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Metastable Fluid Flow Described via a Discrete-Velocity Coagulation–Fragmentation Model

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A discrete-velocity Boltzmann model is introduced. It is based on two principles: (i) clusters of particles move in $\mathbb{R}^3$ with seven fixed momenta; (ii) clusters may gain or lose particles according to the rules of Becker–Döring cluster equations. The model provides a kinetic representation of evaporation and condensation. The model is used to obtain macroscopic fluid equations which are valid into the metastable fluid regime, $0 \leqslant \rho < \rho_S + O(\mu^\sigma)$, where σ is any positive number, μ is the inelastic Knudsen number, and ρ_S is the saturation density.

KEY WORDS: Boltzmann equation; evaporation; condensation; cluster; nucleation; shock wave; metastability.

INTRODUCTION

The purpose of this paper is to use a discrete-velocity kinetic model for a gas exhibiting coagulation and fragmentation as a mechanism for deriving the approximate fluid mechanical equations for isothermal metastable fluid flow. It is shown that a careful application of Penrose's construction of metastable states for the space-homogeneous Becker–Döring cluster equations[12] can be used to derive the macroscopic fluid equations for small Knudsen number.

Recall that the space-homogeneous Lebowitz–Penrose version of the Becker–Döring cluster equations[11, 13] describe the dynamics of quantities

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$N^\alpha(t)$, $\alpha=1, 2,\ldots$, which represent the concentration of α-particle clusters or droplets in a condensing vapor. The equations are

$$\frac{dN^\alpha(t)}{dt} = \mathscr{T}_{\alpha-1}(\mathbf{N}) - \mathscr{T}_\alpha(\mathbf{N}), \qquad \alpha = 2, 3,\ldots$$

$$\frac{dN^1(t)}{dt} = -\mathscr{T}_1(\mathbf{N}) - \sum_{\alpha=1}^{\infty} \mathscr{T}_\alpha(\mathbf{N})$$

where

$$\mathscr{T}_\alpha(\mathbf{N}) = a_\alpha N^1(t)\, N^\alpha(t) - b_{\alpha+1} N^{\alpha+1}(t)$$

and N^α denote the components of the infinite vector $\mathbf{N}$. The equilibrium solutions are $N^\alpha_{\text{eq}} = \mathscr{Q}_\alpha z^\alpha$, where $\mathscr{Q}_\alpha = \prod_{r=2}^{\alpha} (a_{r-1}/b_r)$, $\alpha = 2, 3,\ldots$, $\mathscr{Q}_1 = 1$. The density $\rho(t)$ of vapor is given by $\rho(t) = \sum_{\alpha=1}^{\infty} \alpha N^\alpha(t)$ and hence at equilibrium the density is given by $\rho_{\text{eq}} = \sum_{\alpha=1}^{\infty} \alpha \mathscr{Q}_\alpha (N^1)^\alpha$. The case of interest here is when this power series has a finite positive radius of convergence z_S, for then the maximum equilibrium density is given by $\rho_S = \sum_{\alpha=1}^{\infty} \alpha \mathscr{Q}_\alpha (z_S)^\alpha$.

For plausible assumptions on the kinetic coefficients a_α, b_α refs. 1, 2, and 14 have shown that for initial data $N^\alpha(0)$ with $\rho_0 = \sum_{\alpha=1}^{\infty} \alpha N^\alpha(0)$ we have $\rho(t) = \rho_0$ and:

(i) If $0 \leqslant \rho_0 \leqslant \rho_S$, then $\mathbf{N}(t) \to \mathbf{N}_{\text{eq}}$ in X at $t \to \infty$.

(ii) If $\rho_0 > \rho_S$, then $\mathbf{N}(t) \overset{*}{\rightharpoonup} \mathbf{N}_{\text{eq}}$ as $t \to \infty$.

Here X is the Banach space given by $\{N^\alpha,\ \alpha = 1, 2,\ldots, \|\mathbf{N}\| < \infty\}$, $\|\mathbf{N}\| = \sum_{\alpha=1}^{\infty} \alpha\, |N^\alpha|$, and "$\to$" denotes "strong" convergence, while "$\overset{*}{\rightharpoonup}$" denotes "weak *" convergence.

We thus see that equilibrium states strongly attract initial data for $0 \leqslant \rho_0 \leqslant \rho_S$, but for $\rho_0 > \rho_S$ mass conservation $\|\rho(t)\| = \|\rho_0\|$ precludes strong decay and only the weak decay (ii) is obtained. This result suggests the existence of states with $\rho_0 > \rho_S$ which, while not equilibria, possess exceptionally long lifetimes in the region $\rho(t) = \rho_0 > \rho_S$. Such a class of solutions was discussed by Penrose.[12] He proved that there is indeed a class of "metastable" solutions of the Becker–Döring equations with $N_1(0) - z_S$ small and positive which take an exponentially long time to decay to their asymptotic steady states [as predicted by (ii) above]. (An "exponentially long time" means a time that increases more rapidly than any negative power of the given value $\rho_0 - \rho_S$ as $\rho_0 - \rho_S \to 0$.)

A key element of Penrose's construction of the initial data for his metastable states is the use of data that yield the "collision" terms on the right-hand sides of the Becker–Döring equations exponentially small in $N^1 - z_S$. Hence a natural guess is that if one imbeds the Becker–Döring equations into a discrete-velocity Boltzmann structure, a similar construction to Penrose's will give a class of "metastable" states for which the collision terms are asymptotically small in some sense similar to what Penrose has provided. In fact, the model given here provides such states, which are termed "approximate Maxwellians," and the smallness of the collision terms is shown to occur in terms of the convenient macroscopic variables $\rho - \rho_S$, i.e., the collision terms are exponentially small in $\rho - \rho_S$ as $\rho - \rho_S \to 0$.

The existence of such "approximate Maxwellian" metastable states for the Boltzmann structure allows the derivation of the macroscopic fluid equations even into the region $\rho > \rho_S$. The method used is a variant of the Chapman–Enskog expansion and yields the viscous Navier–Stokes equations and a relation for the pressure p which are formally valid in the region $0 \leqslant \rho \leqslant \rho_S + O(\mu^\sigma)$, where σ is any positive number and μ is a Knudsen number. (A schematic diagram for the pressure as a function of density ρ for slow flow is given in Fig. 1.) Thus, in the hydrodynamic limit as $\mu \to 0+$ the domain of validity of the fluid equations contracts to the region $0 \leqslant \rho \leqslant \rho_S$. That is, on the mesoscopic time scale of the kinetic equations metastability is meaningful and the Navier–Stokes equations govern the flow, but on the hydrodynamic time scale obtained as $\mu \to 0+$ the Euler equations govern, but only for states ρ, $0 \leqslant \rho \leqslant \rho_S$. This is another way of looking at the decay results (i), (ii) above.

The results indicate that metastability is a dynamic phenomenon and the dynamics of spatially segregated phases (stable vapor and metastable vapor) can be studied on the kinetic time scale via the viscous Navier–Stokes equations.

The model itself has been presented in refs. 15 and 16 and is based on two simple concepts:

(i) Individual particles coagulate into clusters and these clusters may themselves fragment according to the rules of the Becker–Döring cluster equations.

(ii) Clusters move with seven fixed momenta.

Unlike the usual discrete-velocity models of the Boltzmann equation such as the Broadwell model, the gas at hand is a gas of clusters with no *a priori* bound on the cluster size. Basic ideas used in the construction of the model come from refs. 3 and 8–10.

The model is also motivated by the paper of Chen *et al.*[6] which derived a lattice gas model for a gas admitting coagulation and fragmentation. While lattice gas models are advantageous for machine computation, they do not process the analytical simplicity of discrete-velocity models.

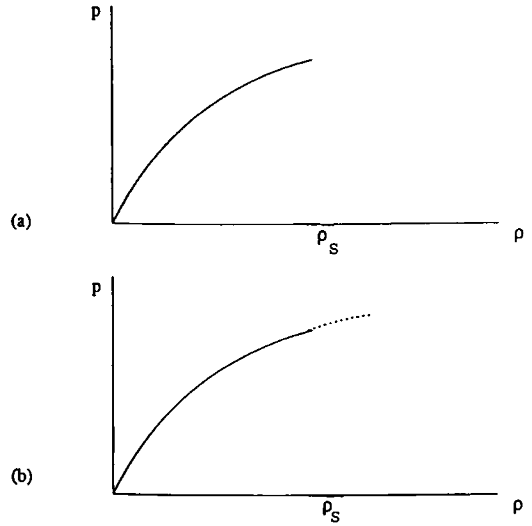

Fig. 1. (a) Fluid dynamic limit: The inelastic Knudsen number is $\mu = 0$ and the compressible "Euler" equations apply for $0 \leqslant \rho \leqslant \rho_S$, where ρ_S is the saturation density of the gas. (b) Small inelastic Knudsen number $\mu > 0$: The compressible viscous "Navier–Stokes" equations apply for $0 \leqslant \rho < \rho_S + O(\mu^\sigma)$, where σ is any positive number, i.e., into the supersaturated metastable regime.

This seems particularly important for the goal at hand, which requires rather careful mathematical analysis. It is here that the simplicity of the model becomes valuable. Further work will provide generalizations to continuous velocity and perhaps lattice gas models.

The paper is organized as follows. Section 2 derives the mathematical model. Section 3 provides the relevant information on the kinetic coefficients and the crucial concept of critical cluster size. Section 4 introduces the idea of an approximate Maxwellian which is the distribution function capable of describing both stable and metastable fluid states. Section 5 derives the "Navier–Stokes" equations valid on $0 \leqslant \rho < \rho_S + O(\mu^\sigma)$, where σ is any positive number, μ the inelastic Knudsen number, and ρ_S the saturation density, via a modification of the Chapman–Enskog expansion.

Notation. $(n_j^\alpha)^p$ means the quantity n_j^α raised to the pth power; $\mathbf{n}$ means the tensor whose components are n_j^α, while $\mathbf{n}^\alpha$ denotes the vector whose components are n_j^α for each fixed α, $\alpha = 1, 2,\ldots$. Finally, $\mathbf{i}, \mathbf{j}, \mathbf{k}$ denotes the standard set of mutually perpendicular unit vectors in $\mathbb{R}^3$.

2. THE MATHEMATICAL MODEL

We consider a discrete-velocity gas of identical particles each of mass m contained in $\mathbb{R}^3$. A point in $\mathbb{R}^3$ is identified with its Euclidean coordinates (x, y, z). The particles are grouped into clusters possessing 1, 2,..., particles. The clusters move with fixed momenta $\mathbf{P}_1 = mc\mathbf{i}$, $\mathbf{P}_2 = -\mathbf{P}_1$, $\mathbf{P}_3 = mc\mathbf{j}$, $\mathbf{P}_4 = -\mathbf{P}_3$, $\mathbf{P}_5 = mc\mathbf{k}$, $\mathbf{P}_6 = -\mathbf{P}_5$, $\mathbf{P}_0 = \mathbf{0}$. A cluster made up of α-particles will be called an α-cluster. It is clear that an α-cluster with momentum $\mathbf{P}_j$ has velocity $\mathbf{v}_j^\alpha = \mathbf{P}_j/m\alpha$.

As the clusters move, they collide in a binary fashion. Both elastic and inelastic collisions are allowed. In terms of the momenta $\mathbf{P}_i$ the elastic collisions will be represented by $\mathbf{P}_1 + \mathbf{P}_2 = \mathbf{P}_3 + \mathbf{P}_4 = \mathbf{P}_5 + \mathbf{P}_6$, while the inelastic collisions are defined by $\mathbf{P}_1 + \mathbf{P}_2 = \mathbf{P}_0$, $\mathbf{P}_3 + \mathbf{P}_4 = \mathbf{P}_0$, $\mathbf{P}_5 + \mathbf{P}_6 = \mathbf{P}_0$, $\mathbf{P}_i + \mathbf{P}_0 = \mathbf{P}_i$ $(i = 1,..., 6)$. Notice that elastic collisions conserve mass, momentum, and energy; inelastic collisions conserve mass and momentum, but not energy. We denote by $n_j^\alpha(x, y, z, t)$ the number density of α-clusters with momentum $\mathbf{P}_j$ at a point (x, y, z) at time $t > 0$, i.e., the number of clusters in this class per unit volume.

A collision of an α-cluster with momentum $\mathbf{P}_i$ and a β-cluster with momentum $\mathbf{P}_j$ which yields a δ-cluster with momentum $\mathbf{P}_k$ and a γ-cluster with momentum $\mathbf{P}_l$ will be represented by $(n_i^\alpha, n_j^\beta) \to (n_k^\delta, n_l^\gamma)$. This notation allows us to write the allowable elastic collisions as follows.

1. *Mechanical collisions*: $(n_1^\alpha, n_2^\alpha) \to (n_1^\alpha, n_2^\alpha)$ (prob. 1/3), (n_3^α, n_4^α) (prob. 1/3), (n_5^α, n_6^α) (prob. 1/3), with similar statements for (n_3^α, n_4^α), (n_5^α, n_6^α).

2. *Exchange collisions*: (a) "head-on" collisions: $(n_1^\alpha, n_2^\beta) \to (n_1^\alpha, n_2^\beta)$ (prob. 1/6), (n_1^β, n_2^α) (prob. 1/6), (n_3^α, n_4^β) (prob. 1/6), (n_3^β, n_4^α) (prob. 1/6), (n_5^α, n_6^β) (prob. 1/6), (n_5^β, n_6^α) (prob. 1/6) with similar statements for (n_3^α, n_4^β) and (n_5^α, n_6^β). (b) "Angle" collisions: $(n_1^\alpha, n_3^\beta) \to (n_1^\alpha, n_3^\beta)$ (prob. 1/2), (n_1^β, n_3^α) (prob. 1/2), with similar statements for (n_1^α, n_5^β) and (n_3^α, n_5^β).

We only allow inelastic collisions of the Becker–Döring type,[11–13] i.e., where an α-cluster may gain or lose a 1-cluster in coagulation or fragmentation, respectively. The coagulation of a 1-cluster with momentum $\mathbf{P}_j$ to form an $(\alpha + 1)$-cluster with momentum $\mathbf{P}_k$ is represented as $(n_i^1, n_j^\alpha) \to (n_k^{\alpha+1})$, while the fragmentation of an $(\alpha + 1)$-cluster with momentum $\mathbf{P}_k$ into a 1-cluster with momentum $\mathbf{P}_i$ and an α-cluster with momentum $\mathbf{P}_j$ will be denoted by $(n_k^{\alpha+1}) \to (n_i^1, n_j^\alpha)$. With this notation the allowable Becker–Döring inelastic collisions are as follows.

1. "Head-on" coagulation: $(n_1^1, n_2^{\alpha-1})$, $(n_1^{\alpha-1}, n_2^1)$, $(n_3^1, n_4^{\alpha-1})$, $(n_3^{\alpha-1}, n_4^1)$, $(n_5^1, n_6^{\alpha-1})$, $(n_5^{\alpha-1}, n_6^1) \to (n_0^\alpha)$.

2. "Moving cluster coagulates with rest cluster": $(n_j^1, n_0^{\alpha-1})$, $(n_j^{\alpha-1}, n_0^1) \to n_j^\alpha$, $j = 1, 2,..., 6$.

3. Reversal of 1 (fragmentation): $(n_0^\alpha) \to (n_1^1, n_2^{\alpha-1})$ (prob. 1/6), $(n_1^{\alpha-1}, n_2^1)$ (prob. 1/6), $(n_3^1, n_4^{\alpha-1})$ (prob. 1/6), $(n_3^{\alpha-1}, n_4^1)$ (prob. 1/6), $(n_5^1, n_6^{\alpha-1})$ (prob. 1/6), $(n_5^{\alpha-1}, n_6^1)$ (prob. 1/6) if $\alpha > 2$; $(n_0^2) \to (n_1^1, n_2^1)$ (prob. 1/3), (n_3^1, n_4^1) (prob. 1/3), (n_5^1, n_6^1) (prob. 1/3).

4. Reversal of 2 (fragmentation): $(n_j^\alpha) \to (n_j^1, n_0^{\alpha-1})$ (prob. 1/2), $(n_0^1, n_j^{\alpha-1})$ (prob. 1/2), $j = 1, 2,..., 6$ if $\alpha > 2$; $(n_j^2) \to (n_j^1, n_0^1)$, $j = 1, 2,..., 6$.

The rate equations governing the motion of clusters are given by the transport equations

$$\frac{\partial n_j^\alpha}{\partial t} + \mathbf{v}_j^\alpha \cdot \nabla n_j^\alpha = E_j^\alpha(\mathbf{n}) + I_j^\alpha(\mathbf{n}), \qquad j = 0, 1, 2,..., 6, \quad \alpha = 1, 2,...$$

The calculation of E_j^α has been given in refs. 9 and 10 according to the rules of discrete-velocity kinetic theory, i.e., these terms are proportional to collisional cross-sectional areas, the relative velocity of the particles before collision, the probability of each admissible collision, and both number densities of the colliding particles.

