

Original Research Article

Modified Chachiyo Formula for Correlation Energy of Uniform Electron Gas

Udomsilp Pinsook^{*}

Department of Physics, Faculty of Science, Chulalongkorn University, Bangkok 10330, THAILAND

^{*}Corresponding author E-mail: udomsilp.p@chula.ac.th

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Abstract

In 2016, Chachiyo proposed a simple expression with only three parameters for the correlation energy of the uniform electron gas in the range of $1 \leq r_s \leq 100$. This so-called Chachiyo formula gives reasonable agreement with the results from the quantum Monte-Carlo simulations by Ceperley and Alder (CA). In this paper, I propose the TCUP formula, written as;

$$\varepsilon_c(r_s) = -0.0311 \log \left(1 + \frac{28.1660}{r_s} - \frac{19.5920}{r_s^{3/2}} + \frac{20.5613 \exp(0.9529 \sqrt{r_s} - 0.3444 r_s)}{r_s^2} \right)$$

This is a modification of the Chachiyo formula, which can reproduce the two well-known analytic limits, i.e. the high-density and the low-density limits. In addition, this modified formula gives an excellent fitting to the Perdew-Zunger data which combines the results of CA and the analytic high-density limit. Thus, it covers the more extending range of $0.01 \leq r_s \leq 100$.

Keywords: Correlation energy, Uniform electron gas, Chachiyo formula, TCUP formula

The correlation energy of the electron gas arises from the many-body interaction among the electrons. In the uniform electron gas, it can be calculated from the expression;

$$\varepsilon_c = \varepsilon_{GS} - \varepsilon_{HF} \quad (1)$$

where ε_c is the average correlation energy per electron. ε_{GS} is the average ground state energy per electron and ε_{HF} is the average Hartree-Fock energy per electron. The density of the electron gas can be defined by $n = \left(\frac{4}{3}\pi r_s^3\right)^{-1}$, which is the ratio of one electron and the average spherical volume with radius r_s . The parameter r_s is also known as the Wigner-Seitz radius. The energy in this paper is expressed in Rydberg (Ry) unit, unless otherwise stated, and r_s is in the unit of Bohr radius a_0 .

There exist two analytic limits of ε_c in the paramagnetic (PM) state at the low and high density, i.e. $r_s \gg 1$ and $r_s \ll 1$, respectively. The evaluation of ε_c in the low-density limit was pioneered by Wigner [1]. He proposed that the electron gas at very low density undergoes a phase transition into a regular lattice. Then he used the Wigner-Seitz cell to calculate ε_{GS} of the system. At that time, ε_{HF} was already known. Thus, Eq. (1) can be fulfilled. Later, there exist several correction terms from the electron lattice vibration as well [2]. At the present, the most acceptable analytic expression of ε_c in the low-density limit can be written as [1,2]

$$\varepsilon_c(r_s) = -\frac{0.8757}{r_s} + \frac{2.65}{r_s^{3/2}} - \frac{2.94}{r_s^2} \quad (2)$$

The evaluation of ε_c in the high-density limit was pioneered by Gell-mann and Brueckner [3]. They used quantum field theory (QFT) to study nuclear matters. By using a perturbation method, Gell-mann and Brueckner were able to obtain two of the leading terms of ε_c . More elaborate works have been carried out by using the QFT perturbation series and Feynman diagrams [4]. The widely accepted analytic expression of ε_c in the high-density limit can be written as [3,4],

$$\varepsilon_c(r_s) = -0.094 + 0.0622 \log(r_s) - 0.036 r_s + 0.018 r_s \log(r_s) \quad (3)$$

The great challenge comes from the fact that these two analytic limits, Eq. (2) and (3), do not join smoothly. Applications in density functional theory (DFT) need a smooth function of ε_c in a wide range of r_s . There have been numerous ways to find ε_c in between these two analytic limits, such as performing even more elaborate QFT works or resorting to a non-perturbative method. The most important non-perturbative method is the quantum Monte-Carlo (QMC) method which performs numerical path integration of the known Hamiltonian of the uniform electron gas in the range of $1 \leq r_s \leq 100$ [5]. It is regarded as the most accurate method for the correlation

energy of the uniform electron gas. The QMC method can be used to show how the correlation energy in the different limits can join each other. However, the results are still only discrete numerical data points.

