Liquid-gas phase transition to first order of an argon-like fluid modeled by the hard-core similar Sutherland potential

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Abstract
In this paper, an argon-like canonical system is studied. We introduce five hypothesis to deal with the total potential of the system. Then the balanced liquid-gas coexistence phenomenon is analyzed. Good equations of state and phase diagram are given.

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1 INTRODUCTION

Much attention has been paid in recent years to the hard core Yukawa (HCY) potential as a model for the pair interactions of fluids[1]. The liquid state theories such as the Mean Spherical Approximation (MSA)[2] and the Self Consistent Ornstein-Zernike Approximation (SCOZA) are proposed. Recent studies of the HCY fluid can be found in [2,3] and references therein. Worthwhile similar hard-core Sutherland potential

\[ u_{ij} = \begin{cases} +\infty, & r_{ij} < 2r_0; \\
-\frac{B}{r_{ij}^3}, & r_{ij} \geq 2r_0. 
\end{cases} \]  

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was less investigated. \( i \) and \( j \) sign two particles, \( r_{ij} \) is their distance, and \( r_0 \) is interpreted here to be the radius of a hard core. \( B \) is a constant. When \( \xi = 1 \), this potential is Sutherland potential. It is generally accepted that \( \xi > 1 \). In this paper, we mention out five hypothesis to deal with the total potential energy of a balanced canonical system, on the basis of which we obtain the canonical partition function by integrating under the help of Eq.(1).

In section 2 of this paper, the equation of state and the chemical potential are gotten by thermodynamical formulas. So do their reduced forms. Phase diagrams under five similar hard-core Sutherland potentials are illustrated.

In section 3 of this paper, We conclude that there is no proper similar hard-core Sutherland potential which can bring completely right interpretation to the balanced liquid-gas coexistence phenomenon by the theory in this paper. We find that a similar Sutherland potential with its exponent \( \xi = 1.8 \) leads to more accurate forecast not only to the critical coefficient of argon but also to the phase diagram and the relation of pressure versus temperature.

2 THEORY

2.1 The Canonical Partition Function

The case of a balanced argon-like gases canonical system in 3 dimension space is considered firstly. The average volume \( v \) of each particle is defined as

\[
v = \frac{V}{N} = \frac{1}{n},
\]

where \( n \) is the particle number density, \( V \) is the volume of the system, and \( N \) is the particle number of the system.

Suppose that volume \( v \) is a three dimensional sphere with its radius \( r_D \). Then we have

\[
\frac{4\pi}{3} r_D^3 = \frac{V}{N} = \frac{1}{n}.
\]

It is natural that \( r_D > r_0 \). They are illustrated clearly in Fig.(1).

As far as the potential energy is considered, we mention out such hypotheses as

1. The potential energy \( u_{ij} \) of two particles with their distance \( r_{ij} \) is described by Eq.(1).
2. \( U = NU_i \).
3. \( U_{i \rightarrow j} = U_{j \rightarrow i} = \frac{1}{2} U_{ij} \).

\( U_{i \rightarrow j} \) is the potential energy contribution of particle \( j \) to the total potential energy of particle \( i \), and \( U_{i \rightarrow j} \) is the potential energy contribution of particle \( i \)
to the total potential energy of particle \(j\). The potential energy \(U_{ij}\) is divided to these two particles equably. Thus,

\[
U_i = \left( U_{1 \rightarrow i} + U_{2 \rightarrow i} + \ldots + U_{(i-1) \rightarrow i} + U_{(i+1) \rightarrow i} + U_{(i+2) \rightarrow i} + \ldots + U_{N \rightarrow i} \right)
\]

\[
= \frac{1}{2} \left( U_{1i} + U_{2i} + \ldots + U_{(i-1)i} + U_{(i+1)i} + U_{(i+2)i} + \ldots + U_{Ni} \right). \quad (4)
\]

4. When the thermal fluctuation is omitted:

\[
U_{1 \rightarrow i} + U_{2 \rightarrow i} + \ldots + U_{(i-1) \rightarrow i} + U_{(i+1) \rightarrow i} + U_{(i+2) \rightarrow i} + \ldots + U_{N \rightarrow i} = \int_V u_{j \rightarrow i} d^3r_{ij} = \frac{1}{2} \int_V u_{ij} d^3r_{ij}
\]

When the idea that the total potential energy is divided to each particle equably is accepted, the total potential energy of arbitrary particle \(i\) keeps the same in the case we discussed. Our case is a balanced argon-like gases canonical system. When the thermal fluctuation is omitted, we do think that the sum of the potential energy contributions of \((N-1)\) particles to particle \(i\) is equal to the potential energy contribution of particle \(j\) to particle \(i\) in the universe space of this balanced system. \(r_1\) is the down limit of the integration and \(r_2\) is the up limit of the integration.