Clearly $E_0^\alpha = 0$, $\alpha = 1,...,$ and we record the set of E_j^α by their appropriate formulas:

$$\begin{aligned}
E_j^\alpha(\mathbf{n}) = {}& \frac{4}{3}\frac{\sigma_{\alpha\alpha} c}{\alpha}\left(n_{j+2}^\alpha n_{j+3}^\alpha + n_{j+4}^\alpha n_{j+5}^\alpha - 2n_j^\alpha n_{j+1}^\alpha\right) \\
& + \sum_{\beta \neq \alpha}^{\infty} \frac{c\sigma_{\alpha\beta}}{6}\left(\frac{1}{\alpha} + \frac{1}{\beta}\right)\left(n_j^\beta n_{j+1}^\alpha + n_{j+2}^\alpha n_{j+3}^\beta + n_{j+2}^\beta n_{j+3}^\alpha\right. \\
& \left. + n_{j+4}^\alpha n_{j+5}^\beta + n_{j+4}^\beta n_{j+5}^\alpha - 5n_j^\alpha n_{j+1}^\beta\right) \\
& + \sum_{\beta \neq \alpha}^{\infty} c\sigma_{\alpha\beta} R_{\alpha\beta}\left\{n_j^\beta \sum_{k=2}^{5} n_{j+k}^\alpha - n_j^\alpha \sum_{k=2}^{5} n_{j+k}^\beta\right\}, \qquad j = 1, 3, 5 \\
E_j^\alpha(\mathbf{n}) = {}& \frac{4}{3}\frac{\sigma_{\alpha\alpha} c}{\alpha}\left(n_{j+1}^\alpha n_{j+2}^\alpha + n_{j+3}^\alpha n_{j+4}^\alpha - 2n_{j-1}^\alpha n_j^\alpha\right) \\
& + \sum_{\beta \neq \alpha}^{\infty} \frac{c\sigma_{\alpha\beta}}{6}\left(\frac{1}{\alpha} + \frac{1}{\beta}\right)\left(n_{j-1}^\alpha n_j^\beta + n_{j+1}^\alpha n_{j+2}^\beta + n_{j+1}^\beta n_{j+2}^\alpha\right. \\
& \left. + n_{j+3}^\alpha n_{j+4}^\beta + n_{j+3}^\beta n_{j+4}^\alpha - 5n_j^\alpha n_{j-1}^\beta\right) \\
& + \sum_{\beta \neq \alpha}^{\infty} c\sigma_{\alpha\beta} R_{\alpha\beta}\left\{n_j^\beta \sum_{k=1}^{4} n_{j+k}^\alpha - n_j^\alpha \sum_{k=1}^{4} n_{j+k}^\beta\right\}, \qquad j = 2, 4, 6
\end{aligned}$$

where $n_{j+6} = n_j$ in the above formulas.

Here $\sigma_{\alpha\beta}$ and $cR_{\alpha\beta}$ denote the collisional cross-sectional area and magnitude of relative velocities in a (n_j^α, n_k^β) collision, respectively. An estimate of $\sigma_{\alpha\beta}$ is easily obtained by noting that it is proportional to $(r_\alpha + r_\beta)^2$, with r_α, r_β the radii of α- and β-clusters, respectively. Since the volume of an α-cluster is $\frac{4}{3}\pi\alpha r_1^3$, the radius of a spherical α-cluster is $r_1 \alpha^{1/3}$ and hence $\sigma_{\alpha\beta}$ is proportional to $r_1^2(\alpha^{1/3} + \beta^{1/3})^2$. To compute $R_{\alpha\beta}$ we note that the velocity of an n_j^α cluster is $\mathbf{P}_j/m\alpha$ and that of an n_k^β cluster is $\mathbf{P}_k/m\beta$. Hence $R_{\alpha\beta} = (1/\alpha + 1/\beta)$ for "head-on" collisions and $R_{\alpha\beta} = (1/\alpha^2 + 1/\beta^2)^{1/2}$ for collisions at a right angle.

The inelastic collision terms I_j^α are computed according to the rules of proportionality to the probability of each admissible collision and the number densities of coagulating or fragmenting clusters. We then see that, according to our list of inelastic collisions, $I_j^\alpha(\mathbf{n})$ satisfy the following relations.

For $2 \leqslant \alpha$:

$$
\begin{aligned}
I_j^\alpha(\mathbf{n}) = {} & a_{\alpha-1,1}^{0,j} n_j^1 n_0^{\alpha-1} + (1-\delta_{\alpha 2})\, a_{\alpha-1,1}^{j,0} n_j^{\alpha-1} n_0^1 - b_\alpha^j n_j^\alpha \\
& + (-a_{\alpha,1}^{j,j+1} n_j^\alpha n_{j+1}^1 - a_{\alpha,1}^{j,0} n_j^\alpha n_0^1 \\
& + \tfrac{1}{2} b_{\alpha+1}^j n_j^{\alpha+1} + \tfrac{1}{6} b_{\alpha+1}^0 n_0^{\alpha+1}), \qquad j = 1, 3, 5 \\
I_j^\alpha(\mathbf{n}) = {} & a_{\alpha-1}^{0,j} n_0^{\alpha-1} + (1-\delta_{\alpha 2})\, a_{\alpha-1,1}^{j,0} n_j^{\alpha-1} n_0^1 - b_\alpha^j n_j^\alpha \\
& + (-a_{\alpha,1}^{j,j-1} n_j^\alpha n_{j-1}^1 - a_{\alpha,1}^{j,0} n_j^\alpha n_0^1 \\
& + \tfrac{1}{2} b_{\alpha+1}^j n_j^{\alpha+1} + \tfrac{1}{6} b_{\alpha+1}^0 n_0^{\alpha+1}), \qquad j = 2, 4, 6 \\
I_0^\alpha(\mathbf{n}) = {} & -b_\alpha^0 n_0^\alpha + a_{\alpha-1,1}^{2,1} n_2^{\alpha-1} n_1^1 + a_{\alpha-1,1}^{4,3} n_4^{\alpha-1} n_3^1 \\
& + a_{\alpha-1,1}^{6,5} n_6^{\alpha-1} n_5^1 + (1-\delta_{\alpha 2})(a_{1,\alpha-1}^{2,1} n_2^1 n_{\alpha-1}^1 \\
& + a_{1,\alpha-1}^{4,3} n_4^1 n_3^{\alpha-1} + a_{1,\alpha-1}^{6,5} n_6^1 n_5^{\alpha-1}) \\
& + \left(-n_0^\alpha \sum_{k=1}^{6} a_{\alpha,k}^{0,1} n_k^1 + \tfrac{1}{2} \sum_{k=1}^{6} b_{\alpha+1}^k n_k^{\alpha+1}\right)
\end{aligned}
$$

For $\alpha = 1$:.

$$
\begin{aligned}
I_j^1(\mathbf{n}) = {} & -\sum_{\beta=1}^{\infty} \Bigg\{ a_{1,\beta}^{j,j+1} n_j^1 n_{j+1}^\beta + a_{1,\beta}^{j,0} n_j^1 n_0^\beta \\
& - (1+\delta_{\beta 1}) \left(\frac{1}{2} b_{1+\beta}^j n_j^{1+\beta} + \frac{b_{1+\beta}^0}{6} n_0^{1+\beta} \right) \Bigg\}, \qquad j = 1, 3, 5
\end{aligned}
$$

$$I_j^1(\mathbf{n}) = -\sum_{\beta=1}^{\infty} \left\{ a_{1,\beta}^{j,j+1} n_j^1 n_{j-1}^{\beta} + a_{1,\beta}^{j,0} n_j^0 n_0^{\beta} - (1+\delta_{\beta 1}) \left(\frac{1}{2} b_{1+\beta}^{j} n_j^{1+\beta} + \frac{b_{1+\beta}^{0}}{6} n_0^{1+\beta} \right) \right\}, \qquad j=2,4,6$$

$$I_0^1(\mathbf{n}) = -\sum_{\beta=1}^{\infty} \left\{ n_0^1 \sum_{k=1}^{6} a_{1,\beta}^{0,k} n_k^{\beta} - \frac{1}{2}(1+\delta_{\beta 1}) \sum_{k=1}^{6} b_{1+\beta}^{k} n_k^{1+\beta} \right\}$$

The $a_{\alpha,\beta}^{j,k}$ and $b_{\alpha+1}^{j}$ are positive kinetic rate coefficients for coagulation and fragmentation, respectively. The $a_{\alpha,\beta}^{j,k}$ satisfy the principle of detailed balance $a_{\alpha,\beta}^{j,k} = a_{\beta,\alpha}^{j,k} = a_{\alpha,\beta}^{k,j}$.

Here δ_{ij} denotes the Kronecker delta ($\delta_{ij}=1$, $i=j$; $\delta_{ij}=0$, $i \neq j$). It enters the formulas to prevent double counting of collisions when $\alpha = 2$.

It is a straightforward exercise to show

$$\sum_{j=1}^{6} E_j^{\alpha} = 0, \qquad \alpha = 1,\ldots$$

$$\sum_{\alpha=1}^{\infty} E_j^{\alpha} - E_{j+1}^{\alpha} = 0, \qquad j=1,3,5$$

$$\sum_{\alpha=1}^{\infty} \alpha \sum_{j=0}^{6} I_j^{\alpha} = 0$$

$$\sum_{\alpha=1}^{\infty} I_j^{\alpha} - I_{j+1}^{\alpha} = 0, \qquad j=1,3,5$$

Next define the quantity

$$J_{\alpha}(\mathbf{n}) = n_0^{\alpha} \sum_{k=1}^{6} a_{\alpha,1}^{k,0} n_k^1 + (1-\delta_{\alpha 1})\, n_0^1 \sum_{k=1}^{6} a_{\alpha,1}^{k,0} n_k^{\alpha} + (1-\delta_{\alpha 1}) \sum_{k=1,3,5} a_{\alpha,1}^{k,k+1} n_k^{\alpha} n_{k+1}^1 + \sum_{k=2,4,6} a_{\alpha,1}^{k,k+1} n_k^{\alpha} n_{k+1}^1 - \sum_{k=0}^{6} b_{\alpha+1}^{k} n_k^{\alpha+1} \qquad \text{for} \quad 1 \leqslant \alpha$$

It then follows that

$$\sum_{j=0}^{6} I_j^{\alpha} = J_{\alpha-1} - J_{\alpha}, \qquad 2 \leqslant \alpha$$

Let us define

$$n_j^\alpha = \bar{n}_j^\alpha \bar{\rho}$$

where $\bar{\rho}$ is a typical value of the density $\sum_{j=0}^{6} \sum_{\alpha=1}^{\infty} \alpha n_j^\alpha$, with $x = \bar{x}L$, $y = \bar{y}L$, $z = \bar{z}L$, and $ct = \bar{t}L$, where L is a typical macroscopic length. Hence $\bar{n}_j^\alpha$, $\bar{x}$, $\bar{y}$, $\bar{z}$ are dimensionless quantities. Then, for example, we see that

$$\frac{\partial \bar{n}_1^\alpha}{\partial \bar{t}} + \frac{1}{\alpha} \frac{\partial \bar{n}_1^\alpha}{\partial \bar{x}} = \frac{L}{c\bar{\rho}} (E_1^\alpha + I_1^\alpha)$$

Hence we have

$$\frac{\partial \bar{n}_1^\alpha}{\partial \bar{t}} + \frac{1}{\alpha} \frac{\partial \bar{n}_1^\alpha}{\partial \bar{x}} = L r_1^2 \bar{\rho} (\bar{E}_1^\alpha) + \frac{L}{c\bar{\rho}} I_1^\alpha$$

where we define $\bar{E}_1^\alpha$ as E_1^α with $c = 1$, n_j^α replaced by $\bar{n}_j^\alpha$, and $r_1 = 1$ in the definition of $\sigma_{\alpha\beta}$. Next set $\varepsilon = 1/L r_1^2 \bar{\rho}$, which is the dimensionless elastic Knudsen number.

I_1^α possesses both coagulation and fragmentation coefficients. It is natural to assume that the coagulation coefficients $a_{\alpha,\beta}^{j,k}$ scale in a similar way to the elastic collision coefficients, i.e., they are proportional to $c r_1^2$. By consistency we then assume that the fragmentation coefficients $b_{\alpha+1}^j$ are proportional to $c r_1^2 \bar{\rho}$.

Thus, setting

$$a_{\alpha,1}^{j,k} = c r_1^2 \bar{a}_{\alpha,1}^{j,k} \frac{\varepsilon}{\mu}$$

$$b_{\alpha+1}^j = c r_1^2 \bar{\rho} \bar{b}_{\alpha+1}^j \frac{\varepsilon}{\mu}$$

where μ is the dimensionless inelastic Knudsen number, and $\bar{I}_1^\alpha$ is I_1^α with n_j^α replaced by $\bar{n}_j^\alpha$, $a_{\alpha,1}^{j,k}$, $b_{\alpha+1}^j$ replaced by the above definitions, we see that

$$\frac{\partial \bar{n}_1^\alpha}{\partial \bar{t}} + \frac{1}{\alpha} \frac{\partial \bar{n}_1^\alpha}{\partial \bar{x}} = \frac{\bar{E}_1^\alpha}{\varepsilon} + \frac{\bar{I}_1^\alpha}{\mu}$$

Finally, we drop the overbars and we have derived the system

$$\frac{\partial n_1^\alpha}{\partial t} + \mathbf{V}_j^\alpha \cdot \nabla n_j^\alpha = \frac{E_j^\alpha}{\varepsilon} + \frac{I_j^\alpha}{\mu} \qquad (2.1)$$

$$\mathbf{V}_j^\alpha = \frac{\mathbf{e}_j}{\alpha}, \qquad j = 1,..., 6 \qquad (2.2)$$

$$\mathbf{e}_1 = \mathbf{i}, \qquad \mathbf{e}_3 = \mathbf{j}, \qquad \mathbf{e}_5 = \mathbf{k}, \qquad \mathbf{e}_{j+1} = -\mathbf{e}_j, \quad j = 1, 3, 5 \qquad (2.3)$$

Next define

$$N^\alpha \doteq \sum_{j=0}^{6} n_j^\alpha$$

$$N^\alpha u^\alpha \doteq \frac{1}{\alpha}(n_1^\alpha - n_2^\alpha)$$

$$N^\alpha v^\alpha \doteq \frac{1}{\alpha}(n_3^\alpha - n_4^\alpha)$$

$$N^\alpha w^\alpha \doteq \frac{1}{\alpha}(n_5^\alpha - n_6^\alpha)$$

$$\mathbf{u}^\alpha \doteq (u^\alpha, v^\alpha, w^\alpha)$$

$$\rho \doteq \sum_{\alpha=1}^{\infty} \alpha N^\alpha$$

$$\rho u \doteq \sum_{\alpha=1}^{\infty} \alpha N^\alpha u^\alpha = \sum_{\alpha=1}^{\infty} (n_1^\alpha - n_2^\alpha)$$

$$\rho v \doteq \sum_{\alpha=1}^{\infty} \alpha N^\alpha v^\alpha = \sum_{\alpha=1}^{\infty} (n_3^\alpha - n_4^\alpha)$$

$$\rho w \doteq \sum_{\alpha=1}^{\infty} \alpha N^\alpha w^\alpha = \sum_{\alpha=1}^{\infty} (n_5^\alpha - n_6^\alpha)$$

$$\mathbf{u} = (u, v, w)$$

Then the equations for transport of α-clusters are

$$\frac{\partial}{\partial t}(N^\alpha) + \operatorname{div}(N^\alpha \mathbf{u}^\alpha) = \frac{J_{\alpha-1} - J_\alpha}{\mu}, \qquad 2 \leqslant \alpha \tag{2.4}$$

$$\frac{\partial}{\partial t} N^1 + \operatorname{div}(N^1 \mathbf{u}^1) = \frac{-J_1 - \sum_{\alpha=1}^{\infty} J_\alpha}{\mu} \tag{2.5}$$

the equation for conservation of mass is

$$\frac{\partial}{\partial t}\rho + \operatorname{div}(\rho \mathbf{u}) = 0 \tag{2.6}$$

and the equations for conservation of linear momentum are

$$\begin{aligned}
&\frac{\partial}{\partial t}(\rho u) + \frac{\partial}{\partial x} \sum_{\alpha=1}^{\infty} \frac{1}{\alpha}(n_1^\alpha + n_2^\alpha) = 0 \\
&\frac{\partial}{\partial t}(\rho v) + \frac{\partial}{\partial y} \sum_{\alpha=1}^{\infty} \frac{1}{\alpha}(n_3^\alpha + n_4^\alpha) = 0 \\
&\frac{\partial}{\partial t}(\rho w) + \frac{\partial}{\partial z} \sum_{\alpha=1}^{\infty} \frac{1}{\alpha}(n_5^\alpha + n_6^\alpha) = 0
\end{aligned} \tag{2.7}$$

[We note that (2.4) is derived by Friedlander[7] from the "general dynamic equation for the continuous distribution function." Friedlander's presentation possesses mass but not momentum conservation, and coagulation but not fragmentation kinetics. See also ref. 4 for a related approach.]

