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# Study of the Discontinuity of the Exchange-Correlation Potential in an Exactly Soluble Case

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**ABSTRACT:** It was found by Perdew et al. (Phys Rev Lett, 1982, 49, 1691) and by Sham and Schlüter (Phys Rev Lett. 1983, 51, 1884) that the exact Kohn–Sham exchange–correlation potential of an open system may jump discontinuously as the particle number crosses an integer, with important physical consequences. Recently, Sagvolden and Perdew (Phys Rev A 2008, 77, 012517) have analyzed the discontinuity of the exchange–correlation potential as the particle number crosses one, with an illustration that uses a model density for the  $H^-$  ion. In this work, we extend their analysis to the case in which the external potential is the simple harmonic confinement, choosing spring-constant values for which the two-electron hamiltonian has an analytic solution. This way, we can obtain the exact, analytic, exchange and correlation potentials for particle number fluctuating between zero and two, illustrating the discontinuity as the particle number crosses one without introducing any model or approximation. We also discuss exchange and correlation separately. © 2009 Wiley Periodicals, Inc. Int J Quantum Chem 109: 2410–2415, 2009

**Key words:** density functional theory; electronic correlation; exchange–correlation potentials; harmonic traps

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## 1. Introduction

**K**ohn–Sham (KS) density functional theory (DFT) (see, e.g., [1]) is a successful method for electronic structure calculations, thanks to its unique combination of low computational cost and reasonable accuracy. In the Kohn–Sham formalism, the total energy of a many-electron system in the external potential  $\hat{V}_{\text{ext}} = \sum_i v_{\text{ext}}(\mathbf{r}_i)$  is rewritten as a functional of the one-electron density  $\rho(\mathbf{r})$ ,

$$E[\rho] = T_s[\rho] + \int d\mathbf{r} v_{\text{ext}}(\mathbf{r})\rho(\mathbf{r}) + U[\rho] + E_{\text{xc}}[\rho]. \quad (1)$$

In Eq. (1),  $T_s[\rho]$  is the kinetic energy of a noninteracting system of fermions (usually called KS system) having the same one-electron density  $\rho$  of the physical, interacting system. The Hartree energy  $U[\rho]$  is the classical repulsion energy,  $U[\rho] = 1/2 \int d\mathbf{r} \int d\mathbf{r}' \rho(\mathbf{r})\rho(\mathbf{r}')/|\mathbf{r} - \mathbf{r}'|$ , and the exchange-correlation functional  $E_{\text{xc}}[\rho]$  must be approximated. Minimization of Eq. (1) with respect to the orbitals forming the KS determinant lead to the KS equations. Thus, instead of the physical problem, in KS DFT we solve the hamiltonian of a model system of noninteracting fermions in the one-body local potential  $\hat{V}_{\text{KS}} = \sum_i v_{\text{KS}}(\mathbf{r}_i)$ , with

$$v_{\text{KS}}(\mathbf{r}) = v_{\text{ext}}(\mathbf{r}) + v_{\text{H}}(\mathbf{r}) + v_{\text{xc}}(\mathbf{r}) \quad (2)$$

$$v_{\text{H}}(\mathbf{r}) = \int d\mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \quad (3)$$

$$v_{\text{xc}}(\mathbf{r}) = \frac{\delta E_{\text{xc}}[\rho]}{\delta \rho(\mathbf{r})}, \quad (4)$$

and we recover the energy of the physical, interacting system, through the sum of the two functionals  $U[\rho] + E_{\text{xc}}[\rho]$ .

In Refs. [2–4] an analysis of KS theory for systems with fluctuating particle number lead to the conclusion that the exact exchange-correlation potential  $v_{\text{xc}}(\mathbf{r})$  may jump discontinuously by a spatial-independent constant as the particle number crosses an integer, with important physical consequences. In the last years there has been new interest in the derivative discontinuity of  $E_{\text{xc}}[\rho]$  (see, e.g., [5–11]), and its existence has been questioned in Ref. [12]. In a recent paper, Sagvolden and Perdew [13] have given further support to the assumptions used

to find the exchange-correlation potential discontinuity, and they have illustrated the discontinuity when the particle number crosses one, using a model density for the  $\text{H}^-$  ion. They have also rigorously proved that the von Weizsäcker functional,

$$T_{\text{vw}}[\rho] = \frac{1}{2} \int d\mathbf{r} |\nabla \sqrt{\rho(\mathbf{r})}|^2 \quad (5)$$

is the correct  $T_s[\rho]$  for a system with particle number  $N \leq 2$ .

