Quasi-Particle Picture for Monatomic Gases

K. Bärwinkel, J. Schnack¹, U. Thelker²

Universität Osnabrück, Fachbereich Physik
Barbarastr. 7, D-49069 Osnabrück

Abstract

A quasi-particle theory for monatomic gases in equilibrium is formulated and evaluated to yield the exact virial contributions to the thermodynamic state functions in lowest order of the density. Van der Waals blocking has necessarily to be accounted for in occupation number statistics. The quasi-particle distribution function differs from the Wigner function by a bilinear functional thereof. The progress made so far is promising with respect to a corresponding version of kinetic theory.

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1 Introduction and summary

The quasi-particle concept serves as a camouflage of interaction in many-body systems. The aim is to explain a macroscopic system in equilibrium as an ensemble of noninteracting and countable units called ”quasi-particles”. Non-equilibrium may then be described by a kinetic theory for these quasi-particles.

In this article the interaction-free theory is occupation number statistics which yields the entropy as a functional of the quasi-particle distribution function \( f \). Interaction between gas atoms is taken care of by modelling the elementary cell volume and by suitable constraints for \( f \).

Our theory reproduces the exact quantum mechanical density corrections to the distribution function and to equilibrium thermodynamics. The only partial success of related attempts (e.g. [1]) stems from an insufficient handling of the combinatorial entropy, i.e. the neglect of van der Waals blocking.

¹ Email: jschnack@uos.de, WWW: http://obelix.physik.uni-osnabrueck.de/~schnack
² present address: Westfalenring 58, D-49565 Bramsche
2 One-particle distribution function

The one-particle distribution function \( f(\vec{p}_1, \vec{r}, t) \) of gas kinetics enables the calculation of densities (in position space) for macroscopic quantities by taking moments over momentum space. We insist the two simplest ones be the number density \( n \)

\[
n(\vec{r}, t) = \int d^3p_1 \, f(\vec{p}_1, \vec{r}, t) \tag{1}
\]

and the density of linear momentum \( \vec{\pi} \)

\[
\vec{\pi}(\vec{r}, t) = \int d^3p_1 \, \vec{p}_1 \, f(\vec{p}_1, \vec{r}, t) . \tag{2}
\]

Any justification of gas kinetics within the frame of a more fundamental theory starts with a definition of \( f \) which one is free to choose, provided the interpretation of the moments (1) and (2) is correct. Our quantum statistical ansatz reads

\[
f(\vec{p}_1, \vec{r}, t) := f_W(\vec{p}_1, \vec{r}, t) + \Psi(\vec{p}_1, \vec{r}, t) \tag{3}
\]

where \( f_W \) is the Wigner distribution function

\[
f_W(\vec{p}_1, \vec{r}, t) = (2\pi\hbar)^{-3} \int d^3r' \left( \psi^\dagger(\vec{r} + \frac{\vec{r'}}{2}, t)\psi(\vec{r} - \frac{\vec{r'}}{2}, t) \right) \exp\left\{ \frac{i}{\hbar} \vec{p}_1 \cdot \vec{r}' \right\} \tag{4}
\]

and \( \Psi \) is a bilinear functional thereof

\[
\Psi(\vec{p}_1, \vec{r}, t) := \int d^3p_2 d^3p'_1 d^3p'_2 \, E(\vec{p}, \vec{p}') \delta(\vec{P} - \vec{P}') \left[ f_W(\vec{p}_1, \vec{r}, t)f_W(\vec{p}_2, \vec{r}, t) - f_W(\vec{p}'_1, \vec{r}, t)f_W(\vec{p}'_2, \vec{r}, t) \right]
+ \int d^3p_2 d^3p'_1 d^3p'_2 \, O(\vec{p}, \vec{p}') \delta(\vec{P} - \vec{P}') \left[ f_W(\vec{p}_1, \vec{r}, t)f_W(\vec{p}_2, \vec{r}, t) + f_W(\vec{p}'_1, \vec{r}, t)f_W(\vec{p}'_2, \vec{r}, t) \right] ,
\]

with \( \vec{P}, \vec{P}' \) and \( \vec{p}, \vec{p}' \) denoting centre-of-mass and relative momenta throughout the article

\[
\vec{P} = \vec{p}_1 + \vec{p}_2 , \quad \vec{p} = \frac{1}{2} (\vec{p}_1 - \vec{p}_2) . \tag{6}
\]