Another simplest way is to find a good interpolation scheme, and yet containing most of the known physics properties [6,7]. Recently, Chachiyo [8] has proposed a new interpolation scheme of ε_c which composes of only three parameters as;

$$\varepsilon_c(r_s) = A \log(1 + \frac{B_1}{r_s} + \frac{C_1}{r_s^2}) \quad (4)$$

where $A = (\log 2 - 1)/\pi^2$, B_1 , and C_1 are the three parameters. Chachiyo suggested that $B_1 = 20.4562557 a_0$ and $C_1 = 20.4562557 a_0^2$ for the electron gas in the PM state [8]. This formula is a very smooth function with smooth derivative which is suitable for the applications in DFT [9,10]. Furthermore, it gives a reasonable comparison with the QMC results of Ceperley and Alder (CA) in the range of $1 \leq r_s \leq 100$ [5]. Eq. (4) together with Chachiyo's original parameters shall be referred to as the Chachiyo formula from now on.

Modified Chachiyo Formula

The aim of this paper is to extract the known physics properties from the Chachiyo formula and to propose a modified expression which contains most of the information from Eq. (2) and (3). At this stage, it is essential to introduce the modified expression.

$$\varepsilon_c(r_s) = A \log(1 + \frac{B_1}{r_s} + \frac{B_2}{r_s^{3/2}} + \frac{C_1 \exp(-\frac{B_2}{C_1} \sqrt{r_s} + C_2 r_s + C_3 r_s \log(r_s))}{r_s^2}) \quad (5)$$

where B_i , and C_j are the modified parameters. The reason for extending the number of parameters will be elaborated in the subsequent paragraphs. For, $B_2 = 0$, and $C_2 = C_3 = 0$, this equation reduces straightforwardly to Eq. (4). It is common to set $A = (\log 2 - 1)/\pi^2$ which is derived directly from the QFT [6-8].

By taking the limit $r_s \gg 1$ of Eq. (5), I can estimate ε_c as;

$$\varepsilon_c(r_s) \approx \frac{AB_1}{r_s} + \frac{AB_2}{r_s^{3/2}} + O(r_s^{-2}) \quad (6)$$

By comparing with the analytic low-density limit, Eq. (2), I can derive;

$$B_1 = -\frac{0.8757}{A} = 28.1660 a_0 \quad (7)$$

and,

$$B_2 = \frac{2.65}{A} = -85.2345 a_0^{3/2} \quad (8)$$

By having B_2 in Eq. (5), I can express physics of the low density limit up to the order of $r_s^{-3/2}$. The term of order r_s^{-2} depended on the exponential term, $\exp(C_2 r_s + C_3 r_s \log(r_s))$, which is strongly related to the high density limit. I will show later in Eq. (11) and (12) that, in the exact limits, $C_2 < 0$ and $C_3 < 0$. Therefore, this exponential term goes to zero at large r_s . Thus, the coefficient of r_s^{-2} in Eq. (2) cannot be obtained from the exact limit of Eq. (5). This is the limitation of Eq. (5). If $C_2 > 0$, which may occur in the later fitting process, there will be incorrect mixing between low-density and high-density limits, see Eq. (11). If $C_3 > 0$, the exponential term would also diverge at large r_s . In this case, I might need to remove that problematic parameter from Eq. (5). This is also another limitation.

By taking the limit $r_s \ll 1$ of Eq. (5), I can estimate ε_c as;

$$\varepsilon_c(r_s) \approx A \log(C_1) - 2A \log(r_s) + A \left(\frac{B_1}{C_1} + \frac{B_2^2}{2C_1^2} + C_2 \right) r_s + AC_3 r_s \log(r_s) + O(r_s^{3/2}) \quad (9)$$

