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DSMC Collision Model for the Lennard-Jones Potential: Efficient Algorithm and Verification

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An efficient algorithm to implement elastic scattering using the Lennard-Jones (LJ) intermolecular potential in the direct simulation Monte Carlo (DSMC) method is presented. The exact elastic scattering angle for the LJ intermolecular potential obtained by numerical integration is used to construct a piecewise polynomial representation in terms of two collision parameters - the reduced impact parameter and the reduced relative energy. The 5th degree polynomials representation is obtained using the Chebyshev basis. The implementation valid for reduced relative energies ranging from 0.001 to 10.0 is verified by DSMC simulations of subsonic and supersonic Couette flow of Argon at temperatures of 273 K and 40 K and is shown to accurately reproduce the viscosity variation with temperature that corresponds to the LJ intermolecular potential. The LJ collision model is formulated in non-dimensional coordinates and is applicable to arbitrary gas species. For the Couette flow problem, the algorithm is shown to have a computational cost comparable to the variable hard sphere (VHS) model that is widely used in DSMC simulations.

I. Introduction

The direct simulation Monte Carlo (DSMC) technique is a powerful numerical method for solving non-equilibrium gas flow problems encountered in high-altitude aerothermodynamics, vacuum technology and in microsystems. The DSMC technique uses a stochastic approach to solve the Boltzmann equation by simulating the interaction between molecules based on a specified molecular model. First implementations of the DSMC method applied a simple hard-sphere interaction model. The hard sphere model, however, leads to a square-root dependence of viscosity coefficient on temperature which deviates from experimental observations for common gases. A variable hard sphere (VHS) model was proposed by Bird¹ that results in a more general power-law viscosity variation with temperature. The simplified VHS model has its own limitations due to the fact that it does not account for the attractive force between molecules at large distances and assumes infinite repulsive force for close approach. One of the main advantages of the VHS model is that it is easy to implement and not computationally intensive making it a more attractive option than some of the more realistic models that include the attractive force between molecules. In a number of applications the detailed collision dynamics that includes the attractive interactions is important. Examples include flows with large temperature variations such as supersonic plume expansions in vacuum as encountered in space propulsion and in low-pressure deposition of thin film materials.

There have been a number of variations of the VHS model proposed in the past such as the variable soft sphere (VSS),² generalized hard sphere (GHS),³ generalized soft sphere (GSS),⁴ but the parameters of all these models are determined in such a way that they match observed transport properties and not based on fundamental physical principles. Other methods include the μ -DSMC method⁵ that can reproduce arbitrary viscosity variation with temperature. There have also been a number of studies⁶⁻⁹ that dealt with the implementation of various realistic interaction potentials in DSMC simulations. Davis et.al⁶ used a look-up table to obtain deflection angles based on the Morse potential. The Lennard-Jones (LJ) potential interaction was used by Koura and Matsumoto⁷ to investigate the velocity distribution functions within an argon shock wave at free-stream temperatures much lower than the potential well depth of argon. Dimpfl

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et.al⁸ used the Born-Mayer potential with a hard sphere scattering kernel to develop what is referred to as the Extended-VHS (EVHS) model to deal with hypersonic gas flow modeling. More recently, Valentini and Schwartzentruber⁹ performed large scale molecular dynamics simulations to revisit the computation of velocity distribution functions within an argon shock wave. They reported significant differences between the DSMC computations with the VHS model and the molecular dynamics simulations with the LJ potential.

For a large class of problems involving a wide range of temperatures, the transport properties of gases are well represented using an LJ potential. The attractive component of the force between neutral molecules becomes important at low temperatures for most of the common gases and at moderate temperatures for metal vapors that have a reasonably deep potential well. Also, most of the molecular models mentioned earlier derive their parameters by fitting to transport coefficients obtained using the LJ potential. In spite of the accuracy of the LJ potential, one of the main reasons for not using the LJ potential in DSMC simulations is the computational cost involved in computing the elastic scattering using a numerical integration procedure for every collision. The main goal of this paper is to present and perform verifications of a DSMC implementation of the LJ potential interaction between molecules by obtaining the scattering angle as a polynomial approximation of the collision parameters. Such an implementation ensures that the increase in accuracy due to accounting for the attractive component of force is not hampered by a significant increase in computational cost. It should be mentioned that a look-up table approach can be used to implement the LJ potential collisions but is not chosen here because a method that involves computation would be more efficient than a method that involves frequent memory access from a large two-dimensional array as in the case of a look-up table approach.

The remainder of the paper is organized as follows: Section II provides the necessary background about the LJ potential, Section III describes the methodology used to obtain a polynomial approximation for the LJ scattering angle, Section IV presents the results and discussion with the conclusions summarized in Section V.

