Fluid analysis of positrons in soft matter

G J Boyle¹, R D White¹, R E Robson¹,²
¹ ARC Centre for Antimatter-Matter Studies, School of Engineering and the Physical Science, James Cook University, Townsville 4811, QLD, Australia
² ARC Centre for Antimatter-Matter Studies, Research School of Physical Sciences and Engineering, Australian National University, 2600 Canberra, Australia
E-mail: gregory.boyle@jcu.edu.au

Abstract.
Improvements to Positron Emission Tomography (PET) technology depend to a large extent on obtaining a better understanding of positron transport processes in soft condensed matter. The most economically computational, physically tenable method of carrying out such an investigation is the fluid approach, in which the Boltzmann kinetic equation is replaced by a hierarchy of low order, velocity moment equations, in which collision terms are approximated by extending ‘momentum transfer theory’ to allow for coherent scattering arising from the structure of the medium. We show how these fluid equations furnish structure-modified versions of well known semi-empirical relations linking transport coefficients, and give numerical calculations for simple models.

1. Introduction
Positron Emission Tomography (PET) has become a well established and powerful, non-invasive tool for measuring biological processes in humans and animals [1]. However, most current models do not systematically address the inherent structure of the medium. This is critical as the biological materials that constitute the background media for PET procedures consist of soft condensed matter involving structure. The program at James Cook University aims to address this issue, and, although the present work involving the calculation of universal transport coefficients is not directly applicable to PET, it does represent a first step toward a complete transport model valid in structured media.

The approach to including structure in the framework of modern kinetic theory was proposed by Cohen and Lekner in 1967 [2], using a modified differential cross-section given by Van Hove [3]. Such microscopic information alone is insufficient for applications to the macroscopic world. The Boltzmann equation represents the fundamental equation from which macroscopic transport properties of interest, such as drift velocity and mean energy, can be determined [4]. However, the integro-differential nature of Boltzmann’s equation makes finding a general solution for the velocity distribution function a formidable task.

The approach undertaken in this paper involves approximate ‘moment’ representations of the Boltzmann equation, otherwise known as the ‘fluid description’ [5]. The set of moment or balance equations can be found by multiplying the Boltzmann equation by some property of the swarm velocity, Ψ(v), and integrating over all velocities. Different forms chosen for Ψ(v) yield the balance equations for the different moments. Momentum transfer theory (MTT) [4]
can be used to facilitate analytic solutions and simplify computations for the resulting coupled non-linear differential equations.

In Section 2 the fluid treatment of the new kinetic equation accounting for the structure of the medium are undertaken using MTT. For a special case, the generalized Wannier energy relation \[4\] and a criterion for the phenomena of structure-induced negative differential conductivity (NDC) \[6\] are derived analytically. In Section 3 a numerical scheme is used to solve the balance equations in soft-condensed matter. The Percus-Yevick model \[7\] is used to simulate structure, and investigate the effect of volume fractions on the spatially homogeneous transport coefficients.

2. Theory

2.1. Boltzmann equation and the material structure

The fundamental equation from which macroscopic transport properties can be determined is the Boltzmann equation,

\[
\left( \frac{\partial}{\partial t} + v \cdot \nabla + \frac{F}{m} \cdot \frac{\partial}{\partial v} \right) f = -J(f),
\]

where \(f\) is the phase-space distribution function, \(v\) is the velocity and \(m\) is mass, all relating to the swarm particle, and \(F\) refers to the external forces applied to the system. The right hand side is the eponymous Boltzmann collision integral and describes the effect of collisions on the velocity distribution function \(f\) at a fixed position and velocity. The collision integral accounts for the different processes, and generally the structure of the medium is only included via the elastic component, which is modified to account for coherent scattering. Robson and White \[8\] give derivations for the first and second order differential cross-sections valid in soft condensed media, and found an expression for the collisional rate of change involving the dynamic structure factor \(S(K, \Omega)\), where \(K\) and \(\Omega\) are the momentum and energy exchanges during a collision. In the ‘single-scatterer approximation’ \(S(K, \Omega)\), which is the Fourier transform of the Van Hove \[3\] space-time pair correlation function, contains all the necessary information about the excitations of the system \[2\]. The structure factor is purely determined by the medium, and can be found in neutron or x-ray scattering experiments. The integral quantity,