The modified term $C_2 r_s + C_3 r_s \log(r_s)$, which appears in the exponential term in Eq. (5), plays an important role in Eq. (9). The term $-\frac{B_2}{C_1} \sqrt{r_s}$ in Eq. (5) is also essential. Without this term, there will appear an extra term $\frac{B_2}{C_1} \sqrt{r_s}$ in Eq. (9) with no supporting physics and does not match with any term in the analytic limits. By comparing Eq. (9) with the analytic high-density limit, Eq. (3), I can derive;

$$C_1 = \exp\left(-\frac{0.094}{A}\right) = 20.5613 a_0^2 \quad (10)$$

and

$$C_2 = -\frac{0.036}{A} - \frac{B_1}{C_1} - \frac{B_2^2}{2C_1^2} = -8.8040 a_0^{-1} \quad (11)$$

and,

$$C_3 = \frac{0.018}{A} = -0.5790 a_0^{-1} \quad (12)$$

By using C_1 , C_2 , and C_3 from Eq. (10), (11) and (12), respectively, Eq. (9) is exactly equal to Eq. (3) up to the order of $r_s \log(r_s)$. It is worth noting that C_2 in Eq. (11) behaves as a mixing regulator which controls the mixing between the parameters of the low-density limit

B_i and of high-density limits C_1 . The most influential term is $-\frac{B_2^2}{2C_1}$, as the terms $-\frac{0.036}{A}$ and $-\frac{B_1}{C_1}$ almost cancel, and this also implies that if $B_2 \neq 0$, the analytic C_2 will always be negative.

In order to investigate the limits of Eq. (4), I apply the condition that $B_2 = 0$, and $C_2 = C_3 = 0$ to Eq. (6) and (9). Then Eq. (6) reduces to [11];

$$\varepsilon_c(r_s) \approx -\frac{AB_1}{r_s} + O(r_s^{-2}) \quad (13)$$

It can express physics of the low-density limit up to the order of r_s^{-1} only. This is one of the limitations of Eq. (4). Next, Eq. (9) reduces to [11];

$$\varepsilon_c(r_s) \approx A \log(C_1) - 2A \log(r_s) + \frac{AB_1}{C_1} r_s + O(r_s^2) \quad (14)$$

By comparing Eq. (14) with Eq. (3), I found also that;

$$\frac{B_1}{C_1} = -\frac{0.036}{A} = 1.1579 a_0^{-1} \quad (15)$$

This is equivalent to solving Eq. (11) with the conditions $B_2 = 0$ and $C_2 = 0$. Accidentally, the ratio in Eq. (15) is close, but not equal, to unity. Thus, it implies that $B_1 \approx (1 + \Delta)C_1$ could be a reasonable approximation. Chachiyo [8] suggested that $\Delta = 0 a_0^{-1}$, whereas Karasiev [12] suggested that $\Delta = 0.0627 a_0^{-1}$. For the exact conditions of the Chachiyo formula, i.e. $\Delta = 0 a_0^{-1}$ and $C_1 = 20.4562557 a_0^2$ [8], Eq. (14) becomes $\varepsilon_c(r_s) \approx -0.094 + 0.0622 \log(r_s) - 0.031 r_s$ which closely resembles Eq. (3). Thus, the Chachiyo formula gives excellent description to the high-density limit. The major drawback is that Eq. (13) becomes $\varepsilon_c(r_s) = -\frac{0.6360}{r_s}$ which is quite different from the leading terms of Eq. (2). Furthermore, the Δ approximation is inconsistent with Eq. (7) and (10), which gives $\frac{B_1}{C_1} = \frac{28.1660}{20.5613} = 1.3699 a_0^{-1}$ or $\Delta = 0.3699 a_0^{-1}$. I shall refer to this problem as the $\frac{B_1}{C_1}$ inconsistency. If this inconsistency remains, both analytic limits, Eq. (2) and (3) cannot be achieved simultaneously. This is also the major limitation of Eq. (4) where the parameter set $\{A, B_1, C_1\}$ is not large enough to cover all the physics properties of Eq. (2) and (3). I will show later that the $\frac{B_1}{C_1}$ inconsistency cannot be cured by fitting. One way to cure this inconsistency is to solve Eq. (11) with $C_2 \neq 0$.