In this work we repeat a similar analysis for electronic systems with particle number fluctuating between zero and two when the external potential is harmonic,  $v_{\text{ext}}(\mathbf{r}) = 1/2 \omega^2 r^2$ . Taut [14] has shown that the corresponding hamiltonian for  $N = 2$  electrons is analytically soluble for some special values of  $\omega$ , which means that, in such special cases, we can calculate analytically the exact interacting density and the exact exchange-correlation potential [15]. This way, we can illustrate the derivative discontinuity without relying on any approximation. The paper is organized as follows. In Section 2, we report the equations used to extract the exact exchange and correlation potentials from Taut's [14] analytical solutions. The corresponding results are shown and analyzed in Section 3, and the last Section IV is devoted to concluding remarks.

## 2. Theory

We consider a system with particle number fluctuating between 0 and 2, in the harmonic external potential  $v_{\text{ext}}(\mathbf{r}) = 1/2 \omega^2 r^2$ . This system can be thought as arising from identical quantum traps whose centers are separated by a very large distance [16]. If the energy of the physical system is convex as a function of the integer particle number  $M$ , i.e., if  $E(M) \leq (E(M+1) + E(M-1))/2$ , then the system with noninteger particle number  $N$  is an ensemble of only the two systems with integer-particle number  $M$  and  $M+1$  such that  $M < N < M+1$  [2]. In the case treated here,  $v_{\text{ext}}(\mathbf{r}) = 1/2 \omega^2 r^2$ , the energy of the noninteracting system is convex, and the electron-electron interaction seems to make it strictly convex (see, e.g., the results for two-dimensional harmonic traps of Refs. [17] or the three-dimensional case treated in Ref. [18]). The density of the system with particle number  $0 \leq N \leq 2$ ,  $\rho_N(\mathbf{r}) = \rho_N(r)$ , is then equal to [2, 13]

$$\rho_N(\mathbf{r}) = \begin{cases} N\rho_1(\mathbf{r}) & 0 \leq N \leq 1 \\ (2-N)\rho_1(\mathbf{r}) + (N-1)\rho_2(\mathbf{r}) & 1 < N \leq 2 \end{cases} \quad (6)$$

The exact Kohn–Sham potential is, up to a constant, given by the functional derivative with respect to  $\rho_N(\mathbf{r})$  of the von Weizsäcker functional of Eq. (5),

$$v_{\text{KS}}(\mathbf{r}) = \frac{\nabla^2 \sqrt{\rho_N(\mathbf{r})}}{2\sqrt{\rho_N(\mathbf{r})}} + \text{const.} \quad (7)$$

The Hartree potential  $v_{\text{H}}(r)$  can be easily calculated by plugging Eq. (6) into Eq. (3). The exchange and correlation potential  $v_{\text{xc}}(r)$  is then calculated from Eq. (4),  $v_{\text{xc}}(r) = v_{\text{KS}}(r) - v_{\text{H}}(r) - v_{\text{ext}}(r)$ . Setting  $v_{\text{xc}}(r \rightarrow \infty) = 0$  for any fixed  $N$  determines the arbitrary constant in the exchange–correlation potential.

