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12. Grandcanonical Ensemble

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Abstract

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Grandcanonical ensemble [tln60]

Consider an open classical system (volume V , temperature T , chemical potential μ). The goal is to determine the thermodynamic potential $\Omega(T, V, \mu)$ pertaining to that situation, from which all other thermodynamic properties can be derived.

A quantitative description of the grandcanonical ensemble requires a set of phase spaces Γ_N , $N = 0, 1, 2, \dots$ with probability densities $\rho_N(\mathbf{X})$. The interaction Hamiltonian for a system of N particles is $H_N(\mathbf{X})$.

Maximize Gibbs entropy $S = -k_B \sum_{N=0}^{\infty} \int_{\Gamma} d^{6N} X \rho_N(\mathbf{X}) \ln[C_N \rho_N(\mathbf{X})]$

subject to the three constraints

- $\sum_{N=0}^{\infty} \int_{\Gamma_N} d^{6N} X \rho_N(\mathbf{X}) = 1$ (normalization),
- $\sum_{N=0}^{\infty} \int_{\Gamma_N} d^{6N} X \rho_N(\mathbf{X}) H_N(\mathbf{X}) = \langle H \rangle = U$ (average energy),
- $\sum_{N=0}^{\infty} \int_{\Gamma_N} d^{6N} X \rho_N(\mathbf{X}) N = \langle N \rangle = \mathcal{N}$ (average number of particles).

Apply calculus of variation with three Lagrange multipliers:

$$\begin{aligned} \delta \left[\sum_{N=0}^{\infty} \int_{\Gamma_N} d^{6N} X \{ -k_B \rho_N \ln[C_N \rho_N] + \alpha_0 \rho_N + \alpha_U H_N \rho_N + \alpha_N N \rho_N \} \right] &= 0 \\ \Rightarrow \sum_{N=0}^{\infty} \int_{\Gamma_N} d^{6N} X \delta \rho_N \{ -k_B \ln[C_N \rho_N] - k_B + \alpha_0 + \alpha_U H_N + \alpha_N N \} &= 0 \\ \Rightarrow \{ \dots \} = 0 \Rightarrow \rho_N(\mathbf{X}) = \frac{1}{C_N} \exp \left(\frac{\alpha_0}{k_B} - 1 + \frac{\alpha_U}{k_B} H_N(\mathbf{X}) + \frac{\alpha_N}{k_B} N \right). \end{aligned}$$

Determine the Lagrange multipliers $\alpha_0, \alpha_U, \alpha_N$:

$$\exp \left(1 - \frac{\alpha_0}{k_B} \right) = \sum_{N=0}^{\infty} \frac{1}{C_N} \int_{\Gamma_N} d^{6N} X \exp \left(\frac{\alpha_U}{k_B} H_N(\mathbf{X}) + \frac{\alpha_N}{k_B} N \right) \equiv Z,$$

$$\sum_{N=0}^{\infty} \int_{\Gamma_N} d^{6N} X \rho_N(\mathbf{X}) \{ \dots \} = 0 \Rightarrow S - k_B + \alpha_0 + \alpha_U U + \alpha_N \mathcal{N} = 0$$

$$\Rightarrow U + \frac{1}{\alpha_U} S + \frac{\alpha_N}{\alpha_U} \mathcal{N} = \frac{k_B}{\alpha_U} \ln Z.$$

Compare with $U - TS - \mu\mathcal{N} = -pV = \Omega \Rightarrow \alpha_U = -\frac{1}{T}, \alpha_N = \frac{\mu}{T}$.

Grand potential: $\Omega(T, V, \mu) = -k_B T \ln Z = -pV$.

Grand partition function: $Z = \sum_{N=0}^{\infty} \frac{1}{C_N} \int_{\Gamma_N} d^{6N} X e^{-\beta H_N(\mathbf{X}) + \beta \mu N}, \beta = \frac{1}{k_B T}$.

Probability densities: $\rho_N(\mathbf{X}) = \frac{1}{Z C_N} e^{-\beta H_N(\mathbf{X}) + \beta \mu N}$.