\[
S(K) \equiv \int_{-\infty}^{\infty} d\Omega \ S(K, \Omega)
\]

represents the static structure factor, which is evaluated at \(K = 2k \sin(\chi/2)\) \[8\], i.e. the momentum transfer in an elastic collision in which the positron is scattered through an angle \(\chi\). For the isotropic scattering considered in this work, the momentum transfer cross section can be written as \(\tilde{\sigma}_m(v) = \sigma_m(v)s(v)\), where \(\sigma_m(v)\) is the momentum transfer cross section for single particle scattering, and \(s(v)\) is the angle-integrated structure factor given by

\[
s(v) = \frac{1}{2} \int_{-1}^{1} S \left( \frac{2mv}{\hbar} \sin \left( \frac{\chi}{2} \right) \right) (1 - \cos \chi) d(\cos \chi).
\]

which also plays an important role in this discussion.

2.2. Fluid treatment

A full and general solution to (1) is formidable. A fluid equation treatment can yield approximate absolute results and relationships between physically measurable properties. Here the problem of solving the Boltzmann equation for \(f\) in phase space is replaced by a low order set of approximate equations for the moments of \(f\) \[5, 9\]. The set of moment equations is found by multiplying the modified Boltzmann equation (1) by an arbitrary property of swarm particle velocity, \(\Psi(v)\),
and integrating over all velocities. Momentum transfer theory provides a way of evaluating the collision terms \[4\] with good qualitative and quantitative results. Essentially, the mathematical form of expressions derived for the constant collision frequency model are assumed to carry over to the general, energy-dependent collision frequency case. The momentum and energy balance equations can then be found \[10\]. The homogeneous balance equations represent coupled first order differential equations, which can only be solved analytically in some special cases.

2.3. Generalized Wannier energy relation and structure-induced NDC

The simple case of a steady, spatially uniform swarm undergoing elastic collisions and subject to an electric field only is now considered. All time and space derivatives vanish, and the momentum and energy balance take the form,

\[
eE = \mu \tilde{\nu}_m(\epsilon) W, \tag{4}
\]

\[
\epsilon = \frac{3}{2} k_B T_0 + \frac{1}{2} m_0 W^2 \frac{\tilde{\nu}_m}{\nu_m}. \tag{5}
\]

where \(e\) is the charge, \(E\) is the electric field, \(\mu\) is the reduced mass, \(m_0\) is the neutral mass, \(W\) is the flux drift velocity, \(\epsilon\) is the mean energy, \(T_0\) is the neutral temperature, and \(\nu_m\) and \(\tilde{\nu}_m\) are the elastic momentum collision frequencies, the latter for coherent scattering. Equation (5) is a generalization of the well known Wannier energy relation in gas transport theory derived by Wannier in 1953 \[4\]. If the interparticle spacing is large compared to the de B\'ogie wavelength of the positron then the angle-integrated structure factor \(s(\epsilon) \rightarrow 1\), and subsequently \(\tilde{\nu}_m \rightarrow \nu_m\) reducing to the dilute gas case expressions.

Another important phenomena that is predicted by transport theory is negative differential conductivity (NDC). NDC is characterized by a decrease in the drift velocity \(W\) despite an increase in the magnitude of applied electric field \(E\), i.e. \(\frac{dW}{dE} < 0\). It is well known in gas transport theory that, for some energy profiles, the inclusion of inelastic or non-conservative scattering cross sections can cause regions of NDC \[11, 12\]. For a condensed media there is a new type of NDC which is purely a consequence of the medium structure \[6\]. For the cold gas approximation \((T = 0)\) it can be shown that,

\[
\frac{1}{m_0W} \left( \frac{\tilde{\nu}_m}{\nu_m} \right)^{-1} \left[ 1 - \frac{d \ln(\frac{\tilde{\nu}_m}{\nu_m})}{d \ln \epsilon} \right] \frac{d \epsilon}{d \ln E} = \frac{dW}{d \ln E}. \tag{6}
\]

By considering the signs of the constituents of the left hand side and right hand side of (6) it is evident that for structure-induced NDC to occur, then the inequality

\[
\frac{d \ln(\frac{\tilde{\nu}_m}{\nu_m})}{d \ln \epsilon} > 1, \tag{7}
\]

must be satisfied.