If we define the macroscopic symmetric tensor Π by

$$\begin{aligned}
\Pi_{xx} &= -\rho u^2 + \sum \frac{1}{\alpha}(n_1^\alpha + n_2^\alpha) \\
\Pi_{yy} &= -\rho v^2 + \sum \frac{1}{\alpha}(n_3^\alpha + n_4^\alpha) \\
\Pi_{zz} &= -\rho w^2 + \sum \frac{1}{\alpha}(n_5^\alpha + n_6^\alpha) \\
\Pi_{xy} &= -\rho uv \\
\Pi_{xz} &= -\rho uw \\
\Pi_{yz} &= -\rho vw
\end{aligned}$$

then the conservation of linear momentum may be expressed in the familiar form

$$\frac{\partial}{\partial t}(\rho \mathbf{u}) + \operatorname{div}(\rho \mathbf{u} \otimes \mathbf{u} + \mathbf{\Pi}) = 0 \tag{2.8}$$

The hydrodynamic pressure is $p = \frac{1}{3}$ trace $\mathbf{\Pi}$, so that

$$p \doteq \frac{1}{3}\left[-\rho\, |\mathbf{u}|^2 + \sum_{\alpha=1}^{\infty} \frac{1}{\alpha} \sum_{j=1}^{6} n_j^\alpha \right]$$

We define the *elastic* and *inelastic Maxwellian states* as to those $\mathbf{n}$ for which all $E_j^\alpha = 0$ and $I_j^\alpha = 0$, respectively. The elastic Maxwellian states have been computed in ref. 10, p. 66, and have the form

$$\begin{aligned}\mathbf{n}^\alpha = {} & (0, e^{c_1}, e^{-c_1}, e^{c_2}, e^{-c_2}, e^{c_3}, e^{-c_3})\, C^\alpha \\ & + (n_0^\alpha, 0, 0, 0, 0, 0, 0), \qquad \alpha = 1, 2,... \end{aligned} \tag{2.9}$$

where the parameters $C^\alpha > 0$, c_1, c_2, c_3 may depend on (x, y, z, t). Notice since n_0^α does not enter the elastic collision terms, it is as yet unconstrained. We define $\mathbf{c} = (c_1, c_2, c_3)$.

The inelastic Maxwellians are not so readily identified. What is rather straightforward to do is to obtain the inelastic Maxwellians which are also elastic Maxwellians. For convenience we call such states simply Maxwellians.

For simplicity we will only consider the case

$$\begin{aligned} a_\beta^{j,k} &= a_\beta, \qquad j, k = 0, 1,..., 6, \quad 1 \leqslant \beta \\ b_\beta^j &= b_\beta, \qquad j = 1,..., 6, \quad 2 \leqslant \beta \\ b_\beta^0 &= 3b_\beta, \qquad 2 \leqslant \beta \end{aligned}$$

The reason for the value 3 in last assumption becomes clear in the computation of the inelastic Maxwellians. In fact, any positive constant would suffice with a corresponding change in our formulas.

As shown in Appendix A, *the Maxwellian states are given by*

$$\mathbf{n}^\alpha = (1, e^{c_1}, e^{-c_1}, e^{c_2}, e^{-c_2}, e^{c_3}, e^{-c_3})\, Q_\alpha (n_0^1)^\alpha \tag{2.10}$$

where

$$Q_\alpha = 2^{\alpha - 2} \frac{a_{\alpha-1} \cdots a_1}{b_\alpha \cdots b_2}, \qquad \alpha = 2, 3,...; \qquad Q_1 = 1 \tag{2.11}$$

Next define the series

$$\mathscr{P}_k(z) \doteq \sum_{\alpha=1}^{\infty} \alpha^k Q_\alpha (z)^\alpha, \qquad k = -1, 0, 1 \tag{2.12}$$

We then see that on Maxwellians the four macroscopic variables ρ, ρu, ρv, ρw can be related to the four Maxwellian parameter c_1, c_2, c_3, n_0^1 by the relations

$$\begin{aligned} \rho &= (1 + 2\cosh c_1 + 2\cosh c_2 + 2\cosh c_3)\, \mathscr{P}_1(n_0^1) \\ \rho u &= 2\sinh c_1\, \mathscr{P}_0(n_0^1) \\ \rho v &= 2\sinh c_2\, \mathscr{P}_0(n_0^1) \\ \rho w &= 2\sinh c_3\, \mathscr{P}_0(n_0^1) \end{aligned} \tag{2.13}$$

We also see that the hydrostatic pressure p satisfies

$$p + \tfrac{1}{3}\rho\, |\mathbf{u}|^2 = \tfrac{2}{3}(\cosh c_1 + \cosh c_2 + \cosh c_3)\, \mathscr{P}_{-1}(n_0^1) \tag{2.14}$$

3. KINETIC COEFFICIENTS AND CRITICAL CLUSTERS

So far we have not made any restrictions beyond positivity on the kinetic coefficients a_β, b_β. Following Penrose,[12] we now make the following physically motivated assumptions:

(i) There exist positive constants A, A', κ, with $0<\kappa<1$, such that

$$A' < a_\beta < A\beta^\kappa \qquad (\beta = 1, 2,...) \tag{3.1}$$

(ii) We have

$$\lim_{\beta\to\infty} \frac{b_{\beta+1}}{b_\beta} = 1 \tag{3.2}$$

(iii) The sequence b_β/a_β is monotonic decreasing, with a positive limit which we call z_S, i.e.,

$$\frac{b_{\beta+1}}{a_{\beta+1}} \leqslant \frac{b_\beta}{a_\beta} \qquad (\beta = 2, 3,...) \tag{3.3}$$

$$\lim_{\beta\to\infty} \frac{b_\beta}{2a_\beta} = z_S > 0 \tag{3.4}$$

(iv) The sequence b_β/a_β converges to its limit like a negative power of β, but not as rapidly as β^{-1}; that is, there exist positive constants γ, γ', G, G' satisfying

$$0 < \gamma < 1, \qquad 0 < \gamma'$$

such that

$$z_S \exp(G\beta^{-\gamma}) < \frac{b_\beta}{2a_\beta} < z_S \exp(G'\beta^{-\gamma'}) \tag{3.5}$$

One consequence of the assumptions (i)–(iv) is that the series

$$\sum_{\alpha=1}^{\infty} \alpha Q_\alpha(z)^\alpha \tag{3.6}$$

has by (3.2), (3.4) the positive radius of convergence

$$\lim_{\beta\to\infty} \frac{b_{\beta+1}}{2a_\beta} = z_S$$

Moreover, (3.5) and Theorem 2 of ref. 10 imply that the series (3.6) actually converges when $z = z_S$. Its sum will be denoted by ρ'_S:

$$\rho'_S = \sum_{\alpha=1}^{\infty} \alpha Q_\alpha (z_S)^\alpha < \infty \tag{3.7}$$

We observe that on Maxwellians the density is well defined and given by

$$\rho = (1 + 2\cosh c_1 + 2\cosh c_2 + 2\cosh c_3)\, \mathscr{P}_1(n_0^1)$$

when $0 \leqslant n_0^1 \leqslant z_S$. The maximum value of this quantity for fixed $\mathbf{c}$ is then

$$\rho_S = (1 + 2\cosh c_1 + 2\cosh c_2 + 2\cosh c_3)\, \rho'_S \tag{3.8}$$

This sum is usually interpreted as the *density of saturated vapor* and for convenience we will not subsequently denote its dependence on $\mathbf{c}$. For $0 \leqslant n_0 \leqslant z_S$, $\rho\mathbf{u}$ and p are also well defined. On macroscopic equilibrium Maxwellians the hydrostatic pressure satisfies

$$p = 2\mathscr{P}_{-1}(n_0^1), \qquad 0 \leqslant n_0^1 \leqslant z_S \tag{3.9}$$

From the chain rule and the positivity of the kinetic coefficients $dp/d\rho = (dp/dn_0^1)/(d\rho/dn_0^1)$ we trivially observe that p is a monotone increasing function of ρ. Of course we can eliminate n_0^1 from (3.8), (3.9) to find p explicitly as a function of ρ in terms of a power series in ρ:

$$p = 2[\tfrac{1}{7}\rho - \tfrac{3}{2}\cdot\tfrac{1}{49} Q_2 \rho^2 + \cdots] \tag{3.10}$$

which is valid for $0 \leqslant \rho \leqslant \rho_S$, $c_1 = c_2 = c_3 = 0$.

For fixed $z > 0$ one could define the critical cluster size α^* as that value of α that minimizes the quantity $Q_\alpha z^\alpha$, i.e., for a Maxwellian state given by (2.10) we will have

$$n_j^{\alpha^*} \leqslant n_j^\beta, \qquad \beta \neq \alpha^* \tag{3.11}$$

where $0 \leqslant j \leqslant 6$. For computational convenience, however, we follow Penrose[12] and define the critical cluster size α^* as that value of α that minimizes the quantity $a_\alpha Q_\alpha(z)^\alpha$, i.e., for a Maxwellian state given by (2.10) we have

$$a_{\alpha^*} n_j^{\alpha^*} \leqslant a_\beta n_j^\beta, \qquad \beta \neq \alpha^* \tag{3.12}$$

where $0 \leqslant j \leqslant 6$. We then readily see that $a_\alpha Q_\alpha z^\alpha$ is monotonically decreasing in α for z fixed when

$$a_\alpha Q_\alpha(z)^\alpha \leqslant a_{\alpha-1} Q_{\alpha-1}(z)^{\alpha-1}$$

or equivalently

$$2^{\alpha-2} \frac{a_\alpha \cdots a_1}{b_\alpha \cdots b_2} (z)^\alpha \leqslant 2^{\alpha-3} \frac{a_{\alpha-1} \cdots a_1}{b_{\alpha-1} \cdots b_2} (z)^{\alpha-1}$$

that is, those α for which $z \leqslant \frac{1}{2} b_\alpha / a_\alpha$. On the other hand, $a_\alpha Q_\alpha z^\alpha$ will be monotonically increasing in α for z fixed for those α for which

$$a_{\alpha+1} Q_{\alpha+1} z^{\alpha+1} \geqslant a_\alpha Q_\alpha z^\alpha$$

i.e., those α for which $z \geqslant \frac{1}{2} b_{\alpha+1} / a_{\alpha+1}$. Hence, since b_α / a_α is monotone decreasing by (iii), we see for α^* that

$$\frac{1}{2} \frac{b_{\alpha^*+1}}{a_{\alpha^*+1}} \leqslant z \leqslant \frac{1}{2} \frac{b_{\alpha^*}}{a_{\alpha^*}} \tag{3.13}$$

we have $a_\alpha Q_\alpha z^\alpha$ decreasing for $1 \leqslant \alpha < \alpha^*$, $a_\alpha Q_\alpha z^\alpha$ increasing for $\alpha^* < \alpha$. For fixed z the α^* satisfying (3.13) thus defines the critical cluster size (in the sense of Penrose). We take $b_1 = \infty$ to take care of the case when $z > b_2/2a_2$. Since for $0 < z < z_S$ the ratio test tells us that $\alpha^* = \infty$, we know that as $z \searrow z_S$, $\alpha^* \to \infty$. One may think of the cluster with $\alpha = \alpha^*$ as a condensation nucleus from which larger supersaturated vapor phase clusters form. Of course this computation only makes sense on Maxwellians, but it allows us to identify clusters with $1 \leqslant \alpha < \alpha^*$ as being vapor clusters and clusters with $\alpha^* \leqslant \alpha$ as being supersaturated vapor clusters. In particular the above designation allows us to quantify the supersaturated vapor and vapor components of our gas.

We also note that along elastic Maxwellians an H-theorem can be proved. This is given in Appendix B.

4. APPROXIMATE MAXWELLIANS

In Section 2 we defined a Maxwellian state $\mathbf{n} = (n_j^\alpha)$ as an elastic Maxwellian which is also an inelastic Maxwellian, i.e., a solution to (i) $E_j^\alpha(\mathbf{n}) = 0$, $1 \leqslant \alpha$, (ii) $I_0^\alpha(\mathbf{n}) = 0$, $1 \leqslant \alpha$, (iii) $0 = J_{\alpha-1}(\mathbf{n}) - J_\alpha(\mathbf{n})$, $2 \leqslant \alpha < \infty$, and (iv) $0 = -J_1(\mathbf{n}) - \sum_{\alpha=1}^{\infty} J_\alpha(\mathbf{n})$. As noted in ref. 12, nucleation theory tends to regard $J_\alpha(\mathbf{n})$ taking on a common α-independent value as more sensible thermodynamic equilibrium in the space-homogeneous case. For example, such a state, if it was also an elastic Maxwellian, would satisfy (i), (iii). Leaving aside for a moment condition (ii), we see that the classical theory cannot satisfy (iv), which followed from conservation of mass. Hence we can either disregard mass conservation, which seems unphysical, or introduce a new idea suggested by Penrose's theory and define an *approximate Maxwellian state.* An approximate Maxwellian state will satisfy (i) exactly, but (ii)–(iv) will be satisfied approximately when $\rho(\mathbf{n}) > \rho_S$ and exactly when $0 \leqslant \rho(\mathbf{n}) \leqslant \rho_S$. [Notice since (i) is satisfied exactly, the parameter $\mathbf{c}$ is determined by virtue of $\mathbf{n}$ being a fixed elastic Maxwellian.] Thus, for unsaturated and saturated states approximate Maxwellians will be true Maxwellians. *The importance of approximate Maxwellians is that they define metastable states for* $\rho_S < \rho$.

Before giving a precise definition of approximate Maxwellian, we recall some terminology used by Penrose.[12]

Definition 4.1. Let $q(z)$ represent any quantity depending on z. Then "$q(z)$ exponentially small in $(z - z_S)$" means for all positive m we have $q(z) = O(z - z_S)^m$, i.e., $q(z)/(z - z_S)^m$ is bounded as $z \searrow z_S$.

The expression "$q(z)$ is at most algebraically large in $(z - z_S)$" means for some positive m we have $q(z) = O(z - z_S)^{-m}$.

We can now define an approximate Maxwellian.

Definition 4.2. An *approximate Maxwellian* $\mathbf{n}$ is (i) an elastic Maxwellian with associated density $\rho(\mathbf{n})$ so that (ii) $I_0^\alpha(\mathbf{n})$, $1 \leqslant \alpha$, (iii) $J_{\alpha-1}(\mathbf{n}) - J_\alpha(\mathbf{n})$, $2 \leqslant \alpha$, (iv) $J_1(\mathbf{n}) + \sum_{\alpha=1}^{\infty} J_\alpha(\mathbf{n})$ are identically zero when $0 \leqslant \rho(\mathbf{n}) \leqslant \rho_S$ and exponentially small in $\rho(\mathbf{n}) - \rho_S$ as $\rho(\mathbf{n}) \searrow \rho_S$. Furthermore, $J_\alpha(\mathbf{n}) = 0$, $1 \leqslant \alpha$, $0 \leqslant \rho(\mathbf{n}) \leqslant \rho_S$.

For $\rho > \rho_S$ the approximate Maxwellian states are defined as the *metastable states.* Again recall that since $\mathbf{n}$ is a fixed elastic Maxwellian, $\mathbf{c}$ is fixed for $\rho(\mathbf{n})$ and ρ_S.

Approximate Maxwellians have now been defined. The next step is to construct such states. This is done in the following lemmas and theorem.

Lemma 4.3. Set

$$J(z) = \left\{ \frac{1}{a_1(z)^2} + \sum_{\alpha=2}^{\infty} \frac{1}{a_\alpha Q_\alpha(z)^{\alpha+1}} \right\}^{-1}$$

$$f^1(z) = z$$

$$f^\alpha(z) = Q_\alpha(z)^\alpha J(z) \left\{ \sum_{\beta=\alpha}^{\infty} \frac{1}{a_\beta Q_\beta(z)^{\beta+1}} \right\}, \qquad z > z_S$$

We now follow Penrose[12] and use his concept of a space-homogeneous metastable equilibrium as the basis for our candidate for an approximate Maxwellian. Specifically we set

$$g^{\alpha}(z)=\begin{cases} Q_{\alpha}(z)^{\alpha}, & 1\leqslant\alpha, \quad 0\leqslant z\leqslant z_S \\ f^{\alpha}(z), & \alpha\leqslant\alpha^*, \quad z>z_S \\ Q_{\alpha}(z_S)^{\alpha}, & \alpha>\alpha^*, \quad z>z_S \end{cases} \tag{4.1}$$

where α^* (which depends on z) is the critical cluster size defined in Section 3.