Grandcanonical ensemble in quantum mechanics:

$$Z = \text{Tr} e^{-\beta(H - \mu N)}, \quad \rho = \frac{1}{Z} e^{-\beta(H - \mu N)}, \quad \Omega = -k_B T \ln Z.$$

Derivation of thermodynamic properties from grand potential:

$$S = - \left(\frac{\partial \Omega}{\partial T} \right)_{V, \mu}, \quad p = - \left(\frac{\partial \Omega}{\partial V} \right)_{T, \mu}, \quad \mathcal{N} = \langle N \rangle = - \left(\frac{\partial \Omega}{\partial \mu} \right)_{T, V}.$$

Relation between canonical and grandcanonical partition functions:

$$Z = \sum_{N=0}^{\infty} e^{\mu N / k_B T} Z_N = \sum_{N=0}^{\infty} z^N Z_N, \quad z \equiv e^{\mu / k_B T} \text{ (fugacity)}.$$

Open system of indistinguishable noninteracting particles:

$$Z_N = \frac{1}{N!} \tilde{Z}^N, \quad Z = \sum_{N=0}^{\infty} \frac{1}{N!} z^N \tilde{Z}^N = e^{z \tilde{Z}}$$

$$\Rightarrow \Omega = -k_B T \ln Z = -k_B T z \tilde{Z}.$$

Thermodynamic properties of the classical ideal gas in the grandcanonical ensemble are calculated in exercise [tex94].

[tex94] Classical ideal gas (grandcanonical ensemble)

Consider a classical ideal gas [$H_N = \sum_{i=1}^N (p_i^2/2m)$] in a box of volume V in equilibrium with heat and particle reservoirs at temperature T and chemical potential μ , respectively.

(a) Show that the grand partition function is $Z = \exp(zV/\lambda_T^3)$, where $z = \exp(\mu/k_B T)$ is the fugacity, and $\lambda_T = \sqrt{h^2/2\pi m k_B T}$ is the thermal wavelength.

(b) Derive from Z the grand potential $\Omega(T, V, \mu)$, the entropy $S(T, V, \mu)$, the pressure $p(T, V, \mu)$, and the average particle number $\langle N \rangle = \mathcal{N}(T, V, \mu)$.

(c) Derive from these expressions the familiar results for the internal energy $U = \frac{3}{2}\mathcal{N}k_B T$, and the ideal gas equation of state $pV = \mathcal{N}k_B T$.

Solution:

Density fluctuations and compressibility [tln61]

Average number of particles in volume V :

$$\mathcal{N} = \langle N \rangle = \sum_{N=0}^{\infty} \frac{1}{Z C_N} \int_{\Gamma_N} d^{6N} X N e^{-\beta H_N(\mathbf{x}) + \beta \mu N} = \frac{1}{Z \beta} \frac{\partial Z}{\partial \mu} = \frac{1}{\beta} \frac{\partial}{\partial \mu} \ln Z.$$

Fluctuations in particle number (in volume V):

$$\langle N^2 \rangle - \langle N \rangle^2 = \frac{1}{Z \beta^2} \frac{\partial^2 Z}{\partial \mu^2} - \left[\frac{1}{Z \beta} \frac{\partial Z}{\partial \mu} \right]^2 = \frac{1}{\beta^2} \frac{\partial^2 \ln Z}{\partial \mu^2} = \frac{1}{\beta^2} \frac{\partial(\beta \langle N \rangle)}{\partial \mu} = k_B T \left(\frac{\partial \mathcal{N}}{\partial \mu} \right)_{TV}.$$

Here we use $Z = Z(\beta, V, \mu)$.

$$\text{Gibbs-Duhem: } d\mu = \frac{V}{\mathcal{N}} dp - \frac{S}{\mathcal{N}} dT \Rightarrow \left(\frac{\partial \mu}{\partial(V/\mathcal{N})} \right)_T = \frac{V}{\mathcal{N}} \left(\frac{\partial p}{\partial(V/\mathcal{N})} \right)_T.$$

$$\text{For } V = \text{const: } \frac{\partial}{\partial(V/\mathcal{N})} = \frac{\partial \mathcal{N}}{\partial(V/\mathcal{N})} \frac{\partial}{\partial \mathcal{N}} = -\frac{\mathcal{N}^2}{V} \frac{\partial}{\partial \mathcal{N}}.$$