5. \(r_1 = r_D, \ r_2 = +\infty\).

The down limit is supposed to be \(r_D\). And the up limit is supposed to be \(+\infty\). From Eq. (1), we know that \(r_1\) should take the value of \(r_0\), which is the radius of the hard core. \(r_1 = r_D\) is equal to such an idea that integration \(I_1 = \int_{r_D}^{r_2} u_{ij} 4\pi r_{ij}^2 dr_{ij} \ll I_2 = \int_{r_D}^{r_2} u_{ij} 4\pi r_{ij}^2 dr_{ij}\). Hence \(I_1\) is not taken into account in the calculation of potential. \(r_D > r_0\) is illustrated in Fig. (1).

From hypotheses (1-5), we have the result below

\[
U = NU_i = N \left( U_{1 \rightarrow i} + U_{2 \rightarrow i} + \ldots + U_{(i-1) \rightarrow i} + U_{(i+1) \rightarrow i} + \ldots + U_{N \rightarrow i} \right)
\]

\[
= N \frac{1}{2} \int_V u_{ij} d^3r_{ij}. \quad (5)
\]

We define

\[
U_{IJ} = \int_V u_{ij} d^3r_{ij}, \quad (6)
\]

and integrate it as follows

\[
U_{IJ} = \int_V u_{ij} d^3r_{ij} = \int_{r_D}^{+\infty} u_{ij} 4\pi r_{ij}^2 dr_{ij}
\]

\[
= \int_{r_D}^{+\infty} \left( -\frac{B}{r_{ij}^{3\xi}} \right) 4\pi r_{ij}^2 dr_{ij}
\]

\[
= -\frac{4\pi B}{3 - 3\xi} (r_{ij})^{3\xi} \bigg|_{r_D}^{+\infty}
\]
Here we applied $r_D > r_0, \xi > 1$.

The Hamiltonian $H$ reads

$$H = U + \sum_{i=1}^{N} \frac{p_i^2}{2m},$$

where $m$ is the mass of one particle, $p_i$ is the momentum of particle $i$.

Thus canonical partition function can be solved as

$$Q = \frac{1}{N!\lambda^3N} \int \exp(-\beta U) d^{3N}r,$$

with thermal wavelength $\lambda = \frac{\hbar}{\sqrt{2\pimk_BT}}$, $k_B$ is the Boltzmann constant and $z_N(T,V)$ is called position partition function normally.

Inputting Eq.(5) to Eq.(10), we get

$$Q = \frac{1}{N!\lambda^3N} \int \exp(-\beta N\frac{4\pi B}{3-3\xi}(r_D)^{3-3\xi}) d^{3N}r$$

$$= \frac{1}{N!\lambda^3N} \int \exp(-\beta N\frac{2\pi B}{3-3\xi}(\frac{3}{4\pi})(1-\xi)n^{(-1+\xi)}) d^{3N}r$$

$$= \frac{1}{N!\lambda^3N} \int \exp(\beta B'Nn^{(-1+\xi)}) d^{3N}r$$

$$= \frac{1}{N!\lambda^3N} \int \exp(\beta B'Nn^{\sigma}) d^{3N}r$$

$$= \frac{1}{N!\lambda^3N} \exp(\beta B'Nn^{\sigma}) \int d^{3N}r$$

$$= \frac{1}{N!\lambda^3N} \exp(\beta B'Nn^{\sigma}) V_f^{N},$$

with $\sigma = \xi - 1$, $B' = -\frac{2\pi B}{3-3\xi}(\frac{3}{4\pi})(1-\xi)$ and $V_f$ is the free volume. $n$ is the particle number density which keeps conservation when a balanced canonical system is considered. And we do not consider the fluctuation of $n$ in this paper. For the
existence of the radius \( r_0 \) of the hard core, each particle excludes a volume \( \frac{4\pi r_0^3}{3} \) and hence for \( N \) particles the excluded volume is \( \frac{4\pi r_0^3}{3} N \). The free volume, therefor \( V_f = V - Nb \) with \( b = \frac{4\pi r_0^3}{3} \). Thus Eq. (11) reads

\[
Q = \frac{1}{N!\lambda^{3N}} \exp(-\beta B'Nn^\sigma)(V - Nb)^N.
\]  

