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Non-Fourier Heat-Flux Law for Diatomic Gases

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Nomenclature

$E^{\text{int}}(v, r)$ = internal energy of the reduced mass
$E^{\text{kin}}(v)$ = kinetic part of the internal energy
$E^{\text{pot}}(r)$ = potential part of the internal energy
$E^{\text{tr}}(V)$ = translational energy of the center of mass
$E(V, v, r)$ = total energy, $E^{\text{tr}}(V) + E^{\text{int}}(v, r)$
$\{\hat{e}_l\}$ = set of linearly independent basis
$f^{(1)}(\boldsymbol{R}, \boldsymbol{V}, t)$ = local equilibrium distribution function for the center of mass:

$$n_M(\boldsymbol{R}, t)[M\beta(\boldsymbol{R}, t)/2\pi]^{\frac{3}{2}} \times \exp\left\{-[M\beta(\boldsymbol{R}, t)/2][\boldsymbol{V} - \boldsymbol{u}_M(\boldsymbol{R}, t)]^2\right\}$$

$f^{(1)}(\boldsymbol{R}, \boldsymbol{v}, t)$ = local equilibrium distribution function for the reduced mass:

$$n_\mu(\boldsymbol{R}, t)[\mu\beta(\boldsymbol{R}, t)/2\pi]^{\frac{3}{2}} \times \exp\left\{-[\mu\beta(\boldsymbol{R}, t)/2][\boldsymbol{v} - \boldsymbol{u}_\mu(\boldsymbol{R}, t)]^2\right\}$$

$g(|\boldsymbol{r}|)$ = radial or pair distribution function, $\exp[\beta W(r)]$
$\boldsymbol{J}(t)$ = heat flux
k_B = Boltzmann constant
M = total mass of the diatomic molecule
$n_M(\boldsymbol{R}, t)$ = number density of the total mass
$n_\mu(\boldsymbol{R}, t)$ = number density of the reduced mass
$\hat{n}$ = unit vector
q = real number
$\boldsymbol{R}$ = position vector of the center of mass
$\boldsymbol{r}$ = position vector of the relative distance between two point particles
r_m = distance between point particles at which their potential energy is minimum
T = temperature
t = time
$\boldsymbol{u}_M(\boldsymbol{R}, t)$ = center-of-mass average velocity vector, $\langle \boldsymbol{V} \rangle$
$\boldsymbol{u}_\mu(\boldsymbol{R}, t)$ = reduced-mass average velocity vector, $\langle \boldsymbol{v} \rangle$
$\boldsymbol{V}$ = velocity of the center of mass
$\boldsymbol{v}$ = velocity of the relative position vector between two point particles
$W(r)$ = potential energy of the average force a particle experiences
β = $1/k_B T$
β^*_{dia} = Eucken number for diatomic gases:

$$\left[\beta^*_{\text{tr}} - \frac{(5 - 3\gamma)}{3(\gamma - 1)}\beta^*_{\text{int}}\right]$$

β^*_{int} = Eucken number caused by internal states:

$$3\left[1 - \frac{\mu(\delta v)^2}{3k_B T} + \frac{2\langle E^{\text{int}} W(r)\rangle}{3\langle E^{\text{int}}\rangle k_B T}\right]$$

β^*_{tr} = Eucken number caused by translational motion, $3[1 - M(\delta V)^2/3k_B T]$
Γ = arbitrary volume over which the relative coordinate is integrated
γ = ratio of heat capacities
Δ = variable's nonequilibrium instantaneous departure from its equilibrium value
$\Delta\boldsymbol{a}$ = total area:

$$\int_{\text{open surface}} \mathrm{d}\boldsymbol{a}$$

δ = fluctuation in a variable
∂_t = derivative with respect to time, $\partial/\partial t$
ϵ = perturbation
θ = time as an integration variable
λ = thermal conductivity of diatomic gases, $\beta^*_{\text{dia}}\langle E^{\text{tr}}\rangle u/2T\Delta a$
λ_{int} = thermal conductivity caused by internal motion, $\beta^*_{\text{int}}\langle E^{\text{int}}\rangle u/2T\Delta a$
λ_{tr} = thermal conductivity caused by translational motion, $\beta^*_{\text{tr}}\langle E^{\text{tr}}\rangle u/2T\Delta a$
μ = reduced mass of the diatomic molecule
σ = physical constant, $\beta^*_{\text{dia}}\langle E^{\text{tr}}\rangle/3\Delta a$
τ = relaxation time
$\langle\,\rangle$ = nonequilibrium ensemble average
$\langle\,\rangle_e$ = equilibrium ensemble average