Unlike Eq. (4), it is clearly seen through Eq. (6)–(12) that Eq. (5) has a potential to reproduce both of the analytic limits, Eq. (2) and (3), simultaneously. Next, I will show how well Eq. (5) can represent the data from the QMC method. In this paper, I use ε_c data points from the works of Perdew and Zunger (CAPZ) [6]. The CAPZ data points combine the QMC results from the CA work [5] with

the results from the analytic high-density limit. It covers a wider range of $0.01 \leq r_s \leq 100$. The numerical data points are shown in the Appendix. I intend not to use the CA data directly because it covers the range of $1 \leq r_s \leq 100$ only. It does not contain the data in the high-density limit. As Eq. (5) contains the high-density terms explicitly, I would also like to see how well it can represent the high-density limit.

Fitting Results and Discussion

In order to measure the accuracy of several models, I define;

$$\chi^2 = \frac{1}{N-M} \sum_i^N (\varepsilon_c^{Model}(r_{s,i}) - \varepsilon_c^{CAPZ}(r_{s,i}))^2 \quad (16)$$

and perform the least squares fit between a selected model and the CAPZ data points [6], $N = 13$. M is the number of fitting parameters, for example if I set B_1 and C_1 as the fitting parameters of Eq. (4), then $M = 2$, and if I set all B_i and C_j as the fitting parameters of Eq. (5), then $M = 5$. From Eq. (16), I can estimate $\chi^2 \approx \sigma^2$, where σ is the standard deviation. The CAPZ data is in the units of eV and accurate to the second decimal place [6], thus I am looking for $\sigma \lesssim 0.005$ eV.

For the PM state, several sets of the parameters are summarized in Table 1.

Table 1 The parameters of the correlation energy in the paramagnetic (PM) state. I use $A = \frac{\log 2 - 1}{\pi^2} (Ry)$.

	Eq. (4) with Eq. (7), (10)	Chachiyo formula [8]	Eq. (4) with full fitting	Eq. (5) with Eq. (7), (8), (10)–(12)	TCUP formula (Eq. (17))	Eq. (5) with full fitting
$B_1(a_0)$	28.1660	20.4562557	21.7291	28.1660	28.1660	25.0494
$B_2(a_0^{3/2})$	0	0	0	-85.2345	-19.5920	-18.5019
$C_1(a_0^2)$	20.5613	20.4562557	22.4063	20.5613	20.5613	21.9395
$C_2(a_0^{-1})$	0	0	0	-8.8040	-0.3444	-0.2739
$C_3(a_0^{-1})$	0	0	0	-0.5790	0.0	0.0279
M	-	1	2	-	2	5
σ	0.056	0.028	0.009	***	0.014	0.006

***The parameter values from the analytic limits give negative argument in one of the logarithmic terms in the intermediate range of r_s .

The results in Table 1 also show that the analytically derived values of the parameters are not suitable for the interpolation. For example, Eq. (4) with the analytic parameters Eq. (7) and (10) gives a large $\sigma_{PM} = 0.056$ eV. With Chachiyo's original parameters [8], $M = 1$, Eq. (4) gives $\sigma_{PM} = 0.028$ eV. The reason is that ε_c in the region of intermediate values of r_s is not well represented by either Eq. (2) or (3). In order to provide accurate description of CAPZ, some parameters need to be smeared out via the fitting process [6]. For example, Eq. (4) with the least squares fit of B_1 and C_1 gives $\sigma_{PM} = 0.009$ eV, which is close to the target $\sigma_{PM} \lesssim 0.005$ eV. This is a very impressive fitting result. I shall refer to this result as the full fitting of Eq. (4). In this scheme, I find that the $\frac{B_1}{C_1}$ inconsistency still remains, i.e. the full fitting gives $\frac{B_1}{C_1} < 1$. By substituting the full fitting parameters of Eq. (4) in Table 1 into Eq. (14), I find that $\varepsilon_c(r_s) \approx -0.097 + 0.0622 \log(r_s) - 0.030 r_s$, which is still in good agreement with the leading terms of the analytic high density limit. However, Eq. (13) becomes $\varepsilon_c(r_s) = -\frac{0.6756}{r_s}$, which is still not much improved. At this point, it seems that if I insist to keep the accuracy, one of the analytic limits will be lost, and vice versa.