Splitting the potential  $v_{\text{xc}}(r)$  into exchange and correlation is subtle. When  $0 \leq N \leq 1$  we know that  $v_{\text{x}}(r)$  must exactly cancel the Hartree potential,  $v_{\text{x}}(r) = -v_{\text{H}}(r)$ , and that, since the correlation energy is zero,  $v_{\text{c}}(r) = 0$ . For  $1 \leq N \leq 2$  the sum  $U[\rho_N] + E_{\text{x}}[\rho_N]$  should be equal to the expectation of the electron–electron repulsion  $\hat{V}_{\text{ee}}$  over the noninteracting ensemble density matrix that yields  $\rho_N(r)$ . The noninteracting density matrix that yields  $\rho_N(r)$  and corresponds to the kinetic energy functional of Eq. (5) is

$$\Gamma_0 = (2-N)|\Phi_1\rangle\langle\Phi_1| + (N-1)|\Phi_2\rangle\langle\Phi_2|, \quad (8)$$

with

$$\Phi_1(r) = \sqrt{\frac{\rho_N(r)}{N}} \quad (9)$$

$$\Phi_2(r_1, r_2) = \sqrt{\frac{\rho_N(r_1)}{N}} \sqrt{\frac{\rho_N(r_2)}{N}} \quad (10)$$

thus

$$U[\rho_N] + E_{\text{x}}[\rho_N] = \text{Tr}(\Gamma_0 \hat{V}_{\text{ee}}) = \frac{2(N-1)}{N^2} U[\rho_N]. \quad (11)$$

Unlike the Hartree functional  $U[\rho_N]$  and the noninteracting kinetic energy functional  $T_{\text{vW}}[\rho_N]$  of Eq. (5), which have a simple explicit dependence on  $\rho_N(\mathbf{r})$  alone, we see from Eq. (11) that the exchange functional  $E_{\text{x}}[\rho_N]$  also explicitly depends on the particle number  $N$ . If we take the functional derivative  $\delta/\delta\rho(\mathbf{r})$  of both sides of Eq. (11) at fixed constant  $N$ , the

exchange potential is not discontinuous at  $N = 1$ . If, instead, we allow little variations of  $N$  with respect to  $\rho_N(\mathbf{r})$ , by taking into account that  $N = \int d\mathbf{r} \rho_N(\mathbf{r})$ , we obtain, for the whole range  $0 \leq N \leq 2$ ,

$$v_{\text{x}}(r) = \begin{cases} -v_{\text{H}}(r) & 0 \leq N \leq 1 \\ -\frac{(N^2 - 2N + 2)}{N^2} v_{\text{H}}(r) + \frac{2(2-N)}{N^3} U[\rho_N] & 1 < N \leq 2 \end{cases} \quad (12)$$

Equation (12) shows that, within the definition of exchange of Eq. (11) and allowing variations of  $N$  with respect to  $\rho$ , the discontinuity of the exchange potential at  $N = 1$  is a spatially independent constant equal to

$$v_{\text{x}}(r)|_{N \rightarrow 1^+} - v_{\text{x}}(r)|_{N \rightarrow 1^-} = 2U[\rho_1]. \quad (13)$$

Exact exchange in an open system of fluctuating electron number as also been widely discussed in Ref. [19], where only the energy, and not the potential, has been analyzed.

When the external potential is harmonic,  $v_{\text{ext}}(r) = 1/2 \omega^2 r^2$ , we have  $\rho_1(r) = \omega^{3/2}/\pi^{3/2} e^{-\omega r^2}$ . The ground-state wavefunction of the hamiltonian with  $N = 2$  electrons has the form

$$\Psi(\mathbf{r}_1, \mathbf{r}_2) = \eta(R) \psi(r_{12}), \quad R = \frac{|\mathbf{r}_1 + \mathbf{r}_2|}{2}, \quad r_{12} = |\mathbf{r}_2 - \mathbf{r}_1|. \quad (14)$$