II. Theory & Background

The LJ potential is one of the intermolecular potentials that accounts for the long range attractive force between neutral molecules and is given by

$$U(r) = 4\epsilon_{LJ} \left[\left(\frac{\sigma_{LJ}}{r} \right)^{12} - \left(\frac{\sigma_{LJ}}{r} \right)^6 \right] \quad (1)$$

where ϵ_{LJ} is the potential well depth and σ_{LJ} corresponds to the distance at which the potential energy becomes zero. The viscosity variation with temperature T , obtained using the LJ potential is given by¹⁰

$$\mu = \frac{5}{8\sigma_{LJ}^2} \sqrt{\frac{mkT}{2\pi}} \frac{V}{W^{(2)}(2)} \quad (2)$$

where m is the molecular mass, k is the Boltzmann's constant, V and $W^{(2)}(2)$ are functions of kT/ϵ_{LJ} tabulated in the work of Hirschfelder et. al.^{10,11} Figure 1 compares the viscosity variation of Argon obtained from experiments,¹² predictions using the LJ potential, the most commonly used VHS model for Argon and a curve fit obtained by Maitland and Smith¹³ using data from a number of experiments. The curve fit is expected to lead to error of less than 1 % in the temperature range 80 - 600 K. As can be seen, the viscosity variation obtained using the LJ potential parameters agrees extremely well with the experiments and the Maitland-Smith curve fit whereas the VHS model's performance improves with increase in temperature. This is mainly due to the fact that the VHS model does not account for the attractive component of the force which becomes important at low temperatures. It should be mentioned that use of a different VHS model in the low temperature regime will lead to good agreement with the experimental data but a single VHS model will never be able to provide an accurate description of the viscosity behavior over a wide range of temperatures.

The single parameter that completely describes an elastic collision between two atoms or molecules by relating the pre-collisional and post-collisional velocities is the scattering angle, χ , which strongly depends

Figure 1. Comparison of viscosity obtained using the VHS model, LJ potential, Maitland-Smith fit,¹³ and experiments by Kestin et.al¹²

on the nature of intermolecular forces between the two molecules. χ depends on the relative energies of the two colliding molecules (ϵ) and the impact parameter (b). For the LJ potential, the scattering angle χ is given by⁷

$$\chi = \pi - 2\sqrt{1 + cz - (1 + c)z^2} \int_0^1 \{1 - [1 + cz - (1 + c)z^2]u^2 + czu^6 - (1 + c)z^2u^{12}\}^{-1/2} du \quad (3)$$

where $c = (2/\epsilon^*)[1 + \sqrt{1 + \epsilon^*}]$, $z = (4/c\epsilon^*)(\sigma_{LJ}/r_0)^6$, and $u = r_0/r$ where r_0 is the distance of closest approach between the two molecules. Here $\epsilon^* = \epsilon/\epsilon_{LJ}$ is the reduced relative energy and $b^* = b/\sigma_{LJ}$ is referred to as the reduced impact parameter. Varying u from 0 to 1 varies r from ∞ to r_0 . z in the above integral depends on b^* and ϵ^* and is obtained by solving the implicit equation

$$b^* = \left(\frac{4}{c\epsilon^*}\right)^{1/6} \sqrt{1 + cz - (1 + c)z^2} z^{-1/6} \quad (4)$$

For a given value of b^* and ϵ^* , Eq. (4) is solved using a bisection method and the solution for z is used to compute χ by numerical integration of Eq. (3). The numerical integration is performed using the Gauss-Chebyshev quadrature method. The integral in Eq.(3) is written as

$$\int_0^1 I(u)du = \sum_{k=1}^M \frac{1}{2} w_k I\left(\frac{y_k + 1}{2}\right) \sqrt{1 - y_k^2} \quad (5)$$

where w_k are the weights for Gauss-Chebyshev quadrature, y_k are the zeros of the M^{th} degree Chebyshev polynomial ϕ_M and I is the integrand in Eq.(3) given by

$$I(u) = \{1 - [1 + cz - (1 + c)z^2]u^2 + czu^6 - (1 + c)z^2u^{12}\}^{-1/2} \quad (6)$$

The zeros, y_k , of ϕ_M are given by

$$y_k = \cos\left(\frac{(2k + 1)\pi}{2M}\right) \quad (7)$$

and the weights are all equal and are given by

$$w_k = \frac{\pi}{M} \quad (8)$$

For results presented in this work, the value of M was used as 200 with further increase in the number of quadrature points leading to negligible difference in the computed value of χ . It should be mentioned that any other numerical integration scheme such as Gauss-Legendre could have been used and would have given the same result as long as sufficient number of points was used.

