Precise calculations of the effective-range expansion parameters for the Lennard-Jones potential

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Several effective-range-theory parameters for the Lennard-Jones 12-6 potential needed in helium equation-of-state calculations are determined analytically. Though the results are not expressed in terms of known functions, we show how they may be computed at machine precision. The power expansion of these parameters in terms of the coupling constant of the attractive part of the potential is also determined through the first fourteen terms.

I. INTRODUCTION

The possibility of constructing the equation of state of liquid helium from the well-known low-density expansions has recently been pointed out for both bosons and fermions. It is based on a perturbation scheme about the fluid with repulsive interactions, instead of the usual one about the ideal gas, to which is then applied extrapolation to moderate densities by Padé approximants and generalizations thereof.

The perturbation scheme, which really goes back to van der Waals and which has already been very successfully applied in classical fluids, suggests itself in the quantum fluid case by the solution of the following difficulty. The low-density expansion of the ground-state energy per particle for a Bose gas of particles of mass \( m \) with pair interactions \( V(r) \) which gives rise to an \( S \)-wave scattering length \( a \) is given by the non-power-series expansion

\[
\frac{E}{N} = \frac{2\pi \hbar^2}{m \rho a} \left[ 1 + C_1 (\rho a^3)^{1/2} + C_2 \rho a^3 \ln(\rho a^3) + C_3 \rho a^3 + \cdots \right],
\]

where \( \rho \) is the number density and \( C_1 \) and \( C_2 \) are pure numbers. The pair interaction between helium atoms is sufficiently attractive so that \( a < 0 \), and this clearly introduces complex quantities in (1) which cannot be easily interpreted. To avoid this problem one can define the dimensionless attractive coupling parameter \( \lambda \) such that

\[
V(r, \lambda) = \begin{cases} V(r), & r < R \\ \lambda V(r), & r \geq R \end{cases}
\]

where \( V(r) \) is the original intermolecular potential and \( R \) is the separation at which it vanishes, i.e., \( V(R) = 0 \). The scattering length \( a(\lambda) \), which is a space integral over \( V(r, \lambda) \) can then be expanded in powers of \( \lambda \) about \( \lambda = 0 \) so that

\[
a(\lambda) = a_0 + a_1 \lambda + a_2 \lambda^2 + \cdots.
\]

When this is substituted back into (1) the energy can be expressed as a double series in \( \rho \) and \( \lambda \). It is still irregular in \( \rho \) but is now a regular (power) series in \( \lambda \). The energy is now real for all \( \lambda \geq 0 \) and has been cast into a perturbation scheme about the purely repulsive \( \lambda = 0 \) fluid.

In the fermion case the known low-density expansion contains the scattering length \( a \) as well as other scattering parameters like the \( S \)-wave effective range \( r_0 \), the \( P \)-wave scattering length \( A_1(0) \), and a higher-order \( S \)-wave quantity called \( A_0^+(0) \) which is potential shape dependent. The usual technique of determining these parameters, except the last one mentioned, for a given specific potential \( V(r) \), is to determine the low-energy partial-wave shifts \( \delta_i(k) \) by numerical integration of the Schrödinger equation for positive energies and then fitting these to determine indirectly, by graphical procedures, the parameters \( A_i(0) \) (equal to \( a \) for \( l = 0 \)) and \( r_1 \) through the effective range expansion

\[
k^{2l+1} \cot \delta_i(k) \approx -\frac{1}{A_i(0)} + \frac{1}{2} r_1 k^2 + O(k^4) \quad \text{as } k \to 0.
\]

This technique is not very accurate in practice, but the direct procedure based on the fact that \( a, r_0, A_1(0), \) etc., can be expressed as simple integrals over the zero-energy radial wave function times the potential has recently been shown to be superior for determining their value at fixed \( \lambda \) but cumbersome for calculating the derivatives in \( \lambda \) as in Eq. (3).

The aim of this paper is to show that for the Lennard-Jones 12-6 potential these expansions can be determined with machine precision through algebraic methods. In Sec. II we determine the low-energy parameters for the full interaction. Sec. III is devoted to the calculation of the above-mentioned expansion coefficients, and Sec. IV contains some conclusions and comments.

