A Kinetic Model for Fluid-Wall Interaction

Aldo Frezzotti and Livio Gibelli

Dipartimento di Matematica del Politecnico di Milano

Piazza Leonardo da Vinci 32 - 20133 Milano, Italy

July 11th 2007

Summary

- Motivation

- Kinetic model structure and basic equations.

- Application to test problems and comparison with MD results

- Concluding remarks

Motivation

☞ The traditional approach to gas-surface interaction modeling is based on phenomenological scattering kernels in the form (Maxwell, Cercignani-Lampis...)

$$(\boldsymbol{v} \circ \boldsymbol{n}) f(\boldsymbol{r}, \boldsymbol{v}|t) = \int_{\boldsymbol{v}_1 \circ \boldsymbol{n} < 0} K_w(\boldsymbol{v}_1 \to \boldsymbol{v}) |\boldsymbol{v}_1 \circ \boldsymbol{n}| f(\boldsymbol{r}, \boldsymbol{v}_1|t) \, d\boldsymbol{v}_1 \qquad \boldsymbol{v} \circ \boldsymbol{n} > 0 \qquad (1)$$

- Advantages: theoretical and computational simplicity

- Disadvantages: they contain a few free parametes (typically accommodation coefficients). Their relationships with fundamental interaction properties (intermolecular potentials) can be determined with great difficulty. More often they are fitted to match experimental results, but tuning has to be repeated if physical conditions change.

☞ Modeling gas-surface interaction by solving the microscopic dynamics of wall and gas atoms/molecules is possible by Molecular Dynamics (MD) techniques.

- Advantages: model is based on fundamental interaction properties.

- Disadvantages: only numerical approach is possible; computationally expensive

A Kinetic Model ?

☞ Computational efforts can be reduced by hybrid simulation methods in which DSMC is used to describe collisions in the gas phase whereas MD use is limited to describe interactions among wall atoms/molecules and interactions between wall and gas atoms/molecules [Yamamoto (2006), Nedea et al. (2006)].

• For rarefied gas flows hybrid methods are still quite demanding in terms of computational resources.

☞ An alternative approach to fluid-wall interaction can be based on the use of a kinetic equation which describes "collisions" between fluid and wall atoms/molecules.

• Advantages: based on fundamental interaction properties, solved by DSMC (no hybrid method is necessary), approximate solutions possible.

• Disadvantages: complex mathematical structure, numerical treatment necessary.

☞ **The idea is not new**

1. Cercignani (1988) has shown that Cercignani-Lampis model can be derived by modeling the motion of gas molecules adsorbed into the solid matrix by a Fokker-Planck equation.

2. More recently, Pozhar and Gubbins (1991,1993) have proposed a rather general kinetic formulation of non-uniform dense fluids in which the fluid-wall interaction is described by a collision integral.

☞ **However**

1. The Fokker-Planck equation does not take into account non-local effects of molecular interaction which play a very important role in the vicinity of the solid boundary.

2. Non-local effects are correctly included in the treatment described by Pozhar and Gubbins, but wall molecules are given an infinite mass thus limiting the model capabilities in predicting the correct momentum and energy exchanges between the wall and the fluid phase.

Kinetic Model Structure and Basic Equations

- We consider a monatomic gas whose atoms have mass m_1

- The gas phase interacts with walls whose atoms have mass m_2

- It is assumed that atoms in the gas and solid phases interact pairwise through a potential $\phi_{12}(\rho)$ in the form

$$\phi_{12}(\rho) = \begin{cases} +\infty & \rho < \sigma_{12} \\ -\overline{\phi}_{12}\left(\frac{\rho}{\sigma_{12}}\right)^{-\gamma_{12}} & \rho \geq \sigma_{12} \end{cases} \tag{2}$$

- $\phi_{12}(\rho)$ depends only on the distance ρ between the centers of two interacting atoms and it results from the superposition of a repulsive hard sphere potential and a soft tail.

- The hard sphere diameter is defined as $\sigma_{12} = \frac{\sigma_1 + \sigma_2}{2}$, being σ_1 and σ_2 the hard sphere diameters of gas and wall atoms, respectively.

- The behavior of the soft potential tail is determined by the right limit of $\phi_{12}(\rho)$ at $\rho = \sigma_{12}$, $-\overline{\phi}_{12}$, and by the value of the positive exponent γ_{12}.

Neglecting atomic interactions in the gas phase, the following equation holds

$$
\frac{\partial f_1}{\partial t} + \boldsymbol{v}_1 \circ \nabla_{\boldsymbol{r}_1} f_1 = \nabla_{\boldsymbol{v}_1} \circ \left[\int_{\rho > \sigma_{12}} \hat{\boldsymbol{k}} \frac{d\phi_{12}}{d\rho} f_{12}(\boldsymbol{r}_1, \boldsymbol{v}_1, \boldsymbol{r}_2, \boldsymbol{v}_2 | t) \, d\boldsymbol{r}_2 \, d\boldsymbol{v}_2 \right] +
$$

$$
\sigma_{12}^2 \int \left[f_{12}(\boldsymbol{r}_1, \boldsymbol{v}_1^*, \boldsymbol{r}_1 + \sigma_{12}\hat{\boldsymbol{k}}, \boldsymbol{v}_2^* | t) - f_{12}(\boldsymbol{r}_1, \boldsymbol{v}_1, \boldsymbol{r}_1 - \sigma_{12}\hat{\boldsymbol{k}}, \boldsymbol{v}_2 | t) \right] (\boldsymbol{v}_r \circ \hat{\boldsymbol{k}})^+ \, d\boldsymbol{v}_2 \, d^2\hat{\boldsymbol{k}}
$$

$$(3)$$

- $f_1(\boldsymbol{r}_1, \boldsymbol{v}_1 | t)$ denotes the one-particle distribution function of velocities $\boldsymbol{v}_1$ of gas atoms at spatial location $\boldsymbol{r}_1$ at time t.

- $f_{12}(\boldsymbol{r}_1, \boldsymbol{v}_1, \boldsymbol{r}_2, \boldsymbol{v}_2 | t)$ is the distribution function of pairs formed by one gas atom, having position $\boldsymbol{r}_1$ and velocity $\boldsymbol{v}_1$, and one wall atom located at $\boldsymbol{r}_2$ with velocity $\boldsymbol{v}_2$.

- The first integral at $r.h.s$ of Eq. (3) represents the soft tail contribution to the rate of change of f_1, being $\rho = \|\boldsymbol{r}_2 - \boldsymbol{r}_1\|$ and $\hat{\boldsymbol{k}}$ the unit vector $\frac{\boldsymbol{r}_1 - \boldsymbol{r}_2}{\rho}$.

- The contribution of hard collisions is given by the second integral where $\boldsymbol{v}_1^*$ and $\boldsymbol{v}_2^*$ are the post-collisional velocity vectors in a hard sphere impact, $\boldsymbol{v}_r$ is the relative velocity $\boldsymbol{v}_2 - \boldsymbol{v}_1$. The integral over $\hat{\boldsymbol{k}}$ is limited to the hemisphere where the condition $\boldsymbol{v}_r \circ \hat{\boldsymbol{k}} > 0$ holds.

☞ Within the limits of the assumptions described above, Eq.(3) is exact but not closed, since it also involves the pair distribution function $f_{12}(\boldsymbol{r}_1, \boldsymbol{v}_1, \boldsymbol{r}_2, \boldsymbol{v}_2|t)$.