Form Table 1, I find also that the analytically derived values of the parameters of Eq. (5) are not suitable for the interpolation. This is because they impose too much constraint, so that the logarithmic term is diverged in the intermediate range. However, Eq. (5) contains more added parameters, which are suitable for the adjustment purpose. Some leading terms of the analytic limits could be restored on the expense of these added parameters. Of course, the lowest $\sigma_{PM} = 0.006$ eV comes from the full fitting of Eq. (5), using all B_i and C_j as the fitting parameters, and hence $M = 5$. The fitting results are reported in Table 1. The full fitting gives $\varepsilon_c(r_s) \approx -\frac{0.7788}{r_s} + \frac{0.58}{r_s^{3/2}}$ in the low-density limit and $\varepsilon_c(r_s) \approx -0.096 + 0.0622 \log(r_s) - 0.038 r_s - 0.001 r_s \log(r_s)$ in the high-density limit. I find that, in this fitting scheme, B_2 and C_2 are much smaller than that from Eq. (8) and (11). This is due to the smearing effect. Most of all, $C_3 > 0$, and consequently the $r_s \log(r_s)$ term has incorrect sign. It may also cause a divergence beyond $r_s = 100$. Thus, Eq. (5) with this set of parameters might not be applicable outside the fitting range. If Eq. (5) with these full fitting parameters is used in an application that the electron density is confined within the fitting range, i.e. $0.01 \leq r_s \leq 100$, it will be very accurate. However, if it is used for any prediction beyond $r_s = 100$, this might fail. It is worth noting that the magnitude of C_3 is very much smaller than that from Eq. (12) due to the smearing effect as well. It plays only an insignificant role. Therefore, this is the reason to omit C_3 in the next task.

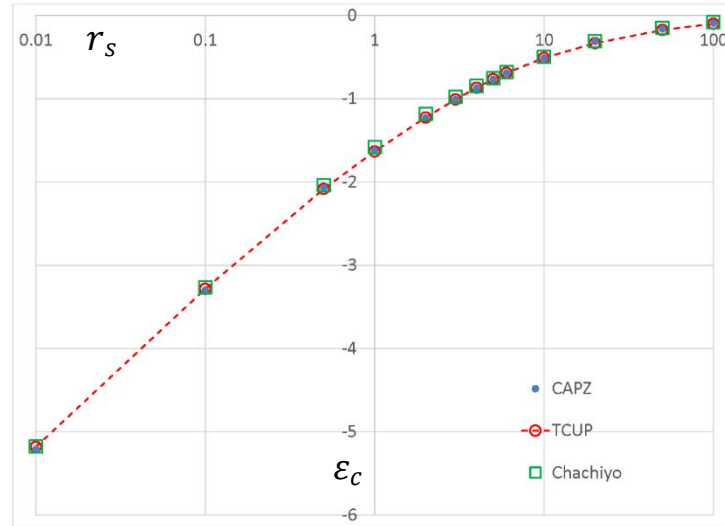


Figure 1 Shows the correlation energy in the paramagnetic (PM) state as a function of the Wigner-Seitz radius. The results are from the CAPZ data [6] (filled circles), the Chachiyo formula [8] (opened squares) and the TCUP formula, Eq. (17), (opened circles with dashed line)

If I would like Eq. (5) to reproduce both of the exact limits, I can choose to fix B_1 and C_1 to be equal to the value from Eq. (7) and (10). I still can perform the least square fitting by using B_2 , C_2 and C_3 as the fitting parameters. However, as discussed earlier, C_3 plays only an insignificant role, hence I choose to set $C_3 = 0$. Thus, in this fitting scheme, only B_2 and C_2 are the fitting parameters, and hence $M = 2$. The fitting still gives reasonable results with $\sigma_{PM} = 0.014$. The parameters are reported in Table 1. I shall refer to this formula as the TCUP formula [13]. The TCUP formula can be explicitly expressed as;

$$\varepsilon_c(r_s) = -0.0311 \log \left(1 + \frac{28.1660}{r_s} - \frac{19.5920}{r_s^{3/2}} + \frac{20.5613 \exp(0.9529 \sqrt{r_s} - 0.3444 r_s)}{r_s^2} \right) \quad (17)$$

Furthermore, I substitute the TCUP parameters into Eq. (6), and find that;

$$\varepsilon_c(r_s) \approx -\frac{0.8757}{r_s} + \frac{0.61}{r_s^{3/2}}, \quad (18)$$

and Eq. (9) becomes;

$$\varepsilon_c(r_s) \approx -0.094 + 0.0622 \log(r_s) - 0.046 r_s + 0.0 r_s \log(r_s). \quad (19)$$

The leading terms of Eq. (18) and (19) are indeed exact, compared with those of Eq. (2) and (3). By trading the accuracy, I gain the exact limits. The problem of the $\frac{B_1}{C_1}$ inconsistency is now lifted. Furthermore, this formula can be used in an application that the electron density may not be confined in the fitting range. The TCUP formula gives the power of extrapolation as well.