The scattering angle for the widely used VHS model is given by¹

$$\chi = 2 \cos^{-1} \left(\frac{b}{d} \right) \quad (9)$$

where d the diameter varies with the relative velocity between the colliding molecules as¹

$$d = d_{\text{ref}} (c_{r,\text{ref}}/c_r)^{(\omega-0.5)} \quad (10)$$

Since there is no general relation between the parameters of the LJ potential and the VHS model, comparisons of the scattering angle for the two cases can be made only for a given gas. Figure 2 shows contours of the scattering angle obtained using the VHS model and the LJ potential computed using parameters for Argon. The relevant parameters were $\sigma_{LJ} = 3.405 \times 10^{-10} \text{ m}$, $\epsilon_{LJ} = 0.0103 \text{ eV}$, $d_{\text{ref}} = 4.17 \times 10^{-10} \text{ m}$ at 273 K , and $\omega = 0.81$. As can be observed, there are significant differences for a range of b and ϵ values. The differences are pronounced for low energy collisions as can be observed from the contour lines close to the x-axis and also for long-range collisions corresponding to values of $b > d$. Figure 3 shows the variation of χ as a function of b^* for four different values of ϵ^* in order to compare the LJ potential and VHS model scattering angles more rigorously. Since the VHS model is a purely repulsive model, negative values of scattering angle are not obtained while the LJ potential has a shallow well for χ even at relative energies of $\epsilon^* = 5.0$.

The mean translation energy of collisions in the center of mass frame for VHS model is given by Bird¹ as

$$\bar{E}_t = (2.50 - \omega)kT. \quad (11)$$

For a temperature of 273 K , the mean translation energy of collisions is 0.0397 eV and corresponds to $\epsilon^* = 3.86$ for Argon. On the other hand, for a temperature of 40 K , the mean translation energy of collisions is 0.0058 eV corresponding to $\epsilon^* = 0.57$. The effect of negative scattering angles due to the attractive component of the force between molecules will be more important at 40 K when compared to 273 K .

Figure 2. Comparison of scattering angle contours for the LJ potential and the VHS model.

Figure 3. Variation of χ as a function of b^* for various values of ϵ^* .

III. Polynomial Approximation of Lennard-Jones Scattering Angle

As described earlier, this work aims to construct a polynomial approximation of the scattering angle, χ , in terms of the reduced impact parameter (b^*) and reduced relative energy (ϵ^*) for use in DSMC simulations. In order to construct the polynomial approximation of the LJ scattering angle, the Chebyshev polynomials are chosen as the basis functions though any other set of basis functions will work equally well. The Chebyshev polynomials are defined in the domain $[-1, 1]$ and since both b^* and ϵ^* take a different range of values, a transformation that converts $b^* \in [b_{\min}^*, b_{\max}^*]$ to $\hat{b}^* \in [-1, 1]$ and $\epsilon^* \in [\epsilon_{\min}^*, \epsilon_{\max}^*]$ to $\hat{\epsilon}^* \in [-1, 1]$ is performed. The transformations are given by

$$\hat{b}^* = \frac{2b^* - (b_{\min}^* + b_{\max}^*)}{b_{\max}^* - b_{\min}^*} \quad (12)$$

$$\hat{\epsilon}^* = \frac{2\epsilon^* - (\epsilon_{\min}^* + \epsilon_{\max}^*)}{\epsilon_{\max}^* - \epsilon_{\min}^*} \quad (13)$$

Therefore, the scattering angle will be represented as

$$\chi = \sum_{i=0}^N \sum_{j=0}^{N-i} \hat{\chi}_{ij} \phi_i(\hat{b}^*) \phi_j(\hat{\epsilon}^*) \quad (14)$$

where N is the order of the approximation, ϕ_i is the Chebyshev polynomial of degree i and $\hat{\chi}_{ij}$ are the coefficients. In this work, the value of N is chosen as 5 and is shown to be adequate to accurately represent the LJ potential scattering angle. The coefficients $\hat{\chi}_{ij}$ are obtained using the orthogonality of the basis functions and are given by

$$\hat{\chi}_{ij} = \int_{-1}^1 \int_{-1}^1 \chi(\hat{b}^*) \phi_j(\hat{\epsilon}^*) \frac{1}{\sqrt{1-\hat{b}^{*2}}} \frac{1}{\sqrt{1-\hat{\epsilon}^{*2}}} d\hat{b}^* d\hat{\epsilon}^* / \|\phi_i\|^2 \|\phi_j\|^2 \quad (15)$$