• Closure is obtained by the following assumptions on pair correlations:

1. The first term at *r.h.s* of Eq. (3) gives the the total force exterted by wall atoms on gas atoms when the interatomic distance is larger than σ_{12}. Since spatial correlations are expected to decay rapidly, it is not unreasonable to ignore correlations completely by writing

$$f_{12}(\boldsymbol{r}_1, \boldsymbol{v}_1, \boldsymbol{r}_2, \boldsymbol{v}_2|t) = f_1(\boldsymbol{r}_1, \boldsymbol{v}_1|t) f_2(\boldsymbol{r}_2, \boldsymbol{v}_2|t) \tag{4}$$

2. In the hard sphere collision integral, short range correlations are taken into account but, following Enskog, they are assumed to be velocity independent. Accordingly, the pair distribution function at contact is written as:

$$f_{12}(\boldsymbol{r}_1, \boldsymbol{v}_1, \boldsymbol{r}_1 - \sigma_{12}\hat{\boldsymbol{k}}, \boldsymbol{v}_2|t) = \chi_{12}(\boldsymbol{r}_1, \boldsymbol{r}_1 - \sigma_{12}\hat{\boldsymbol{k}}) f_1(\boldsymbol{r}_1, \boldsymbol{v}_1|t) f_2(\boldsymbol{r}_1 - \sigma_{12}\hat{\boldsymbol{k}}), \boldsymbol{v}_2|t) \tag{5}$$

In Eq. (5), $\chi_{12}(\boldsymbol{r}_1, \boldsymbol{r}_1 - \sigma_{12})$ is the pair correlation function at contact, which takes spatial correlation and excluded volume effects into account[Enskog (1921)].

☞ A rigourous derivation of χ_{12} would be difficult, therefore:

- it is assumed that excluded volume effects are determined solely by wall molecules through their number density $n_2(\boldsymbol{r}_2)$.

- The specific form of $\chi_{12}(n_2)$ is taken from an approximate expression for the contact value of the pair correlation function of a single component hard sphere gas in uniform equilibrium:

$$\chi_{12}(n_2) = \frac{1}{2}\frac{2 - \eta_2}{(1 - \eta_2)^3}, \qquad \eta_2 = \frac{\pi}{6}n_2\sigma_2^3 \qquad (6)$$

  where η_2 is the reduced density (or volume fraction) in the solid phase.

- The expression given above is easily derived from the approximate equation of state of a hard sphere gas proposed by Carnahan and Starling (1969).

- Although Eq. (6) provides a very accurate approximation of the contact value of the uniform equilibrium pair correlation function in a single component hard sphere gas, its use in the present context is highly questionable.

☞ No explicit assumption is made about the interaction among wall atoms.

- It is assumed that walls are in a prescribed state of equilibrium which is not altered by the interaction with the gas phase.

- Hence, the velocity distribution function f_2 will take the following form

$$f_2(\boldsymbol{r}_2, \boldsymbol{v}_2) = \frac{n_2(\boldsymbol{r}_2)}{[2\pi R_2 T_2(\boldsymbol{r}_2)]^{3/2}} \exp\left\{-\frac{[\boldsymbol{v}_2 - \boldsymbol{u}_2(\boldsymbol{r}_2)]^2}{2R_2 T_2(\boldsymbol{r}_2)}\right\} \tag{7}$$

being $n_2(\boldsymbol{r}_2)$, $T_2(\boldsymbol{r}_2)$ and $\boldsymbol{u}_2(\boldsymbol{r}_2)$ the wall atoms number density, temperature and mean velocity, respectively. The gas constant R_2 is defined as $\frac{k_B}{m_2}$, where k_B is the Boltzmann constant.

Substituting the expressions in Eqs. (4,5) into Eq.(3) leads to following linear integro-differential equation for f_1:

$$\frac{\partial f_1}{\partial t} + \boldsymbol{v}_1 \circ \nabla_{\boldsymbol{r}_1} f_1 + \frac{\boldsymbol{F}_{12}}{m_1} \circ \nabla_{\boldsymbol{v}_1} f_1 = \sigma_{12}^2 \int \left\{\chi_{12}\left[n_2(\boldsymbol{r}_1 + \sigma_{12}\hat{\boldsymbol{k}})\right] f_1(\boldsymbol{r}_1, \boldsymbol{v}_1^*|t) f_2(\boldsymbol{r}_1 + \sigma_{12}\hat{\boldsymbol{k}}, \boldsymbol{v}_2^*|t) - \right.$$

$$\left.\chi_{12}\left[n_2(\boldsymbol{r}_1 - \sigma_{12}\hat{\boldsymbol{k}})\right] f_1(\boldsymbol{r}_1, \boldsymbol{v}_1|t) f_2(\boldsymbol{r}_1 - \sigma_{12}\hat{\boldsymbol{k}}, \boldsymbol{v}_2|t)\right\} (\boldsymbol{v}_r \circ \hat{\boldsymbol{k}})^+ d\boldsymbol{v}_2 \, d^2\hat{\boldsymbol{k}} \tag{8}$$

$$\boldsymbol{F}_{12}(\boldsymbol{r}_1) = -\int_{\rho > \sigma_{12}} \frac{\boldsymbol{r}_1 - \boldsymbol{r}_2}{\rho} \frac{d\phi_{12}}{d\rho} n_2(\boldsymbol{r}_2) \, d\boldsymbol{r}_2 \tag{9}$$

Test Problems

P1 Heat transport in a collisionless gas confined between two parallel plates kept at different temperatures.

P2 Couette flow in a collisionless gas

to obtain

- Profiles of macroscopic quantities

- Shapes of velocity distribution function

- Accommodation coefficients for :
    1. kinetic energy and normal momentum (from problem P1)
    2. tangential momentum accommodation coefficient (from problem P2)

1D Equation for Test Problems

- In both problems listed above a collisionless gas fills the gap between two parallel infinite plates represented by the following density profile:

$$
n_2(z_1) = \begin{cases} n_w & |z_1| > L_z \\ 0 & |z_1| \le L_z \end{cases} \tag{10}
$$

  n_w being the constant value of the number density of wall atoms.

- The equilibrium velocity distribution function of wall atoms is given by the following expressions

$$
f_2(z_1, \boldsymbol{v}_2) = \begin{cases} \frac{n_w}{(2\pi R_2 T_L)} \exp\left[-\frac{\boldsymbol{v}_2^2}{2 R_2 T_L}\right] & z_1 \le -L_z \\ \frac{n_w}{(2\pi R_2 T_R)} \exp\left[-\frac{\boldsymbol{v}_2^2}{2 R_2 T_R}\right] & z_1 \ge L_z \end{cases} \quad \text{Heat Trasport}
$$

$$
f_2(z_1, \boldsymbol{v}_2) = \begin{cases} \frac{n_w}{(2\pi R_2 T_L)} \exp\left[-\frac{(\boldsymbol{v}_2 - u_L \hat{\boldsymbol{x}})^2}{2 R_2 T_L}\right] & z_1 \le -L_z \\ \frac{n_w}{(2\pi R_2 T_R)} \exp\left[-\frac{(\boldsymbol{v}_2 + u_L \hat{\boldsymbol{x}})^2}{2 R_2 T_L}\right] & z_1 \ge L_z \end{cases} \quad \text{Couette Flow}
$$

- In the simple one-dimensional geometry considered here, the force field $\boldsymbol{F}_{12}$ reduces to the component $F_z^{(12)}(z_1)$ normal to the plates.