Introduction

PREVIOUSLY a derivation of the Cattaneo's heat-flux law, which is a non-Fourier heat-flux law, was given for monoatomic gases.[1] The derivation of Cattaneo's heat-flux equation for diatomic gases will emphasize the validity of the method used before for monoatomic gases, which is an important thing to do before the method is applied to the far-from-equlibrium case, and will demonstrate the phenomenon of finite speed heat propagation for a different kind of gas, namely the diatomic gas. Therefore, the derivation of Cattaneo's heat-flux equation for diatomic gases is presented here. Under special conditions, which include being at the critical temperature, the method used before proved very successful here in the case of diatomic gases. However, the distribution function that

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corresponds to the diatomic gas is taken as a product between the distribution function of the total mass of the diatomic molecule, the distribution function of the reduced mass of the diatomic molecule, and a function that represents the radial distribution function.[2] Pair distribution functions in the form of a product between two distribution functions representing each particle and a radial distribution function have been extensively used before[3] and were mainly used to study correlations.[4] When the radial distribution function equals unity for all values of the radial distances between the two particles, one gets the Vlasov equation, which resembles the collisionless Boltzmann equation.[5] Sometimes the correct form of the pair distribution function is reached a posteriori, depending on the final numerical results.[3] Writing the pair distribution function in a product form has been applied in this Note because of the nature of diatomic molecules. Moreover, when a diatomic gas at the critical temperature is considered here, the value of the Eucken number will be 1.875 in comparison to the value obtained by Eucken (see Ref. 6), which is 1.94.

Derivation of Cattaneo's Heat Flux for Diatomic Gases

In the case of diatomic molecules, one must write the energy as[2] $E = E^{\mathrm{tr}} + E^{\mathrm{int}}$. For a constant value of γ, the two energies at equilibrium are related by[7]

$$\langle E^{\mathrm{int}}\rangle_e = \frac{(5-3\gamma)}{3(\gamma-1)}\langle E^{\mathrm{tr}}\rangle_e \tag{1}$$

and because local equilibrium is considered here, Eq. (1) can be applied. The relation $E^{\mathrm{int}} = [(5-3\gamma)/3(\gamma-1)]E^{\mathrm{tr}}$ can be immediately reached from Eq. (1). The equation for heat flux is written as $\boldsymbol{J} = E\boldsymbol{V} = (E^{\mathrm{tr}} + E^{\mathrm{int}})\boldsymbol{V}$, which with the preceding relation can be written as $\boldsymbol{J}(t) = [1 + (5-3\gamma)/3(\gamma-1)]E^{\mathrm{tr}}\boldsymbol{V}(t)$. For diatomic gases $\gamma = 1.4$. In view of the fact that, for bound states, which resemble diatomic molecules, the internal energy is negative,[7] the equation for the heat flux per unit volume will be given by

$$\boldsymbol{J}(t) = \tfrac{1}{3}[MV^2(t)/2]\boldsymbol{V}(t)$$

where $V = |\boldsymbol{V}|$. With the aid of the Onsager hypothesis,[1] the following equation can be obtained:

$$\langle\Delta\boldsymbol{J}(t)\rangle = \langle\boldsymbol{J}(t)\rangle = \frac{\beta\epsilon}{3}\sum_{l=1}^{3}\langle\delta\boldsymbol{J}(0)\cdot\delta\boldsymbol{J}(t)\rangle_e\hat{e}_l$$

$$\leq \beta\epsilon\left(\frac{M}{6}\right)^2\frac{1}{3}\sum_{l=1}^{3}\sum_{i=1}^{3}\langle V_i(0)V_i(t)\rangle_e^3\hat{e}_l \tag{2}$$

which is true at all times, where it is assumed that $\epsilon \propto \nabla T$. The conservation of energy equation can be written as[1]

$$\int_0^t \mathrm{d}\theta\boldsymbol{J}(V,v,\theta)\cdot\Delta\boldsymbol{a} = -E(V,v) \tag{3}$$