$$\text{For } \mathcal{N} = \text{const: } \frac{\partial}{\partial(V/\mathcal{N})} = \frac{\partial V}{\partial(V/\mathcal{N})} \frac{\partial}{\partial V} = \mathcal{N} \frac{\partial}{\partial V}.$$

$$\Rightarrow -\frac{\mathcal{N}^2}{V} \left(\frac{\partial \mu}{\partial \mathcal{N}} \right)_{TV} = V \left(\frac{\partial p}{\partial V} \right)_{T\mathcal{N}} \Rightarrow \left(\frac{\partial \mu}{\partial \mathcal{N}} \right)_{TV} = \frac{V}{\mathcal{N}^2} \kappa_T^{-1}.$$

$$\text{Compressibility: } \kappa_T \equiv -\frac{1}{V} \left(\frac{\partial V}{\partial p} \right)_{T\mathcal{N}}.$$

$$\text{Fluctuations in particle number: } \langle N^2 \rangle - \langle N \rangle^2 = \frac{\mathcal{N}^2}{V} k_B T \kappa_T.$$

An alternative expression for $\langle N^2 \rangle - \langle N \rangle^2$ is calculated in exercise [tex95].

The density fluctuations for a classical ideal gas are calculated in exercise [tex96].

At the critical point of a liquid-gas transition, the isotherm has an inflection point with zero slope ($\partial p / \partial V = 0$), implying $\kappa_T \rightarrow \infty$. The strongly enhanced density fluctuations are responsible for critical opalescence.

[tex95] Density fluctuations in the grandcanonical ensemble

Consider a system of indistinguishable particles in the grandcanonical ensemble. Derive the following two expressions for the fluctuations in the number of particles N for an open system of volume V in equilibrium with heat and particle reservoirs at temperature T and chemical potential μ , respectively:

$$\langle N^2 \rangle - \langle N \rangle^2 = z \frac{\partial}{\partial z} z \frac{\partial}{\partial z} \ln Z = k_B T V \frac{\partial^2 p}{\partial \mu^2},$$

where $z = \exp(\mu/k_B T)$ is the fugacity, $p(T, V, \mu) = -(\partial \Omega / \partial V)_{T\mu} = -\Omega/V$ is the pressure, and $\Omega(T, V, \mu) = -k_B T \ln Z$ is the grand potential.

Solution:

[tex96] Density fluctuations and compressibility of the classical ideal gas

(a) Use the results of [tex94] and [tex95] to show that the variance of the number of particles in a classical ideal gas (open system) is equal to the average number of particles:

$$\langle N^2 \rangle - \langle N \rangle^2 = \langle N \rangle = \mathcal{N}.$$

(b) Use this result to show that the isothermal compressibility of the classical ideal gas is $\kappa_T = 1/p$.

Solution:

[tex103] Energy fluctuations and thermal response functions

(a) Show that the following relation holds between the energy fluctuations in the microscopic ensemble and the heat capacity of a system described by a microscopic Hamiltonian H :

$$\langle (H - \langle H \rangle)^2 \rangle = k_B T^2 C_V.$$

(b) Prove the following relation in a similar manner:

$$\langle (H - \langle H \rangle)^3 \rangle = k_B^2 \left[T^4 \left(\frac{\partial C_V}{\partial T} \right)_V + 2T^3 C_V \right].$$

(c) Determine the relative fluctuations as measured by the quantities $\langle (H - \langle H \rangle)^2 \rangle / \langle H \rangle^2$ and $\langle (H - \langle H \rangle)^3 \rangle / \langle H \rangle^3$ for the classical ideal gas with N atoms.

Solution:

Microscopic states of ideal quantum gases [tln62]

Hamiltonian: $\hat{H}_N = \sum_{\ell=1}^N \hat{h}_\ell.$

1-particle eigenvalue equation: $\hat{h}_\ell |\mathbf{k}_\ell\rangle = \epsilon_\ell |\mathbf{k}_\ell\rangle.$

N -particle eigenvalue equation: $\hat{H}_N |\mathbf{k}_1, \dots, \mathbf{k}_N\rangle = E_N |\mathbf{k}_1, \dots, \mathbf{k}_N\rangle.$

Energy: $E_N = \sum_{\ell=1}^N \epsilon_\ell, \quad \epsilon_\ell = \frac{\hbar^2 \mathbf{k}_\ell^2}{2m}.$