The above integration is performed using multi-variate Gauss-Chebyshev numerical integration using a Smolyak sparse grid as described in detail by Gerstner et.al¹⁴ and the above integral can be written as

$$\hat{\chi}_{ij} = \sum_{k=L}^{L+1} \sum_{k_1=1}^k \sum_{i_1=1}^{k_1} \sum_{i_2=1}^{k-k_1} (-1)^{l+1-k} \chi_{k_1} w_{i_1} w_{i_2} \phi_i(y_{i_1}) \phi_j(y_{i_2}) / \|\phi_i\|^2 \|\phi_j\|^2 \quad (16)$$

where $_{k_1} w_{i_1}$ and $_{k_1} y_{i_1}$ refer to i_1^{th} weight and zero respectively of the Chebyshev polynomial of degree k_1 . L is the order of the Gauss-Chebyshev integration and for results obtained in this work, $L = 80$ was used. $\|\phi_i\|^2$ can be evaluated analytically and is given by

$$\|\phi_i\|^2 = \begin{cases} \pi & \text{if } i = 0; \\ \frac{\pi}{2} & \text{if } i \neq 0. \end{cases} \quad (17)$$

The value of χ in Eq.(15) is evaluated at b^* and ϵ^* corresponding to $\hat{b}^* = _{k_1} y_{i_1}$ and $\hat{\epsilon}^* = _{k_2} y_{i_2}$ using the numerical integration described in Section II.

However, obtaining a single set of coefficients $\hat{\chi}_{ij}$ and hence one accurate polynomial representation in the entire domain of interest is not possible due to the presence of the region of orbiting collisions where the value of χ diverges to $-\infty$. Since orbiting collisions are not included in the collisions computed in DSMC, it is not necessary to obtain polynomial approximations for the scattering angle in the region of orbiting collisions. Also, even in regions of non-orbiting collisions it can be seen that the form of χ is significantly different for $\epsilon^* < 0.8$ and for $\epsilon^* > 0.8$. For $\epsilon^* < 0.8$, the presence of a region of orbiting collisions results in χ decreasing steeply as b^* tends to the value at which orbiting collisions begin. Therefore, we construct a piecewise polynomial approximation for $\chi(b^*, \epsilon^*)$ by dividing the $b^* - \epsilon^*$ domain into four regions the boundaries of which are described below.

For a given value of ϵ^* , the range of values of interest of b^* is divided into two regions $[b_{\min}^*, b_1^*(\epsilon^*)]$ and $[b_2^*(\epsilon^*), B^*]$. Here, $b_1^*(\epsilon^*)$ is the minimum value of b^* for which $\chi(b_1^*, \epsilon^*) \rightarrow -\infty$ and $b_2^*(\epsilon^*)$ is the maximum value of b^* for which $\chi(b_2^*, \epsilon^*) \rightarrow -\infty$. The above definition would yield values of $b_1^*(\epsilon^*)$ and $b_2^*(\epsilon^*)$ only for $\epsilon^* < 0.8$ since there are no orbiting collisions for $\epsilon^* > 0.8$. Therefore, for $\epsilon^* > 0.8$, the above definitions of b_1^* and b_2^* are slightly modified as follows. For $\epsilon^* > 0.8$, $b_1^*(\epsilon^*) = b_2^*(\epsilon^*)$ is defined as the value of b^* for which $\chi(b^*, \epsilon^*)$ reaches a minimum value.

The values of $b_1^*(\epsilon^*)$ and $b_2^*(\epsilon^*)$ can be obtained by using the above definitions for any given value of ϵ^* . However, for computing these values during every collision in a DSMC simulation, polynomial approximations of these boundaries are required. These approximations are again based on Chebyshev polynomials and since the Chebyshev polynomials are defined in the domain $[-1, 1]$, a transformation from $\epsilon^* \in [\epsilon_{\min}, \epsilon_{\max}]$ is required. The polynomial approximation is given by

$$b_1^* = \sum_{i=0}^{N_{b_1}} \hat{b}_{1i} \phi_i(\tilde{\epsilon}^*) \quad (18)$$

The above expression can be modified by expanding the Chebyshev polynomials and grouping terms to obtain

$$b_1^* = \sum_{i=0}^{N_{b_1}} \tilde{b}_{1i} \tilde{\epsilon}^{*i} \quad (19)$$

The values of both $b_1^*(\epsilon^*)$ and $b_2^*(\epsilon^*)$ vary rapidly for small values of ϵ^* and hence, in order to obtain an accurate representation of $b_1^*(\epsilon^*)$ using a reasonable value for N_{b_1} , the range of ϵ^* is divided into 4 different regions with different values for the coefficients $\tilde{b}_{1i}$. It was observed that using $N_{b_1} = 9$ was sufficient to represent b_1^* . The transformation from ϵ^* to $\tilde{\epsilon}^*$ also depends on the actual value of ϵ^* and the functional form of the transformation is presented below.

