = 10$. We find that in all the cases, and with different N, the Hartree energy is very close to our suggested bound. However, it is noteworthy that our estimate does not serve as a tighter upper bound (rather as an approximation for it), as for for harmonic dots we obtain values above E_H.

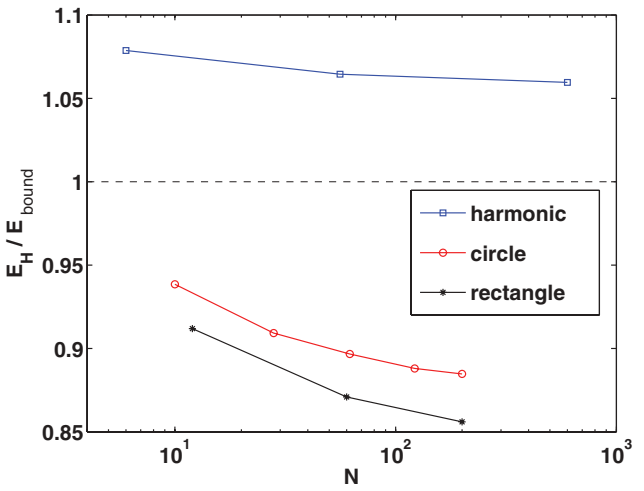

FIG. 1. (Color online) Hartree energy E_H from a self-consistent 2D-LDA calculation with respect to our estimate for the upper bound $E_{\rm bound}$, i.e., the right-hand side of Eq. (4) with $C=\frac{16}{3\sqrt{\pi}}$. Results as a function of N are shown for three different 2D systems.

B. Self-consistent scheme

First, for large N we have $\sqrt{N-1}\approx\sqrt{N}$, and therefore—and according to the above analysis—Eq. (1) can be modified as follows:

$$E_{\rm tot}^{\alpha}[\rho(\mathbf{r})]=T_{\rm TF}[\rho(\mathbf{r})]+\alpha W[\rho(\mathbf{r})]+\int d\mathbf{r}\,\rho(\mathbf{r})\,v_{\rm ext}(\mathbf{r}), \qquad (6)$$

where $\alpha=\frac{8}{3}(\frac{2}{\pi})^{3/2}\approx 1.35453$. It is apparent that the modified form fails to to recover the size consistency of the TF approximation. An immediate consequence (that can be verified analytically for, e.g., rectangular quantum dots) is that the fundamental relation $\mu = dE_{\rm tot}/dN$ is not fulfilled.

In order to find the ground-state density we have to minimize Eq. (6) for a fixed number of particles. We may first express the total energy in a single integral as

$$\begin{aligned}E_{\rm tot}^{\alpha}[\rho(\mathbf{r})]&=\int d\mathbf{r}\,F[\rho(\mathbf{r})]\\&=\int d\mathbf{r}\left[\frac{\pi}{2}\rho^2(\mathbf{r})+\frac{\pi\alpha}{2}\sqrt{\frac{N-1}{2}}\,\rho^{3/2}(\mathbf{r})\right.\\&\quad\left.+\rho(\mathbf{r})\,v_{\rm ext}(\mathbf{r})\right].\end{aligned} \qquad (7)$$

We have to find a stationary value for the functional $F[\rho(\mathbf{r})]$ with respect to variations in $\rho(\mathbf{r})$. To take the electron number conservation into account, we introduce another functional

$G[\rho(\mathbf{r})] = \rho(\mathbf{r})$ so that

$$\int d\mathbf{r}\, G[\rho(\mathbf{r})] = N. \tag{8}$$

This constraint introduces a Lagrange multiplier μ in the variational equation, which can be written as

$$\frac{dF}{d\rho} - \mu \frac{dG}{d\rho} = 0. \tag{9}$$

Substituting F and G to this equation yields

$$\pi\rho(\mathbf{r}) + \frac{3\pi\alpha}{4}\sqrt{\frac{N-1}{2}}\rho^{1/2}(\mathbf{r}) + v_{\text{ext}}(\mathbf{r}) - \mu = 0. \tag{10}$$