The Lennard-Jones potential is given by

\[
V(r) = 4\varepsilon \left( \frac{\sigma}{r} \right)^{12} - 2 \left( \frac{\sigma}{r} \right)^6,
\]

where \( \varepsilon = 10.22 \) K and \( \sigma = 2.556 \) \AA. The particles
considered have $\frac{h^2}{m(\text{He})} = 12.120904 \text{ K}\dot{\text{A}}^2$ and $\frac{h^2}{m(\text{He})} = 16.085775 \text{ K}\dot{\text{A}}^2$. We use natural units, i.e., lengths are measured in units of $\sigma$. Our results for $a$ and $r_0$ must be multiplied by $\sigma$ to have them in $\text{Å}$, and $A_4(0)$ and $A_5(0)$, to be defined later on, must be multiplied by $\sigma^5$. In such units the potential becomes zero at $x = 1$, and the dimensionless potential is thus given by

$$U(x) = \frac{4e^2m\sigma^2}{h^2} \left( \frac{1}{x^{12}} - \frac{1}{x^6} \right).$$

**II. EFFECTIVE-RANGE EXPANSION PARAMETERS FOR THE FULL INTERACTION**

As is well known, the effective-range-theory parameters can be defined in the following form: the $S$-wave radial function at zero energy and regular at $x = 0$ behaves, at large distances, as

$$u(x) \rightarrow x - a \quad \text{as} \quad x \rightarrow \infty,$$

(4)

$a$ being the $S$-wave scattering length. The two conditions, $u(0) = 0$ and Eq. (4), completely determine the regular solution of the equation

$$\frac{d^2 u(x)}{dx^2} = U(x)u(x)$$

(5)

and once $u(x)$ is known the effective range may be computed by means of the integral

$$r_0 = \frac{2}{a^2} \int_0^\infty dx \left[ (x - a)^2 - u^2(x) \right].$$

(6)

Another $S$-wave parameter of interest is the shape-dependent quantity

$$A_0(0) = -\frac{1}{2} \int_0^\infty dx \frac{x}{U(x)}u(x).$$

(7)

We now look for solutions of Eq. (5) of the form

$$u(x) = x + \sum_{n=0}^{\infty} \frac{A_n}{x^n}.$$  

(8)

The differential equation (5) has two singular points at 0 and at $\infty$. The proposed ansatz, Eq. (8), corresponds to a solution around the point at infinity, which is a regular point, and this solution is everywhere convergent except at $x = 0$. Substitution of Eq. (8) in Eq. (5) gives rise to the three-term recurrence relation

$$n(n + 1)A_n = V_0(A_{n-10} - A_{n-90}),$$

(9)

where $V_0 = 4e^2\sigma^2m/h^2$ and $A_{-1} = 1$ and $A_{-n} = 0$, $n = 2, 3, \ldots$. The first coefficients of the expansion (8) are given by

$$A_{-1} = 1,$$

$$A_0 = \text{undetermined} = -a,$$

$$A_1 = 0,$$

$$A_2 = 0,$$

$$A_3 = -V_0/12,$$

$$A_4 = -V_0A_0/20,$$

$$\ldots$$

Note that all odd coefficients are independent of $a$ but even coefficients are linear in $A_0 = -a$. If the even coefficients are redefined so as to factor $A_0$, $u(x)$ can be put in the form

$$u(x) = x + \sum_{n=3}^{\infty} \frac{A_n}{x^n} = A_0 \left[ 1 + \sum_{n=4}^{\infty} \frac{\tilde{A}_n}{x^n} \right].$$

(11)

$\tilde{A}_n \equiv A_n/A_0$, $n = 4, 6, 8, \ldots$, and the value of $A_0$ follows from the condition $u(x) = 0$ for $x$ very small. This is the only approximation involved in the determination of the wave function with the boundary condition (4), and is a consequence of the fact that the expansion (8) is not valid at $x = 0$. However, this approximation is harmless because the presence of the repulsive core at short distances makes $u(x)$ very small in a large region beyond $x = 0$. Taking $u(x) = 0$ at $x = 0.4$, e.g., already gives very good results.