The even kernel

\[
E(\vec{p}, \vec{p}') = E(\vec{p}', \vec{p}) = E(-\vec{p}, -\vec{p}') \tag{7}
\]

and the odd kernel
\[ O(\vec{p}, \vec{p}') = -O(\vec{p}', \vec{p}) = O(-\vec{p}, -\vec{p}') \] (8)

remain to be specified. The structure of \( \Psi \) reminds of Boltzmann's collision integral although there is no \( \delta \)-function for the kinetic energies of relative motion \( E_p, E_{p'} \) which are defined by

\[ E_p = \frac{\vec{p}^2}{2m_{rel}} = \frac{\vec{p}^2}{m}. \] (9)

In any case equations (1) and (2) hold:

\[ n(\vec{r}, t) = \langle \psi^\dagger(\vec{r}, t) \psi(\vec{r}, t) \rangle = \int d^3p_1 \, f(\vec{p}_1, \vec{r}, t) = \int d^3p_1 \, f_W(\vec{p}_1, \vec{r}, t) \] (10)

and

\[ \pi(\vec{r}, t) = \int d^3p_1 \, p_1 \, f(\vec{p}_1, \vec{r}, t) = \int d^3p_1 \, p_1 \, f_W(\vec{p}_1, \vec{r}, t). \] (11)

There is not only a quantum statistical motivation to the definition (3), but it also allows a quasi-particle interpretation.

3 Quantum statistical background

Explicit expressions for the kernels \( E \) and \( O \) follow from the theory of Kadanoff and Baym [2], from which in the spatially homogeneous case and in \( \mathcal{T} \)-matrix approximation the kinetic equation

\[ \partial_t (f_W + \Psi) = J_B[f_W] \] (12)

is obtained [3,4] with \( J_B[f_W] \) as Boltzmann’s collision integral depending on \( f_W \). Here \( \Psi \) is just the functional (5) with the kernels

\[ O(\vec{p}, \vec{p'}) = \frac{1}{4}(2\pi\hbar)^3 \mathcal{P}(E_p - E_{p'}) \]

\[ \left\{ |\langle \vec{p} | \mathcal{T}_{\pm}(E_{p'} + i\epsilon) | \vec{p}' \rangle|^2 - |\langle \vec{p'} | \mathcal{T}_{\pm}(E_p + i\epsilon) | \vec{p} \rangle|^2 \right\} \] (13)

and

3
\[ E(\vec{p}, \vec{p}') = \pi (2\pi \hbar)^3 \delta (E_p - E_{p'}) \]
\[
\frac{1}{4} \frac{1}{(2\pi \hbar)^3} \mathcal{P}'(E_p - E_{p'}) \left\{ \left| \langle \vec{p} \right| \mathcal{T}_\pm (E_{p'} + i\epsilon) \left| \vec{p}' \right\rangle \right|^2 + \left| \langle \vec{p}' \right| \mathcal{T}_\pm (E_p + i\epsilon) \left| \vec{p} \right\rangle \right|^2 \right\} .
\]

The \( \mathcal{T} \)-matrix occurring here is the properly symmetrized momentum representation of the two-particle operator

\[ \mathcal{T}(z) = V - V \frac{1}{H - z} V , \quad \mathcal{T}'(z) = \frac{d}{dz} \mathcal{T}(z) , \] (15)

with \( H = H_{\text{kin}} + V \) being the Hamiltonian of relative motion. \( \mathcal{P} \) is the principle value distribution and \( \mathcal{P}' \) it’s derivative. The upper sign in \( \mathcal{T}_\pm \) (and elsewhere) refers to bosons and the lower sign to fermions.