After taking the statistical average corresponding to a pair of particles, one gets

$$\int_0^t \mathrm{d}\theta\langle\boldsymbol{J}(\boldsymbol{R},\theta,t)\rangle\cdot\Delta\boldsymbol{a} = -\langle E(\boldsymbol{R},t)\rangle \tag{4}$$

where the average is taken using the pair distribution function given by $f^{(2)}(\boldsymbol{R},\boldsymbol{V},\boldsymbol{r},\boldsymbol{v},t) = f^{(1)}(\boldsymbol{R},\boldsymbol{V},t)f^{(1)}(\boldsymbol{R},\boldsymbol{v},t)g(|\boldsymbol{r}|)$. Therefore,

$$\langle E(\boldsymbol{R},t)\rangle = \frac{1}{2n}\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}\int_{\Gamma}\mathrm{d}^3r\,\mathrm{d}^3V\,\mathrm{d}^3v\,E(V,v,r)$$

$$\times f^{(2)}(\boldsymbol{R},\boldsymbol{V},\boldsymbol{r},\boldsymbol{v},t) \tag{5}$$

where $n_M = n_\mu = n$. Setting $W(r) = W(r_m)$ and assuming the relation

$$E^{\mathrm{int}}(v,r_m) = E^{\mathrm{int}}(v)\exp[\beta W(r_m)], \qquad \int_\Gamma \mathrm{d}^3r g(r_m) = \frac{1}{n}$$

(see Appendix), the right-hand side of Eq. (5) is written as

$$2\langle E(\boldsymbol{R},t)\rangle = \int_{-\infty}^{+\infty}\mathrm{d}^3V\left(\frac{M\beta}{2\pi}\right)^{\frac{3}{2}}E^{\mathrm{tr}}(V)$$

$$\times\exp\left\{-\frac{\beta M}{2\pi}[\boldsymbol{V}-\boldsymbol{u}_M(\boldsymbol{R},t)]^2\right\} + \int_{-\infty}^{+\infty}\mathrm{d}^3v\left(\frac{\mu\beta}{2\pi}\right)^{\frac{3}{2}}E^{\mathrm{int}}(v)$$

$$\times\exp\left\{-\frac{\beta\mu}{2\pi}[\boldsymbol{v}-\boldsymbol{u}_\mu(\boldsymbol{R},t)]^2\right\}\exp[\beta W(r_m)] \tag{6}$$

It can be shown,[1] from Eq. (2), that after taking ∂_t of Eq. (4) one can get

$$\left[\int_0^\tau \partial_t\langle\boldsymbol{J}(\boldsymbol{R},\theta,t)\rangle\,\mathrm{d}\theta + \langle\boldsymbol{J}(\boldsymbol{R},t,t)\rangle\right]\cdot\Delta\boldsymbol{a} = -\partial_t\langle E(\boldsymbol{R},t)\rangle \tag{7}$$

where the energies in Eq. (7) are twice as much as the energies in Eq. (4). A direct evaluation of the right-hand side of Eq. (7) will give

$$\partial_t\langle E(\boldsymbol{R},t)\rangle = \langle E^{\mathrm{tr}}(V)\tfrac{3}{2}(M/2\pi)(2\pi/M\beta)\partial_t\beta\rangle$$

$$+\langle E^{\mathrm{int}}(v)\tfrac{3}{2}(\mu/2\pi)(2\pi/\mu\beta)\partial_t\beta\rangle$$

$$+\langle\beta M E^{\mathrm{tr}}(V)[\boldsymbol{V}-\boldsymbol{u}_M(\boldsymbol{R},t)]\cdot\partial_t\boldsymbol{u}_M\rangle$$

$$-\langle E^{\mathrm{tr}}(V)(M/2)[\boldsymbol{V}-\boldsymbol{u}_M(\boldsymbol{R},t)]^2\partial_t\beta\rangle$$

$$+\langle\beta\mu E^{\mathrm{int}}(v)[\boldsymbol{v}-\boldsymbol{u}_\mu(\boldsymbol{R},t)]\cdot\partial_t\boldsymbol{u}_\mu\rangle$$

$$-\langle E^{\mathrm{int}}(v)(\mu/2)[\boldsymbol{v}-\boldsymbol{u}_\mu(\boldsymbol{R},t)]^2\partial_t\beta\rangle + \langle E^{\mathrm{int}}(v)(\partial_t\beta)W(r_m)\rangle \tag{8}$$

The Euler equation for the evolution of the temperature T is given by

$$\partial_t T = -\boldsymbol{u}\cdot\nabla T - 2T(\nabla\cdot\boldsymbol{u})/3$$

where $\boldsymbol{u} = n_M M\boldsymbol{u}_M(\boldsymbol{R},t) + n_\mu\mu\boldsymbol{u}_\mu(\boldsymbol{R},t)/(n_M M + n_\mu\mu)$ is the stream velocity.[2] The evolution of β will be given by

$$\partial_t\beta = (1/k_B T^2)\boldsymbol{u}\cdot\nabla T + (1/k_B T^2)\tfrac{2}{3}(T\nabla\cdot\boldsymbol{u}) \tag{9}$$