$$
F_z^{(12)}(z_1) = 2\pi\overline{\phi}_{12} \left[\sigma_{12}^{\gamma_{12}} \int_{|z_1-z_2|>\sigma_{12}} \frac{(z_2-z_1)n_2(z_2)}{|z_1-z_2|^{\gamma_{12}}} \, dz_2 + \int_{|z_1-z_2|\leq\sigma_{12}} (z_2-z_1)n_2(z_2) \, dz_2 \right]
$$

$$(11)$$

- Since walls cannot produce spatial gradients of fluid properties along x_1 or y_1 directions, the following one-dimensional form of Eq. (8) can be used to study the problem:

$$
\frac{\partial f_1}{\partial t} + v_{1z}\frac{\partial f_1}{\partial z_1} + \frac{F_z^{(12)}(z_1)}{m_1}\frac{\partial f_1}{\partial v_{1z}} = C_{12}(f_2, f_1) \tag{12}
$$

$$
C_{12}(f_2, f_1) = \sigma_{12}^2 \int \left\{ \chi_{12}\left[n_2(z_1+\sigma_{12}k_z)\right] f_2(z_1+\sigma_{12}k_z, \boldsymbol{v}_2^*) f_1(z_1, \boldsymbol{v}_1^*|t) - \right.
$$
$$
\left. \chi_{12}\left[n_2(z_1-\sigma_{12}k_z)\right] f_2(z_1-\sigma_{12}k_z, \boldsymbol{v}_2) f(z_1, \boldsymbol{v}_1|t) \right\} (\boldsymbol{v}_r \circ \hat{\boldsymbol{k}})^+ d\boldsymbol{v}_2 d^2\hat{\boldsymbol{k}} \tag{13}
$$

Numerical Method

☞ Eq. (12) has been solved numerically by a variant of a DSMC scheme for the non-linear Enskog and Enskog-Vlasov kinetic equation [Frezzotti (1997)].

- Steady solutions of Eq. (12) are sought as long time limit of unsteady solutions.

- Each simulation is started by filling the gap between the walls uniformly with N_p particles whose velocities are distributed according with a Maxwellian having temperature T_L and bulk velocity equal to zero.

- After the onset of steady flow conditions, particle properties are sampled and time averaged to obtain the macroscopic quantities of interest as well as the distribution function.

☞ It should be observed that the existence of steady solutions is not obvious because, in general, gas atoms can diffuse through the solid under the action of random collisions.However,the equation of balance of linear momentum shows that the non-local structure of $C_{12}(f_2, f_1)$ produces a net force where n_2 profile is not uniform.

- The force generated by hard sphere collisions repels atoms approaching the walls. The intensity of repulsion is proportional to $n_w \chi_{12}(n_w)$ and it becomes strong enough to confine the fluid within the gap, when n_w has the typical values of a solid substance.

Comparison with MD results

- The results obtained from the numerical solution of Eq. 12 have been compared to the results of MD studies of test problems (1) and (2).

- Numerical data have been collected from the following investigations of rare gases (Xe and Ar) flows in contact with metal plates (Pt):

1. K. Yamamoto and K. Yamashita, *Analysis of the Couette flow based on the molecular dynamics study for gas-wall interaction*, Rarefied Gas Dynamics 20 (1997).

2. K. Yamamoto, *Slightly rarefied gas flow over a smooth platinum surface*, Rarefied Gas Dynamics 22 (2001)

3. K. Yamamoto and H. Takeuchi and T. Hyakutake, *Effect of wall characteristics on the behaviors of reflected gas molecules in a thermal problem*, Rarefied Gas Dynamics 23 (2003).

4. Bing-Yang Cao et al. *Temperature dependence of the tangential momentum accommodation coefficient for gases*, Applied Physics Letters **86**, 091905 (2005).

Warnings!

- Interaction potentials used in MD studies are not the same used to derive the kinetic model

- Yamamoto adopted a Morse potentials in the form

$$\phi_{(Xe-Pt)}(\rho) = \epsilon_{(Xe-Pt)} \left\{ exp\left[-2\sigma_{(Xe-Pt)}(\rho-\rho_0)\right] - 2exp\left[-\sigma_{(Xe-Pt)}(\rho-\rho_0)\right]\right\} \tag{14}$$

$$\phi_{(Ar-Pt)}(\rho) = \epsilon_{(Ar-Pt)} \left\{ exp\left[-2\sigma_{(Ar-Pt)}(\rho-\rho_0)\right] - 1\right\}^2 \tag{15}$$

- Interactions among wall atoms simulated through Lennard-Jones potential.

- Gas phase not collisionless $Kn = 0.2 - 10$. DSMC scheme adopted to account for atomic interactions in the gas. Small effects of Kn on accomodation coefficients.

- In Ref. [4] Lennard-Jones potential has been used to describe $Ar - Pt$ interaction (with a very different $\epsilon_{(Ar-Pt)}$). Walls simulated by arrays of non-interacting atoms, individually tethered to fixed points by springs.

- Gas phase not collisionless. MD algorithm adopted to to account for atomic interactions in the gas

Model Computational Settings

If σ_1, $\sigma_1/\sqrt{R_1 T_w}$ and m_1 are adopted as units of lenght, time and mass, solutions of Eq. (12) can be shown to depend on the following non-dimensional parameters:

$$\eta_w = \frac{\pi \sigma_2^3 n_w}{6}, \qquad \frac{\overline{\phi}_{12}}{k_B T_w}, \qquad \gamma_{12} \qquad \frac{\sigma_2}{\sigma_1}, \qquad \frac{m_2}{m_1}, \qquad \frac{T_L}{T_w}, \qquad \frac{T_R}{T_L}, \qquad S_w = \frac{u_w}{\sqrt{2 R_1 T_w}}.$$

| System	η_w	$\dfrac{\overline{\phi}_{12}}{k_B T_w}$	$\dfrac{\sigma_2}{\sigma_1}$	$\dfrac{m_2}{m_1}$	$\dfrac{T_L}{T_w}$	$\dfrac{T_R}{T_L}$	S_w
Problem 1 - Heat Transport - $T_w = 300°K$, $\gamma_{12} = 6$							
$Xe - Pt$	0.5, 0.7	1.4896, 1.064	1.0	1.486	1.0	1.2 - 4.0	0.0
$Ar - Pt$	0.5, 0.7	0.6286, 0.449	1.0	4.883	1.0	1.2 - 4.0	0.0
Problem 2 - Couette Flow A - $T_w = 300°K$, $\gamma_{12} = 6$							
$Xe - Pt$	0.5, 0.7	0.0, 1.4896, 1.064	1.0	1.486	1.0	1.0	0.1 - 2.5
$Ar - Pt$	0.5, 0.7	0.0, 0.6286, 0.449	1.0	4.883	1.0	1.0	0.1 - 0.5
Problem 2 - Couette Flow B - $T_w = 300°K$, $\gamma_{12} = 6$							
$Ar - Pt$	0.7	0.2158, 0.449	1.0	4.883	0.333 - 1.333	1.0	0.224