$$\begin{aligned}
\tilde{\epsilon}^* &= \frac{2\epsilon^* - 0.011}{0.009} & \epsilon^* \in [0.001, 0.01] \\
&= \frac{2\epsilon^* - 0.21}{0.19} & \epsilon^* \in (0.01, 0.2] \\
&= \frac{2\epsilon^* - 1}{0.6} & \epsilon^* \in (0.2, 0.8] \\
&= \frac{2\epsilon^* - 10.8}{9.2} & \epsilon^* \in (0.8, 10]
\end{aligned}$$

It can be seen that the above transformations are of the form

$$\tilde{\epsilon}^* = \frac{2\epsilon^* - (\epsilon_{min}^* + \epsilon_{max}^*)}{\epsilon_{max}^* - \epsilon_{min}^*} \quad (20)$$

for each of the 4 regions. The coefficients $\hat{b}_{1i}$ are obtained using the orthogonal property of Chebyshev polynomials and is given by

$$\hat{b}_{1i} = \int_{-1}^1 b_1^* \phi_i(\tilde{\epsilon}^*) \frac{1}{\sqrt{1 - \tilde{\epsilon}^{*2}}} d\tilde{\epsilon}^* / \|\phi_i\|^2 \quad (21)$$

The above integration is performed using Gauss-Chebyshev quadrature and the final expression for $\hat{b}_{1i}$ is given by

$$\hat{b}_{1i} = \sum_{k=1}^M b_1^* \phi_i(y_k) w_k / \|\phi_i\|^2 \quad (22)$$

where y_k and w_k are the zeros and weights as defined in Section II. Once the integration is performed, the coefficients $\hat{b}_{1i}$ are converted to $\tilde{b}_{1i}$ by expanding the Chebyshev polynomials and are summarized in Table 1

Table 1. Coefficients $\tilde{b}_{1i}^*$ for computing b_1^* using Eq.(19)

Region \ i	0	1	2	3	4	5	6	7	8	9
$\epsilon^* \in [0.001, 0.01]$	4.1224	-0.5639	0.2543	-0.1340	0.2010	-0.1914	-0.1752	0.1932	0.2014	-0.1769
$\epsilon^* \in (0.01, 0.2]$	2.5172	-0.3920	0.1476	-0.0012	0.4577	-0.6883	-0.7430	1.05	0.6109	-0.7050
$\epsilon^* \in (0.2, 0.8]$	1.9200	-0.2097	0.0656	-0.0310	0.0152	-0.0061	0.0007	-0.0037	0.0028	-0.0001
$\epsilon^* \in (0.8, 10.0]$	1.2967	-0.0821	0.0228	-0.0446	0.3270	-0.2403	-0.5108	0.4464	0.3660	-0.3339

A similar representation is obtained for $b_2^*(\epsilon^*)$ and is given by

$$b_2^* = \sum_{i=0}^{N_{b_2}} \tilde{b}_{2i} \tilde{\epsilon}^{*i} \quad (23)$$

with the coefficients for $N_{b_2} = 9$ presented in Table 2. The definition of $\tilde{\epsilon}^*$ for computing b_2^* is same as that used to compute b_1^* . It should be remembered that, for $\epsilon^* > 0.8$, $b_1^*(\epsilon^*)$ and $b_2^*(\epsilon^*)$ are the same. Figure III shows the polynomial approximation of $b_1^*(\epsilon^*)$ and $b_2^*(\epsilon^*)$ along with $B^*(\epsilon^*)$ which corresponds to the maximum value of b^* for which $\chi(b^*, \epsilon^*) = 0.1$ and can be obtained using small-angle approximation⁷ as

$$B^* = \max \left[\left(\frac{0.4\pi}{\epsilon^*} \right)^{1/6}, \left(\frac{0.6\pi}{\epsilon^*} \right)^{1/12} \right] \quad (24)$$

As mentioned earlier, the piecewise polynomial approximation for $\chi(b^*, \epsilon^*)$ is obtained in four different domains with different values of coefficients $\hat{\chi}_{ij}$ in each domain. The four different domains are given by

Table 2. Coefficients $\tilde{b}_{2_i}^*$ for computing b_2^* using Eq.(23)