In equilibrium, quantum statistical mechanics yields the density expansion for \( f = f_W + \Psi \), which up to second order reads

\[ f_{eq}(\vec{p}_1) = \frac{n}{(2\pi m\kappa T)^{3/2}} e^{-\frac{\vec{p}_1^2}{2m\kappa T}} (1 + n(2B(T) + \phi(\vec{p}_1))) , \] (16)

with \( \kappa \) being Boltzmann’s constant and

\[ \phi(\vec{p}_1) = \pm \lambda^3(T) e^{-\frac{\vec{p}_1^2}{2m\kappa T}} - \lambda^3(T) \int d^3 p_2 e^{-\frac{\vec{p}_2^2}{2m\kappa T}} \left\{ \tilde{F}(p) + \tilde{G}(p) \right\} . \]

Here we have introduced the thermal wave length

\[ \lambda = \frac{2\pi \hbar}{\sqrt{2\pi m\kappa T}} . \] (17)

The quantities \( \tilde{F} \) and \( \tilde{G} \) as well as the second virial coefficient \( B(T) \) are given as functionals of the \( \mathcal{T} \)-matrix:

\[ \tilde{F}(p) = \frac{1}{2} (2\pi \hbar)^3 \operatorname{Re} \left( \langle \vec{p} \right| \mathcal{T}_\pm (E_p + i\epsilon) \left| \vec{p} \rangle \right) , \] (18)

\[ \tilde{G}(p) = \frac{\pi}{2} (2\pi \hbar)^3 \int d^3 q \quad \delta (E_p - E_q) \]
\[
\times \frac{1}{4} \frac{1}{(2\pi \hbar)^3} \mathcal{P}'(E_q - E_{p'}) \left\{ \left| \langle \vec{q} \right| \mathcal{T}_\pm (E_q + i\epsilon) \left| \vec{p} \right\rangle \right|^2 + \left| \langle \vec{p} \right| \mathcal{T}_\pm (E_q + i\epsilon) \left| \vec{q} \right\rangle \right|^2 \right\} . \] (19)
\[ B(T) = B_0(T) + B_1(T) + B_2(T) \]  

\[ B_0(T) = \mp 2^{-5/2} \lambda^3, \quad B_1(T) = \frac{\langle \langle \tilde{F} \rangle \rangle}{\kappa T}, \quad B_2(T) = \langle \langle \tilde{G} \rangle \rangle, \]  

where \( \langle \langle \cdot \rangle \rangle \) denotes the thermal average, e.g.

\[ \langle \langle \tilde{F} \rangle \rangle = \frac{\int d^3 p \exp\{-\frac{E_p}{\kappa T}\} \tilde{F}(p)}{\int d^3 p \exp\{-\frac{E_p}{\kappa T}\}}. \]  

These formulae are exact if the two-particle interaction does not allow any bound states. \( B_1 \) essentially accounts for long-range attraction and \( B_2 \) for hard repulsion, this correspondence being most striking in the van der Waals limit (cf. section 6).

### 4 Quasi-particle picture for equilibrium

We just work out the usual idea:

1. The entropy density \( s \) is represented as a functional of the one-particle distribution function. This is an outcome of occupation number statistics (combinatorial entropy)

\[ s = -\kappa \int d^3 p_1 \left[ f(p_1) \ln \left( v_{el}(p_1) f(p_1) \right) \right. \]

\[ \left. \mp \left( \frac{1}{v_{el}(p_1)} \pm f(p_1) \right) \ln \left( 1 \pm v_{el}(p_1) f(p_1) \right) \right] \]  

with \( v_{el} \) as the volume of an elementary cell in six-dimensional \( \mu \)-space, \( v_{el} \) accommodating one single-particle quantum state. Eq. (22) is well known as a standard result for non-interacting particles. However, the choice of \( v_{el} \) and the constraints for \( f \) may provide a camouflage of interaction.

2. For equilibrium the distribution function \( f \) is the one which minimizes \( s \) subject to appropriate constraints.