Heat Transfer between Parallel Plates, Xe-Pt
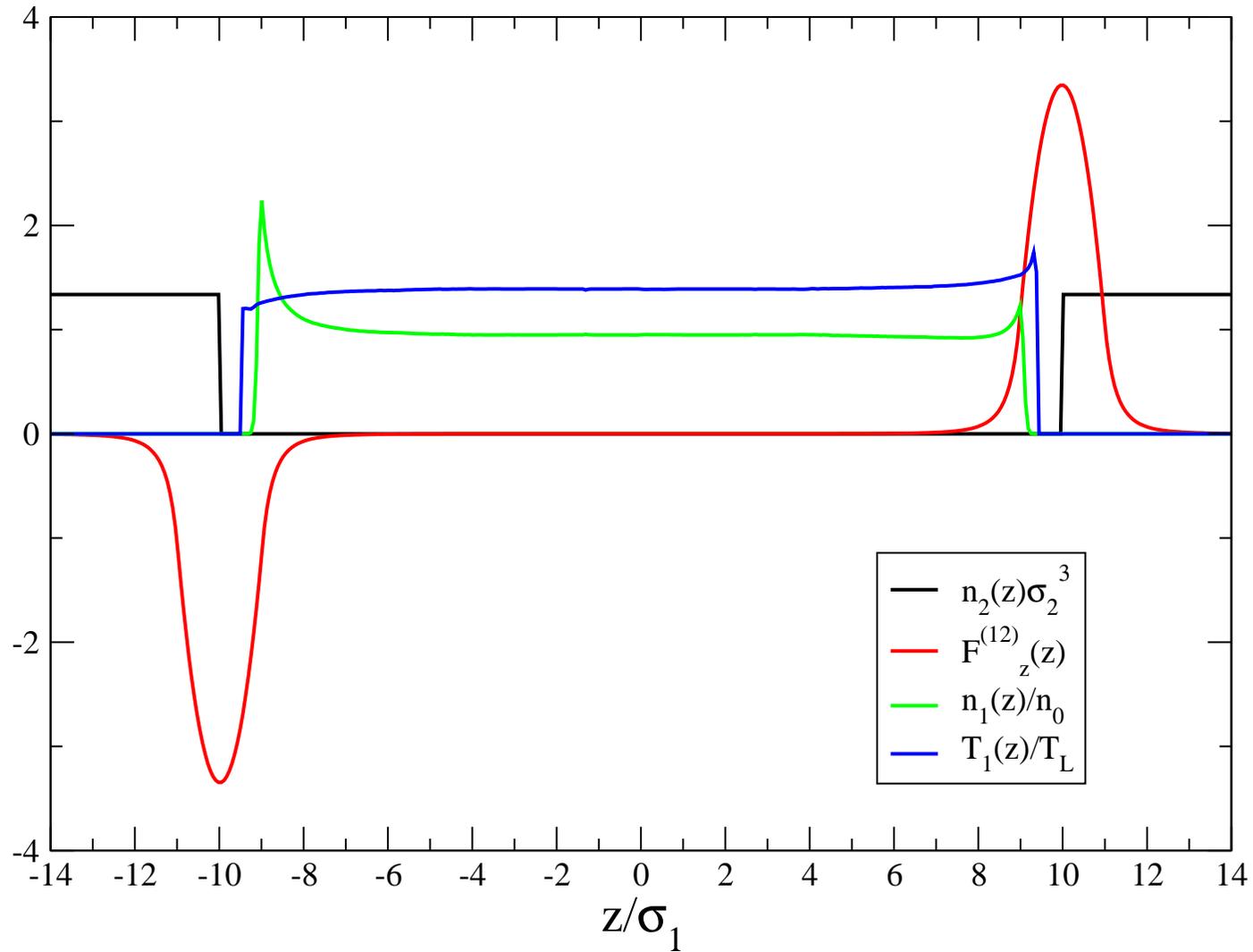
$T_R/T_L=2.0$, $m_2/m_1=1.486$, $\sigma_1=\sigma_2=1$, $\phi_{12}=1.064k_BT_L$, $L_z=10\sigma_1$, $\eta_w=0.7$