Region \ i	0	1	2	3	4	5	6	7	8	9
$\epsilon^* \in [0.001, 0.01]$	15.4308	-6.3136	3.4828	-1.8501	4.5174	-5.2192	-5.0253	6.3418	5.3530	-5.2582
$\epsilon^* \in (0.01, 0.2]$	3.7283	-1.6027	0.4159	0.8131	4.9915	-8.1469	-9.0145	13.0152	6.9851	-8.3878
$\epsilon^* \in (0.2, 0.8]$	1.9877	-0.3913	0.2676	-0.0054	-0.0961	-0.3098	0.1862	0.3236	-0.0631	-0.1444

$$\text{Domain 1 (non - glancing, slow)} : [0, 0.98b_1^*(\epsilon^*)] \times [0.001, 0.8] \quad (25)$$

$$\text{Domain 2 (non - glancing, fast)} : [0, 0.98b_1^*(\epsilon^*)] \times [0.8, 10.0] \quad (26)$$

$$\text{Domain 3 (glancing, slow)} : [1.02b_2^*(\epsilon^*), B^*(\epsilon^*)] \times [0.2, 0.8] \quad (27)$$

$$\text{Domain 4 (glancing, fast)} : [1.02b_2^*(\epsilon^*), B^*(\epsilon^*)] \times (0.8, 10.0] \quad (28)$$

It can be observed that the four domains described above do not include $0.98b_1^*(\epsilon^*) < b^* \leq b_1^*(\epsilon^*)$ and $b_2^*(\epsilon^*) \leq b^* < 1.02b_2^*(\epsilon^*)$. For the range $0.98b_1^*(\epsilon^*) < b^* \leq b_1^*(\epsilon^*)$, $\chi(b^*, \epsilon^*) = \chi(0.98b_1^*, \epsilon^*)$ and for the range $b_2^*(\epsilon^*) \leq b^* < 1.02b_2^*(\epsilon^*)$, $\chi(b^*, \epsilon^*) = \chi(1.02b_2^*, \epsilon^*)$. The coefficients obtained for the four domains in the $b^* - \epsilon^*$ are summarized in Tables 3-6. The coefficients corresponding to a given region are used in Eq.(14) to obtain the value of $\chi(b^*, \epsilon^*)$. The values of $\hat{b}^*$ and $\hat{\epsilon}^*$ are obtained from b^* and ϵ^* using domain dependent transformations of the form

$$\hat{b}^* = \frac{2b^* - (b_{\max}^* + b_{\min}^*)}{b_{\max}^* - b_{\min}^*} \quad (29)$$

$$\hat{\epsilon}^* = \frac{2\epsilon^* - (\epsilon_{\max}^* + \epsilon_{\min}^*)}{\epsilon_{\max}^* - \epsilon_{\min}^*} \quad (30)$$

The specific transformations for each domain are obtained by using the relevant $b_{\max}^*$, $b_{\min}^*$, $\epsilon_{\max}^*$ and $\epsilon_{\min}^*$. For example, the transformation for domain 1 would be

$$\hat{b}^* = \frac{2b^* - 0.98b_1^*(\epsilon^*)}{0.98b_1^*(\epsilon^*)} \quad (31)$$

$$\hat{\epsilon}^* = \frac{2\epsilon^* - (0.8 + 0.001)}{0.8 - 0.001} \quad (32)$$

In order to compare the values of χ obtained using the piecewise polynomial approximation and the exact values obtained using numerical integration, the variation of χ as a function of b^* is compared for four different values of ϵ^* . As can be seen in Figure 5, there is very good agreement between the exact scattering angles computed by numerical integration and those obtained using the polynomial approximation. The only regions where the agreement is not so great is in the vicinity of the orbiting boundary which forms a very small fraction of the collision cross-section $\pi B^{*2} \sigma_{LJ}^2$. The algorithm to compute the scattering angle given b^* and ϵ^* is summarized below in Algorithm 1

Algorithm 1 Algorithm to compute the value of LJ scattering angle for given b^* and ϵ^* using the polynomial approximation

- 1: Compute $b_1^*(\epsilon^*)$ and $b_2^*(\epsilon^*)$ using Eq.(19) and Eq.(23) with coefficients from Tables 1 and 2.
 - 2: Find the domain in which (b^*, ϵ^*) lies with the domains as defined in Eq.(25).
 - 3: Compute $\hat{b}^*$ and $\hat{\epsilon}^*$ using the transformation (Eq.(29)) relevant to the domain in which (b^*, ϵ^*) lies.
 - 4: Compute χ using Eq.(14) and coefficients $\hat{\chi}_{ij}$ from one of the Tables 3, 4, 5 and 6.
-

IV. Results & Discussion

The polynomial approximation obtained in Section III for the LJ scattering angle is verified by using it in DSMC simulations of compressible subsonic and supersonic Couette flow problems similar to those used

Figure 4. Polynomial approximations of b_1^* , and b_2^* compared with B^{*7} obtained using the small angle approximation

Table 3. Coefficients for computing χ in domain 1: $[0, 0.98b_1^*(\epsilon^*)] \times [0.001, 0.8]$