The center-of-mass wavefunction  $\eta(R)$  is a simple ground-state three-dimensional harmonic oscillator state. Taut [14] has shown that for some special values of  $\omega$  also the relative wavefunction  $\psi(r_{12})$  has an analytic form. For some of these special  $\omega$ -values we have calculated the corresponding electronic density  $\rho_2(r)$ ,

$$\rho_2(r) = C \frac{e^{-\omega r^2}}{2\omega r} \int_0^\infty dr_{12} r_{12} t(r_{12})^2 (e^{-\omega(r-r_{12})^2} - e^{-\omega(r+r_{12})^2}), \quad (15)$$

where  $t(x) = e^{1/2\omega x^2} \psi(x)$ , and  $C$  is a normalization constant. The function  $t(x)$  is a polynomial of some finite order  $n$  (depending on  $\omega$ ), so that the corresponding  $\rho_2(r)$  is completely analytical (see Appendix A). This allows us to obtain the exact exchange–correlation potentials up to any large  $r$  without introducing any approximation. The large- $r$  part of the potentials is crucial to illustrate the derivative discontinuity.

Notice that similar calculations of the exact exchange-correlation potential and of other DFT-related properties for the  $N = 2$  system have been carried out in the past, mainly aimed at comparing the exact potentials with current approximations (see e.g., [15, 20, 21]). Here, our aim is to calculate the exact potentials for  $0 \leq N \leq 2$  to study their discontinuity as the particle number crosses one.

### 3. Results

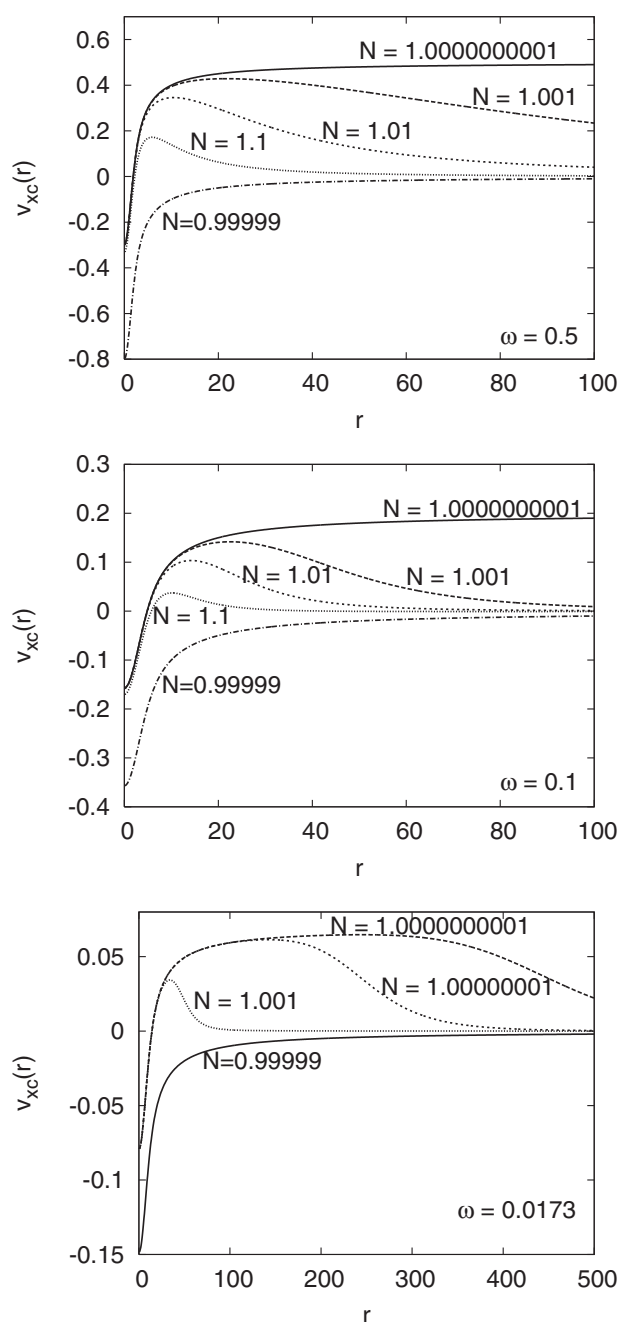
We considered three cases for which  $\psi(r_{12})$  of Eq. (14) is analytical:  $\omega = 1/2$ ,  $\omega = 1/10$ , and  $\omega = (35 - 2\sqrt{57})/712 \approx 0.0173$  (see Appendix A). The exact exchange-correlation potentials for  $N$  slightly below and slightly above 1 are reported in Figure 1. The qualitative behavior is similar to the one reported by Sagvolden and Perdew [13] obtained from the model density of the  $\text{H}^-$  ion: as  $N \rightarrow 1^+$  the exchange-correlation potential is, over a larger and larger range, more and more equal to the  $N \rightarrow 1^-$  potential plus a constant. The magnitude of the constant is, as expected [2-4],  $I - A$ , where  $I$  is the ionization potential and  $A$  the electron affinity. For closed-shell two-electron systems this amounts to  $I - A = E_2 - 2E_1$ , where  $E_2$  is the energy of the system with  $N = 2$  electrons and  $E_1$  is the energy for  $N = 1$ . Figure 1 clearly shows that, as argued in Refs. [2-4, 13], for any fixed given  $N$  (above or below 1)