3. The constraints are a given number density \( n \),

\[ n = \int d^3 p_1 f(p_1), \]  

and a given energy density \( u \),

\[ u = \int d^3 p_1 \varepsilon(p_1) f(p_1). \]  

The quasi-particle interpretation is now introduced by way of ansatz (eqs. (25), (26)), its aim being to account for interaction effects in lowest order of the density. Strong repulsion
reduces the freely accessible volume for gas particles. Because of this effect, called "van der Waals blocking", more than just \((2\pi\hbar)^3\) is needed as an elementary cell volume

\[ v_{el}(\bar{p}_1) = (2\pi\hbar)^3 \left[ 1 + \int d^3p_2 \, G(|\bar{p}|) \, f(\bar{p}_2) \right]. \tag{25} \]

Also, an interacting gas particle carries with it a correlation cloud giving rise to an interactive contribution which changes the kinetic energy of a particle into the energy of a quasi particle

\[ \varepsilon(\bar{p}_1) = \frac{\bar{p}_1^2}{2m} + \int d^3p_2 \, F(|\bar{p}|) \, f(\bar{p}_2). \tag{26} \]

For our variational problem the functions \(G\) and \(F\) are considered as given though, for the time being, unknown. They will be determined afterwards by comparing the equilibrium solution \(f = f_{eq}\) with the corresponding expression from many-body quantum theory. The resulting thermodynamics then serves as a further touchstone of the quasi-particle interpretation.

The solution of the variational problem is obviously equivalent to

\[ \left( \frac{\delta s}{\delta f(\bar{p})} \right)_{f = f_{eq}} = \frac{1}{\vartheta} \left( \frac{\delta u}{\delta f(\bar{p})} - \alpha \frac{\delta n}{\delta f(\bar{p})} \right)_{f = f_{eq}} \tag{27} \]

with \(\vartheta\) and \(\alpha\) as Lagrange parameters due to the the constraints for \(f\). Comparison with the thermodynamic identity for the entropy \(S\) at constant volume

\[ dS = \frac{1}{T} (dU - \mu dN) \tag{28} \]

reveals that \(\vartheta\) means the temperature, \(\vartheta = T\), and \(\alpha\) means the chemical potential, \(\alpha = \mu\). According to (27) \(f = f_{eq}\) is equivalently determined by the fixed-point equation

\[ f(\bar{p}_1) = \frac{\zeta}{v_{el}(\bar{p}_1)} \frac{\exp \left\{ -\beta \left( \frac{\bar{p}_1^2}{2m} + K(\bar{p}_1) \right) \right\}}{1 + \zeta \exp \left\{ -\beta \left( \frac{\bar{p}_1^2}{2m} + K(\bar{p}_1) \right) \right\}} \tag{29} \]

with

\[ \beta = \frac{1}{\kappa T}, \quad \zeta = \exp \{\beta \mu\} \tag{30} \]

and
\[ K(\vec{p}_1) = \int d^3p_2 \left\{ 2f(\vec{p}_2)F(\{|\vec{p}|\}) \pm \frac{(2\pi\hbar)^3}{\beta v_{el}^2(\vec{p}_2)} \ln \left( 1 \pm v_{el}(\vec{p}_2)f(\vec{p}_2) \right) G(\{|\vec{p}|\}) \right\}. \] (31)

5 Lowest-order density corrections

The fugacity \( \zeta \) can be given as a power series in \( n \)

\[ \zeta = n\lambda^3 (1 + 2nB(T) + \cdots) \] (32)

and the fixed point equation (29) may be iterated starting off with a Maxwellian normalized to \( n \). This yields a density expansion according to which – apart from third and higher order contributions – one regains eq. (16) for \( f \), but now with

\[ \phi(\vec{p}_1) = \pm \lambda^3 e^{-\frac{\beta}{2}E_{p1}} - \frac{2\lambda^3}{(2\pi\hbar)^3} \int d^3p_2 e^{-\frac{\beta}{2}E_{p2}} \left\{ \beta F(\{|\vec{p}|\}) + G(\{|\vec{p}|\}) \right\}. \] (33)

Therefore, the quasi-particle picture independently explains eq. (16), if and only if

\[ F(p) = \tilde{F}(p) \] (34)

and

\[ G(p) = \tilde{G}(p) \] (35)

Having determined \( f \) up to second order in \( n \), we deduce the entropy with its lowest order density corrections from eq. (22):

\[ s = n\kappa \left\{ \frac{5}{2} - \ln \left( n\lambda^3 \right) - n \left[ B(T) + TB'(T) \right] \right\}, \] (36)

with \( B(T) \) defined by eqs. (20). Analogously evaluating eq. (24), we obtain the two leading contributions to the energy density:

\[ u = n\kappa T \left\{ \frac{3}{2} - nTB'(T) \right\}. \] (37)
The last two results imply the pressure equation of state

\[
p = n \left( \frac{\partial u}{\partial n} \right)_{s/n} - u = n\kappa T \{1 + nB(T)\}.
\]  

(38)

The exact density corrections, i.e. virial contributions have thus been obtained.