Heat Transfer between Parallel Plates, Xe-Pt

$T_R/T_L=2.0$, $m_2/m_1=1.486$, $\sigma_1=\sigma_2=1$, $\phi_{12}=1.064k_BT_L$, $L_z=10\sigma_1$, $\eta_w=0.7$
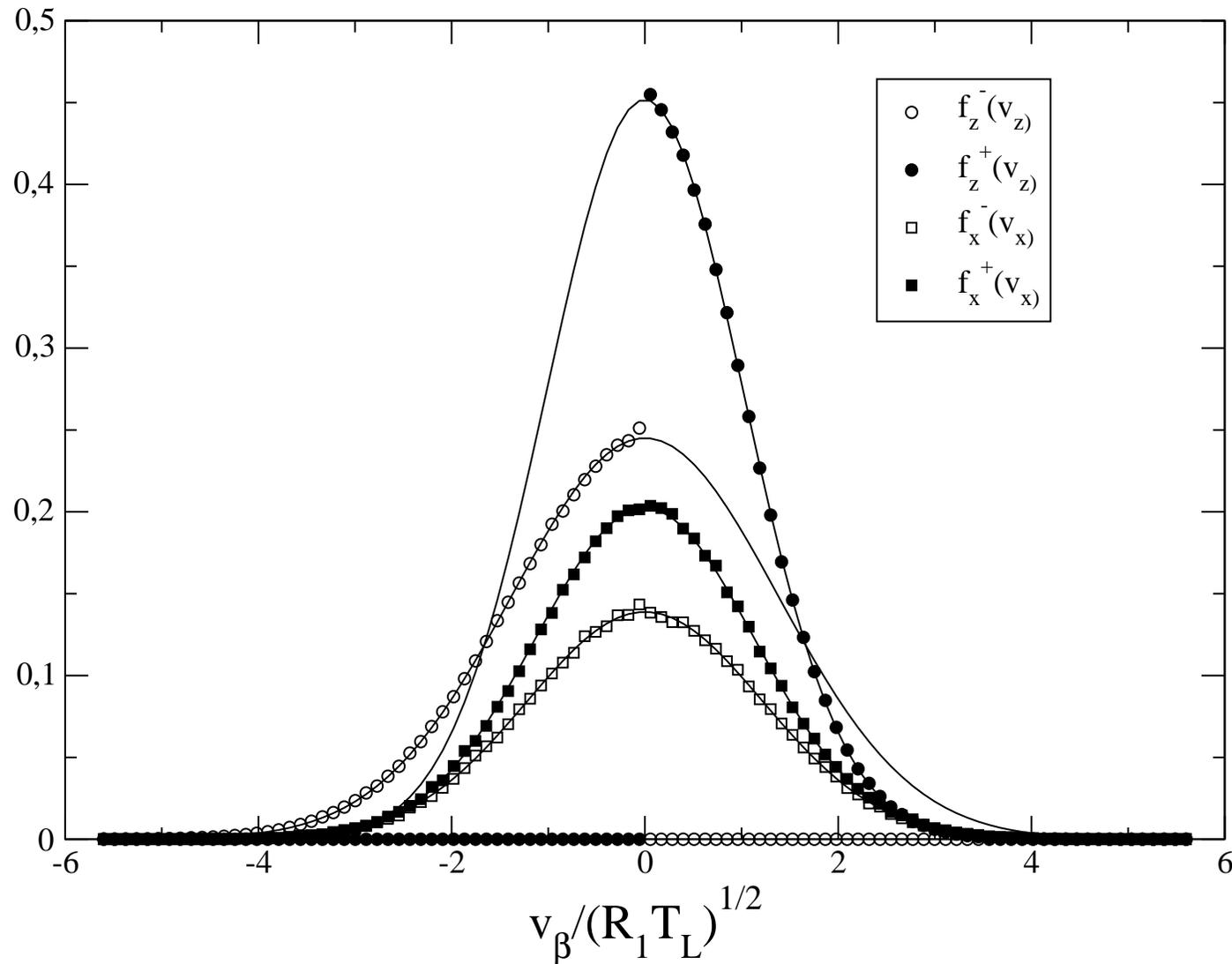
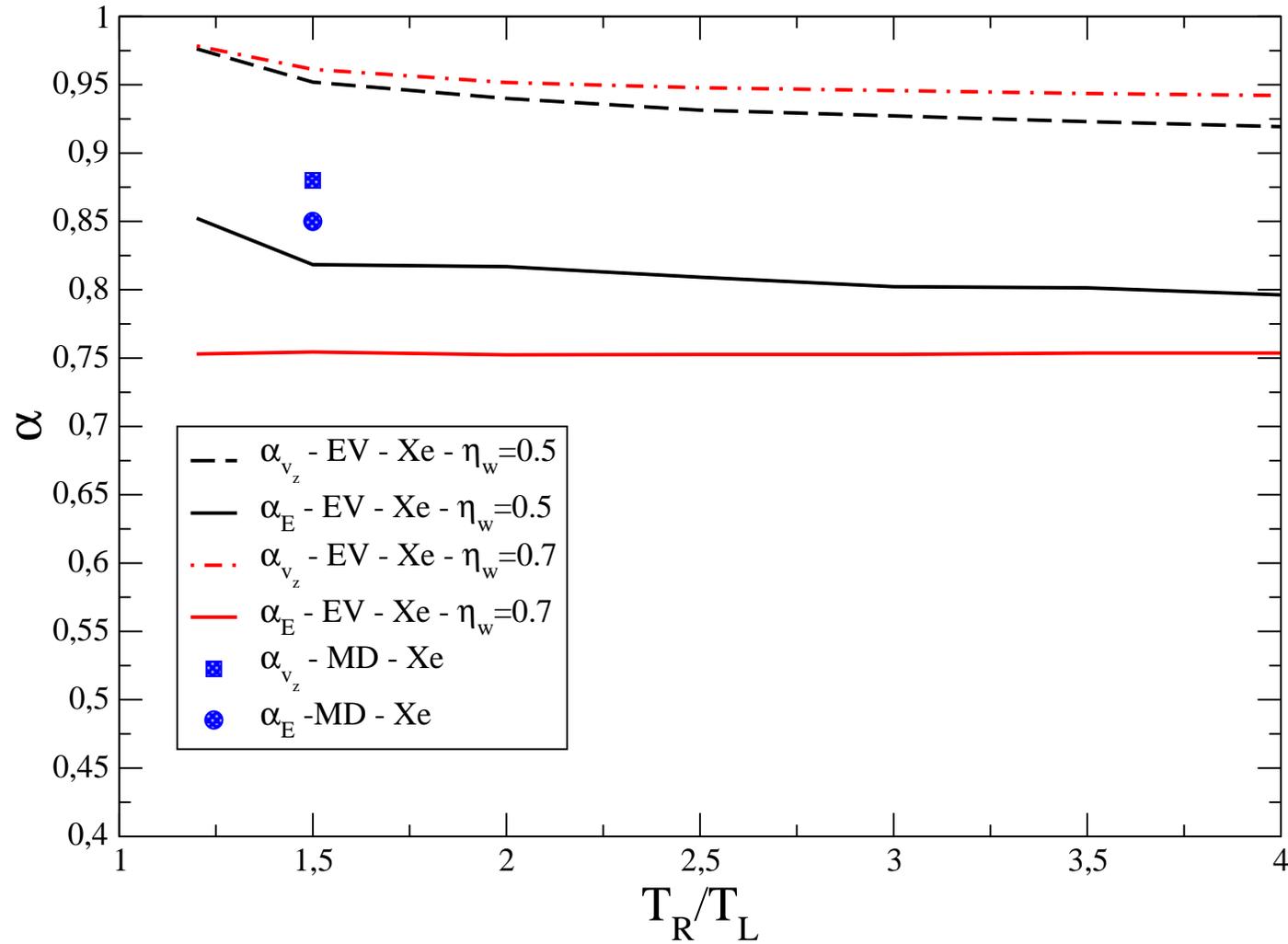Heat Transfer between Parallel Plates
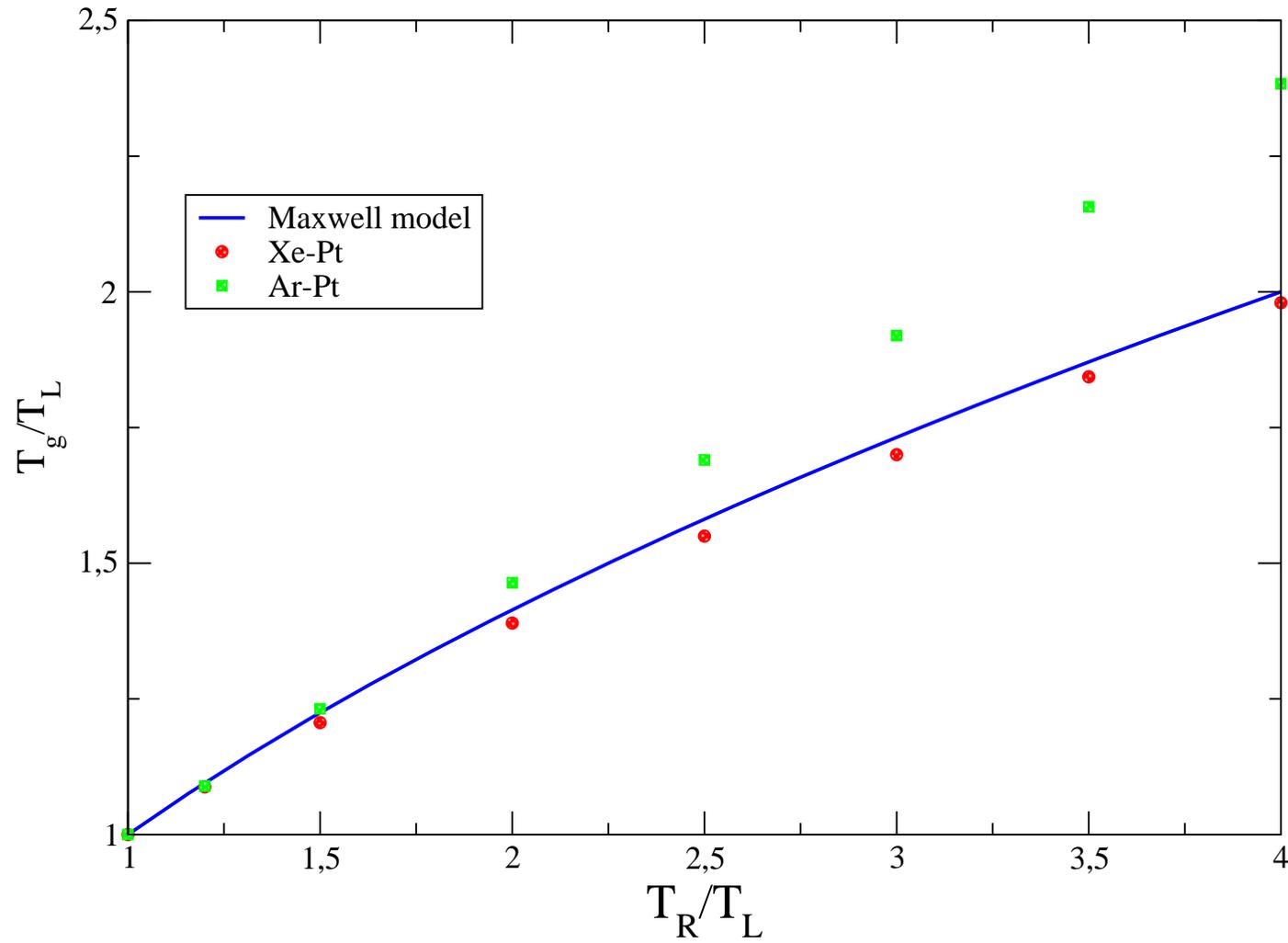
Accommodation coefficients