$$\lim_{r \rightarrow \infty} v_{\text{xc}}(r) = 0, \quad (16)$$

but

$$\lim_{r \rightarrow \infty} \lim_{N \rightarrow 1^+} v_{\text{xc}}(r) = I - A. \quad (17)$$

Notice that when  $\omega = 1/2$  and  $\omega = 1/10$  the  $N = 1.0000000001$   $v_{\text{xc}}(r)$  starts to decay to zero for distances much larger than those considered in the figure. The case of the ensemble H and  $\text{H}^-$  studied by Sagvolden and Perdew [13] resembles most closely to the last case considered here,  $\omega \approx 0.0173$ . This is due to the fact that the  $N = 2$   $\text{H}^-$  system is considerably correlated. In the case treated here,  $v_{\text{ext}}(r) = 1/2 \omega^2 r^2$ , the  $N = 2$  system becomes more and more correlated as  $\omega \rightarrow 0$ . This is also consistent with our previous work [22], in which we compared the dependence on  $N$  (in the range  $1 \leq N \leq 2$ ) of the noninteracting kinetic energy of the



**FIGURE 1.** The exact exchange-correlation potential for a system with particle number  $N$  fluctuating between zero and two in the external potential  $1/2 \omega^2 r^2$ , for three values of  $\omega$ . The magnitude of the discontinuity at  $N = 1$  is equal to  $E_2 - 2E_1$ . We have for  $\omega = 1/2$   $E_2 - 2E_1 = 1/2$ , for  $\omega = 1/10$   $E_2 - 2E_1 = 1/5$ , and for  $\omega \approx 0.0173$   $E_2 - 2E_1 = 4\omega \approx 0.06938$ .

He series with the one of the Hooke's atom series, finding a resemblance of the  $\text{H}^-$  case (He series) with the low- $\omega$  case (Hooke's series).

## 4. Conclusions

The discontinuity of the exact Kohn–Sham exchange-correlation potential was argued in Refs. [2–4], and is known to have very important physical consequences. In particular, the absence or the underestimation of this discontinuity in local and semilocal functionals explains why they often produce a qualitatively incorrect dissociation limit for nonequilibrium nuclear positions and why they underestimate charge-transfer excitation energies in time-dependent DFT. In this work, we have calculated the exact Kohn–Sham exchange and correlation potentials as the particle number crosses 1 for a system which is analytically soluble. This way, we have illustrated, at least in one case, the existence of this discontinuity without relying on any approximation, thus providing further support to the assumptions that were used for its prediction.

## Appendix A: Analytical Densities for $N = 2$

As shown by Taut [14], for a set of special values of  $\omega$  the ground-state wavefunction of the two-electron hamiltonian has the form

$$\Psi(r_1, r_2, r_{12}) = \tilde{C} e^{-\frac{\omega}{2}(r_1^2 + r_2^2)} t(r_{12}), \quad (\text{A1})$$