(12)

2.2 The Equation of State

We can get the equation of state as follows

\[
P = k_B T \left( \frac{\partial \ln Q}{\partial V} \right)_{N,T} = \frac{Nk_BT}{V - Nb - \sigma B'n^{\sigma+1}}.
\]  

(13)

When we choose \( \sigma = 1 \), the equation of state is written as

\[(P + B'n^2)(v - b) = k_BT.
\]  

(14)

This is just the form of the VDW equation of state.

Here we sign the pressure of the gases \( P_1 \), the pressure of the liquids \( P_2 \), the chemical potential of the gases \( \mu_1 \), the chemical potential of the liquids \( \mu_2 \), the critical temperature \( T_c \), the critical pressure \( P_c \), the critical particle number density \( n_c \), the particle number density of the gases \( n_1 \), the particle number density of the liquids \( n_2 \), the reduced temperature \( T^* \), the reduced particle number density of the gases \( n_1^* \), the reduced particle number density of the liquids \( n_2^* \), the reduced pressure of the gases \( P_1^* \), the reduced pressure of the liquids \( P_2^* \), the particle number of the gases \( N_1 \), the particle number of the liquids \( N_2 \), the volume of the gases \( V_1 \), and the volume of the liquids \( V_2 \). The relations between them are

\[T^* = \frac{T}{T_c},\]
\[n_1 = \frac{N_1}{V_1},\]
\[n_2 = \frac{N_2}{V_2},\]
\[n_1^* = \frac{n_1}{n_c},\]
\[n_2^* = \frac{n_2}{n_c},\]
\[P_1^* = \frac{P_1}{P_c},\]
\[P_2^* = \frac{P_2}{P_c}.
\]  

(15) - (21)

At critical point, the function \( P = P(N,V,T) \) has such qualities as

\[
\left. \frac{\partial P}{\partial V} \right|_{T_c} = 0,
\]

(22)
Thus we get the critical data by solving Eq.(22) and Eq.(23). They are

\[ n_c = \frac{\sigma}{(\sigma + 2)b} \]  

\[ k_B T_c = \sigma(\sigma + 1)(\frac{2}{\sigma + 2})^2 B' n_c^\sigma. \]  

\[ P_c = \frac{\sigma^2}{\sigma + 2} B' n_c^{\sigma+1}, \]  

\[ \frac{n_c k_B T_c}{P_c} = \frac{4(\sigma + 1)}{\sigma(\sigma + 2)}. \]  

When \( \sigma = 1 \), they read

\[ n_c = \frac{1}{3b}. \]  

\[ k_B T_c = \frac{8B'}{27b}. \]  

\[ P_c = \frac{B'}{27b^2}. \]  

\[ C = \frac{n_c k_B T_c}{P_c} = 8/3. \]  

We are very familiar with these results which are just the reduced data from the VDW equation of state. \( C \) is the critical coefficient. When \( \sigma = 0.7432 \), \( C = 3.4201 \).

Inputting Eq.(24-26) to Eq.(13), we get the reduced equation of state

\[ P^* = \frac{4n^* T^* (\sigma + 1)}{((\sigma + 2) - n^* \sigma) \sigma} - \frac{(\sigma + 2) n^* (\sigma + 1)}{\sigma}. \]  

### 2.3 The Chemical Potential

Here we can solve the chemical potential as follows

\[ \mu = -k_B T \left( \frac{\partial \ln Q}{\partial N} \right)_{V,T} \]

\[ = -B'(\sigma + 1) n^\sigma + k_B T \ln \left( \frac{n}{1 - nb} \right) + \frac{n k_B T b}{1 - nb} - \frac{3 k_B T}{2} \ln \frac{2 \pi m k_B T}{h^2}. \]  