We now state the main result of this section.

Theorem 4.4. The state

$$\mathbf{m}^{\alpha}=(1, e^{d_1}, e^{-d_1}, e^{d_2}, e^{-d_2}, e^{d_3}, e^{-d_3})\, g^{\alpha}(m_0^1) \tag{4.2}$$

is an approximate Maxwellian.

The proof will be based on a lemma and theorem of Penrose[12] and some additional lemmas. It is given in Appendic C.

5. THE CHAPMAN–ENSKOG EXPANSION

In this section we perform the Chapman–Enskog expansion to obtain the fluid dynamic approximation to the kinetic equations for μ small. The presentation follows the formulation given in the paper of Chen *et al.*[5] The computation will show that ρ, $\rho\mathbf{u}$ satisfy the "Navier–Stokes"-like system

$$\frac{\partial\rho}{\partial t}+\operatorname{div}(\rho\mathbf{u})=0 \tag{5.1}$$

$$\frac{\partial(\rho u)}{\partial t}+\frac{\partial}{\partial x}\sum_{\alpha=1}^{\infty}\frac{m_1^{\alpha}+m_2^{\alpha}}{\alpha}=O(\mu) \tag{5.2}$$

$$\frac{\partial(\rho v)}{\partial t}+\frac{\partial}{\partial y}\sum_{\alpha=1}^{\infty}\frac{m_3^{\alpha}+m_4^{\alpha}}{\alpha}=O(\mu) \tag{5.3}$$

$$\frac{\partial(\rho w)}{\partial t}+\frac{\partial}{\partial z}\sum_{\alpha=1}^{\infty}\frac{m_5^{\alpha}+m_6^{\alpha}}{\alpha}=O(\mu) \tag{5.4}$$

where $\hat{\mathbf{m}}(\rho, \rho\mathbf{u})$ is the approximate Maxwellian given in Theorem 4.4 expressed in terms of the macroscopic quantities ρ, $\rho\mathbf{u}$. The "Navier–

Stokes"-like system (5.1)–(5.4) will be valid into the saturated regime as long as $0 \leqslant \rho \leqslant \rho_S + O(\mu^\sigma)$, where σ is any positive constant. For slow flow (5.2)–(5.4) will deliver an equation of state schematically suggested by Fig. 1. In the fluid mechanical limit $\mu \to 0+$ the domain of validity of (5.1)–(5.4) collapses to the states for which $0 \leqslant \rho \leqslant \rho_S$.

The expansion procedure is a bit tedious, but important, since it shows the crucial role of the approximate Maxwellian. We give it below.

Assume that we have let $\varepsilon \to 0+$ in (2.1), so that the ε-limit microscopic state $\mathbf{n}$ is an elastic Maxwellian and satisfies (2.5)–(2.8). Next we assume the n_j^α can be expressed in terms of the macroscopic variables ρ, $\rho\mathbf{u}$, where $\mathbf{u}$ is the velocity vector (u, v, w), i.e., $n_j^\alpha = K_j^\alpha(\rho, \rho\mathbf{u})$. We write the macroscopic conservation laws of mass and momentum (2.7), (2.8) as

$$\frac{\partial \rho}{\partial t} + \operatorname{div}(\rho\mathbf{u}) = 0 \tag{5.5}$$

$$\frac{\partial(\rho u)}{\partial t} + \frac{\partial}{\partial x} \sum_{\alpha=1}^{\infty} \left(\frac{K_1^\alpha + K_2^\alpha}{\alpha}\right) = 0 \tag{5.6}$$

$$\frac{\partial(\rho v)}{\partial t} + \frac{\partial}{\partial y} \sum_{\alpha=1}^{\infty} \left(\frac{K_3^\alpha + K_4^\alpha}{\alpha}\right) = 0 \tag{5.7}$$

$$\frac{\partial(\rho w)}{\partial t} + \frac{\partial}{\partial z} \sum_{\alpha=1}^{\infty} \left(\frac{K_5^\alpha + K_6^\alpha}{\alpha}\right) = 0 \tag{5.8}$$

plus the kinetic relations

$$\frac{\partial K^\alpha}{\partial t} + \operatorname{div}(K^\alpha \mathbf{u}^\alpha) = \frac{J_{\alpha-1} - J_\alpha}{\mu} \tag{5.9}$$

$$\frac{\partial K^1}{\partial t} + \operatorname{div}(K^1 \mathbf{u}^1) = \frac{-J_1 - \sum_{\alpha=1}^{\infty} J_\alpha}{\mu} \tag{5.10}$$

$$\frac{\partial K_0^\alpha}{\partial t} = \frac{I_0^\alpha}{\mu} \tag{5.11}$$

where $K^\alpha = \sum_{j=0}^{6} K_j^\alpha$, $\mathbf{u} = (u, v, w)$, $\mathbf{u}^\alpha = (u^\alpha, v^\alpha, w^\alpha)$.

Since K_j^α depend on x, y, z, t through ρ, $\rho\mathbf{u}$, we can use the chain rule to compute (5.9), (5.10) via (5.5)–(5.8):

$$
\begin{aligned}
&-\frac{\partial K^\alpha}{\partial \rho}(\operatorname{div}(\rho \mathbf{u}))-\frac{\partial K^\alpha}{\partial(\rho u)}\left(\frac{\partial}{\partial x}\sum_{\alpha=1}^{\infty}\left(\frac{K_1^\alpha+K_2^\alpha}{\alpha}\right)\right)\\
&\quad-\frac{\partial K^\alpha}{\partial(\rho v)}\left(\frac{\partial}{\partial y}\sum_{\alpha=1}^{\infty}\left(\frac{K_3^\alpha+K_4^\alpha}{\alpha}\right)\right)\\
&\quad-\frac{\partial K^\alpha}{\partial(\rho w)}\left(\frac{\partial}{\partial z}\sum_{\alpha=1}^{\infty}\left(\frac{K_5^\alpha+K_6^\alpha}{\alpha}\right)\right)+\frac{\partial}{\partial x}\left(\frac{K_1^\alpha-K_2^\alpha}{\alpha}\right)\\
&\quad+\frac{\partial}{\partial y}\left(\frac{K_3^\alpha-K_4^\alpha}{\alpha}\right)+\frac{\partial}{\partial z}\left(\frac{K_5^\alpha-K_6^\alpha}{\alpha}\right)\\
&\quad=\begin{cases}\dfrac{J_{\alpha-1}(\mathbf{n})-J_\alpha(\mathbf{n})}{\mu}, & 2\leqslant\alpha\\[2ex] \dfrac{-J_1(\mathbf{n})-\sum_{\alpha=1}^{\infty}J_\alpha(\mathbf{n})}{\mu}, & \alpha=1\end{cases}
\end{aligned}
\tag{5.12}
$$

$$
\begin{aligned}
&-\frac{\partial K_0^\alpha}{\partial \rho}(\operatorname{div}(\rho \mathbf{u}))-\frac{\partial K_0^\alpha}{\partial(\rho u)}\left(\frac{\partial}{\partial x}\sum_{\alpha=1}^{\infty}\left(\frac{K_1^\alpha+K_2^\alpha}{\alpha}\right)\right)\\
&\quad-\frac{\partial K_0^\alpha}{\partial(\rho v)}\left(\frac{\partial}{\partial y}\sum_{\alpha=1}^{\infty}\left(\frac{K_3^\alpha+K_4^\alpha}{\alpha}\right)\right)\\
&\quad-\frac{\partial K_0^\alpha}{\partial(\rho w)}\left(\frac{\partial}{\partial z}\sum_{\alpha=1}^{\infty}\left(\frac{K_5^\alpha+K_6^\alpha}{\alpha}\right)\right)\\
&\quad=\frac{I_0^\alpha(\mathbf{n})}{\mu}
\end{aligned}
\tag{5.13}
$$

Now we expand $K_j^\alpha(\rho, \rho\mathbf{u})$ in an asymptotic expansion in μ:

$$K_j^\alpha(\rho, \rho\mathbf{u}) = m_j^\alpha(\rho, \rho\mathbf{u}) + \mu w_j^\alpha(\rho, \rho\mathbf{u}) + \cdots \tag{5.14}$$

where $\mathbf{m}$ is the approximate Maxwellian given by (4.2). It is easy to see that $\mathbf{m}$ can be expressed in the macroscopic quantities ρ, $\rho\mathbf{u}$: we just set

$$\rho = (1 + 2\cosh d_1 + 2\cosh d_2 + 2\cosh d_3) \sum_{\alpha=1}^{\infty} \alpha g^\alpha(m_0^1) \tag{5.15}$$

$$\rho u = 2\sinh d_1 \sum_{\alpha=1}^{\infty} g^\alpha(m_0^1) \tag{5.16}$$

$$\rho v = 2\sinh d_2 \sum_{\alpha=1}^{\infty} g^\alpha(m_0^1) \tag{5.17}$$

$$\rho w = 2\sinh d_3 \sum_{\alpha=1}^{\infty} g^\alpha(m_0^1) \tag{5.18}$$

Equations (5.16)–(5.18) define d_1, d_2, d_3 in terms of $\rho\mathbf{u}$, m_0^1. Substitution of these relations into (5.15) defines m_0^1 in terms of ρ, $\rho\mathbf{u}$. Hence we obtain the functional relations

$$d_i = \hat{d}_i(\rho, \rho\mathbf{u}), \qquad m_0^1 = \hat{m}_0^1(\rho, \rho\mathbf{u}), \qquad i = 1, 2, 3 \tag{5.19}$$

and also

$$m^\alpha = [1 + 2\cosh \hat{d}_1(\rho, \rho\mathbf{u}) + 2\cosh \hat{d}_2(\rho, \rho\mathbf{u}) + 2\cosh \hat{d}_3(\rho, \rho\mathbf{u})] \times g^\alpha(\hat{m}_0^1(\rho, \rho\mathbf{u}))$$

where $m^\alpha = \sum_{j=0}^6 m_j^\alpha$.

Our goal now is to find $w_j^\alpha(\rho, \rho\mathbf{u})$ by substituting the expansion (5.14) into (5.12), (5.13) and matching terms of order one.

First we must determine the structure of w_j^α. Since $\mathbf{n}$ is an elastic Maxwellian, it satisfies (2.9), while $\mathbf{m}$ satisfies (4.2). Hence

$$\mathbf{n} = (n_0^\alpha, C^\alpha e^{c_1}, C^\alpha e^{-c_1}, C^\alpha e^{c_2}, C^\alpha e^{-c_2}, C^\alpha e^{c_3}, C^\alpha e^{-c_3}) \tag{5.20}$$

$$\mathbf{m} = (1, e^{d_1}, e^{-d_1}, e^{d_2}, e^{-d_2}, e^{d_3}, e^{-d_3})\, g^\alpha(m_0^1) \tag{5.21}$$

with

$$n_0^\alpha = g^\alpha + \mu k^\alpha + O(\mu^2), \qquad C^\alpha = g^\alpha + \mu h^\alpha + O(\mu^2), \qquad c_i = d_i + \mu l_i + O(\mu^2)$$
$$i = 1, 2, 3, \qquad \alpha \geqslant 1$$

Then we see

$$C^\alpha e^{c_i} = g^\alpha e^{d_i} + \mu e^{d_i}(h^\alpha + g^\alpha l_i) + O(\mu^2)$$
$$C^\alpha e^{-c_i} = g^\alpha e^{-d_i} + \mu e^{-d_i}(h^\alpha - g^\alpha l_i) + O(\mu^2)$$
$$\mathbf{w} = (k^\alpha, e^{d_1}(h^\alpha + g^\alpha l_1), e^{-d_1}(h^\alpha - g^\alpha l_1), e^{d_2}(h^\alpha + g^\alpha l_2), e^{-d_2}(h^\alpha - g^\alpha l_2), e^{d_3}(h^\alpha + g^\alpha l_3), e^{-d_3}(h^\alpha - g^\alpha l_3)) \tag{5.22}$$

and with

$$\mathscr{R} = 2(l_1 \sinh d_1 + l_2 \sinh d_2 + l_3 \sinh d_3)$$

we have

$$C^\alpha S_\mathbf{c} = g^\alpha S_\mathbf{d} + \mu(h^\alpha S_\mathbf{d} + g^\alpha \mathscr{R}) + O(\mu^2)$$
$$S_\mathbf{c} = 2(\cosh c_1 + \cosh c_2 + \cosh c_3) \tag{5.23}$$
$$S_\mathbf{d} = 2(\cosh d_1 + \cosh d_2 + \cosh d_3)$$

Next we recall that along an elastic Maxwellian **n**

$$J_\alpha(\mathbf{n}) = S_c a_\alpha n_0^\alpha C^1 + S_c(1-\delta_{\alpha 1})\, C^\alpha a_\alpha n_0^1 + (6-3\delta_{\alpha 1})\, a_\alpha C^1 C^\alpha - b_{\alpha+1} C^{\alpha+1} S_c - 3b_{\alpha+1} n_0^{\alpha+1}$$

while

$$J_\alpha(\mathbf{m}) = S_d a_\alpha g^\alpha g^1 + S_d(1-\delta_{\alpha 1})\, g^\alpha a_\alpha g^1 + (6-3\delta_{\alpha 1})\, a_\alpha g^1 g^\alpha - b_{\alpha+1} g^{\alpha+1} S_d - 3b_{\alpha+1} g^{\alpha+1}$$

which when combined with (5.23) yields

$$\frac{J_\alpha(\mathbf{n}) - J_\alpha(\mathbf{m})}{\mu} = \delta J_\alpha(\mathbf{m}) + O(\mu), \qquad \alpha \geqslant 1 \tag{5.24}$$

where

$$\begin{aligned}\delta J_\alpha(\mathbf{m}) = {} & S_d a_\alpha (g^\alpha h^1 + k^\alpha g^1) + \mathcal{R} a_\alpha (g^\alpha g^1) \\ & + S_d a_\alpha (1-\delta_{\alpha 1})(h^\alpha g^1 + g^\alpha h^1) + \mathcal{R} a_\alpha (1-\delta_{\alpha 1})(g^\alpha g^1) \\ & + (6-3\delta_{\alpha 1})\, a_\alpha (h^1 g^\alpha + h^\alpha g^1) - b_{\alpha+1} S_d h^{\alpha+1} \\ & - b_{\alpha+1} \mathcal{R} g^{\alpha+1} - 3b_{\alpha+1} k^{\alpha+1}.\end{aligned}$$

We then see that

$$\frac{J_{\alpha-1}(\mathbf{n}) - J_\alpha(\mathbf{n})}{\mu} = \frac{J_{\alpha-1}(\mathbf{m}) - J_\alpha(\mathbf{m})}{\mu} + \delta J_{\alpha-1}(\mathbf{m}) - \delta J_\alpha(\mathbf{m}) + O(\mu), \qquad \alpha \geqslant 2 \tag{5.25}$$

and

$$\frac{-J_1(\mathbf{n}) - \sum_{\alpha=1}^{\infty} J_\alpha(\mathbf{n})}{\mu} = \frac{-J_1(\mathbf{m}) - \sum_{\alpha=1}^{\infty} J_\alpha(\mathbf{m})}{\mu} - \delta J_1(\mathbf{m}) - \sum_{\alpha=1}^{\infty} \delta J_\alpha(\mathbf{m}) + O(\mu). \tag{5.26}$$

Similarly we know that along an elastic Maxwellian **n** and for $\alpha \geqslant 2$

$$I_0^\alpha(\mathbf{n}) = -n_0^\alpha (C^1 S_c a_\alpha + 3b_\alpha) + (6-3\delta_{\alpha 2})\, a_{\alpha-1} C^{\alpha-1} C^1 + \tfrac{1}{2} b_{\alpha+1} C^{\alpha+1} S_c$$

while for an approximate Maxwellian **m**

$$I_0^\alpha(\mathbf{m}) = -g^\alpha (g^1 S_d a_\alpha + 3b_\alpha) + (6-3\delta_{\alpha 2})\, a_{\alpha-1} g^{\alpha-1} + \tfrac{1}{2} b_{\alpha+1} g^{\alpha+1} S_d$$

We again use (5.23) to find

$$\frac{I_0^\alpha(\mathbf{n})-I_0^\alpha(\mathbf{m})}{\mu}=\delta I_0^\alpha(\mathbf{m})+O(\mu) \tag{5.27}$$

where

$$\begin{aligned}\delta I_0^\alpha(\mathbf{m})= &-S_{\mathbf{d}} a_\alpha(g^\alpha h^1+k^\alpha g^1)-\mathscr{R} a_\alpha(g^\alpha g^1)-3b_\alpha k^\alpha \\ &+(6-3\delta_{\alpha 2})\, a_{\alpha-1}(g^{\alpha-1}h^1+h^{\alpha-1}g^1)+\tfrac{1}{2}b_{\alpha+1}h^{\alpha+1}S_{\mathbf{d}} \\ &+\tfrac{1}{2}b_{\alpha+1}\mathscr{R}g^{\alpha+1}\end{aligned}$$

For $\alpha=1$ we note that

$$I_0^1(\mathbf{n})=-S_{\mathbf{c}}\sum_{\beta=1}^{\infty}\{6n_0^1 a_\beta C^\beta-3(1+\delta_{\beta 1})\, b_{1+\beta}C^{1+\beta}\}$$

$$I_0^1(\mathbf{m})=-S_{\mathbf{d}}\sum_{\beta=1}^{\infty}\{6g^1 a_\beta g^\beta-3(1+\delta_{\beta 1})\, b_{1+\beta}g^{1+\beta}\}$$

and hence

$$\frac{I_0^1(\mathbf{n})-I_0^1(\mathbf{m})}{\mu}=\delta I_0^1(\mathbf{m})+O(\mu) \tag{5.28}$$

where

$$\begin{aligned}\delta I_0^1(\mathbf{m})= &-S_{\mathbf{d}}\sum_{\beta=1}^{\infty}\{6a_\beta(g^\beta k^1+k^\beta g^1)-3(1+\delta_{\beta 1})\, b_{1+\beta}h^{1+\beta}\} \\ &-\mathscr{R}\sum_{\beta=1}^{\infty}\{6a_\beta g^\beta g^1-3(1+\delta_{\beta 1})\, b_{1+\beta}g^{1+\beta}\}\end{aligned}$$

Notice $\delta J_\alpha(\mathbf{m})$, $\delta I_0^\alpha(\mathbf{m})$ are linear in h^α, k^α, $\mathscr{R}$.

Now expand the left-hand sides of (5.12), (5.13) about $\mathbf{m}$ to see that

$$\text{l.h.s.}(5.12)\doteq r^\alpha+O(\mu)$$

where

$$\begin{aligned}r^\alpha \doteq &-\frac{\partial m^\alpha}{\partial \rho}(\operatorname{div}(\rho\mathbf{u}))-\frac{\partial m^\alpha}{\partial(\rho u)}\left(\frac{\partial}{\partial x}\sum_{\alpha=1}^{\infty}\frac{m_1^\alpha+m_2^\alpha}{\alpha}\right) \\ &-\frac{\partial m^\alpha}{\partial(\rho v)}\left(\frac{\partial}{\partial y}\sum_{\alpha=1}^{\infty}\frac{m_3^\alpha+m_4^\alpha}{\alpha}\right) \\ &-\frac{\partial m^\alpha}{\partial(\rho w)}\left(\frac{\partial}{\partial z}\sum_{\alpha=1}^{\infty}\frac{m_5^\alpha+m_6^\alpha}{\alpha}\right)+\frac{\partial}{\partial x}\left(\frac{m_1^\alpha-m_2^\alpha}{\alpha}\right) \\ &+\frac{\partial}{\partial y}\left(\frac{m_3^\alpha-m_4^\alpha}{\alpha}\right)+\frac{\partial}{\partial z}\left(\frac{m_5^\alpha-m_6^\alpha}{\alpha}\right), \qquad \alpha\geqslant 1\end{aligned} \tag{5.29}$$

$$m^{\alpha} = (1 + 2\cosh d_1 + 2\cosh d_2 + 2\cosh d_3)\, g^{\alpha}(m_0^1), \qquad \alpha \geqslant 1$$

$$\mathbf{m} = (1, e^{d_1}, e^{-d_1}, e^{d_2}, e^{-d_2}, e^{d_3}, e^{-d_3})\, g^{\alpha}(m_0^1)$$

$$m_0^1 = \hat{m}_0^1(\rho, \rho\mathbf{u}), \qquad d_i = \hat{d}_i(\rho, \rho\mathbf{u}), \qquad i = 1, 2, 3$$

Notice that r^{α} is a function of ρ, $\rho\mathbf{u}$, and their first derivatives in x, y, z. Similarly we find

$$\text{l.h.s.}(5.13) = s^{\alpha} + O(\mu)$$

where

$$\begin{aligned} s^{\alpha} \doteq & -\frac{\partial g^{\alpha}}{\partial \rho}(\operatorname{div}(\rho\mathbf{u})) - \frac{\partial g^{\alpha}}{\partial(\rho u)}\left(\frac{\partial}{\partial x}\sum_{\alpha=1}^{\infty}\frac{m_1^{\alpha} + m_2^{\alpha}}{\alpha}\right) \\ & - \frac{\partial g^{\alpha}}{\partial(\rho v)}\left(\frac{\partial}{\partial y}\sum_{\alpha=1}^{\infty}\frac{m_3^{\alpha} + m_4^{\alpha}}{\alpha}\right) \\ & - \frac{\partial g^{\alpha}}{\partial(\rho w)}\left(\frac{\partial}{\partial z}\sum_{\alpha=1}^{\infty}\frac{m_5^{\alpha} + m_6^{\alpha}}{\alpha}\right), \qquad \alpha \geqslant 1 \end{aligned} \tag{5.30}$$

Again note that s^{α} is a function of ρ, $\rho\mathbf{u}$, and their first derivatives in x, y, z.

We now insert (5.25)–(5.30) into (5.12), (5.13) to find

$$r^{\alpha} = \frac{J_{\alpha-1}(\mathbf{m}) - J_{\alpha}(\mathbf{m})}{\mu} + \delta J_{\alpha-1}(\mathbf{m}) - \delta J_{\alpha}(\mathbf{m}) + O(\mu), \qquad \alpha \geqslant 2 \tag{5.31}$$

$$r^1 = \frac{-J_1(\mathbf{m}) - \sum_{\alpha=1}^{\infty} J_{\alpha}(\mathbf{m})}{\mu} - \delta J_1(\mathbf{m}) - \sum_{\alpha=1}^{\infty} \delta J_{\alpha}(\mathbf{m}) + O(\mu) \tag{5.32}$$

$$s^{\alpha} = \frac{I_0^{\alpha}(\mathbf{m})}{\mu} + \delta I_0^{\alpha}(\mathbf{m}) + O(\mu), \qquad \alpha \geqslant 1 \tag{5.33}$$

It is at this point where **m** *being an approximate Maxwellian comes to the fore.* Simply put, since **m** is an approximate Maxwellian, then (i) for $0 \leqslant \rho \leqslant \rho_S$, all the numerators in the $O(1/\mu)$ terms of (5.31)–(5.33) are identically zero, and (ii) for $\rho_S \leqslant \rho$, the numerators in the $O(1/\mu)$ terms of (5.27)–(5.29) are uniformly in α exponentially small in $\rho - \rho_S$.

But (ii) means that the numerators are bounded by const $\cdot\, (\rho - \rho_S)^j$ for all positive j if $0 \leqslant \rho - \rho_S \leqslant \theta$ for some fixed $\theta > 0$ sufficiently small.

So if we enforce the restriction

$$\rho - \rho_S \leqslant \text{const} \cdot (\mu)^{\sigma} \qquad \text{for } \textit{any} \quad \sigma > 0, \qquad \text{const} > 0 \tag{5.34}$$

we will have $p - p_S \leqslant \theta$ when μ is sufficiently small and the numerators in the $O(1/\mu)$ terms will be smaller than *any* positive integer order of μ.

In summary, if (5.34) *holds, the* $O(1/\mu)$ *terms can be neglected in* (5.31)–(5.33) *even though* **m** *is not a true Maxwellian.*

We now equate order-one terms in (5.31)–(5.33) to obtain

$$r^\alpha = \delta J_{\alpha-1}(\mathbf{m}) - \delta J_\alpha(\mathbf{m}), \qquad \alpha \geqslant 2 \tag{5.35}$$

$$r^1 = -\delta J_1(\mathbf{m}) - \sum_{\alpha=1}^{\infty} \delta J_\alpha(\mathbf{m}) \tag{5.36}$$

$$s^\alpha = \delta I_0^\alpha(\mathbf{m}), \qquad \alpha \geqslant 1 \tag{5.37}$$

which is an infinite system of linear algebraic equations in $\mathscr{R}$, (h^α, k^α), $1 \leqslant \alpha$.

We solve (5.35) recursively to find

$$\delta J_\alpha(\mathbf{m}) = \delta J_1(\mathbf{m}) - \sum_{j=1}^{\alpha} r^j, \qquad \alpha \geqslant 2 \tag{5.38}$$

Equation (5.38) suggests setting

$$\delta J_\alpha(\mathbf{m}) = \sum_{j=\alpha+1}^{\infty} r^j, \qquad \alpha \geqslant 1 \tag{5.39}$$

which yields

$$\delta J_1(\mathbf{m}) = \sum_{j=2}^{\infty} r^j \tag{5.40}$$

Hence (5.34) is satisfied. Substitution of (5.39), (5.40) yields

$$r^1 = -\sum_{j=2}^{\infty} r^j - \sum_{\alpha=1}^{\infty} \sum_{j=\alpha+1}^{\infty} r^j \tag{5.41}$$

which is equivalent to

$$\sum_{\alpha=1}^{\infty} \alpha r^\alpha = 0 \tag{5.42}$$

But a direct calculation of the left-hand side of (5.42) using the definitions of r^α and ρ shows that (5.42) does indeed hold to order one in μ. [It is also easy to see from (5.35) and (5.36) that (5.42) is a consistency condition for solvability.] Hence, if $\delta J_\alpha(\mathbf{m})$ satisfies (5.39), Eqs. (5.35), (5.36) are satisfied automatically.

The next step is to solve (5.37), (5.39) for $\mathscr{R}$, h^α, k^α as functions of ρ, $\rho\mathbf{u}$, and their spatial derivatives. This step would involve a computation of a four-parameter family of solutions with the additional computation of an approximate solution (similar to an approximate Maxwellian) for $0 \leqslant \rho - \rho_S \leqslant \text{const} \cdot (\mu)^\sigma$. Enforcement of the requirement that the approximate Maxwellian $\mathbf{m}$ yield the macroscopic density and momenta (5.15)–(5.18) requires that $\mathbf{w}$ makes no contribution to these macroscopic quantities:

$$\sum_{\alpha=1}^{\infty} \alpha[k^\alpha + (2\cosh d_1 + 2\cosh d_2 + 2\cosh d_3)\, h^\alpha + (2l_1 \sinh d_1 + 2l_2 \sinh d_2 + 2l_3 \sinh d_3)\, g^\alpha] = 0 \tag{5.43}$$

$$\sinh d_i \sum_{\alpha=1}^{\infty} h^\alpha + l_i(\cosh d_i) \sum_{\alpha=1}^{\infty} g^\alpha = 0, \qquad i = 1, 2, 3 \tag{5.44}$$

For example, if the four free parameters were h^1, l_1, l_2, l_3, then (5.43)–(5.44) would provide a system of four nonhomogeneous linear equations to determine their values. For the purposes of this paper we do not attempt this rather tedious algebraic computation, but skip to the immediate consequence of the Chapman–Enskog expansion. That is the observation that substitution of the Chapman–Enskog expansion (5.14) [which is formally valid under assumption (5.34)] into the macroscopic balance laws (5.5)–(5.8) yield the approximate (i.e., viscous compressible Navier–Stokes-like) system of balance laws for mass and momentum:

$$\frac{\partial \rho}{\partial t} + \operatorname{div}(\rho\mathbf{u}) = 0 \tag{5.45}$$

$$\frac{\partial(\rho u)}{\partial t} + \frac{\partial}{\partial x} \sum_{\alpha=1}^{\infty} \frac{m_1^\alpha + m_2^\alpha}{\alpha} = O(\mu) \tag{5.46}$$

$$\frac{\partial(\rho v)}{\partial t} + \frac{\partial}{\partial y} \sum_{\alpha=1}^{\infty} \frac{m_3^\alpha + m_4^\alpha}{\alpha} = O(\mu) \tag{5.47}$$

$$\frac{\partial(\rho w)}{\partial t} + \frac{\partial}{\partial z} \sum_{\alpha=1}^{\infty} \frac{m_5^\alpha + m_6^\alpha}{\alpha} = O(\mu) \tag{5.48}$$

where $m_i^\alpha = \hat{m}_i^\alpha(\rho, \rho\mathbf{u})$.

The main point is that this system is formally valid when $\rho - \rho_S \leqslant \text{const} \cdot (\mu)^\sigma$ for *any* $\sigma > 0$, const > 0. Hence (5.45)–(5.48) are valid into the supersaturated metastable regime $\rho_S < \rho$ up to a class of densities of size $\rho_S + O(\mu^\sigma)$ in μ, where σ is *any* positive number. As in Section 3, the hydrostatic pressure can be computed on the equilibrium ($\mathbf{u}=0$) approximate Maxwellian

$$\rho = 7 \sum_{\alpha=1}^{\infty} \alpha g^{\alpha}(m_0^1) \tag{5.49}$$

$$p = 2 \sum_{\alpha=1}^{\infty} \frac{g^{\alpha}(m_0^1)}{\alpha} \tag{5.50}$$

Since g^{α} is monotone increasing in m_0^1, we see that g, ρ are monotone increasing and hence p is monotone increasing in ρ. Unlike the analytic case in Section 3 where $0 \leqslant \rho \leqslant \rho_S$, the definition of $g^{\alpha}(m_0^1)$ suggests loss of analyticity at $\rho = \rho_S$, i.e., $m_0^1 = z_S$. Notice that, of course, the equation of state into the metastable regime is an extension of the equation of state already given for $0 \leqslant \rho \leqslant \rho_S$. Thus we have recovered Fig. 1. The use of a discrete-velocity model limits the ability to find a true equation of state independent of $\mathbf{u}$. Hopefully this is balanced by the conceptual simplicity of the ideas.

As noted earlier, (5.34) coupled with (5.45)–(5.48) suggests quite nicely the qualitative nature of the fluid dynamical limit $\mu \to 0+$. That is, as $\mu \to 0+$, the domain of validity of the expansion (5.34) collapses to $\rho - \rho_S \leqslant 0$ and we can only recover the inviscid conservation laws of mass and momentum in the stable subsaturated regime. The model thus implies that inviscid gas dynamics is not appropriate for mixed unsaturated and supersaturated flow. Classical inviscid shocks between unsaturated and supersaturated phases are impossible and viscous diffuse waves are the appropriate mechanism.

APPENDIX A. COMPUTATION OF MAXWELLIANS

We compute the Maxwellians as follows: On a Maxwellian we must have $J_{\alpha-1} - J_{\alpha} = 0$, $2 \leqslant \alpha$, and $0 = -J_1 - \sum_{\alpha=1}^{\infty} J_{\alpha}$, $I_0^{\alpha} = 0$, $1 \leqslant \alpha$, combined with the constraint of the state being an elastic Maxwellian. These equalities of course imply $I_0^{\alpha} = 0$, $J_{\alpha} = 0$, $1 \leqslant \alpha$. Now substitute the known form of an elastic Maxwellian (2.10) into this set of equations. We find that the equation $I_0^{\alpha} = 0$, $2 \leqslant \alpha$, implies

$$-n_0^{\alpha}(C^1 S_c a_{\alpha} + b_{\alpha}^0) + (6 - 3\delta_{\alpha 2})\, a_{\alpha-1} C^{\alpha-1} C^1 + \tfrac{1}{2} b_{\alpha+1} C^{\alpha+1} S_c = 0, \qquad 2 \leqslant \alpha \tag{A.1}$$

and that $J_\alpha = 0$, $1 \leqslant \alpha$, implies

$$S_c a_\alpha n_0^\alpha C^1 + S_c(1-\delta_{\alpha 1})\, C^\alpha a_\alpha n_0^1 + (6 - 3\delta_{\alpha 1})\, a_\alpha C^\alpha C^1 - b_{\alpha+1} C^{\alpha+1} S_c - b_{\alpha+1}^0 n_0^{\alpha+1} = 0, \qquad 1 \leqslant \alpha \tag{A.2}$$

where $S_c \doteq 2\cosh c_1 + 2\cosh c_2 + 2\cosh c_3$. Since S_c is arbitrary, we see that

$$-n_0^\alpha C^1 a_\alpha + \tfrac{1}{2} b_{\alpha+1} C^{\alpha+1} = 0, \qquad 2 \leqslant \alpha \tag{A.3}$$

$$-n_0^\alpha b_\alpha^0 + C^1(6 - 3\delta_{\alpha 2})\, a_{\alpha-1} C^{\alpha-1} = 0, \qquad 2 \leqslant \alpha \tag{A.4}$$

$$a_\alpha n_0^\alpha C^1 + (1-\delta_{\alpha 1})\, C^\alpha a_\alpha n_0^1 - b_{\alpha+1} C^{\alpha+1} = 0, \qquad 1 \leqslant \alpha \tag{A.5}$$

$$C^1(6 - 3\delta_{\alpha 1})\, a_\alpha C^\alpha - b_{\alpha+1}^0 n_0^{\alpha+1} = 0, \qquad 1 \leqslant \alpha \tag{A.6}$$

where obviously (A.4), (A.6) are equivalent.

Equations (A.3), (A.4) imply

$$-n_0^\alpha C^1 a_\alpha + \frac{a_\alpha n_0^\alpha C^1}{2} + \frac{1}{2} C^\alpha a_\alpha n_0^1 = 0, \qquad 2 \leqslant \alpha$$

or equivalently

$$-n_0^\alpha C^1 + C^\alpha n_0^1 = 0, \qquad 2 \leqslant \alpha \tag{A.7}$$

Substitution of (A.7) into (A.4) yields

$$\frac{(6 - 3\delta_{\alpha 2})\, a_{\alpha-1}}{b_\alpha^0} C^{\alpha-1}(C^1)^2 - C^\alpha n_0^1 = 0, \qquad 2 \leqslant \alpha$$

or

$$C^\alpha = \frac{3(2 - \delta_{\alpha 2})\, a_{\alpha-1}(C^1)^2\, C^{\alpha-1}}{n_0^1 b_\alpha^0}, \qquad 2 \leqslant \alpha \tag{A.8}$$

Substitution of $\alpha = 2$ into (A.8) yields of course that

$$C^2 = \frac{3}{n_0^1} \frac{a_1}{b_2^0} (C^1)^3 \tag{A.9}$$

On the other hand, when $\alpha = 1$, (A.5) shows that

$$C^2 = \frac{a_1}{b_2} n_0^1 C^1 \tag{A.10}$$

and hence consistency of (A.9), (A.10) requires

$$(C^1)^2 = \frac{b_2^0}{3b_2}(n_0^1)^2 \tag{A.11}$$

Let us repeat this process. Substitute $\alpha = 3$ into (A.8) and $\alpha = 2$ into (A.5) and equate the two representations for C^3 making use of (A.11). After some straightforward computations we see that the relation

$$\frac{b_2}{b_3} = \frac{b_2^0}{b_3^0} \tag{A.12}$$

must hold. If we continue in this manner, we see that we need

$$\frac{b_\beta}{b_{\beta+1}} = \frac{b_\beta^0}{b_{\beta+1}^0}, \qquad 2 \leqslant \beta \tag{A.13}$$

to have a solution C^α of (A.1), (A.2) which is independent of S. Of course (A.13) can be expressed as

$$\frac{b_\beta}{b_\beta^0} = \frac{b_{\beta+1}}{b_{\beta+1}^0}, \qquad 2 \leqslant \beta$$

i.e., b_β / b_β^0 is a positive constant independent of β. For convenience we chose this constant to be 1/3 so that we have $3b_\beta = b_\beta^0$ for all β. Equation (A.11) then implies the simple relation $C^1 = n_0^1$. (Of course any other choice of the constant will not change our results qualitatively.) Finally, (A.5)–(A.8) yield

$$C^\alpha = n_0^\alpha = Q_\alpha (n_0^1)^\alpha, \qquad 1 \leqslant \alpha \tag{A.14}$$

where

$$Q_\alpha = 2^{\alpha-2} \frac{a_{\alpha-1} \cdots a_1}{b_\alpha \cdots b_2}, \qquad \alpha \geqslant 2$$

$$Q_1 = 1$$

We thus see that the Maxwellian states are given by

$$\mathbf{n}^\alpha = (1, e^{c_1}, e^{-c_1}, e^{c_2}, e^{-c_2}, e^{c_3}, e^{-c_3})\, Q_\alpha (n_0^1)^\alpha \tag{A.15}$$

and hence depend on the parameters $\mathbf{c}$, n_0^1.

APPENDIX B. The *H*-THEOREM

In this appendix we prove that an *H*-theorem holds along elastic Maxwellians.

Set

$$H_j \doteq \sum_{\alpha=1}^{\infty} n_j^\alpha \left[\ln\left(\frac{n_j^\alpha}{Q_\alpha(z_S)^\alpha}\right) - 1 \right], \qquad H \doteq \sum_{j=0}^{6} H_j$$

Then

$$\begin{aligned}
&\frac{\partial H}{\partial t} + \frac{\partial}{\partial x}(H_1 - H_2) + \frac{\partial}{\partial y}(H_3 - H_4) + \frac{\partial}{\partial z}(H_5 - H_6) \\
&\quad = \frac{1}{\mu} \sum_{\alpha=1}^{\infty} \sum_{j=0}^{6} I_j^\alpha \left[\ln \frac{n_j^\alpha}{Q_\alpha(z_S)^\alpha} \right]
\end{aligned}$$

Along an elastic Maxwellian

$$\mathbf{n}^\alpha = (n_0^\alpha, C^\alpha e^{c_1}, C^\alpha e^{-c_1}, C^\alpha e^{c_2}, C^\alpha e^{-c_2}, C^\alpha e^{c_3}, C^\alpha e^{-c_3})$$

we find

$$\begin{aligned}
&\sum_{\alpha=1}^{\infty} \sum_{j=0}^{6} I_j^\alpha \left[\ln\left(\frac{n_j^\alpha}{Q_\alpha(z_S)^\alpha}\right) \right] \\
&\quad = \sum_{\alpha=1}^{\infty} \left\{ \sum_{j=1}^{6} I_j^\alpha \ln\left(\frac{C^\alpha}{Q_\alpha(z_S)^\alpha}\right) + I_0^\alpha \ln\left(\frac{n_0^\alpha}{Q_\alpha(z_S)^\alpha}\right) \right\} \\
&\quad = \sum_{\alpha=2}^{\infty} (J_{\alpha-1} - J_\alpha) \ln\left(\frac{C^\alpha}{Q_\alpha(z_S)^\alpha}\right) \\
&\qquad + \sum_{\alpha=1}^{\infty} I_0^\alpha \left\{ \ln\left(\frac{n_0^\alpha}{Q_\alpha(z_S)^\alpha}\right) - \ln\left(\frac{C^\alpha}{Q_\alpha(z_S)^\alpha}\right) \right\} \\
&\qquad - \left(J_1 + \sum_{\alpha=1}^{\infty} J_\alpha \right) \ln\left(\frac{C^1}{z_S}\right) \\
&\quad = \sum_{\alpha=1}^{\infty} J_\alpha \ln\left(\frac{C^{\alpha+1} Q_\alpha}{Q_{\alpha+1} C^\alpha C^1}\right) + \sum_{\alpha=1}^{\infty} I_0^\alpha \ln\left(\frac{n_0^\alpha}{C^\alpha}\right)
\end{aligned}$$

After some direct manipulations the last expression is explicitly computed and we obtain the following result.

Theorem (*H*-Theorem). Along an elastic Maxwellian the following equality is satisfied:

$$
\begin{aligned}
&\frac{\partial H}{\partial t}+\frac{\partial}{\partial x}(H_1-H_2)+\frac{\partial}{\partial y}(H_3-H_4)+\frac{\partial}{\partial z}(H_5-H_6)\\
&\quad= -\frac{3}{\mu}(b_2n_0^2-a_1(C^1)^2)\ln\left(\frac{b_2n_0^2}{a_1(C^1)^2}\right)\\
&\quad-\frac{S_c}{\mu}(b_2C^2-a_1n_0^1C^1)\ln\left(\frac{b_2C^2}{a_1n_0^1C^1}\right)\\
&\quad-\frac{1}{\mu}\sum_{\alpha=3}^{\infty}(3b_\alpha n_0^\alpha-6a_{\alpha-1}C^1C^{\alpha-1})\ln\left(\frac{b_\alpha n_0^\alpha}{2a_{\alpha-1}C^1C^{\alpha-1}}\right)\\
&\quad-\frac{S_c}{\mu}\sum_{\alpha=3}^{\infty}\left(\frac{b_\alpha}{2}C^\alpha-a_{\alpha-1}n_0^{\alpha-1}C^1\right)\ln\left(\frac{b_\alpha C^\alpha}{2a_{\alpha-1}n_0^{\alpha-1}C^1}\right)\\
&\quad-\frac{S_c}{\mu}\sum_{\alpha=3}^{\infty}\left(\frac{b_\alpha C^\alpha}{2}-a_{\alpha-1}n_0^1C^{\alpha-1}\right)\ln\left(\frac{b_\alpha C^\alpha}{2a_{\alpha-1}n_0^1C^{\alpha-1}}\right)
\end{aligned}
$$

The right side is less than or equal to zero and vanishes if and only if the elastic Maxwellian is also an inelastic Maxwellian.

APPENDIX C. CONSTRUCTION OF APPROXIMATE MAXWELLIAN STATES

Proof of Lemma 4.3. First let us reconsider Eqs. (A.1) and (A.2). Equation (A.1) followed from the fact that $I_0^\alpha(\mathbf{n})=0$, $2\leqslant\alpha$. Now we replace (A.1) by the weaker requirement

$$I_0^\alpha(\mathbf{n})=J(6-S_c) \tag{C.1}$$

for some J independent of α. Notice that at equilibrium $S_c=6$ and $I_0^\alpha=0$ holds identically. Away from equilibrium (C.1) says that $I_0^\alpha(n)$ is zero up to quadratic terms in c_1, c_2, c_3.

Equation (A.2) followed from the fact that $J_\alpha(\mathbf{n})=0$, $1\leqslant\alpha$. Now we replace (A.2) by

$$J_\alpha(\mathbf{n})=J(6+2S_c),\qquad 1\leqslant\alpha \tag{C.2}$$

so that (iii) $J_{\alpha-1}(\mathbf{n})-J_\alpha(\mathbf{n})=0$, $2\leqslant\alpha$, is satisfied.

If we now substitute in the definitions of $I_0^\alpha(\mathbf{n})$, $J_\alpha(\mathbf{n})$ where $\mathbf{n}$ is an elastic Maxwellian, (C.1) and (C.3) become, respectively,

$$-n_0^\alpha(C^1 S_c a_\alpha + b_\alpha^0) + (6 - 3\delta_{\alpha 2})\, a_{\alpha-1} C^{\alpha-1} C^1 + \tfrac{1}{2} b_{\alpha+1} C^{\alpha+1} S_c = J(6 - S_c), \qquad 2 \leqslant \alpha \tag{C.3}$$

$$S_c a_\alpha n_0^\alpha C^1 + S_c(1 - \delta_{\alpha 1})\, C^\alpha a_\alpha n_0^1 + (6 - 3\delta_{\alpha 1})\, a_\alpha C^\alpha C^1 - b_{\alpha+1} C^{\alpha+1} S_c - b_{\alpha+1}^0 n_0^{\alpha+1} = J(6 + 2S_c), \qquad 1 \leqslant \alpha \tag{C.4}$$

We again use the fact that S_c is arbitrary to recover an analogous system to (A.3)–(A.6), i.e.,

$$-n_0^\alpha C^1 a_\alpha + \tfrac{1}{2} b_{\alpha+1} C^{\alpha+1} = -J, \qquad 2 \leqslant \alpha \tag{C.5}$$

$$-n_0^\alpha b_\alpha^0 + C^1(6 - 3\delta_{\alpha 2})\, a_{\alpha-1} C^{\alpha-1} = 6J, \qquad 2 \leqslant \alpha \tag{C.6}$$

$$a_\alpha n_0^\alpha C^1 + (1 - \delta_{\alpha 1})\, C^\alpha a_\alpha n_0^1 - b_{\alpha+1} C^{\alpha+1} = 2J, \qquad 1 \leqslant \alpha \tag{C.7}$$

$$C^1(6 - 3\delta_{\alpha 1})\, a_\alpha C^\alpha - b_{\alpha+1}^0 n_0^{\alpha+1} = 6J, \qquad 1 \leqslant \alpha \tag{C.8}$$

We note that (C.6) and (C.8) are equivalent.

Next multiply (C.5) by two and add to (C.7) to see that

$$n_0^\alpha C^1 = n_0^1 C^\alpha, \qquad \alpha \geqslant 2 \tag{C.9}$$

Substitute this relation into (C.6) to eliminate n_0^α and set $\alpha = 2$ to find

$$C^2 = \frac{3a_1(C^1)^3}{n_0^\alpha b_2^0} - \frac{6JC^1}{b_2^0 n_0^1} \tag{C.10}$$

On the other hand, (C.7) with $\alpha = 1$ implies

$$C^2 = \frac{a_1 n_0^1 C^1}{b_2} - \frac{2J}{b_2} \tag{C.11}$$

Since J is as yet arbitrary, we see

$$\frac{3a_1}{b_2^0}(C^1)^2 = \frac{a_1(n_0^1)^2}{b_2} \tag{C.12a}$$

$$\frac{6C^1}{b_2^0 n_0^1} = \frac{2}{b_2} \tag{C.12b}$$

If we set $b_j^0/b_j = 3\gamma^2$, (C.12a) tells us that $C^1 = \gamma n_0^1$, while (C.12b) says that $\gamma = 1$. So, as in Section 2, we take

$$b_j^0 = 3b_j$$

and we now know that $C^1 = n_0^1$ and hence from (C.9)

$$n_0^\alpha = C^\alpha, \qquad 1 \leqslant \alpha \tag{C.13}$$

Equation (C.11) then tells us that

$$n_0^2 = \frac{a_1}{b_2}(n_0^1)^2 - \frac{2J}{b_2} \tag{C.14}$$

and (C.5) gives the recursion relation

$$-n_0^\alpha n_0^1 a_\alpha + \tfrac{1}{2} b_{\alpha+1} n_0^{\alpha+1} = -J, \qquad 2 \leqslant \alpha \tag{C.15}$$

We solve (C.15) as in Penrose[12]: divide both sides of (C.15) by $a_\alpha Q_\alpha (n_0^1)^{\alpha+1}$ to see that

$$\frac{-n_0^\alpha}{Q_\alpha (n_0^1)^\alpha} + \frac{n_0^{\alpha+1}}{Q_{\alpha+1}(n_0^1)^{\alpha+1}} = -\frac{J}{a_\alpha Q_\alpha (n_0^1)^{\alpha+1}} \tag{C.16}$$

[Here we used the relation $2a_\alpha Q_\alpha = Q_{\alpha+1} b_{\alpha+1}$ for $2 \leqslant \alpha$, where Q_α is defined in (2.11).]

Now sum (C.16) from two to infinity to see that

$$\frac{-n_0^2}{Q_2 (n_0^1)^2} = -J \sum_{\alpha=2}^{\infty} \frac{1}{a_\alpha Q_\alpha (n_0^1)^{\alpha+1}} \tag{C.17}$$

From our assumptions on the kinetic coefficients a_α, b_α of (3.1)–(3.4) we know that $a_\alpha > A' > 0$ and we see that

$$\sum_{\alpha=1}^{\infty} \frac{1}{a_\alpha Q_\alpha (n_0^1)^\alpha} \leqslant \frac{1}{A'} \sum_{\alpha=1}^{\infty} \frac{1}{Q_\alpha (n_0^1)^\alpha}$$

which converges by the ratio test for $n_0^1 > z_S$. So for $n_0^1 > z_S$, (C.17) is well defined, and combining (C.14), (C.16) and $Q_2 b_2 = a_1$, we see that

$$J(n_0^1) = \left\{ \frac{1}{a_1 (n_0^1)^2} + \sum_{\alpha=2}^{\infty} \frac{1}{a_\alpha Q_\alpha (n_0^1)^{\alpha+1}} \right\}^{-1} \tag{C.18}$$

Finally, summing (C.16) from α to infinity, we recover the formula

$$C^{\alpha} = n_0^{\alpha} = f^{\alpha}(n_0^1), \qquad n_0^1 > z_S \tag{C.19}$$

where

$$\begin{aligned} f^1(z) &= z \\ f^{\alpha}(z) &= Q_{\alpha}(z)^{\alpha} J(z) \left\{ \sum_{\beta=\alpha}^{\infty} \frac{1}{a_{\beta} Q_{\beta}(z)^{\beta+1}} \right\}, \qquad z > z_S \end{aligned} \tag{C.20}$$

This proves Lemma 4.3.

Lemma C.1 (Penrose[12]). (i) For each $n_0^1 > z_S$, (C.15) defines a unique bounded solution $f^{\alpha}(n_0^1)$. (ii) For each fixed z, $Q_{\alpha} f^{\alpha}(z)$ decreases monotonically with α. (iii) For fixed α, $f^{\alpha}(z)/z$ increases monotonically with z, and hence $f^{\alpha}(z)$ increases strictly monotonically with z. (iv) The sequence $f^{\alpha}(z)$ has the upper bound $f^{\alpha}(z) \leqslant Q_{\alpha}(z)^{\alpha}$. (v) In the limit $z \searrow z_S$, the sequence $f^{\alpha}(z)$ becomes the equilibrium cluster distribution at

$$z = z_S: \quad \lim_{z \searrow z_S} f^{\alpha}(z) = Q_{\alpha}(z_S)^{\alpha}$$

and hence by (iii), $f^{\alpha}(z) > Q_{\alpha}(z_S)^{\alpha}$.

Theorem C.2 (Penrose[12]). Let z be any number greater than z_S. Then the following results hold: (i) α^* (the critical cluster size) is at most algebraically large in $z - z_S$. (ii) All moments of the equilibrium cluster size distribution converge when

$$z = z_S: \quad \sum_{\alpha=1}^{\infty} \alpha^n Q_{\alpha}(z_S)^{\alpha} < \infty \qquad (n = 0, 1, 2, \ldots)$$

(iii) The quantities $J^*(z)$ defined by $J^*(z) = a_{\alpha^*} Q_{\alpha^*} z^{\alpha^*+1}$ and $J(z)$ defined by (C.18) are exponentially small in $z - z_S$. (iv) The ratio $\rho^*(z)/J^*(z)$, $\rho^*(z) = \sum_{\alpha=\alpha^*+1}^{\infty} \alpha Q_{\alpha}(z_S)^{\alpha}$ is at most algebraically large in $z - z_S$; moreover, $\rho^*(z)$, being the product of $\rho^*(z)/J^*(z)$ (at most algebraically large) and $J^*(z)$ (exponentially small) is exponentially small in $z - z_S$.

Lemma C.3. $a_{\alpha-1} Q_{\alpha-1}/a_{\alpha} Q_{\alpha} > z_S$ for all α, $\alpha \geqslant 1$, and $a_{\alpha} Q_{\alpha}(z_S)^{\alpha}$ is monotone decreasing in α.

Proof. From the definition of Q_{α},

$$\frac{b_{\alpha}}{2a_{\alpha}} = \frac{2a_{\alpha-1} Q_{\alpha-1}}{2a_{\alpha} Q_{\alpha}} = \frac{a_{\alpha-1} Q_{\alpha-1}}{a_{\alpha} Q_{\alpha}} \tag{C.21}$$

From (3.3), (3.4) we know that $b_\alpha/2a_\alpha > z_S$, $\lim_{\alpha\to\infty}(b_\alpha/2a_\alpha) = z_S$, and b_α/a_α monotone decreasing, and so we have $a_{\alpha-1}Q_{\alpha-1}/a_\alpha Q_\alpha > z_S$ for all α, $\alpha \geqslant 1$. Furthermore, $a_{\alpha-1}Q_{\alpha-1}(z_S)^{\alpha-1}/a_\alpha Q_\alpha(z_S)^\alpha > 1$, so that $a_\alpha Q_\alpha(z_S)^\alpha$ is monotone decreasing in α.

In the next group of lemmas $\mathbf{m}$ denotes the state defined by (4.2), and $S_{\mathbf{d}} = 2\cosh d_1 + 2\cosh d_2 + 2\cosh d_3$.

Lemma C.4. $I_0^\alpha(\mathbf{m})$ is exponentially small in $m_0^1 - z_S$, uniformly in α, $1 \leqslant \alpha$. For $0 \leqslant m_0^1 \leqslant z_S$, $I_0^\alpha(\mathbf{m}) = 0$.

Proof. For $\alpha \geqslant 2$, we know that

$$I_0^\alpha(\mathbf{m}) = -g^\alpha(m_0^1)(m_0^1 S_{\mathbf{d}} a_\alpha + 3b_\alpha) + (6 - 3\delta_{\alpha 2})\, a_{\alpha-1}\, g^{\alpha-1}(m_0^1)\, m_0^1 + \tfrac{1}{2} b_{\alpha+1}\, g^{\alpha+1}(m_0^1)\, S_{\mathbf{d}} \tag{C.22}$$

If in addition $2 \leqslant \alpha \leqslant \alpha^* - 1$, and $g^\alpha(m_0^1) = f^\alpha(m_0^1)$, we can use (C.1) to see that

$$I_0^\alpha(\mathbf{m}) = J(m_0^1)(6 - S_{\mathbf{d}}), \qquad 2 \leqslant \alpha \leqslant \alpha^* - 1 \tag{C.23}$$

[Notice that if $m_0^1 \leqslant z_S$ is a Maxwellian and $I_0^\alpha(\mathbf{m}) = 0$, the second part of the lemma is easily obtained.]

If $\alpha = \alpha^*$ and $m_0^1 > z_S$, we see that

$$I_0^{\alpha^*}(\mathbf{m}) = -f^\alpha(m_0^1)(m_0^1 S_{\mathbf{d}} a_\alpha + 3b_\alpha) + (6 - 3\delta_{\alpha^* 2})\, a_{\alpha^*-1} f^{\alpha^*-1}(m_0^1)\, m_0^1 + \tfrac{1}{2} b_{\alpha^*+1}\, Q_{\alpha^*+1}(z_S)^{\alpha^*+1}\, S_{\mathbf{d}}$$

or by adding and subtracting $\frac{1}{2} b_{\alpha^*+1} S_{\mathbf{d}} f^{\alpha^*+1}(m_0^1)$ and using the definition of $f^\alpha(m_0^1)$,

$$I_0^{\alpha^*}(\mathbf{m}) = J(m_0^1)(6 - S_{\mathbf{d}}) + \tfrac{1}{2} b_{\alpha^*+1} S_{\mathbf{d}}(Q_{\alpha^*+1}(z_S)^{\alpha^*+1} - f^{\alpha^*+1}(m_0^1))$$

This can be rewritten as

$$I_0^{\alpha^*}(\mathbf{m}) = J(m_0^1)(6 - S_{\mathbf{d}}) + S_{\mathbf{d}} a_{\alpha^*}\Big(Q_{\alpha^*}(z_S)^{\alpha^*+1} - \frac{Q_{\alpha^*}}{Q_{\alpha^*+1}} f^{\alpha^*+1}(m_0^1)\Big)$$

Since

$$f^{\alpha^*+1}(m_0^1) \leqslant Q_{\alpha^*+1}(m_0^1)^{\alpha^*+1}$$

(Lemma C.1), we have

$$|I_0^{\alpha^*}(\mathbf{m})| \leqslant J(m_0^1)(6 - S_{\mathbf{d}}) + S_{\mathbf{d}} J^*(m_0^1) + S_{\mathbf{d}} a_{\alpha^*} Q_{\alpha^*}(m_0^1)^{\alpha^*+1}$$

i.e.,

$$|I_0^{\alpha^*}(\mathbf{m})| \leqslant J(m_0^1)(6 - S_\mathbf{d}) + 2S_\mathbf{d} J^*(m_0^1) \tag{C.24}$$

Next we consider the case $\alpha = \alpha^* + 1$. We have

$$\begin{aligned} I_0^{\alpha^*+1}(\mathbf{m}) = &-Q_{\alpha^*+1}(z_S)^{\alpha^*+1}\,(m_0^1 S_\mathbf{d} a_{\alpha^*+1} + 3b_{\alpha^*+1}) \\ &+ (6 - 3\delta_{\alpha^*+1,2})\, a_\alpha f^{\alpha^*}(m_0^1)\, m_0^1 + \tfrac{1}{2} b_{\alpha^*+2} Q^{\alpha^*+2}(z_S)^{\alpha^*+2}\, S_\mathbf{d} \end{aligned}$$

or adding and subtracting

$$f_{\alpha^*+1}(m_0^1)(m_0^1 S_\mathbf{d} a_{\alpha^*+1} + 3b_{\alpha^*+1}) - \tfrac{1}{2} b_{\alpha^*+2} S_\mathbf{d} f^{\alpha^*+2}(m_0^1)$$

we find via (C.1) that

$$\begin{aligned} I_0^{\alpha^*+1}(\mathbf{m}) = &\, J(m_0^1)(6 - S_\mathbf{d}) + (m_0^1 S_\mathbf{d} a_{\alpha^*+1} + 3b_{\alpha^*+1}) \\ &\times (f^{\alpha^*+1}(m_0^1) - Q_{\alpha^*+1}(z_S)^{\alpha^*+1}) \\ &+ \tfrac{1}{2} b_{\alpha^*+2} S_\mathbf{d}(Q^{\alpha^*+2}(z_S)^{\alpha^*+2} - f^{\alpha^*+2}(m_0^1)) \end{aligned}$$

But this then yields

$$\begin{aligned} |I_0^{\alpha^*+1}(\mathbf{m})| \leqslant &\, J(m_0^1)(6 - S_\mathbf{d}) + m_0^1 S_\mathbf{d} a_{\alpha^*+1} f^{\alpha^*+1}(m_0^1) \\ &+ m_0^1 S_\mathbf{d} a_{\alpha^*+1} Q_{\alpha^*+1}(z_S)^{\alpha^*+1} \\ &+ S_\mathbf{d} a_{\alpha^*+1} Q_{\alpha^*+1}(z_S)^{\alpha^*+2} + S_\mathbf{d} a_{\alpha^*+1} \frac{Q_{\alpha^*+1}}{Q_{\alpha^*+2}} f^{\alpha^*+2}(m_0^1) \end{aligned}$$

We now use Lemma C.1(i), (iv) to see that

$$\begin{aligned} |I_0^{\alpha^*+1}(\mathbf{m})| \leqslant &\, J(m_0^1)(6 - S_\mathbf{d}) + m_0^1 S_\mathbf{d} a_{\alpha^*} f^{\alpha^*}(m_0^1) \\ &+ \frac{m_0^1 S_\mathbf{d}}{z_S} a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*+1} \\ &+ S_\mathbf{d} a_{\alpha^*+1} Q_{\alpha^*+1}(z_S)^{\alpha^*+2} + S_\mathbf{d} a_{\alpha^*+1} Q_{\alpha^*+1}(m_0^1)^{\alpha^*+2} \end{aligned}$$

and then apply Lemma C.1 to obtain

$$\begin{aligned} |I_0^{\alpha^*+1}(\mathbf{m})| \leqslant &\, J(m_0^1)(6 - S_\mathbf{d}) + m_0^1 S_\mathbf{d} a_{\alpha^*} f^{\alpha^*}(m_0^1) \\ &+ \frac{m_0^1 S_\mathbf{d} a_\alpha Q_{\alpha^*}(z_S)^{\alpha^*+1}}{z_S} \\ &+ S_\mathbf{d} a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*+1} + \frac{S_\mathbf{d} a_{\alpha^*} Q_{\alpha^*}(m_0^1)^{\alpha^*+2}}{z_S} \end{aligned}$$

Since $m_0^1 > z_S$ we then find via Lemma C.1(iv)

$$|I_0^{\alpha^*+1}(\mathbf{m})| \leqslant J(m_0^1)(6-S_{\mathbf{d}}) + m_0^1 S_{\mathbf{d}} a_{\alpha^*} Q_{\alpha^*}(m_0^1)^{\alpha^*} + \frac{m_0^1 S_{\mathbf{d}} a_{\alpha^*} Q_{\alpha^*}(m_0^1)^{\alpha^*+1}}{z_S} + S_{\mathbf{d}} a_{\alpha^*} Q_{\alpha^*}(m_0^1)^{\alpha^*+1} + \frac{S_{\mathbf{d}} m_0^1 a_{\alpha^*} Q_{\alpha^*}(m_0^1)^{\alpha^*+1}}{z_S}$$

i.e.,

$$|I_0^{\alpha^*+1}(\mathbf{m})| \leqslant J(m_0^1)(6-S_{\mathbf{d}}) + 2S_{\mathbf{d}} J^*(m_0^1) + \frac{2S_{\mathbf{d}} m_0^1}{z_S} J^*(m_0^1) \quad \text{(C.25)}$$

Next we consider $\alpha \geqslant \alpha^* + 2$. In this case

$$I_0^\alpha(\mathbf{m}) = -Q_\alpha(z_S)^\alpha (m_0^1 S_{\mathbf{d}} a_\alpha + 3b_\alpha) + (6 - 3\delta_{\alpha 2})\, a_{\alpha-1} Q_{\alpha-1}(z_S)^{\alpha-1} m_0^1 + \frac{S_{\mathbf{d}}}{2} b_{\alpha+1} Q_{\alpha+1}(z_S)^{\alpha+1}$$

which from the definition of Q_α implies

$$I_0^\alpha(\mathbf{m}) = -m_0^1 S_{\mathbf{d}} a_\alpha Q_\alpha(z_S)^\alpha - 2a_{\alpha-1} Q_{\alpha-1}(z_S)^\alpha + (6-3\delta_{\alpha 2})\, a_{\alpha-1} Q_{\alpha-1}(z_S)^{\alpha-1} m_0^1 + a_\alpha Q_\alpha(z_S)^{\alpha+1} S_{\mathbf{d}}$$

But from Lemma C.3, $a_\alpha Q_\alpha(z_S)^\alpha$ is monotone decreasing and hence

$$|I_0^\alpha(\mathbf{m})| \leqslant m_0^1 S_{\mathbf{d}} a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*} + 2a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*+1} + 6a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*} m_0^1 + a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*+1} S_{\mathbf{d}}$$

But $m_0^1 > z_S$, so we have

$$|I_0^\alpha(\mathbf{m})| \leqslant J^*(m_0^1)(8 + 2S_{\mathbf{d}}) \quad \text{(C.26)}$$

Finally, in the case $\alpha = 1$, we use $f^\alpha(m_0^1) = g^\alpha(m_0^1)$, $1 \leqslant \alpha \leqslant \alpha^*$, and (C.26) to obtain

$$
\begin{aligned}
I_0^1(\mathbf{m}) &= -S_{\mathbf{d}} \sum_{\alpha=1}^{\infty} \{6a_\alpha m_0^1 g^\alpha(m_0^1) - 3(1+\delta_{\alpha 1})\, b_{\alpha+1}\, g^{\alpha+1}(m_0^1)\} \\
&= -S_{\mathbf{d}} \sum_{\alpha=\alpha^*}^{\infty} \{6a_\alpha m_0^1 Q_\alpha(z_S)^\alpha - 3(1+\delta_{\alpha 1})\, b_{\alpha+1}\, Q_{\alpha+1}(z_S)^{\alpha+1}\} \\
&\quad - S_{\mathbf{d}}\{6a_1 m_0^1 g^1(m_0^1) - 6b_2 g^2(m_0^1)\} \\
&\quad - S_{\mathbf{d}} \sum_{\alpha=2}^{\alpha^*-1} \{6a_\alpha m_0^1 g^\alpha(m_0^1) - 3b_{\alpha+1}\, g^{\alpha+1}(m_0^1)\} \\
&= -S_{\mathbf{d}} \sum_{\alpha=\alpha^*}^{\infty} \{6a_\alpha m_0^1 Q_\alpha(z_S)^\alpha - 3(1+\delta_{\alpha 1})\, b_{\alpha+1}\, Q_{\alpha+1}(z_S)^{\alpha+1}\} \\
&\quad - 12 S_{\mathbf{d}} J(m_0^1) - 6S_{\mathbf{d}} J(m_0^1)(\alpha^* - 2)
\end{aligned}
$$

Hence we find, using $b_{\alpha+1} Q_{\alpha+1} = 2a_\alpha Q_\alpha$, $\alpha \geqslant 2$, that

$$
I_0^1(\mathbf{m}) = S_{\mathbf{d}} \alpha^* J(m_0^1) + 6S_{\mathbf{d}} \sum_{\alpha=\alpha^*}^{\infty} \{a_\alpha m_0^1 Q_\alpha(z_S)^\alpha - (1+\delta_{\alpha 1})\, a_\alpha Q_\alpha(z_S)^{\alpha+1}\}
$$

or, since $\alpha^* \geqslant 1$,

$$
\begin{aligned}
I_0^1(\mathbf{m}) &= S_{\mathbf{d}} \alpha^* J(m_0^1) + 6S_{\mathbf{d}}[a_{\alpha^*} m_0^1 Q_{\alpha^*}(z_S)^{\alpha^*} \\
&\quad - (1-\delta_{\alpha^* 1})\, a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*+1}](m_0^1 - z_S)\, 6S_{\mathbf{d}} \sum_{\alpha=\alpha^*+1}^{\infty} a_\alpha Q_\alpha(z_S)^\alpha
\end{aligned}
$$

But since $m_0^1 > z_S$, this immediately yields from (3.1) that

$$
|I_0^1(\mathbf{m})| \leqslant S_{\mathbf{d}} \alpha^* J(m_0^1) + 12 S_{\mathbf{d}} J^*(m_0^1) + 6S_{\mathbf{d}}(m_0^1 - z_S)\, A\rho^*(m_0^1) \quad \text{(C.27)}
$$

If we now apply Theorem C.2 to estimates (C.24)–(C.27), the lemma is proven.

Lemma C.5. $J_{\alpha-1}(\mathbf{m}) - J_\alpha(\mathbf{m})$ is exponentially small in $m_0^1 - z_S$, uniformly in α, $2 \leqslant \alpha$. For $0 \leqslant m_0^1 \leqslant z_S$, $J_\alpha(\mathbf{m}) = 0$.

Proof. We know that

$$
\begin{aligned}
J_\alpha(\mathbf{m}) &= S_{\mathbf{d}}(2-\delta_{\alpha 1})\, a_\alpha g^\alpha m_0^1 + (6 - 3\delta_{\alpha 1})\, a_\alpha g^\alpha m_0^1 \\
&\quad - b_{\alpha+1} g^{\alpha+1} S_{\mathbf{d}} - 3b_{\alpha+1} g^{\alpha+1}
\end{aligned}
$$

Since $g^\alpha(m_0^1) = f^\alpha(m_0^1)$ for $1 \leqslant \alpha \leqslant \alpha^*$, we know from (4.1) that $J_\alpha(\mathbf{m}) = J(m_0^1)(6 + 2S_{\mathbf{d}})$ for $1 \leqslant \alpha \leqslant \alpha^* - 1$, where J is defined by (C.18).

Also notice that when $0 \leqslant m_0^1 \leqslant z_S$, $\mathbf{m}$ is a Maxwellian and the second part of the lemma is proven. For $m_0^1 > z_S$ we then trivially have

$$J_{\alpha-1}(\mathbf{m}) - J_\alpha(\mathbf{m}) = 0, \qquad 1 \leqslant \alpha \leqslant \alpha^* - 1 \tag{C.28}$$

When $\alpha = \alpha^*$ and $m_0^1 > z_S$,

$$\begin{aligned} J_{\alpha^*-1}(\mathbf{m}) - J_{\alpha^*}(\mathbf{m}) = {} & J(m_0^1)(6 + 2S_\mathbf{d}) - 2S_\mathbf{d} a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*+1} \\ & - 6a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*} m_0^1 \\ & + b_{\alpha^*+1} Q_{\alpha^*+1}(z_S)^{\alpha^*+1} S_\mathbf{d} + 3b_{\alpha^*+1} Q_{\alpha^*+1}(z_S)^{\alpha^*+1} \end{aligned}$$

We now use $Q_{\alpha+1} b_{\alpha+1} = 2Q_\alpha a_\alpha$, $\alpha \geqslant 2$, to see

$$\begin{aligned} J_{\alpha^*-1}(\mathbf{m}) - J_{\alpha^*}(\mathbf{m}) = {} & J(m_0^1)(6 + 2S_\mathbf{d}) - 2S_\mathbf{d} a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*} (m_0^1 - z_S) \\ & - 6a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*} (m_0^1 - z_S) \end{aligned}$$

and hence

$$\begin{aligned} |J_{\alpha^*-1}(\mathbf{m}) - J_{\alpha^*}(\mathbf{m})| \leqslant {} & J(m_0^1)(6 + 2S_\mathbf{d}) \\ & + \left(\frac{m_0^1}{z_S} - 1\right)(6 + 2S_\mathbf{d})\, J^*(m_0^1) \end{aligned} \tag{C.29}$$

Next when $\alpha \geqslant \alpha^* + 1$ we find

$$\begin{aligned} J_{\alpha-1}(\mathbf{m}) - J_\alpha(\mathbf{m}) = {} & -2S_\mathbf{d} a_{\alpha-1} Q_{\alpha-1}(z_S)^{\alpha-1} (m_0^1 - z_S) \\ & - 6a_{\alpha-1} Q_{\alpha-1}(z_S)^{\alpha-1} (m_0^1 - z_S) \\ & + 2S_\mathbf{d} a_\alpha Q_\alpha(z_S)^\alpha (m_0^1 - z_S) \\ & + 6a_\alpha Q_\alpha(z_S)^\alpha (m_0^1 - z_S) \end{aligned}$$

i.e.,

$$|J_{\alpha-1}(\mathbf{m}) - J_\alpha(\mathbf{m})| \leqslant (2S_\mathbf{d} + 6)(m_0^1 - z_S)(a_{\alpha-1} Q_{\alpha-1}(z_S)^{\alpha-1} + a_\alpha Q_\alpha(z_S)^\alpha)$$

and by Lemma C.3

$$|J_{\alpha-1}(\mathbf{m}) - J_\alpha(\mathbf{m})| \leqslant 4(S_\mathbf{d} + 3)\left(\frac{m_0^1}{z_S} - 1\right) J^*(m_0^1), \qquad \alpha \geqslant \alpha^* + 1 \tag{C.30}$$

Estimates (C.28)–(C.30) and Theorem C.2 prove the lemma.

Lemma C.6. $J_1(\mathbf{m}) + \sum_{\alpha=1}^\infty J_\alpha(\mathbf{m})$ is exponentially small in $m_0^1 - z_S$. For $0 \leqslant m_0^1 \leqslant z_S$, $J_1(\mathbf{m}) + \sum_{\alpha=1}^\infty J_\alpha(\mathbf{m}) = 0$.

Proof. Since $\mathbf{m}$ is a Maxwellian when $0 \leqslant m_0^1 \leqslant z_S$, the second part of the lemma is trivial. To prove the first part, we note that $J_\alpha(\mathbf{m}) = \mu J(m_0^1)(6 + 2S_\mathbf{d})$, $1 \leqslant \alpha \leqslant \alpha^* - 1$, $m_0^1 > z_S$, so that

$$\begin{aligned} J_1(\mathbf{m}) + \sum_{\alpha=1}^{\infty} J_\alpha(\mathbf{m}) = {} & \alpha^*(6 + 2S_\mathbf{d})\, J(m_0^1) + J_{\alpha^*}(\mathbf{m}) \\ & + 2 \sum_{\alpha=\alpha^*+1}^{\infty} \{2S_\mathbf{d} a_\alpha Q_\alpha(z_S)^\alpha m_0^1 \\ & + 6a_\alpha Q_\alpha(z_S)^\alpha m_0^1 - b_{\alpha+1} Q_{\alpha+1}(z_S)^{\alpha+1} S_\mathbf{d} \\ & - 3b_{\alpha+1} Q_{\alpha+1}(z_S)^{\alpha+1}\} \\ = {} & \alpha^*(6 + 2S_\mathbf{d})\, J(m_0^1) + J_{\alpha^*}(\mathbf{m}) \\ & + (6 + 2S_\mathbf{d})(m_0^1 - z_S) \sum_{\alpha=\alpha^*+1}^{\infty} a_\alpha Q_\alpha(z_S)^\alpha \end{aligned}$$

We now use (3.1) to obtain

$$\begin{aligned} J_1(\mathbf{m}) + \sum_{\alpha=1}^{\infty} J_\alpha(\mathbf{m}) \leqslant {} & \alpha^*(6 + 2S_\mathbf{d})\, J(m_0^1) \\ & + J_{\alpha^*}(\mathbf{m}) + (6 + 2S_\mathbf{d})\, A(m_0^1 - z_S)\, \rho^*(m_0^1) \end{aligned}$$

But since

$$J_{\alpha^*}(\mathbf{m}) = (6 + 2S_\mathbf{d})\, a_{\alpha^*} Q_{\alpha^*}(z_S)^{\alpha^*} m_0^1 - b_{\alpha^*+1}(S_\mathbf{d} + 3)\, Q_{\alpha^*+1}(z_S)^{\alpha^*+1}$$

we see that

$$|J_{\alpha^*}(\mathbf{m})| \leqslant (6 + 2S_\mathbf{d}) \left(\frac{m_0^1}{z_S} - 1\right) J^*(m_0^1), \qquad m_0^1 > z_S$$

Hence

$$\begin{aligned} \left|J_1(m) + \sum_{\alpha=1}^{\infty} J_\alpha(\mathbf{m})\right| \leqslant {} & \alpha^*(6 + 2S_\mathbf{d})\, J(m_0^1) + (6 + 2S_\mathbf{d}) \left(\frac{m_0^1}{z_S} - 1\right) J^*(m_0^1) \\ & + (6 + 2S_\mathbf{d})\, A(m_0^1 - z_S)\, \rho^*(m_0^1) \end{aligned} \tag{C.31}$$

Now use Theorem C.2 and the lemma is proven.

Inspection of Lemmas C.4–C.6 shows that the relevant quantities given are exponentially small in $m_0^1 - z_S$. The definition of approximate Maxwellian was given independently of its structure, i.e., only in terms of the macroscopic perturbation $\rho(\mathbf{n})-\rho_S$. The next lemma shows that for $\mathbf{m}$, our candidate for an approximate Maxwellian given by (4.2), the two perturbations are equivalent.

Lemma C.7. (a) For $0 \leqslant m_0^1 \leqslant z_S$, we have the estimate

$$z_S - m_0^1 \leqslant \rho_S - \rho(\mathbf{m}) \leqslant \text{const} \cdot (z_S - m_0^1)$$

while (b) for $z_S < m_0^1$, we have the bounds

$$m_0^1 - z_S \leqslant \rho(\mathbf{m}) - \rho_S \leqslant \frac{7}{A' z_S}(m_0^1 - z_S)(\alpha^*)^3 J(m_0^1)$$

Proof. (a) If $0 \leqslant m_0^1 \leqslant z_S$,

$$\begin{aligned}
\rho_S - \rho(\mathbf{m}) &= (1 + 2\cosh c_1 + 2\cosh c_2 + 2\cosh c_3) \\
&\quad \times \sum_{\alpha=1}^{\infty} \alpha Q_\alpha ((z_S)^\alpha - (m_0^1)^\alpha) \\
&\leqslant 7 \sum_{\alpha=1}^{\infty} \alpha Q_\alpha (z_S - m_0^1)(z_S^{\alpha-1} + z_S^{\alpha-1} m_0^1 + \cdots + (m_0^1)^{\alpha-1}) \\
&\leqslant 7 \sum_{\alpha=1}^{\infty} \alpha^2 Q_\alpha (z_S - m_0^1)\, z_S^{\alpha-1} \\
&\leqslant 7 \frac{z_S - m_0^1}{z_S} \sum_{\alpha=1}^{\infty} \alpha^2 Q_\alpha z_S^\alpha \leqslant \text{const} \cdot (z_S - m_0^1)
\end{aligned}$$

by Theorem C.2(ii). On the other hand, since

$$\begin{aligned}
\rho_S - \rho(\mathbf{m}) &= (1 + 2\cosh c_1 + 2\cosh c_2 + 2\cosh c_3) \\
&\quad \times \sum_{\alpha=1}^{\infty} \alpha Q_\alpha ((z_S)^\alpha - (m_0^1)^\alpha)
\end{aligned}$$

where all the terms in the sum are nonnegative, we can retain only the first terms to see $\rho_S - \rho(\mathbf{m}) \geqslant (z_S - m_0^1)$.

(b) If $z_S < m_0^1$,

$$\begin{aligned}
\rho(\mathbf{m}) - \rho_S &= (1 + 2\cosh c_1 + 2\cosh c_2 + 2\cosh c_3) \\
&\quad \times \sum_{\alpha=1}^{\infty} \alpha (f^\alpha(m_0^1) - Q_\alpha (z_S)^\alpha)
\end{aligned}$$

so again, since the terms in the series are nonnegative, we can use only the first term to see that $\rho(\mathbf{m}) - \rho_S \geqslant m_0^1 - z_S$. To obtain the bound from above we use the estimate $f^\alpha(m_0^1) \leqslant Q_\alpha(m_0^1)^\alpha$ of Lemma C.1(iv) to find

$$\begin{aligned}
\rho(\mathbf{m}) - \rho_S &\leqslant 7 \sum_{\alpha=1}^{\alpha^*} \alpha Q_\alpha(m_0^1 - z_S)((m_0^1)^{\alpha-1} \\
&\quad + z_S(m_0^1)^{\alpha-2} + \cdots + z_S^{\alpha-1}) \\
&\leqslant 7 \frac{m_0^1 - z_S}{z_S^2} \sum_{\alpha=1}^{\alpha^*} \alpha Q_\alpha((m_0^1)^{\alpha-1} + z_S(m_0^1)^{\alpha-2} + \cdots + z_S^{\alpha-1})\, z_S^2 \\
&\leqslant 7 \frac{m_0^1 - z_S}{z_S^2} \sum_{\alpha=1}^{\alpha^*} \alpha^2 Q_\alpha(m_0^1)^{\alpha+1} \\
&\leqslant 7 \frac{m_0^1 - z_S}{z_S^2} \sum_{\alpha=1}^{\alpha^*} \frac{\alpha^2 a_\alpha Q_\alpha(m_0^1)^{\alpha+1}}{a_\alpha} \\
&\leqslant 7 \frac{m_0^1 - z_S}{A' z_S^2} (\alpha^*)^3 J^*(m_0^1)
\end{aligned}$$

via (3.1) and definition of $J^*(m_0^1)$. The lemma is proven.

Proof of Theorem 4.4. Lemma C.7 and the fact that α^* is at most algebraically large as $m_0^1 > z_S$ and $J^*(m_0^1)$ is exponentially small as $m_0^1 \searrow z_S$ shows that $|m_0^1 - z_S| \leqslant |\rho(\mathbf{m}) - \rho_S|$ and $|\rho(\mathbf{m}) - \rho_S| \leqslant \text{const} \cdot |m_0^1 - z_S|$ for m_0^1 sufficiently close to z_S. Hence, if a quantity is exponentially small with respect to $|m_0^1 - z_S|$, it is exponentially small with respect to $|\rho(\mathbf{m}) - \rho_S|$. Now use Lemmas C.4–C.6 and the theorem is proven.

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REFERENCES

1. J. M. Ball and J. Carr, Asymptotic behaviour of solutions to the Becker–Döring equations for arbitrary initial data, *Proc. R. Soc. Edinburgh A* **1988**:109–116.
2. J. M. Ball, J. Carr, and O. Penrose, The Becker–Döring cluster equations: Basic properties and asymptotic behavior of solutions, *Commun. Math. Phys.* **104**:657–692 (1986).
3. N. Bellomo and L. de Socio, The discrete Boltzmann equation for gas mixtures: A regular space model and a shock wave problem, *Mech. Res. Comm.* **10**:233–238 (1983).
4. J. Brock, Simulation of aerosol dynamics, in *Theory of Dispersed Multiphase Flow*, R. E. Meyer, ed. (Academic Press, New York, 1983), pp. 135–172.

5. G.-Q. Chen, C. D. Levermore, and T.-P. Liu, Hyperbolic conservation laws with stiff relaxation terms and entropy, *Commun. Pure Appl. Math.* **47**:787–830 (1994).
6. S. Chen, H. Chen, G. D. Doolen, Y. C. Lee, H. Rose, and H. Brand, Lattice gas models for nonideal gas fluids, *Physica D* **47**:97–111 (1991).
7. S. K. Friedlander, *Smoke, Dust, and Haze. Fundamentals of Aerosol Behavior* (Wiley, New York, 1977).
8. E. Longo and R. Monaco, On the thermodynamics of discrete models of the Boltzmann equation for gas mixtures, *Transport Theory Stat. Phys.* **17**:423–442 (1988).
9. R. Monaco and M. Pandolfi Bianchi, A discrete velocity model for gases with chemical reactions of disassociation and recombination, in *Advances in Kinetic Theory and Continuum Mechanics*, R. Gatignol and Soubbaramayer, eds. (Springer-Verlag, Berlin, 1991), pp. 169–181.
10. R. Monaco and L. Preziosi, *Fluid Dynamics Applications of the Discrete Boltzmann Equation* (World Scientific, Singapore, 1991).
11. O. Penrose, *Kinetics of Phase Transitions* (Springer-Verlag, Berlin, 1978).
12. O. Penrose, Metastable states for the Becker–Döring cluster equations, *Commun. Math. Phys.* **124**:515–541 (1984).
13. O. Penrose and J. L. Lebowitz, Towards a rigorous molecular theory of metastability, in *Studies in Statistical Mechanics VII (Fluctuation Phenomena)*, E. Montroll and J. L. Lebowitz, eds. (North-Holland, Amsterdam, 1979), pp. 293–340.
14. M. Slemrod, Trend to equilibrium in the Becker–Döring cluster equations, *Nonlinearity* **2**:429–443 (1989).
15. M. Slemrod, M. Grinfeld, A. Qi, and I. Stewart, A discrete velocity coagulation—fragmentation model, *Math. Meth. Appl. Sci.* **18**:959–994 (1995).
16. M. Slemrod and A. Qi, Numerical simulations of cluster formation using a discrete velocity kinetic theory of gases, *Math. Models Meth. Appl. Sci.* **5**:619–640 (1995).