Figure 1 shows the graphs of $\varepsilon_c(r_s)$ in the PM state. The numerical data points are shown in the Appendix. The filled circles are the CAPZ data points [6], the opened circles are from the TCUP formula, and the opened squares are from the Chachiyo formula [8]. The graphical symbols are designed to aid visualization. If the three models are in good agreement, the filled circle, opened circle and opened square symbols at a given r_s will be at the center of each other. From Figure 1, it can be easily seen that the fitting result from TCUP formula is in good agreement with the CAPZ data points [6] in the PM state with $\sigma_{PM} = 0.014$ eV. The Chachiyo formula [8] also gives good agreement with the CAPZ data points [6] with $\sigma_{PM} = 0.028$ eV.

For completion, I consider the ferromagnetic (FM) data [6] as well. For the FM state, Eq. (4) with the Chachiyo's original parameters [8] gives $\sigma_{FM} = 0.014$ eV. Eq. (4) with the least squares fit gives $\sigma_{FM} = 0.006$ eV. The full fitting of Eq. (5) gives an amazingly accurate result with a very low $\sigma_{FM} = 0.004$ eV. The parameters and the results for the FM state are summarized in Table 2.

Table 2 The parameters of the correlation energy in the ferromagnetic (FM) state. I use $A = \frac{\log 2 - 1}{2\pi^2}$ (Ry).

	Chachiyo formula [8]	Eq. (4) with full fitting	Eq. (5) with full fitting
$B_1 (a_0)$	27.4203609	26.2241	34.6064
$B_2 (a_0^{3/2})$	0	0	-27.6895
$C_1 (a_0^2)$	27.4203609	31.5400	31.2835
$C_2 (a_0^{-1})$	0	0	-0.3816
$C_3 (a_0^{-1})$	0	0	0.0441
M	1	2	5
σ	0.014	0.006	0.004

Conclusions

The Chachiyo formula [8] is an elegant model for representing the correlation energy of the uniform electron gas with an economic parameter set. By using the least squares fit, Eq. (4) provides excellent description to the CAPZ data [6] with $\sigma_{PM} = 0.009$ eV and $\sigma_{FM} = 0.006$ eV. However, the function form and the parameter space of the formula prohibits the validation of the analytic limits, Eq. (2) and (3), simultaneously. Thus, I have proposed the modified formula, Eq. (5), which is also in a simple form with smooth

derivative, and it can reproduce the analytic forms of both the high-density and low-density limits. Its fitting is in excellent agreement with the CAPZ data points [6] with $\sigma_{PM} = 0.006$ eV and $\sigma_{FM} = 0.004$ eV. By trading with a little less accuracy, I can gain the exact analytic limits. This leads to the new “TCUP” formula, Eq. (17), which can give reasonable description of the correlation energy of the uniform electron inside the fitting range and also have an extrapolating power outside the fitting range as well. This could be useful for studying some exotic phases, such as Wigner crystallization [1,2].

Acknowledgments

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Appendix numerical data points of the correlation energy, all in units of eV.

r_s	CAPZ (PM) [6]	Chachiyo [8]	TCUP (Eq. (17))
0.01	-5.21	-5.18	-5.18
0.1	-3.30	-3.27	-3.28
0.5	-2.07	-2.04	-2.08
1	-1.62	-1.58	-1.63
2	-1.23	-1.18	-1.22
3	-1.01	-0.98	-1.01
4	-0.87	-0.85	-0.87
5	-0.77	-0.75	-0.77
6	-0.69	-0.68	-0.69
10	-0.51	-0.50	-0.51
20	-0.31	-0.31	-0.33
50	-0.16	-0.15	-0.17
100	-0.09	-0.08	-0.10

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