Inputting Eq.(24-26) to Eq.(33), we get the critical chemical potential. Then the reduced chemical potential is calculated to be

\[ \mu^* = \frac{A_1 + A_2}{A_3}. \]
where $A_1$, $A_2$ and $A_3$ are given by

$$A_1 = \left[ -n^*\sigma + \left( \ln \frac{n^*\sigma}{b(\sigma + 2 - n^*\sigma)} \right) \frac{4\sigma T^*}{(\sigma + 2)^2} + \frac{4\sigma^2 T^* n^*}{(\sigma + 2 - n^*\sigma)(\sigma + 2)^2} \right],$$

(35)

$$A_2 = \left[ -\frac{6T^*\sigma}{(\sigma + 2)^2} \ln \frac{8\pi m B' T^* \sigma^{(\sigma + 1)}(\sigma + 1)}{b^\sigma (\sigma + 2)^{\sigma + 2} h^2} \right],$$

(36)

$$A_3 = \left[ -1 + \left( \ln \frac{\sigma}{2b} + \frac{\sigma}{2} - 3 \frac{1}{2} \ln \frac{8\pi m B' \sigma^{(\sigma + 1)}(\sigma + 1)}{b^\sigma (\sigma + 2)^{\sigma + 2} h^2} \right) \frac{4\sigma}{(\sigma + 2)^2} \right].$$

(37)

### 2.4 Balanced Liquid-gas Coexistence Canonical System

We consider the case of the balanced liquid-gas coexistence canonical argon-like system in 3 dimensional space, which is a unit-two phases system. Here we do think the two phases are described by the same canonical partition function as Eq.(12). There are three balanced conditions which must be satisfied when these two phases are balanced. They are thermal condition

$$T_1 = T_2,$$

(38)

dynamic condition

$$P_1 = P_2,$$

(39)

and phase condition

$$\mu_1 = \mu_2.$$  

(40)

For Eq.(15-21), these three conditions can be expressed in the reduced unit as

$$T_1^* = T_2^*,$$

(41)

$$P_1^* = P_2^*,$$

(42)

$$\mu_1^* = \mu_2^*.$$  

(43)

From Eq.(32), we have

$$P_1^* = \frac{4n_1^* T_1^* (\sigma + 1)}{((\sigma + 2) - n_1^*\sigma)\sigma} - \frac{(\sigma + 2) n_1^*^{(\sigma + 1)}}{\sigma},$$

(44)

$$P_2^* = \frac{4n_2^* T_2^* (\sigma + 1)}{((\sigma + 2) - n_2^*\sigma)\sigma} - \frac{(\sigma + 2) n_2^*^{(\sigma + 1)}}{\sigma}.$$  

(45)

From Eq.(34), we have

$$\mu_1^* = \frac{(A_1)_1 + (A_2)_1}{(A_3)_1},$$

(46)
The solution of Eq.(41) and Eq.(42) is

\[ T_1^* = T_2^* = T^* = \frac{((n_1^*)^{(\sigma+1)} - (n_2^*)^{(\sigma+1)}) (1 - n_c(n_1^*)b) (1 - n_c(n_2^*)b)}{(\sigma + 1)(\frac{2}{\sigma+2})^2 ((n_1^*) - (n_2^*))} \]  

The solution of Eq.(41) and Eq.(43) is

\[ T_1^* = T_2^* = T^* = \frac{((n_1^*)^{\sigma} - (n_2^*)^{\sigma})}{\sigma(\frac{2}{\sigma+2})^2 \ln \left( \frac{n_1^*(1-\mu_c n_1^*)}{n_2^*(1-\mu_c n_2^*)} \right) + \frac{n_c(n_1^*)b}{(1-n_c(n_1^*)b)} - \frac{n_c(n_2^*)b}{(1-n_c(n_2^*)b)}} \]  

Thus we get a function \( f(n_1^*, n_2^*) = 0 \) easily from Eq.(48) and Eq.(49).

\[ f(n_1^*, n_2^*) = \frac{((n_1^*)^{(\sigma+1)} - (n_2^*)^{(\sigma+1)}) (1 - n_c(n_1^*)b) (1 - n_c(n_2^*)b)}{(\sigma + 1)((n_1^*) - (n_2^*))} \]