Equation (11) cannot be straightforwardly used for the calculation of $u(x)$ since some 2000 terms are needed in the expansion and the values of the two terms of right hand side of Eq. (11) are very large. For example, in $^4\text{He}$ at $x = 0.36$ the even sum is $6.062 \times 10^5$ and the odd sum is $-1.418 \times 10^6$. We obtain for $A_0$ the value of $68.984 \times 10^5$ with all digits correct. However, the resulting value of the function at this point turns out to be $-2.06 \times 10^{16}$ and thus certainly erroneous. This is due to the fact that we are computing with 16 digits of precision. This problem propagates at larger values of $x$ and only for $x \geq 0.56$ can the function be correctly computed in this manner.

It is also important to realize the insensitivity of $A_0$ to $x$, for small $x$, being points where the wave function is assumed to vanish: even at $x = 0.60$ the value of $A_0$ is obtained with seven correct decimal figures. Our result for $a = -176.325 \times 290.624$ agrees with that of Ref. 7 in the first 10 digits. We should remark that our errors are solely rounding errors, and the use of a higher precision computer would allow determination of more digits.

The calculation of $r_0$ and $A_0(0)$ must be carried out with the previous remarks in mind. Equations (6) and (7) are thus taken as

$$r_0 = \frac{2}{a^2} \left[ \int_0^{x_1} dx (x-a)^2 \right.$$

$$+ \int_{x_1}^{\infty} dx [(x-a)^2 - u^2(x)] \left. \right],$$

(12)

$$A_0(0) = -\frac{1}{2} \int_0^{\infty} dx x^3u''(x),$$

(13)
i.e., the wave function $u(x)$ is assumed to vanish for $x < x_1$. The value of $x_1$ must be larger than the value of $x_0$ used for the determination of the scattering length, as we have commented above. All integrals appearing in (12) and (13) are computed analytically and the results for $^4\text{He}$ and $^3\text{He}$ are shown in Table I. Note that our less accurate results correspond to $A_n'(0)$ where we have been able to compute only seven decimal digits. This is a consequence of using $x_1 > 0$ in Eqs. (12) and (13), instead of $x_1 = 0$, and we are forced to do that because the wave function is incorrectly computed at small values of $x$.

The importance of the errors induced may be deduced by looking at Fig. 1, where we have plotted $u(x)$ and the absolute value of $u''(x)$. We see that $u(x)$ changes by several orders of magnitude at $x = 0.5$ and 1.0 and because of that the error induced in the calculation of $r_0$ is negligibly small. On the other hand, $u''(x)$ changes approximately by only 3 orders of magnitude from $x = 0.5$ to 1.0 so that the contribution of the neglected integral from $x = 0$ to 0.5 may be expected to be much more important. Given that our expansion does not permit the evaluation at short distances of $u(x)$ there will be a relatively large error for $A_n'(0)$.

In Table I we include other information necessary for the results to be presented in the next section, namely the logarithmic derivative at $x = 1$, the integral of $u^2(x)$ from $x = 0$ to 1 and the integral of $x^3u''(x)$.

The scattering length in the $P$-wave channel may be computed in an analogous way. The $P$-wave radial function $u_1(x)$ is now expanded in the form

$$u_1(x) = \frac{x^2}{3} + \sum_{n=1} B_n \frac{x^n}{n!}$$

so as to satisfy the corresponding differential equation

$$u_1''(x) = V_0 \left[ \frac{1}{x^{12}} - \frac{1}{x^6} \right] u_1(x) + \frac{2}{x^2} u_1(x).$$

The recurrence relation

$$[n(n+1) - 2] B_n = V_0(B_{n-10} - B_{n-4})$$

must hold, with $B_{-n} = 0$, $n = 0, 1, 3, \ldots$ and $B_{-2} = \frac{1}{3}$. The first terms are

$$B_1 = \text{undetermined},$$

$$B_2 = -\frac{V_0}{12},$$

$$B_3 = B_1 = 0,$$

$$B_5 = -\frac{V_0 B_1}{28},$$

and so on.