The following equalities can be justified[1]: $\langle E^j(V^j)\boldsymbol{V}^j\rangle = \langle E^j(V^j)\rangle\langle\boldsymbol{V}^j\rangle$ and $\langle E^j(V^j)^2\rangle = \langle E^j\rangle\langle(V^j)^2\rangle$, where the index j runs over the translational and the internal modes. These equalities will furnish the following equations:

$$\langle E^j\big[\boldsymbol{V}^j - \boldsymbol{u}_j(\boldsymbol{R},t)\big]\rangle = 0 \tag{10}$$

$$\langle E^j(V^j)\big[\boldsymbol{V}^j - \boldsymbol{u}_j(\boldsymbol{R},t)\big]^2\rangle = \langle E^j\rangle[\langle(V^j)^2\rangle - \langle V^j\rangle^2]$$

$$= \langle E^j\rangle(\delta V^j)^2 \tag{11}$$

Substitution of the equalities in Eqs. (9–11) into Eq. (8) gives

$$-\partial_t\langle E(\boldsymbol{R},t)\rangle = -\left(\frac{\beta^*_{\mathrm{tr}}\langle E^{\mathrm{tr}}\rangle}{2T} + \frac{\beta^*_{\mathrm{int}}\langle E^{\mathrm{int}}\rangle}{2T}\right)\boldsymbol{u}\cdot\nabla T$$

$$-\frac{1}{3}(\beta^*_{\mathrm{tr}}\langle E^{\mathrm{tr}}\rangle + \beta^*_{\mathrm{int}}\langle E^{\mathrm{int}}\rangle)\nabla\cdot\boldsymbol{u} \tag{12}$$

From Eq. (1) one can write the local equilibrium equation $\langle E^{\mathrm{int}}\rangle = [(5-3\gamma)/3(\gamma-1)]\langle E^{\mathrm{tr}}\rangle$, which, when substituted into Eq. (12) together with Eq. (7), yields

$$\int_0^\tau \partial_t\langle\boldsymbol{J}(\boldsymbol{R},\theta,t)\rangle\,\mathrm{d}\theta + \langle\boldsymbol{J}(\boldsymbol{R},t,t)\rangle = -(\lambda_{\mathrm{tr}} + \lambda_{\mathrm{int}})\nabla T$$

$$-\frac{\beta^*_{\mathrm{dia}}\langle E^{\mathrm{tr}}\rangle}{3\Delta a}\hat{n}(\nabla\cdot\boldsymbol{u}) \tag{13}$$

or

$$\int_0^\tau \partial_t \langle \boldsymbol{J}(\boldsymbol{R}, \theta, t)\rangle \, \mathrm{d}\theta + \langle \boldsymbol{J}(\boldsymbol{R}, t, t)\rangle = -\lambda \nabla T - \sigma \hat{n}(\nabla \cdot \boldsymbol{u}) \quad (14)$$

where $E^{\text{int}} = -|E^{\text{int}}|$ for bound states.[7] With the use of Ergodic theory,[1] Eq. (14) will immediately lead to

$$\tau \frac{\partial \langle \boldsymbol{J}(\boldsymbol{R}, t)\rangle}{\partial t} + \langle \boldsymbol{J}(\boldsymbol{R}, t, t)\rangle = -\lambda \nabla T - \sigma \hat{n}(\nabla \cdot \boldsymbol{u}) \quad (15)$$