$i \backslash j$	0	1	2	3	4	5
0	1.27435	0.08774	-0.01967	-0.02160	-0.02278	0.05274
1	-2.27964	0.10974	-0.06034	0.05866	-0.01056	
2	0.15218	0.09175	0.09955	0.00496		
3	0.84119	-0.07042	0.10986			
4	-1.49058	-0.12341				
5	-1.79968					

Table 4. Coefficients for computing χ in domain 2: $[0, 0.98b_1^*(\epsilon^*)] \times [0.8, 10.0]$

$i \backslash j$	0	1	2	3	4	5
0	1.55921	0.13722	0.26211	-0.39017	-0.30944	0.35409
1	-1.89659	-0.38254	0.62989	0.41053	-0.46827	
2	-0.17430	-0.54077	-0.38777	0.44926		
3	0.01434	0.50240	-0.51879			
4	-0.03552	0.59225				
5	0.03425					

by Bird¹ to verify the VHS model and Macrossan⁵ to verify the μ -DSMC technique. The DSMC method itself follows the work of Koura and Matsumoto⁷ very closely with the only change being the use of the polynomial approximation for the scattering angle instead of their quadrature method. For case 1 of the two verification cases, the wall temperature was chosen as $T_{\text{wall}} = 273$ K and the initial number density was fixed at $n_1 = 1.4 \times 10^{20} \text{ 1/m}^3$. The velocity of the moving wall was 300 m/s corresponding to a Mach number of $M = 0.97$. The Knudsen number, Kn , of the flow can be defined based on the mean free path λ of the initial state using the mean free path theory¹⁵ and is given by

$$\lambda = \frac{2\mu}{\rho \bar{c}} \quad (33)$$

Table 5. Coefficients for computing χ in domain 3: $[1.02b_2^*(\epsilon^*), b_{\max}^*(\epsilon^*)] \times [0.2, 0.8]$

i \ j	0	1	2	3	4	5
0	-0.22763	-0.07889	0.06166	0.03669	-0.01667	-0.03408
1	0.21366	0.12409	-0.11916	-0.04256	0.02422	
2	-0.03000	-0.08398	-0.05294	0.05609		
3	-0.04152	0.18752	0.08937			
4	-0.11813	-0.1744				
5	0.10782					

Table 6. Coefficients for computing χ in domain 4: $[1.02b_2^*(\epsilon^*), b_{\max}^*(\epsilon^*)] \times (0.8, 10.0]$

i \ j	0	1	2	3	4	5
0	-0.20896	0.02696	-0.09538	0.31894	0.12584	-0.31161
1	0.22267	-0.06108	0.05057	-0.03972	-0.17980	
2	0.05609	-0.04844	-0.25860	0.15113		
3	-0.13698	-0.21699	0.30747			
4	-0.08635	0.18538				
5	0.07488					

where ρ is the initial density and $\bar{c}$ is the mean thermal velocity based on wall temperature. The $Kn = \lambda/L$ for the conditions mentioned earlier is 0.012 and is in the slip regime. The domain was divided into 500 cells which ensures that the cell size is much smaller than the mean free path. The number of particles used was fixed at 10000 and the timestep was chosen as 1×10^{-6} s. Figure 6 compares the variation of normalized density and temperature obtained using the VHS model and the LJ polynomial approximation obtained in this work. The only non-zero shear stress component for the Couette flow is $\tau_{xy} = \tau_{yx}$ which is computed directly by the DSMC simulation as

$$\tau_{DSMC} = \langle \rho c'_x c'_y \rangle \quad (34)$$

where $\langle \rho c'_x c'_y \rangle$ refers to the average value of $\rho c'_x c'_y$. Figure 7(a) compares the shear stress variation in the gap (supposed to be constant across the gap for the Couette flow) obtained using the VHS model and the LJ polynomial approximation and the agreement is excellent once again. Since the viscosity obtained using VHS model and LJ potential agree very well at 273 K there is very little difference in the results obtained using the two models. The average values of shear stress obtained using the VHS model and LJ polynomial approximation are 6.4211×10^{-3} N/m² and 6.3016×10^{-3} N/m² which corresponds to a difference of less than 2 %. In order to verify that the LJ polynomial approximation is good, the viscosity obtained from DSMC simulations is compared to the theoretical viscosity value obtained using the LJ intermolecular potential at the local temperature, T_{DSMC} . The DSMC viscosity is obtained as

$$\mu_{DSMC} = \frac{\tau_{DSMC}}{(dv/dx)_{DSMC}} \quad (35)$$

The agreement between the theoretical LJ viscosity, μ_{LJ} and the DSMC computed viscosity, μ_{DSMC} is very good as can be seen in Figure 7(b). The higher noise level in μ_{DSMC} is because it is obtained using a numerical derivative obtained using DSMC simulations that have some scatter and this scatter is amplified in the numerical derivative $(dv/dx)_{DSMC}$. The above comparison was repeated for case 2 which corresponds to a much lower temperature of 40 K at which differences are expected between the VHS model and the LJ potential. The initial number density of the flow was fixed at $n_2 = 1.4 \times 10^{20}$ 1/m³. The velocity of the moving wall was fixed at 300 m/s corresponding to $M = 2.55$ and hence a significant region of the flow in the gap was supersonic as opposed to the previous case in which the flow never became supersonic in the entire gap. The Kn obtained using the mean free path obtained from Eq.(33) is 0.0051. The flow conditions for both cases are summarized in Table 7.