As before, the undetermined $B_1$ may be factored out to obtain an equation analogous to (11). Note that $B_1 = -A_1(0)$. Values for $A_1(0)$ in $\sigma$ units are also shown in Table I, as are the logarithmic derivatives at $x = 1$ for the $P$-wave solution.

### III. EXPANSION IN POWERS OF THE COUPLING CONSTANT

As mentioned in Sec. I, we have evaluated the expansion of the various low-energy parameters in powers of the coupling constant of the attractive part of the Lennard-Jones potential, Eq. (2). The potential now presents a discontinuity in the derivative at $x = 1$ for $\lambda \neq 1$ so that we must separately solve the differential equation in the two regions $(0,1)$ and $(1,\infty)$ with the same boundary conditions as above, and impose the equality of the logarithmic derivatives of both these solutions at $x = 1$. The value of $(u'/u)_{x=1}$ for the inner solution has been already determined in the preceding section, and presented in Table I. This value is clearly independent of $\lambda$, the coupling parameter of the attractive part.

To express the outer solution in terms of $\lambda$, we use the ansatz

$$u(x,\lambda) = x + \alpha(\lambda) \left[ \sum_{n=0}^{\infty} \sum_{p=0}^{\infty} C_{np} \frac{\lambda^p x^n}{\lambda^{n+p}} \right]$$

and

$$+ \sum_{n=1}^{\infty} \sum_{p=0}^{\infty} C_{np} \frac{\lambda^p x^n}{\lambda^{n+p}}$$

where $\alpha(\lambda)$ is a constant.

### TABLE I. Effective-range parameters corresponding to the full Lennard-Jones interaction in $\sigma$ units.

<table>
<thead>
<tr>
<th></th>
<th>$^4\text{He}$</th>
<th>$^3\text{He}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
<td>-68.984 855 487.5</td>
<td>-2.373 018 484 44</td>
</tr>
<tr>
<td>$r_0$</td>
<td>3.112 979 625 92</td>
<td>6.041 136 050 03</td>
</tr>
<tr>
<td>$A''(0)$</td>
<td>135.886 8</td>
<td>7.167 161</td>
</tr>
<tr>
<td>$u'(1)/u(1)$</td>
<td>5.519 969 750 98</td>
<td>5.178 679 075 73</td>
</tr>
<tr>
<td>$u(1)$</td>
<td>19.751 109 897 3</td>
<td>1.075 404 278 79</td>
</tr>
<tr>
<td>$\int_1^x \frac{u(x) , dx}{x}$</td>
<td>25.199 988 183 2</td>
<td>0.079 330 634 829</td>
</tr>
<tr>
<td>$\int_1^x x^3 u''(x) , dx$</td>
<td>61.266 9</td>
<td>3.002 242</td>
</tr>
<tr>
<td>$A_1(0)$</td>
<td>2.468 607 616 02</td>
<td>1.569 787 729 43</td>
</tr>
<tr>
<td>$u_1'(1)/u(1)$</td>
<td>5.663 518 731 6</td>
<td>5.332 062 458 07</td>
</tr>
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</table>
so that the scattering length as a function of \( \lambda \) is given by \(-\alpha(\lambda)\). Substituting into the differential equation provides the recurrence relation

\[
n(n+1)C_{n,p} = V_0(C_{n-10,p-1} - C_{n-6,p-1})
\]

with the additional conditions \( C_{-n,p} = 0 \) for \( n = 2, 3, \ldots \) and \( C_{-1,p} = \delta_{1,p}, C_{p} = \delta_{p,p} \). Once the coefficients \( C_{np} \) are determined, we may obtain the power-series expansion of \( \alpha(\lambda) \) by imposing equality of logarithmic derivatives, with the result

\[
\frac{[1 - (u'/u_1)] - \sum_{n=odd}^{\infty} \sum_{p=0}^{\infty} [n + (u'/u_1)]C_{np}\lambda^p}{\sum_{n=even}^{\infty} \sum_{p=0}^{\infty} [n + (u'/u_1)]C_{np}\lambda^p}
\]

The power-series expansion for \( \alpha(\lambda) \) is then obtained by dividing the two polynomials. Here \((u'/u_1)\) is the value of the logarithmic derivative for the inner solution.