☞ The accommodation coefficient of the molecular property $\phi(\boldsymbol{v})$ has been computed from the following general expression:

$$\alpha_\psi = \frac{\int_{v_z<0} \psi(\boldsymbol{v})|v_z|f(\boldsymbol{v})\,d\boldsymbol{v} - \int_{v_z>0} \psi(\boldsymbol{v})|v_z|f(\boldsymbol{v})\,d\boldsymbol{v}}{\int_{v_z<0} \psi(\boldsymbol{v})|v_z|f(\boldsymbol{v})\,d\boldsymbol{v} - \int_{v_z>0} \psi(\boldsymbol{v})|v_z|f_w(\boldsymbol{v})\,d\boldsymbol{v}} \qquad (16)$$

☞ The moments appearing in Eq. (16) have been computed by sampling gas particles in the central slice of the computational domain where both the short and long range interaction with walls are negligible

Heat Transfer between Parallel Plates, Xe-Pt
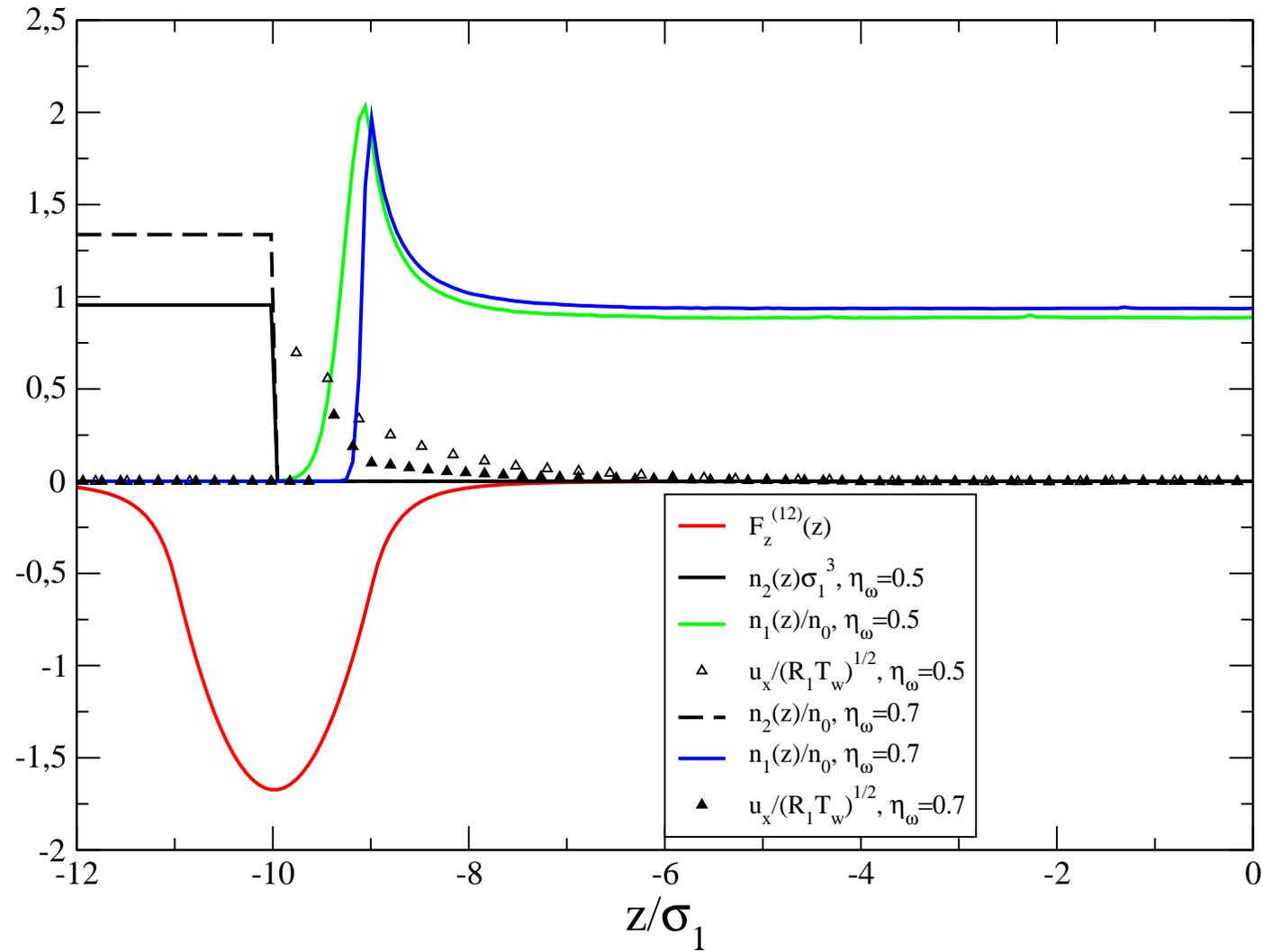
Energy and Normal Momentum Accommodation Coefficients