where  $r_1 = |\mathbf{r}_1|$ ,  $r_2 = |\mathbf{r}_2|$ ,  $r_{12} = |\mathbf{r}_2 - \mathbf{r}_1|$ ,  $t(x)$  is a polynomial of finite order  $n$  (depending on  $\omega$ ), and  $\tilde{C}$  is the normalization constant. The easiest way to obtain the density is to use the coordinates of Coulson and Nielson [23],  $r_1$ ,  $r_2$ , and  $r_{12}$ , for the integration over  $\mathbf{r}_2$ ,

$$\rho_2(r) = C \frac{e^{-\omega r^2}}{r} \int_0^\infty dr_{12} r_{12} t(r_{12})^2 \int_{|r_2 - r_{12}|}^{|r_2 + r_{12}|} dr_2 r_2 e^{-\omega r_2^2}, \quad (\text{A2})$$

where  $C$  is again a normalization constant. Integration over  $r_2$  yields Eq. (15).

### 1. $\omega = 1/2$

The density for  $\omega = 1/2$  has been reported by several authors (see, e.g., Ref. [24]),

$$\rho_2(r) = \frac{e^{-\frac{r^2}{2}}}{4(8 + 5\sqrt{\pi})\pi^{3/2}} \left\{ \sqrt{2\pi} \left[ r^2 + 7 + \frac{4(r^2 + 1)\text{erf}\left(\frac{r}{\sqrt{2}}\right)}{r} \right] + 8e^{-\frac{r^2}{2}} \right\}, \quad (\text{A3})$$

where  $\text{erf}(x)$  is the error function.

### 2. $\omega = 1/10$

$$\rho_2(r) = \frac{e^{-\frac{r^2}{5}}}{25000\pi(61\pi + 48\sqrt{5\pi})r} \left\{ 1000r(r^2 + 45) + 5e^{\frac{r^2}{10}}\sqrt{10\pi} \left[ r(r^4 + 190r^2 + 2875) + 20(r^4 + 50r^2 + 175)\text{erf}\left(\frac{r}{\sqrt{10}}\right) \right] \right\}. \quad (\text{A4})$$

### 3. $\omega = (35 - 2\sqrt{57})/712 \approx 0.0173$

In this case we obtain

$$\rho_2(r) = \frac{2}{c} \omega^{3/2} \tilde{\rho}(\sqrt{\omega}r), \quad (\text{A5})$$

where

$$c = \frac{\pi}{384} [3(67741 + 8855\sqrt{57})\pi + 256(132 + 17\sqrt{57})\sqrt{(70 + 6\sqrt{57})\pi}]. \quad (\text{A6})$$

and

$$\begin{aligned} \tilde{\rho}(y) = & \frac{e^{-2y^2}}{192(-35 + 3\sqrt{57})^5 y} \left\{ y \left[ 8\sqrt{\frac{178}{35 - 3\sqrt{57}}} (8 \right. \right. \\ & \times (-34410443 + 4535711\sqrt{57})y^6 \\ & + 4(-307865169 + 39250957\sqrt{57})y^4 \\ & + (1987335802 - 305824402\sqrt{57})y^2 \\ & + 1407537905\sqrt{57} - 11076745221) + e^{y^2}\sqrt{\pi}(16 \\ & \times (-34410443 + 4535711\sqrt{57})y^8 \\ & + 32(-394348481 + 50982845\sqrt{57})y^6 + 8 \\ & \times (-773680875 + 26538047\sqrt{57})y^4 \end{aligned}$$

$$\begin{aligned}
& + 56(-358975185 + 8100493\sqrt{57})y^2 + 9 \\
& \quad \times (-22356535675 + 2789329039\sqrt{57})) \Big] \\
& + 4e^{y^2} \sqrt{\frac{178\pi}{35 - 3\sqrt{57}}} [16(-34410443 \\
& \quad + 4535711\sqrt{57})y^8 + 32(-85568903 \\
& + 10946667\sqrt{57})y^6 - 56(-53901687 \\
& \quad + 8766619\sqrt{57})y^4 + 72(-272540317 \\
& + 33930025\sqrt{57})y^2 + 33(-412010907 \\
& \quad + 53643311\sqrt{57})] \operatorname{erf}(y) \Big\}. \quad (\text{A7})
\end{aligned}$$

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