To compute \( r_0(\lambda) \) and \( A_0^\prime(0,\lambda) \) we need the wave function \( u(x) \) in both regions. The inner solution is known apart from a multiplicative constant \( D \), i.e., \( u_{inner}(x) = Du(x) \) where \( u(x) \) is the solution determined in Sec. II. The outer solution is fully determined once the appropriate value of \( \alpha(\lambda) \) given in Eq. (19) has been introduced in Eq. (17). The value of \( D \), the multiplicative constant for the inner solution, is obtained from the condition \( u_{inner}(x = 1) = u_{outer}(x = 1) \), with the result

\[
D(\lambda) = \left[ 1 + \alpha(\lambda) \sum_{n=even}^{\infty} \sum_{p=0}^{\infty} C_{np}\lambda^p \right] / u(1).
\]

The appropriate values for \( u(1) \) appear in Table I.

Once the wave function is determined Eqs. (12) and (13) allow determination of \( r_0 \) and \( A_0^\prime(0) \) as a function of \( \lambda \).

### Table III. Same as Table II but for liquid \(^3\)He.

<table>
<thead>
<tr>
<th>( n )</th>
<th>( a )</th>
<th>( r_0 )</th>
<th>( A_0^\prime(0) )</th>
<th>( A_1(0) )</th>
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<tr>
<td>0</td>
<td>0.806 900 565 0</td>
<td>5.374 492 91 \times 10^{-1}</td>
<td>-0.179 694 1</td>
<td>0.175 406 906 3</td>
</tr>
<tr>
<td>1</td>
<td>-0.849 747 620 1</td>
<td>3.612 517 23 \times 10^9</td>
<td>2.884 221 3</td>
<td>-1.323 703 661 6</td>
</tr>
<tr>
<td>2</td>
<td>-0.558 881 136 1</td>
<td>1.177 227 91 \times 10^1</td>
<td>1.102 061 1</td>
<td>-0.253 789 382 4</td>
</tr>
<tr>
<td>3</td>
<td>-0.419 611 977 9</td>
<td>3.039 846 80 \times 10^1</td>
<td>0.799 184 5</td>
<td>-0.098 222 448 9</td>
</tr>
<tr>
<td>4</td>
<td>-0.319 638 034 0</td>
<td>7.041 080 11 \times 10^1</td>
<td>0.606 053 7</td>
<td>-0.049 419 813 9</td>
</tr>
<tr>
<td>5</td>
<td>-0.243 984 941 5</td>
<td>1.533 556 16 \times 10^2</td>
<td>0.462 302 8</td>
<td>-0.016 876 522 5</td>
</tr>
<tr>
<td>6</td>
<td>-0.186 296 393 4</td>
<td>3.211 083 93 \times 10^2</td>
<td>0.352 958 0</td>
<td>-0.007 049 440 3</td>
</tr>
<tr>
<td>7</td>
<td>-0.142 254 933 3</td>
<td>6.542 163 75 \times 10^2</td>
<td>0.269 512 5</td>
<td>-0.002 945 970 6</td>
</tr>
<tr>
<td>8</td>
<td>-0.108 625 958 3</td>
<td>1.306 341 63 \times 10^3</td>
<td>0.205 799 4</td>
<td>-0.001 231 248 6</td>
</tr>
<tr>
<td>9</td>
<td>-0.082 946 954 4</td>
<td>5.620 626 64 \times 10^3</td>
<td>0.151 372 8</td>
<td>-0.000 514 603 3</td>
</tr>
<tr>
<td>10</td>
<td>-0.063 338 438 7</td>
<td>4.898 435 51 \times 10^4</td>
<td>0.119 999 0</td>
<td>-0.000 215 080 7</td>
</tr>
<tr>
<td>11</td>
<td>-0.048 365 343 5</td>
<td>9.596 508 95 \times 10^4</td>
<td>0.091 631 4</td>
<td>-0.000 089 894 0</td>
</tr>
<tr>
<td>12</td>
<td>-0.036 931 861 8</td>
<td>1.830 743 45 \times 10^5</td>
<td>0.069 969 9</td>
<td>-0.000 037 571 6</td>
</tr>
<tr>
<td>13</td>
<td>-0.028 201 235 0</td>
<td>3.468 636 11 \times 10^5</td>
<td>0.053 429 2</td>
<td>-0.000 006 563 3</td>
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<tr>
<td>14</td>
<td>-0.021 534 512 9</td>
<td>6.533 513 15 \times 10^5</td>
<td>0.040 798 6</td>
<td>-0.000 006 563 3</td>
</tr>
</tbody>
</table>