3. Results and discussion

A numerical scheme was developed to solve the coupled non-linear equations (4) and (5).

3.1. Percus-Yevick

In order to investigate the effects of structured media on positron transport, a model for the static structure factor, \(S\), of the medium is required. The model of Percus and Yevick (with the Verlet Weis correction) provides a simple description of the structure behaviour with particularly good agreement for hard spheres \[7, 13\]. The important factor is the volume fraction, \(\Phi\), which is a measure of how tightly packed the particulates of the media are. Figure 1a shows the static
structure value $S(K)$, for different values of $\Phi$. In the limit as $K \to \infty$, $S \to 1$ and the dilute gas case is regained.

The relationship between the angle-integrated structure factor $s$ and energy $\epsilon$ is shown in Figure 1b. It illustrates the oscillatory behaviour caused by structured media, and convergence to the dilute gas-phase-case for high energies. These features are echoed in the behaviour of the transport coefficients below.

3.2. Maxwell Model

The model considered here involves an elastic collision frequency which is energy independent i.e., $\sigma_m = 6\epsilon^{-1/2} \AA^2$, $m_0 = 4$ amu, and $T_0 = 0$ K. This is known as the Maxwell Model.

Figure 2a shows the variation of the mean energy with $E/n_0$ for different volume fractions. The profile for $\Phi = 0$ represents the dilute gas phase. $\tilde{\nu}_m$ is independent of energy and (4) gives $W \propto E$ as shown. When structure is included in the medium, the energy and drift equations become coupled and non-linear which give rise to large deviations from the dilute gas phase case behaviour. At low field strengths, the mean energy increases with the volume fraction, but as $E/n_0$ increases each of the profiles converge on the structureless line. Physically, as the energy of the incoming positron increases, its de Broglie wavelength decreases until it is effectively sampling a single medium particle during a collision and the structure effects are limited.

The variation in drift velocity with $E/n_0$ for various volume fractions is shown in Figure 2b. The larger volume fractions experience larger oscillations away from the dilute gas phase case, and again converge for high field strengths. The profiles in Figure 2b exhibit NDC for volume fractions above some critical value. A condition for the occurrence of this NDC was given
in (7). For the Maxwell model, $\tilde{\nu}_m/\nu_m$ is effectively the angle-integrated structure factor $s$, and Figure 1b shows the log plot of $s$ superimposed with straight lines of slope 1. There are energies for the $\Phi = 0.3$ and $\Phi = 0.4$ profiles for which the slope of $\ln s/\ln \varepsilon$ exceeds 1, but for lower $\Phi$ there are not. This coincides with the occurrence of NDC in Figure 2b for only the $\Phi = 0.3$ and $\Phi = 0.4$ profiles. It can be deduced from (4) that during NDC, a small increase $E$ is accompanied by a rapid increase in $\tilde{\nu}_m$ resulting in an overall decrease in the drift velocity.

4. Conclusion
A fluid equation treatment of swarms in structured media was undertaken. Semi-analytic relations for drift velocity and mean energy were found from moments of the Boltzmann equation using MTT. By considering a simple steady-state situation, analytic expressions for the drift velocity and Wannier energy relation in structured media were derived. A condition for the occurrence of the new phenomena of structure-induced NDC was presented. Using a basic numerical code, the relationships between mean energy and drift velocity were investigated using the Percus-Yevick model to simulate structure. Excellent agreement was found between the numerical results and theory, and structure-induced NDC was clearly demonstrated.

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