which is the non-Fourier heat-flux law for diatomic gases, given that the gas is incompressible. If Eq. (1) is used in the equation for β^*_{int} and the assumption that $W(r_m)$ and $\langle E^{\text{tr}}\rangle$ are uncorrelated is applied, one gets $\beta^*_{\text{int}} = 3\{1 - [\mu(\delta v)^2/3k_B T] + 2W(r_m)/3k_B T\}$. Furthermore, because the pair correlation has a single maximum for low densities that occurs at r_m for the Lennard–Jones potential,[5] which represents gases, one may substitute the value of the Lennard–Jones potential[2] at r_m for $W(r_m)$. Then, assuming it to be at the critical temperature, one gets the equation[2] $W(r_m) = -0.77 k_B T_c$, where the minus sign is caused by the fact that the bound state is considered. Assuming the validity of the equations $u^2_M = (8k_B T_c/\pi M)$, $\langle V^2\rangle = (3k_B T_c/M)$, $u^2_\mu = (8k_B T_c/\pi\mu)$, and $\langle v^2\rangle = (3k_B T_c/\mu)$ at the critical temperature, one gets $\beta^*_{\text{int}} = 1.007$. The value for β^*_{int} obtained by Eucken is unity.[6] On the other hand, $\beta^*_{\text{tr}} = 3 - 3 + (8/\pi) = 2.547$, and $\beta^*_{\text{dia}} = 2.547 - 0.667(1.007) = 1.875$ at $T = T_c$, where the value of γ is taken to be $\gamma = 1.4$ for diatomic gases. The value for β^*_{dia} obtained by Eucken is 1.94. The closeness of these numbers can be taken as an indication of the correct approach. For a van der Waals gas the translational energy will have an extra term that is independent of temperature, which will give the same Eucken number. This could be caused by the fact that the statistical formalism of a diatomic gas takes into account the effects of the van der Waals gas model. For example, the fixed distance between the two point particles generates over time a spherically symmetric shape when all of the rotations are considered. This spherically symmetric shape resembles the volume of a monoatomic van der Waals gas molecule. In addition, the van der Waals gas considers pair potentials, which is the same type of potential that is considered when one deals with the statistical description of diatomic gases.

Appendix: Internal Energy in Exponential Form

In general, the average internal energy is given by

$$\langle E^{\text{int}}\rangle = \langle E^{\text{kin}}\rangle + \langle E^{\text{pot}}\rangle \quad (A1)$$

Because setting $W(r) = W(r_m)$ indicates negligible kinetic energy caused by radial motion, the value of $\langle E^{\text{kin}}\rangle$ equals $k_B T$ at equilibrium and local equilibrium as well. The value of $\langle E^{\text{pot}}\rangle$ is expected to be a multiple of $k_B T$, and one can write

$$\langle E^{\text{int}}\rangle = \langle E^{\text{kin}}\rangle + q\langle E^{\text{kin}}\rangle \quad (A2)$$

where $\langle |E^{\text{pot}}|\rangle = q\langle E^{\text{kin}}\rangle$. If one writes

$$q = \sum_{\ell=1}^{\infty} [\beta W(r_m)]^{\ell}/\ell$$

and chooses the Lennard–Jones potential $W(r_m) = -0.77 k_B T_c$, one gets $q = 0.537$. Therefore, assuming small oscillations about r_m, one gets $\langle E^{\text{kin}}\rangle = k_B T_c$, which will lead to $\langle |E^{\text{pot}}|\rangle = 0.537 k_B T_c$, which is about the average energy contribution from a harmonic oscillator. This is consistent with our assumption to set $W(r) = W(r_m)$, which states that the two particles may be in the neighborhood of a fixed equilibrium distance. Therefore, for temperatures in the neighborhood of T_c, Eq. (A2) can be written as

$$\langle E^{\text{int}}\rangle = \langle E^{\text{kin}}\rangle \exp[\beta W(r_m)] \equiv \langle E^{\text{int}}(v)\rangle \exp[\beta W(r_m)] \quad (A3)$$

From Eq. (A3) one gets $E^{\text{int}}(v, r_m) = E^{\text{int}}(v) \exp[\beta W(r_m)]$. The equation

$$\int_\Gamma \mathrm{d}^3 r g(r_m)$$

can be integrated to give

$$\Gamma g(r_m) = 1/n \quad (A4)$$

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Thermal Contact Conductance of Metal/Polymer Joints

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Introduction

POLYMERS and organic materials are being employed to a greater extent in power generating systems, and with greater use follows an increased interest in the thermal transport properties of polymers. These properties include thermal conductivity, heat capacity, and the thermal contact conductance at the interface with other materials. One important consideration where limited knowledge exists is the heat flow across a metal/polymer interface. Currently, a usable and verifiable model does not exist for predicting the thermal performance of metal/polymer joints.

Marotta and Fletcher[1] measured the thermal conductivity and the thermal contact conductance of several widely available thermoplastic and thermosetting polymers. They compared the experimentally measured data with the current thermal contact models developed for metal/metal contact, such as the elastic contact model developed by Mikic[2] and the plastic contact model developed by Cooper et al. (CMY).[3] The Mikic and CMY models are accepted and proven for metal/metal contacts, but it was observed that a new thermal contact model was needed for a metal in contact with a much softer polymer layer with either finite or infinite length (half-space solution).

Problem Statement

The goal was to obtain a verifiable and usable analytical model for the prediction of thermal joint resistance between a metal and a polymer. The scope of this investigation was limited to assuming

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