For example, the series expansion of $A_0''(0)$ is obtained from
\[
A''(0, \lambda) = -\frac{1}{3} \left[ D(\lambda) \int_0^1 dx \, x^2 u''(x) \right. \\
\left. + \alpha(\lambda) \sum_{n=\text{even}}^\infty \sum_{\rho=0}^n \frac{n(n+1)}{n-2} C_{\alpha \rho} \lambda^\rho \\
\left. + \sum_{n=\text{odd}}^\infty \sum_{\rho=0}^n \frac{n(n+1)}{n-2} C_{\alpha \rho} \lambda^\rho \right].
\]

An analogous algorithm, but slightly more involved, allows the determination of the expansion of $r_0$, as well as the $P$-wave $A_1(0)$ parameter.

The corresponding results for liquid $^4\text{He}$ are shown in Table II, and those of $^3\text{He}$ in Table III. As in the preceding section, the results for $a$ and $r_0$ are computed at machine precision, apart from rounding errors. On the other hand, the coefficients of the expansion of $A_0''(0)$ are less accurate and only 7 or 8 decimals are reliable.

IV. SOME FINAL COMMENTS

In the preceding section we computed algebraically the low-energy scattering parameters for the Lennard-Jones potential, as well as their expansions in terms of the coupling constant of the attractive part of the potential. This has been the basic purpose of this paper.

However, the resulting expansions, particularly in $^4\text{He}$, behave badly. Given that we have not obtained closed expressions for the expansion coefficients, it is not possible to determine the radius of convergence of the expansion. Nevertheless it seems to be small, and most probably much less than 1 (in $\sigma$ units). This suggests carrying out a Padé analysis of the resulting Taylor expansion. The first aim is just to obtain $a$ at $\lambda = 1$ by means of the various $[N/M]$ Padé approximants, and the results shown in Table IV are very gratifying: the $[N/1]$, $[N/2]$, and $[N/3]$ Padés give rather good results for $a$ when $N = 6$.

<table>
<thead>
<tr>
<th>$N$</th>
<th>$0$</th>
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<tr>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<td></td>
<td></td>
<td></td>
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<tr>
<td>1</td>
<td>0.276605</td>
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<td></td>
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<tr>
<td>2</td>
<td>1.205007</td>
<td>-30.861019</td>
<td>-64.959090</td>
<td></td>
</tr>
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<td>8</td>
<td>6.418034</td>
<td>-68.983591</td>
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</table>

These Padé approximants have poles and one is faced with the question of the meaning of these poles. In the case of $[N/1]$ approximants, the pole is located at $\lambda = A_N/A_{N+1}$, where $A_N$ is the $N$th coefficient of the expansion of $a$. A simple calculation using the results of Table II shows that the position of the pole is extremely stable as a function of $N$, the degree of the numerator of the approximants, with the limiting value of 1.013 401 96. The meaning of this pole is well known from the low-energy scattering theory, it is the threshold value for a zero-energy bound state. This approach may hence be a good means of determining the coupling constant of a potential having a zero-energy bound state.

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