Heat Transfer between Parallel Plates, Ar-Pt
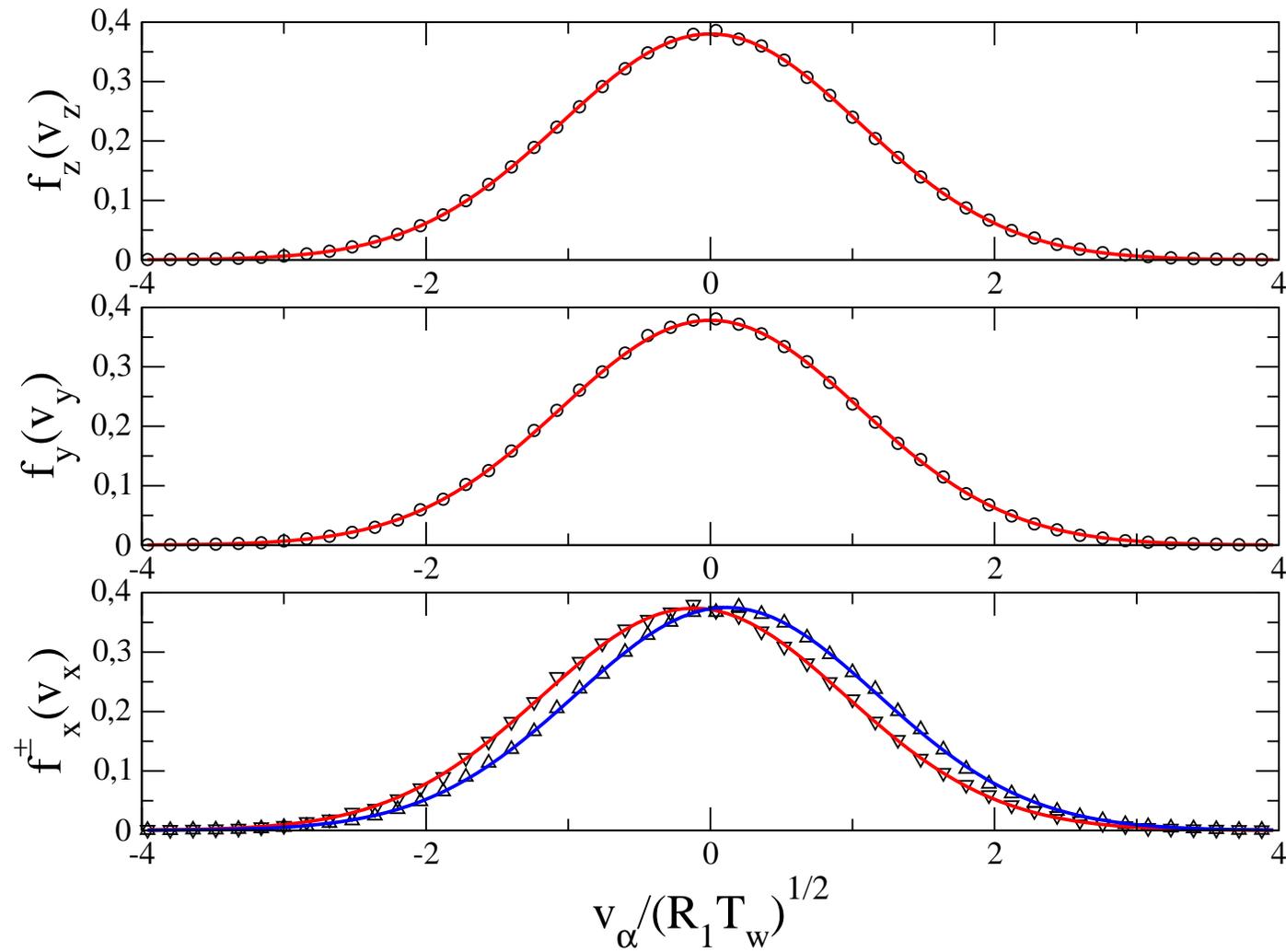
Energy and Normal Momentum Accommodation Coefficients
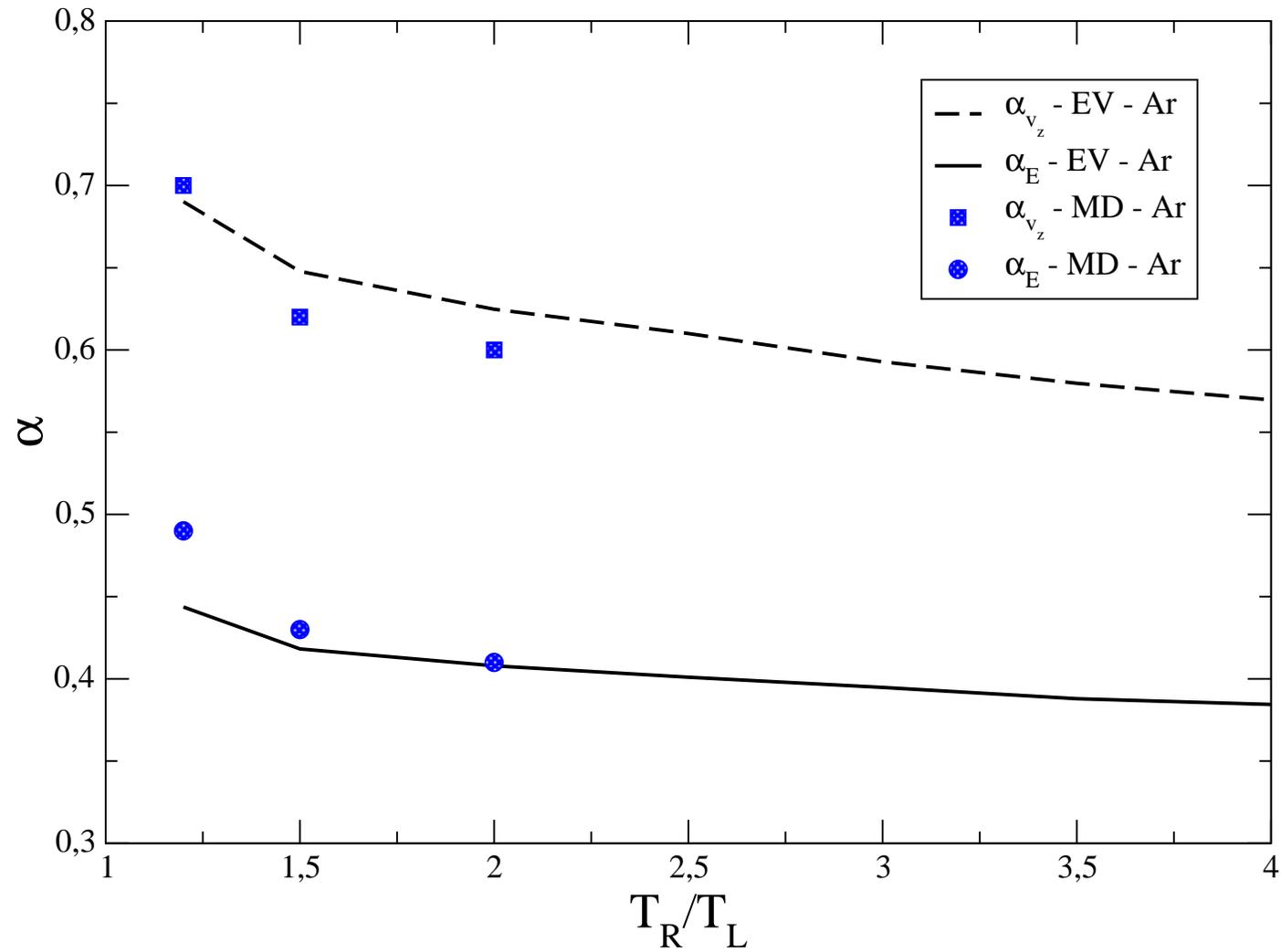
Couette Flow - Xe-Pt
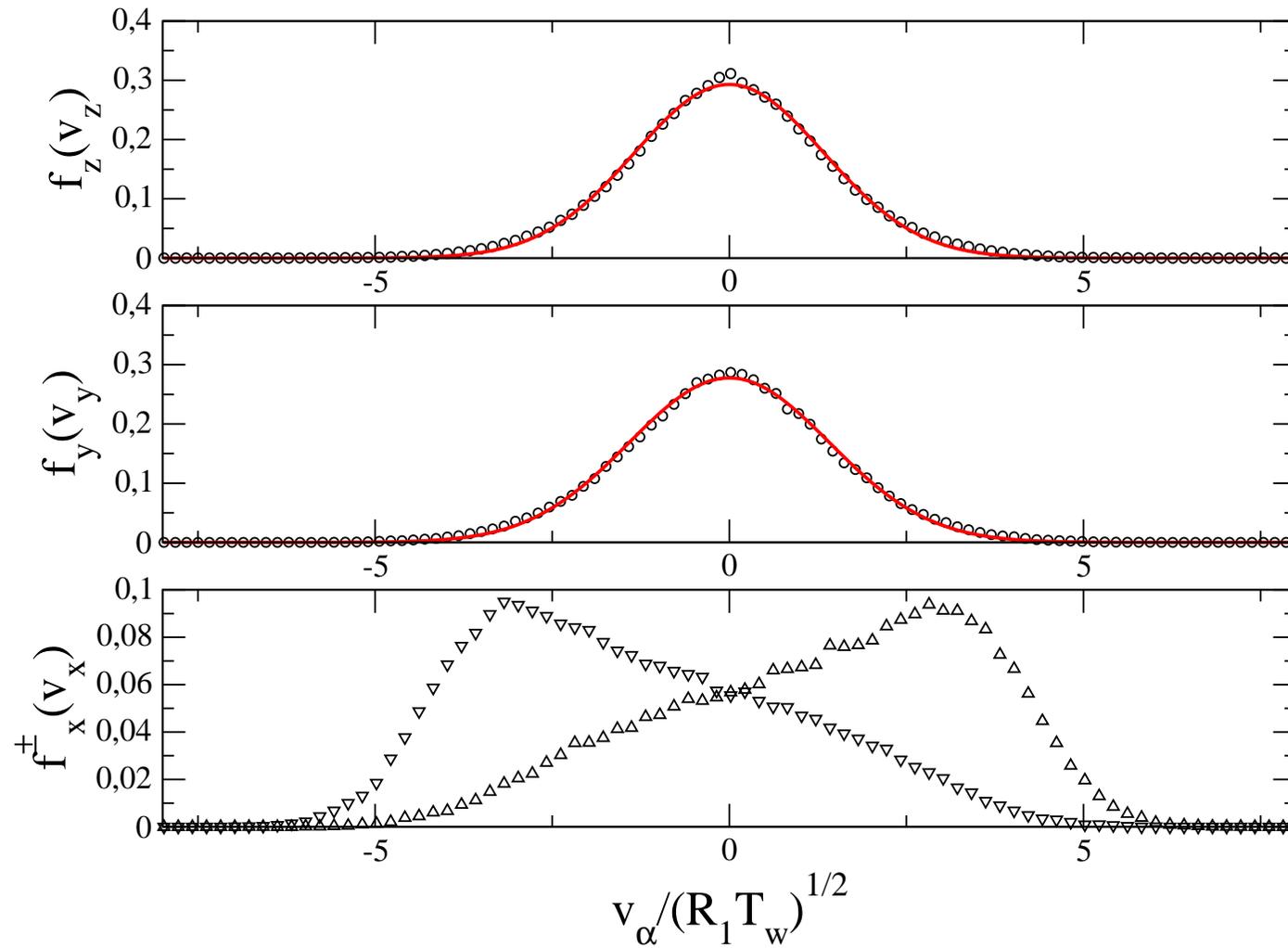
$m_2/m_1=1.486$, $\sigma_1=\sigma_2$, $S_w=0.5$