Figure 5. Comparison of variation of χ as a function of b^* for various values of ϵ^* obtained using the polynomial approximation and the exact numerical integration.

Figure 8 shows a comparison of the temperature and density variation in the gap obtained using the VHS model and the LJ polynomial approximation with no discernible difference in the temperature and density contours. Figure 9(a) compares the shear stress variation in the gap obtained using the two models and the higher VHS viscosity at 40 K leads to higher shear stress value in the gap. The average shear stress obtained using the VHS model and the LJ polynomial approximation are $1.1820 \times 10^{-3} \text{ N/m}^2$ and $1.5956 \times 10^{-3} \text{ N/m}^2$ respectively. Figure 9(b) compares μ_{LJ} with μ_{DSMC} for the supersonic Couette flow and there is excellent agreement within statistical scatter. Figure 10 compares the mean reduced relative energy of collision ($\langle \epsilon^* \rangle$) in the gap for cases 1 and 2. The value of ϵ^* was obtained using Eq.(11) and the local temperature in the gap. As expected, the negative scattering angles due to the attractive component is more important for case 2 where $\langle \epsilon^* \rangle \sim 0.7$.

One of the important aspects to be considered in order to evaluate the applicability of this method is the computational overhead associated with the polynomial computations. The use of pre-computed scattering angles makes the implementation very efficient with insignificant overhead as compared to the VHS model. For the subsonic compressible Couette flow problem, the time taken for 190,020 sampling timesteps using the VHS model was 1910.8 s whereas for the LJ potential polynomial approximation, the time taken was 2701.89 s which is about 41 % higher than the time taken for the VHS model. However, it should be mentioned that most of this increase is contributed by the higher number of collision events due to the long-range nature of the LJ potential. The number of collision events computed for the VHS model was about 161 million whereas for the LJ polynomial approximation, the number of collision events was 379 million which is more than 100 % higher than the number of collision events computed by the VHS model. Therefore,

Table 7. Summary of flow conditions used for the subsonic and supersonic Couette flow problems

	Case 1	Case 2
Wall Temperature, T_{wall} (K)	273	40
Initial Number Density, n ($1/m^3$)	1.4×10^{20}	1.4×10^{20}
Moving Wall Velocity, v_{wall} (m/s)	300	300
Mach number, M	0.97	2.55
Knudsen number, Kn	0.012	0.0051
$\langle \epsilon^* \rangle$ based on wall temperature	3.86	0.57

Figure 6. Comparison of normalized density and temperature variation in the gap obtained using VHS model and the LJ polynomial approximation for subsonic compressible Couette flow of Argon **Figure 7. Comparison of shear stress and viscosity variation in the gap obtained using various methods for subsonic compressible Couette flow of Argon**

Figure 8. Comparison of normalized density and temperature variation in the gap obtained using VHS model and the LJ polynomial approximation for supersonic Couette flow of Argon

Figure 9. Comparison of shear stress and viscosity variation in the gap obtained using various methods for supersonic Couette flow of Argon

Figure 10. Comparison of mean reduced relative energy for cases 1 and 2

the proposed LJ collision model using the polynomial approximation can be applied without significant additional computational cost.

V. Conclusions

An efficient algorithm to compute elastic collisions in DSMC using the Lennard-Jones intermolecular potential was presented. The theory of polynomial approximations was used to represent the scattering angle for the LJ potential using a two-variable polynomial of degree 5. The polynomial approximation of the scattering angle was verified by applying it to subsonic and supersonic Couette flows of Argon. The results obtained using the LJ polynomial approximation were compared with those obtained using the VHS model. The agreement between the two models is within 2% at a temperature of 273 K. On the other hand, at a temperature of 40 K the higher viscosity of the VHS model leads to a shear stress that is about 35 % higher than the LJ shear stress due to the importance of the attractive component of the force between molecules. The DSMC simulations performed using the LJ polynomial approximation were shown to reproduce the theoretical LJ potential viscosity at both 40 K and 273 K. The LJ implementation presented is straightforward to implement in any existing DSMC solver and leads to insignificant increase in computational cost when compared to the VHS model.

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