N -particle product eigenstates: $|\mathbf{k}_1, \dots, \mathbf{k}_N\rangle = |\mathbf{k}_1\rangle \dots |\mathbf{k}_N\rangle.$

Symmetrized states for bosons: $|\mathbf{k}_1, \dots, \mathbf{k}_N\rangle^{(S)}.$

- $N = 2$: $|\mathbf{k}_1, \mathbf{k}_2\rangle^{(S)} = \frac{1}{\sqrt{2}} (|\mathbf{k}_1\rangle |\mathbf{k}_2\rangle + |\mathbf{k}_2\rangle |\mathbf{k}_1\rangle).$

Antisymmetrized states for fermions: $|\mathbf{k}_1, \dots, \mathbf{k}_N\rangle^{(A)}.$

- $N = 2$: $|\mathbf{k}_1, \mathbf{k}_2\rangle^{(A)} = \frac{1}{\sqrt{2}} (|\mathbf{k}_1\rangle |\mathbf{k}_2\rangle - |\mathbf{k}_2\rangle |\mathbf{k}_1\rangle).$

Occupation number representation: $|\mathbf{k}_1, \dots, \mathbf{k}_N\rangle \equiv |n_1, n_2, \dots\rangle.$

Here \mathbf{k}_1 represents the wave vector of the first particle, whereas n_1 refers to the number of particles in the first 1-particle state.

- energy: $\hat{H} |n_1, n_2, \dots\rangle = E |n_1, n_2, \dots\rangle, \quad E = \sum_{k=1}^{\infty} n_k \epsilon_k.$

- number of particles: $\hat{N} |n_1, n_2, \dots\rangle = N |n_1, n_2, \dots\rangle, \quad N = \sum_{k=1}^{\infty} n_k.$

ϵ_ℓ : energy of particle ℓ . ϵ_k : energy of 1-particle state k .

Allowed occupation numbers:

- bosons: $n_k = 0, 1, 2, \dots$
- fermions: $n_k = 0, 1.$

Partition function of ideal quantum gases [tln63]

Canonical partition function: $Z_N = \sum'_{\{n_k\}} \sigma(n_1, n_2, \dots) \exp\left(-\beta \sum_{k=1}^{\infty} n_k \epsilon_k\right)$.

$\sum'_{\{n_k\}}$: sum over all occupation numbers compatible with $\sum_{k=1}^{\infty} n_k = N$.

The statistical weight factor $\sigma(n_1, n_2, \dots)$ is different for fermions and bosons:

- Bose-Einstein statistics: $\sigma_{BE}(n_1, n_2, \dots) = 1$ for arbitrary values of n_k .
- Fermi-Dirac statistics: $\sigma_{FD}(n_1, n_2, \dots) = \begin{cases} 1 & \text{if all } n_k = 0, 1 \\ 0 & \text{otherwise} \end{cases}$.

What is the statistical weight factor for the Maxwell-Boltzmann gas?

$$\begin{aligned} Z_N &= \frac{1}{N!} \tilde{Z}^N = \frac{1}{N!} \left(\sum_{k=1}^{\infty} e^{-\beta \epsilon_k} \right)^N = \frac{1}{N!} \sum'_{\{n_k\}} \frac{N!}{n_1! n_2! \dots} (e^{-\beta \epsilon_1})^{n_1} (e^{-\beta \epsilon_2})^{n_2} \dots \\ &= \sum'_{\{n_k\}} \frac{1}{n_1! n_2! \dots} \exp\left(-\beta \sum_{k=1}^{\infty} n_k \epsilon_k\right). \end{aligned}$$

- Maxwell-Boltzmann statistics: $\sigma_{MB}(n_1, n_2, \dots) = \frac{1}{n_1! n_2! \dots}$.

Grandcanonical partition function:

$$\Rightarrow Z = \sum_{N=0}^{\infty} z^N Z_N = \sum_{\{n_k\}} \sigma(n_1, n_2, \dots) \exp\left(-\beta \sum_{k=1}^{\infty} n_k (\epsilon_k - \mu)\right),$$

where we have used $z^N = (e^{\beta \mu})^N = \exp\left(\beta \mu \sum_{k=1}^{\infty} n_k\right)$.