6 Classical van der Waals approximation

Considering distinguishable particles, one has to neglect quantum statistical contributions. This means

\[
B_0 = 0.
\]  

(39)

Then with (20), (34) and (35)

\[
B(T) = \frac{\langle\langle F \rangle\rangle}{\kappa T} + \langle\langle G \rangle\rangle
\]  

(40)

holds and suggests a comparison with the van der Waals version of the second virial coefficient

\[
B_{vdW}(T) = -\frac{a}{\kappa T} + b,
\]  

(41)

which is readily obtained from the model equation of state

\[
(p + n^2a)(1 - nb) = n\kappa T
\]  

(42)

and the corresponding density expansion

\[
p = n\kappa T \left(1 + n \left[ b - \frac{a}{\kappa T} \right] \right) + o(n^3).
\]  

(43)

The van der Waals limit therefore obviously means

\[
\langle\langle F \rangle\rangle = \text{const} = -a \quad \text{and} \quad \langle\langle G \rangle\rangle = \text{const} = b.
\]  

(44)

This model assumption is actually quite reasonable as we are going to demonstrate for $^4$He atoms interacting via a Lennard-Jones-potential lacking bound states [5]:

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\[ V(r) = 4V_0 \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^6 \right] ; \quad V_0 = 10.22 \text{ K } \sigma = 2.56 \text{ Å} . \] (45)

Because the interaction is radially symmetric the following relations between the (anti-)symmetrized \( T \)-matrix, the scattering amplitude \( f_\pm(p, \theta) \) and phase shifts \( \delta_l(p) \)

\[
\begin{align*}
  f_\pm(p, \theta) &= -\pi^2 \hbar \left( \vec{p} \right) T_\pm \left( E_p + i\epsilon \right) \left( \vec{q} \right) , \quad |\vec{p}| = |\vec{q}| , \quad \vec{p} \cdot \vec{q} = |\vec{p}||\vec{q}| \cos(\theta) \\
  f_\pm(p, \theta) &= \frac{\hbar}{p} \sum'_{l} (2l + 1) e^{i\delta_l(p)} \sin(\delta_l(p)) P(\cos(\theta))
\end{align*}
\] (46)

may be used (see e.g. [6]), where the summation runs over even \( l \) for bosons and odd \( l \) for fermions. \( F(p) \) and \( G(p) \) (compare to eqs. (18) and (19)) can then be expressed in terms of phase shifts which is compatible with the Beth Uhlenbeck result for \( B(T) \) [7]

\[
\begin{align*}
  F(p) &= -\frac{4\pi \hbar^2}{m} f_{\vec{p}}(0) = -\frac{4\pi \hbar^2}{m} \frac{\hbar}{2p} \sum'_{l} (2l + 1) \sin\left[2\delta_l(p)\right] \\
  G(p) &= -\hbar \int d\Omega \text{ Im} \left[ \left(f_{\vec{p}}(\theta)\right)^* \frac{\partial}{\partial p} f_{\vec{p}}(\theta) \right] = -\frac{4\pi \hbar^2}{p^2} \sum'_{l} (2l + 1) \sin^2\left[\delta_l(p)\right] \frac{\partial \delta_l}{\partial p}.
\end{align*}
\] (48)