As this expression is quadratic in $\rho^{1/2}$, we find an explicit expression for the density,

$$\rho(\mathbf{r}) = \left(-\frac{3\alpha}{8}\sqrt{\frac{N-1}{2}} + \frac{1}{2}\sqrt{\left\{\frac{9\alpha^2}{32}(N-1) - \frac{4}{\pi}[v_{\text{ext}}(\mathbf{r}) - \mu]\right\}_+}\right)^2. \tag{11}$$

This shows that the density can be solved instantaneously for any external potential v_{ext} and any N. The only variable to be determined numerically is μ that follows from the normalization condition in Eq. (8). The symbol $[\cdots]_+$ in Eq. (11) represents an additional constraint that no sign changes under the square in Eq. (11) (leading to unphysical "nodal lines" in the density), nor negative values under the square root (leading to complex densities) are allowed. Once $\rho(\mathbf{r})$ is determined from Eq. (11), the total energy is finally obtained from Eq. (6).

Let us emphasize the difference between the present and and the TF approximation in a practical sense. In the latter the variational procedure applied to the total energy leads to an *integral* equation for the density. The TF scheme then transforms into a differential equation (which in 3D leads to the Poisson equation). Instead, our functional is free from this complexity due to the simple expression for the interaction energy [Eq. (3)] in comparison with the Hartree integral utilized by the TF method. Although the Hartree term is simple to calculate in most applications, it may become a bottleneck in large 2D systems. In any case, it is appealing to have a method for the first approximation of the electronic density and energy with a negligible computational cost.

III. APPLICATIONS AND ANALYSIS

Next we test the self-consistent scheme of the previous section for a set of 2D quantum dots and rings including the *e-e* interactions. We use DFT with the 2D-LDA[20] as our reference method; in the range of systems and parameters considered here the LDA has been shown to provide—for the present purpose—sufficiently accurate total energies and densities (see, e.g., Refs. 21 and 22 for quantum dots and rings, respectively). The LDA calculations are performed using the OCTOPUS code[23] as well as another code exploiting circular symmetry.[24]