- $Z_{BE} = \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} \dots \exp\left(-\beta \sum_{k=1}^{\infty} n_k (\epsilon_k - \mu)\right) = \prod_{k=1}^{\infty} (1 - ze^{-\beta \epsilon_k})^{-1}$.
- $Z_{FD} = \sum_{n_1=0}^1 \sum_{n_2=0}^1 \dots \exp\left(-\beta \sum_{k=1}^{\infty} n_k (\epsilon_k - \mu)\right) = \prod_{k=1}^{\infty} (1 + ze^{-\beta \epsilon_k})$.
- $Z_{MB} = \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} \dots \frac{1}{n_1! n_2! \dots} \exp\left(-\beta \sum_{k=1}^{\infty} n_k (\epsilon_k - \mu)\right) = \prod_{k=1}^{\infty} \exp(ze^{-\beta \epsilon_k})$.

Ideal quantum gases: grand potential and thermal averages [tln64]

Grand potential: $\Omega(T, V, \mu) = -k_B T \ln Z = U - TS - \mu \mathcal{N} = -pV$.

- $\Omega_{MB} = -k_B T \sum_{k=1}^{\infty} z e^{-\beta \epsilon_k} = -k_B T \sum_{k=1}^{\infty} e^{-\beta(\epsilon_k - \mu)}$,
- $\Omega_{BE} = k_B T \sum_{k=1}^{\infty} \ln(1 - z e^{-\beta \epsilon_k}) = k_B T \sum_{k=1}^{\infty} \ln(1 - e^{-\beta(\epsilon_k - \mu)})$,
- $\Omega_{FD} = -k_B T \sum_{k=1}^{\infty} \ln(1 + z e^{-\beta \epsilon_k}) = -k_B T \sum_{k=1}^{\infty} \ln(1 + e^{-\beta(\epsilon_k - \mu)})$.

Parametric representation [$a = 1$ (FD), $a = 0$ (MB), $a = -1$ (BE)]:

$$\ln Z = \frac{pV}{k_B T} = \frac{1}{a} \sum_{k=1}^{\infty} \ln(1 + a z e^{-\beta \epsilon_k}).$$

Average number of particles:

$$\mathcal{N} = - \left(\frac{\partial \Omega}{\partial \mu} \right)_{T, V} = \frac{1}{\beta} \left(\frac{\partial \ln Z}{\partial \mu} \right)_{T, V} = \sum_{k=1}^{\infty} \frac{1}{z^{-1} e^{\beta \epsilon_k} + a} = \sum_{k=1}^{\infty} \langle n_k \rangle.$$

Average energy (internal energy):

$$U = - \left(\frac{\partial \ln Z}{\partial \beta} \right)_{z, V} = \sum_{k=1}^{\infty} \frac{\epsilon_k}{z^{-1} e^{\beta \epsilon_k} + a} = \sum_{k=1}^{\infty} \epsilon_k \langle n_k \rangle.$$

Average occupation number of energy level ϵ_k :

$$\langle n_k \rangle = -\beta^{-1} \frac{\partial \ln Z}{\partial \epsilon_k} = \frac{1}{e^{\beta(\epsilon_k - \mu)} + a}.$$

Fluctuations in occupation number [tex110]:

$$\langle n_k^2 \rangle - \langle n_k \rangle^2 = \beta^{-2} \frac{\partial^2 \ln Z}{\partial \epsilon_k^2}.$$

Average occupation numbers for MB, FD, and BE gases [ts135]

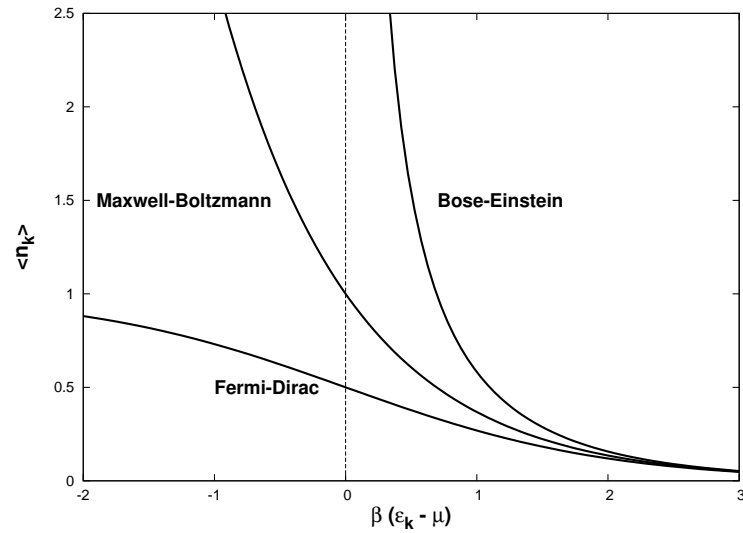
Average occupation number of energy level ϵ_k :