Couette Flow , Ar-Pt - Reduced Distribution Functions

$m_2/m_1 = 4.883,\ \sigma_1 = \sigma_2 = 1,\ \eta_\omega = 0.7,\ \phi_{12} = 0.449\ k_B T_w,\ S_w = 0.5$
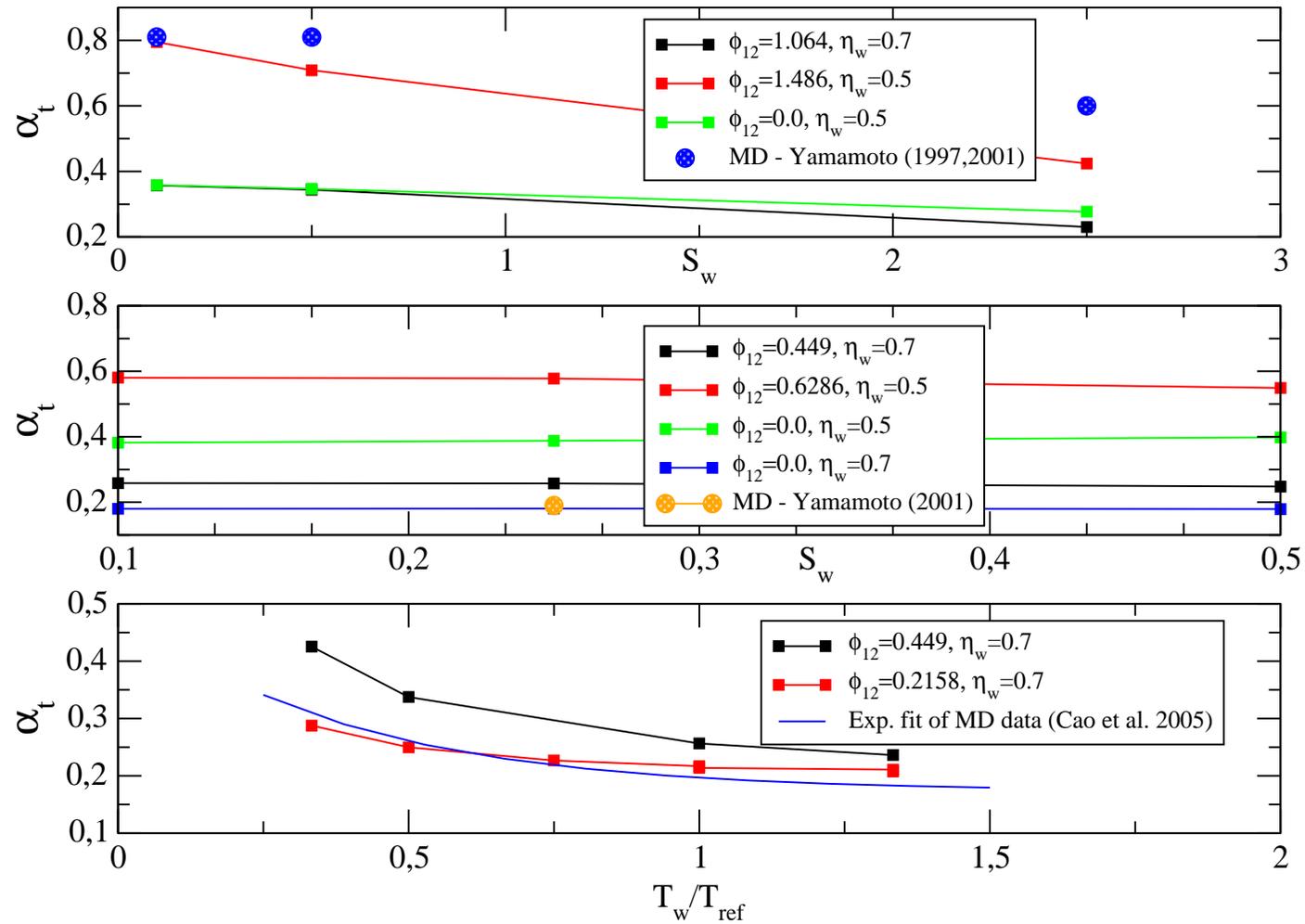
Couette Flow , Xe-Pt - Reduced Distribution Functions

m_2/m_1=1.486, σ_1=σ_2=1, η_ω=0.5, ϕ_{12}=1.4896 $k_B T_w$, S_w=2.5

Tangential Momentum Accommodation Coefficients

Xe-Pt , Ar-Pt

Conclusions

- A simple kinetic model of fluid-wall interaction has been developed along the guidelines of the kinetic theory of dense fluids

- Model predictions for test problems are close to (or not far from) the results of MD simulations.

- Future activity will concentrate on:
  1. Extend the data base of MD simulations
  2. Development of better models for pair correlations.
  3. Coupling with DSMC simulations of gases and liquids [Frezzotti & Gibelli (2006)]
  4. Extension to diatomic gas species.