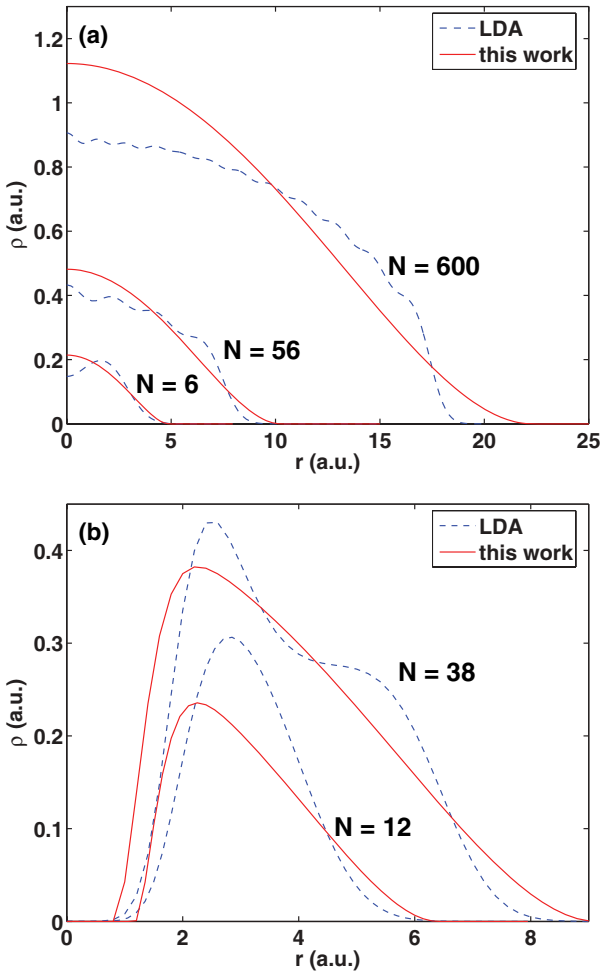

FIG. 2. (Color online) (a) Electron densities in harmonic quantum dots containing $N = 6$, 56, and 600 electrons, respectively. The dashed lines denote the DFT results within the 2D-LDA. The solid lines correspond to the results of the present orbital-free functional. (b) The same as in (a) but for two quantum rings containing $N = 12$ and 38 electrons, respectively.

Figure 2(a) shows the radial densities obtained from Eq. (11) for 2D harmonic quantum dots with $\omega = 0.5$ and $N = 6$, 56, and 600 (solid lines). The dashed lines show the corresponding LDA results. As expected, the present functional cannot describe the shell structure due to the lack of orbitals. However, the overall shape of the density profile is satisfactory in a qualitative fashion, and the correct radial extent of the density profile is obtained in all cases.

In Fig. 2(b) we show the corresponding result for a quantum ring modeled by $v_{\text{ext}}(r) = \omega^2 r^2/2 + V_0 \exp(-r^2/d^2)$, with $\omega = 0.5$, $V_0 = 20$, and $d = 1$. The model potential is the same as the one used in Refs. 22, 25, and 26, the last reference showing direct comparison with an experiment. We find a reasonable qualitative agreement between the present functional and the LDA. The qualitative agreement is similar for both $N = 12$ and $N = 38$.

Apart from densities, it is important to assess the performance of the present functional for total energies. Figure 3

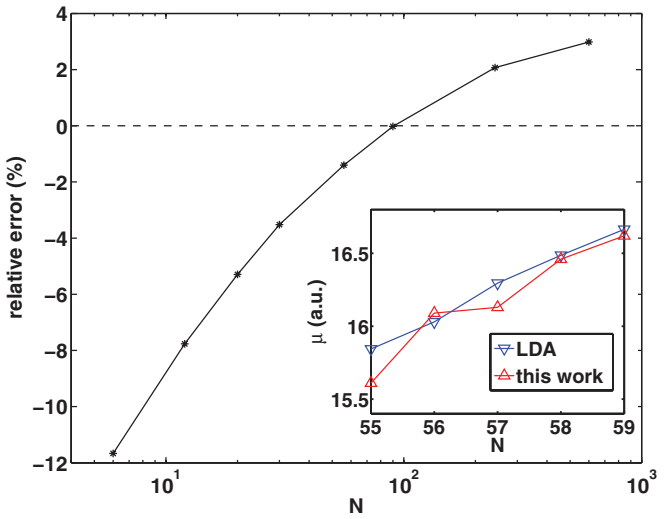

FIG. 3. (Color online) Relative error in the total energies of harmonic quantum dots calculated with our self-consistent scheme with respect to 2D-LDA results. The inset shows the obtained chemical potentials $\mu(N) = E_{\rm tot}(N) - E_{\rm tot}(N-1)$ for $N = 55$–59 in comparison with the 2D-LDA.

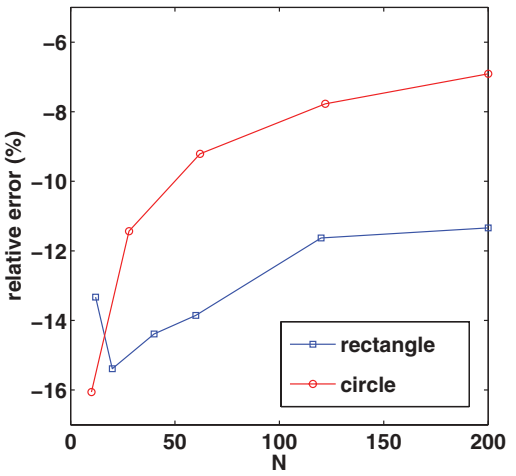

FIG. 4. (Color online) Relative error in the total energies for circular and rectangular hard-wall quantum dots calculated with our self-consistent scheme with respect to 2D-LDA results.

shows the relative total-energy differences from the reference 2D-LDA results for a set of harmonic quantum dots up to $N = 600$. Overall, the accuracy is remarkably good in view of the negligible computational cost. Even for small N the accuracy is well handled, e.g., with $N = 12$ the relative error is below 8%. However, the main interest for practical applications is in the large-N regime. For $N = 600$ our approximation overestimates the total energy only by ∼3%. According to Fig. 3 the error then increases with N, but most likely saturates. It can be extrapolated that for $N \sim 10\,000$ the error of our approximation is still under 5% (note the log-scale in the x axis).

The inset of Fig. 3 shows the obtained (spin-compensated) chemical potentials $\mu(N) = E_{\rm tot}(N) - E_{\rm tot}(N-1)$ for $N =$ 55–59 in harmonic quantum dots. In view of the lack of size consistency (see Sec. II B), our scheme yields surprisingly accurate results in comparison with the 2D-LDA. We point out, however, that due to the lack of orbital dependency (and thus the shell structure) we can only obtain the qualitative behavior of μ without any detailed features.

In Fig. 4 we compare our results for hard-wall circular and rectangular quantum dots with 2D-LDA results. The parameter values are the same as in Sec. II A. Overall, the errors are slightly larger than for harmonic quantum dots. On the other hand, the errors become smaller with N so that we can expect reliable results at least within $N \sim 10^3$–10^4. We point out that real-space 2D-LDA calculations are numerically tedious in those systems. Detailed assessment of our scheme ought to be performed with respect to TF results.

Finally, we discuss the relevance of α in terms of the performance of our scheme. As described in Sec. II A, the inclusion of α results from the limit of a circular and flat 2D density whose Hartree energy is known. Eventually α appears in Eq. (6) as a prefactor of W. Instead, in a previous non-self-consistent formulation[11] α was equal to one on the basis of the Hartree-Fock expression for the pair density. For completeness we have tested our self-consistent scheme also with $\alpha = 1$ and found drastically worse results. For example, for a harmonic quantum dot with $N = 600$ and for a circular hard-wall dot with $N = 200$ the formulation with $\alpha = 1$ yields, in both cases, a 20% overestimation of the total energy. In contrast, and as shown above, the present approach [with $\alpha = \frac{8}{3}(\frac{2}{\pi})^{3/2} \approx 1.35453$] yields respective errors of 3.0% and 6.9%. Therefore, the inclusion of α can be also practically validated.

IV. CONCLUSIONS

We have derived a self-consistent scheme to compute approximate electron densities and total energies for confined (quasi-) two-dimensional (2D) systems. Our scheme can be applied to any number of electrons with a negligible computational cost. In view of its extreme simplicity, we have obtained appealing results for electron densities and total energies in a variety of systems (such as harmonic and hard-wall quantum dots and quantum rings). Preceding the derivation of our self-consistent scheme, we have found a good approximation for an upper bound of the Hartree energy in 2D. The present scheme may be useful in negligible-cost computational investigations of 2D systems such as quantum Hall devices.

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[1] L. Hung and E. A. Carter, Chem. Phys. Lett. **475**, 163 (2009).

[2] G. F. Giuliani and G. Vignale, *Quantum Theory of the Electron Liquid* (Cambridge University Press, New York, 2005).

[3] K. Lier and R. R. Gerhardts, Phys. Rev. B **50**, 7757 (1994); A. Siddiki and R. R. Gerhardts, *ibid.* **68**, 125315 (2003).

[4] Y. A. Wang and E. A. Carter, *Theoretical Methods in Condensed Phase Chemistry*, in series *Progress in Theoretical Chemistry and Physics*, edited by S. D. Schartz (Kluwer, Dordrecht, 2000), pp. 117; V. L. Ligneres and E. A. Carter, *An Introduction to Orbital-free Density Functional Theory* (Springer, Netherlands, 2005).

[5] P. Elliott, D. Lee, A. Cangi, and K. Burke, Phys. Rev. Lett. **100**, 256406 (2008); A. Cangi, D. Lee, P. Elliott, and K. Burke, and E. K. U. Gross, *ibid.* **106**, 236404 (2011).

[6] M. Brack and B. P. van Zyl, Phys. Rev. Lett. **86**, 1574 (2001).

[7] A. Holas, P. M. Kozlowski, and N. H. March, J. Phys. A **24**, 4249 (1991).

[8] L. Salasnich, J. Phys. A: Math. Theor. **40**, 9987 (2007).

[9] A. Putaja, E. Räsänen, R. van Leeuwen, J. G. Vilhena, and M. A. L. Marques, Phys. Rev. B **85**, 165101 (2012).

[10] The situation differs for Fermions with spatially varying effective mass, see K. Berkane and K. Bencheikh, Phys. Rev. A **72**, 022508 (2005).

[11] S. Pittalis and E. Räsänen, Phys. Rev. B **80**, 165112 (2009).

[12] S. Pittalis, E. Räsänen, and E. K. U. Gross, Phys. Rev. A **80**, 032515 (2009).

[13] G. Bilgeç Akyğz, K. Akgüngö, S. Şakiroğlu, A. Siddiki, and İ. Sökmen, Physica E **43**, 1514 (2011).

[14] E. H. Lieb, J. P. Solovej, and J. Yngvason, Phys. Rev. B **51**, 10646 (1995).

[15] J. P. Perdew, A. Ruzsinszky, G. I. Csonka, O. A. Vydrov, G. E. Scuseria, L. A. Constantin, X. Zhou, and K. Burke, Phys. Rev. Lett. **100**, 136406 (2008); P. Elliott and K. Burke, Can. J. Chem. **87**, 1485 (2009); A. Cangi, D. Lee, P. Elliott, and K. Burke, Phys. Rev. B. **81**, 235128 (2010).

[16] R. Pino, A. J. Markvoort, and P. A. J. Hilbers, Physica B **325**, 149 (2003).

[17] E. Räsänen, M. Seidl, and P. Gori-Giorgi, Phys. Rev. B **83**, 195111 (2011).

[18] E. Räsänen, H. Saarikoski, V. N. Stavrou, A. Harju, M. J. Puska, and R. M. Nieminen, Phys. Rev. B **67**, 235307 (2003).

[19] E. Räsänen, A. Harju, M. J. Puska, and R. M. Nieminen, Phys. Rev. B **69**, 165309 (2004).

[20] C. Attaccalite, S. Moroni, P. Gori-Giorgi, and G. B. Bachelet, Phys. Rev. Lett. **88**, 256601 (2002).

[21] H. Saarikoski, E. Räsänen, S. Siljamäki, A. Harju, M. J. Puska, and R. M. Nieminen, Phys. Rev. B **67**, 205327 (2003).

[22] E. Räsänen and M. Aichinger, J. Phys.: Condens. Matter **21**, 025301 (2009).

[23] A. Castro, H. Appel, M. Oliveira, C. A. Rozzi, X. Andrade, F. Lorenzen, M. A. L. Marques, E. K. U. Gross, and A. Rubio, Phys. Status Solidi B **243**, 2465 (2006).

[24] I. Makkonen, M. M. Ervasti, V. J. Kauppila, and A. Harju, Phys. Rev. B **85**, 205140 (2012).

[25] M. Aichinger, S. A. Chin, E. Krotscheck, and E. Räsänen, Phys. Rev. B **73**, 195310 (2006).

[26] E. Räsänen, A. Mühle, M. Aichinger, and R. J. Haug, Phys. Rev. B **84**, 165320 (2011).