If \( \sigma \) is chosen to be a constant, \( n_2^* \) can be gotten in the way of numerical computation by computer when an arbitrary \( n_1^* \) is fixed. Then \( T^* \) is obtained easily from Eq.(48) or Eq.(49). And \( P^* \) is solved by Eq.(32). Table.(1) is the theoretic data when \( \sigma = 0.8 \). Fig.(2) is the correlation of \( n_1^* \) and \( n_2^* \) under different similar hard-core Sutherland potentials signed by different \( \sigma \). Fig.(3) is the phase diagrams corresponding with different \( \sigma \). Fig.(4) is the curve of \( \ln P^* \) versus \( 1/T^* \). In Ref.[4], we have introduced one method called polynomial approximation to simulate the relation of two variables. Here we can get the relation of \( \ln P^* \) versus \( 1/T^* \) by this method when \( \sigma \) is fixed. In Ref.[5], the theoretic relation of \( \ln P^* \) versus \( 1/T^* \) was given by this method in the case of \( \sigma = 1 \).

In 1945, E.A.Guggenheim collected the data of the balanced liquid-gas co-existence system from experiments and gave out the correlation of \( \rho_1^* \), \( \rho_2^* \) and \( T^* \) by the empirical equations[5,6] below

\[ \rho_1^* = 1 + 0.75(1 - T^*) - 1.75(1 - T^*)^{1/3}, \]  

\[ \rho_2^* = 1 + 0.75(1 - T^*) + 1.75(1 - T^*)^{1/3}. \]  

For that

\[ \rho_1^* = mn_1/mn_c = n_1/n_c = n_1^*, \]  

\[ \rho_2^* = mn_2/mn_c = n_2/n_c = n_2^*, \]  

we have

\[ n_1^* = 1 + 0.75(1 - T^*) - 1.75(1 - T^*)^{1/3}, \]  

\[ n_2^* = 1 + 0.75(1 - T^*) + 1.75(1 - T^*)^{1/3}. \]
\[ n^*_2 = 1 + 0.75(1 - T^*) + 1.75(1 - T^*)^{1/3}. \]  
(56)

In Eq.(51-52), \( \rho^*_1 \) is the reduced density of the gases and \( \rho^*_2 \) is the reduced density of the liquids. As far as argon system is concerned, the inaccuracy of these two equations is generally only one or two parts per thousand of \( \rho^*_2 \) or of \( \rho^*_1 \) when \( T^* > 0.60T_c \).[5,6] So, it is acceptable to consider the data of \((T^*, n^*_1(T^*), n^*_2(T^*))\) from Eq.(55-56) as the experimental ones in this temperature region[5].

E.A. Guggenheim gave a numerical analytic result of the relation between the reduced temperature and the reduced pressure from experiments by equation[5,7]

\[ P^*_e = \exp(5.29 - 5.31/T^*), \]  
(57)

which best fits the experimental data for argon when \( T^* > 0.60T_c \) except a tiny region near the critical point[5,7]. Thus it is acceptable to consider the data of \((P^*_e, T^*)\) from Eq.(57) as the experimental ones, too[5]. The data from Eq.(55-57) are illustrated as experimental data in Fig.(2-4) and Table.(2) to compare with the theoretic ones deduced above.

3 CONCLUSIONS AND DISCUSSIONS

In this paper we advanced five hypothesis to deal with the total potential energy of a balanced canonical system. Hypothesis 1-4 are easy to be accepted. The key is hypothesis 5. Actually \( r_1 = r_D \), \( r_2 = +\infty \) is a simple approximation to a concrete system. In labs, the volume of the system we consider is finite: \( r_2 \neq +\infty \). Additionally, only when we consider a static system in 3 dimensional space, can the average volume of a particle be regarded as a sphere. And \( r_D \) is effective. \( r_1 = r_D \) is only a simple effective approximation.

On the basis of the five hypothesis, we got the canonical partition function Eq.(12). Thus all the thermodynamic quantities will be solved by Eq.(12). Following, we analyzed the balanced liquid-gas coexistence canonical argon-like system. The results are partly illustrated in figures and tables. Fig.(3-4) indicates that \( \sigma \) in the region (0,1) leads to more perfect forecast to experimental data. Applying numerical calculation by computer, we find \( \sigma = 0.8 \) is the best among the five values of \( \sigma \) chosen in this paper only when phase diagram and the relation of \( \ln P^* \) versus \( 1/T^* \) are considered together. It is clearly illustrated in Figure.(3-4). Then the similar hard-core Sutherland potential is fixed to be the form of

\[ u_{ij} = \begin{cases} +\infty, & r_{ij} < r_0, \\ -\frac{B}{r_{ij}^{5}}, & r_{ij} \geq r_0. \end{cases} \]  
(58)

In this case, the equation of state is

\[ P = \frac{Nk_BT}{V - N\sigma} - 0.8 \ast B^*n^{1.8}. \]  
(59)

Now the critical coefficient \( C \) is equal to 3.2143. But \( \sigma = 0.8 \) is not the best when the relation of \( n^*_1 \) versus \( n^*_2 \) is considered. Figure.(2) suggests that \( \sigma = 0.9 \) and 1 are better.
Now we will ask whether a proper $\sigma$, which can bring completely right forecast to experiments when the liquid-gas coexistence phenomenon is considered, exists or not by the theory in this paper. Suppose that such a $\sigma = \sigma_0$ exists. Thus Eq.(50) will be right when arbitrary two experimental data terms ($n_1^*, n_2^*$) are considered. From Eq.(55-56), we get two experimental data terms: $(0.1963, 2.0137)|_{T^*=0.8600}$, $(0.3928, 1.6822)|_{T^*=0.9500}$. We input them to Eq.(50) and get the numerical value of $\sigma$ by Matlab software. The result is $\sigma|_{T^*=0.8600} = 0.9872 \neq \sigma|_{T^*=0.9500} = 1.0883$. Thus we conclude such a proper $\sigma$ does not exist. But it does not mean that there exists no value for $\sigma$ corresponding with the potential explaining the experimental data properly. A new theory may be do. Actually, when the liquid-gas phase transiton to second order is considered, our work does not work well for the obvious fluctuation near to the critical point, which is omitted in hypothesis (4). We will discuss it in details in future work. Of course, we see that this work offered the proper critical coefficient $C = 3.4201$ of argon by a simple equation of state with $\sigma = 0.7432$.

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Figure 1: Two radius

Figure 2: The curve of $n^*_1$ versus $n^*_2$. The real one is from Eq.(60-61). The rest are all from Eq.(55) with different $\sigma$: square–$\sigma = 2$; diamand–$\sigma = 1.5$; star–$\sigma = 1$; dot–$\sigma = 0.9$; cross–$\sigma = 0.8$. The star one is the result of the VDW equation of state.
Figure 3: The phase diagrams. The real one is from Eq.(60-61). The rest are from the theoretic data with different $\sigma$: square–$\sigma = 2$; diamond–$\sigma = 1.5$; star–$\sigma = 1$; dot–$\sigma = 0.9$; cross–$\sigma = 0.8$. The star one is the result of the VDW equation of state.
Figure 4: The curve of $\ln P^*$ versus $1/T^*$. The real one is from Eq.(62). The rest are theoretic data with different $\sigma$: square—$\sigma = 2$; diamond—$\sigma = 1.5$; star—$\sigma = 1$; dot—$\sigma = 0.9$; cross—$\sigma = 0.8$. The star one is the result of the VDW equation of state.
Table 1: Theoretic data when $\sigma = 0.8$.

|    | $n_1^*$ | $n_2^*$ | $P^*$ | $T^*$ |
|----|---------|---------|-------|-------|
| $n_1^*$ | 0.0106  | 0.0292  | 0.0535 | 0.0851 | 0.1266 | 0.1827 | 0.2627 | 0.3928 |
| $n_2^*$ | 2.8097  | 2.6574  | 2.5328 | 2.4122 | 2.2860 | 2.1458 | 1.9785 | 1.7521 |
| $P^*$   | 0.0158  | 0.0483  | 0.0931 | 0.1521 | 0.2287 | 0.3280 | 0.4590 | 0.6411 |
| $T^*$   | 0.4911  | 0.5743  | 0.6360 | 0.6906 | 0.7426 | 0.7944 | 0.8484 | 0.9085 |

Table 2: The data from Eq.(55-57).