Figure 1 nicely shows that for a large region of temperatures \( \langle F \rangle \) and \( \langle G \rangle \) may be nearly considered as constants, i.e. not depending on temperature. In this case the single particle energy, the elementary cell volume and the mean energy are easily determined as

\[
\begin{align*}
  \varepsilon(\vec{p}) &= \frac{\vec{p}^2}{2m} - an , \quad v_{el}(\vec{p}) = (2\pi \hbar)^3 \left[ 1 + bn \right] , \quad u = n\kappa T \left\{ \frac{3}{2} - \frac{na}{\kappa T} \right\} .
\end{align*}
\] (49)
7 Physical meaning of the quasi-particles

One may imagine a quasi-particle to be a gas particle together with its surrounding correlation cloud which is described by the radial distribution function $g(r, T)$. This interpretation suggests itself because the virial correction to the energy of the ideal gas (eq. (37)) is mainly determined by $g(r, T)$ in an obvious way. The interpretation is immediately evident in the classical case where

$$B(T) = B_{cl}(T) = -\frac{1}{2} \int d^3 r \ (g_0(r, T) - 1)$$  \hspace{1cm} (50)

with

$$g_0(r, T) = g_{0, cl}(r, T) = \exp \left\{ -\frac{V(r)}{\kappa T} \right\}$$  \hspace{1cm} (51)

as the first term of the density expansion

$$g(r, T) = g_0(r, T) + n g_1(r, T) + n^2 g_2(r, T) + \cdots .$$  \hspace{1cm} (52)

Therefore

$$n^2 \kappa T T B'_{cl}(T) = \frac{n^2}{2} \int d^3 r \ g_0, cl(r, T) V(r)$$  \hspace{1cm} (53)

is the classical virial correction to the internal energy.

In general, however, for a homogeneous system

$$g(r, T) = \frac{1}{n^2} \langle \langle \psi^\dagger(\vec{r}') \psi^\dagger(\vec{r}' + \vec{r}) \psi(\vec{r}' + \vec{r}) \psi(\vec{r}') \rangle \rangle$$

$$= \frac{2}{n} \frac{\delta f_{free}}{\delta V(r)}$$  \hspace{1cm} (54) \hspace{1cm} (55)

with $f_{free}$ being the free energy per particle, whence

$$g_0(r, T) = 2\kappa T \frac{\delta B(T)}{\delta V(r)} .$$  \hspace{1cm} (56)

Then quantum mechanically \cite{8,4}

$$\int d^3 r \ g_0(r, T) V(r) = 2^{3/2} \lambda^3 \int d^3 p \ e^{-\beta E_p} (-\langle \vec{p} | V_\pm | \vec{p} \rangle)^{-} ,$$  \hspace{1cm} (57)
where the scattering eigenstates $|\vec{p}\rangle^{(-)}$ satisfy the Lippmann Schwinger equation which reads in momentum representation

$$
\langle \vec{p}' | \vec{p} \rangle^{(-)} = \delta(\vec{p}' - \vec{p}) - (E_{p'} - E_p - i\epsilon) \langle \vec{p}' | V | \vec{p} \rangle^{(-)} .
$$

The virial correction to the energy density (eqs. (24), (37)) is made up of three terms

$$
n^2 \kappa T TB'(T) = \int d^3 p_1 \frac{\vec{p}_1^2}{2m} \Psi_M(\vec{p}_1) + \int d^3 p_1 d^3 p_2 f_M(\vec{p}_1) f_M(\vec{p}_2) F(p)
+ \int d^3 p_1 \frac{\vec{p}_1^2}{2m} n^2 f_{W,2}(\vec{p}_1) .
$$

Here $f_M$ is the Maxwellian normalized to $n$ and $\Psi_M$ is our functional (5) with $f_W$ replaced by $f_M$. $f_{W,2}$ denotes the second term in the density expansion of the Wigner function

$$
f_W(\vec{p}_1) = f_M(\vec{p}_1) + n^2 f_{W,2}(\vec{p}_1) + \cdots .
$$

Looking more closely at eq. (59) and taking account of (57) one can show that

$$
\int d^3 p_1 \frac{\vec{p}_1^2}{2m} \Psi_M(\vec{p}_1) + \int d^3 p_1 d^3 p_2 f_M(\vec{p}_1) f_M(\vec{p}_2) F(p) = \int d^3 r g_0(r,T)V(r) .
$$

Therefore, in the classical limit the last term in eq. (59) must vanish.

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