$$\langle n_k \rangle = \frac{1}{e^{\beta(\epsilon_k - \mu)} + a}$$

- $a = 1$: Fermi-Dirac gas,
- $a = 0$: Maxwell-Boltzmann gas,
- $a = -1$: Bose-Einstein gas.

Range of 1-particle energies: $\epsilon_k \geq 0$.

BE gas restriction: $\mu \leq 0 \Rightarrow 0 \leq z \leq 1$.



The BE and FD gases are well approximated by the MB gas provided the thermal wavelength $\lambda_T = \sqrt{h^2/2\pi mk_B T}$ is small compared to the average interparticle distance:

$$\beta(\epsilon_k - \mu) \gg 1 \Rightarrow -\beta\mu \gg 1 \Rightarrow z \ll 1.$$

$$[\text{tex94}] \text{ for } \mathcal{D} = 3: \Rightarrow \lambda_T \ll (V/\mathcal{N})^{1/3}.$$

[tex110] Occupation number fluctuations

Consider an ideal quantum gas specified by the grand partition function Z . Start from the expressions

$$\langle n_k^2 \rangle - \langle n_k \rangle^2 = \frac{1}{Z} \beta^{-2} \frac{\partial^2 Z}{\partial \epsilon_k^2} - \left[\frac{1}{Z} \beta^{-1} \frac{\partial Z}{\partial \epsilon_k} \right]^2, \quad \ln Z = \frac{1}{a} \sum_{k=1}^{\infty} \ln(1 + a z e^{-\beta \epsilon_k}),$$

where $a = +1, 0, -1$ represent the FD, MB, and BE cases, respectively, to derive the following result for the relative fluctuations in the occupation numbers:

$$\frac{\langle n_k^2 \rangle - \langle n_k \rangle^2}{\langle n_k \rangle^2} = \frac{1}{\langle n_k \rangle} - a.$$

Note that in the BE (FD) statistics, these fluctuations are enhanced (suppressed) relative to those in the MB statistics.

Solution:

[tex111] **Density of energy levels for ideal quantum gas**

Consider a nonrelativistic ideal quantum gas in \mathcal{D} dimensions and confined to a box of volume $V = L^{\mathcal{D}}$ with rigid walls. Show that the density of energy levels is

$$D(\epsilon) = \frac{L^{\mathcal{D}}}{\Gamma(\mathcal{D}/2)} \left(\frac{m}{2\pi\hbar^2} \right)^{\mathcal{D}/2} \epsilon^{\mathcal{D}/2-1}.$$

Solution:

[tex112] Maxwell-Boltzmann gas in \mathcal{D} dimensions

From the expressions for the grand potential and the density of energy levels of an ideal Maxwell-Boltzmann gas in \mathcal{D} dimensions and confined to a box of volume $V = L^{\mathcal{D}}$ with rigid walls,

$$\Omega(T, V, \mu) = -k_B T \sum_k e^{-\beta(\epsilon_k - \mu)}, \quad D(\epsilon) = \frac{L^{\mathcal{D}}}{\Gamma(\mathcal{D}/2)} \left(\frac{m}{2\pi\hbar^2} \right)^{\mathcal{D}/2} \epsilon^{\mathcal{D}/2-1},$$

derive the familiar results $pV = \mathcal{N}k_B T$ for the equation of state, $C_{V\mathcal{N}} = (\mathcal{D}/2)\mathcal{N}k_B$ for the heat capacity, and $pV^{(\mathcal{D}+2)/\mathcal{D}} = \text{const}$ for the adiabat at fixed \mathcal{N} .

Solution: