Diffusion of muonic atoms

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Transport of muonic hydrogen and deuterium atoms in gaseous hydrogen and deuterium is studied in the diffusion approximation and by means of the multiple-collision expansion. The diffusion coefficient is derived. Numerical results of the time-dependent problem in slab geometry are presented for a number of initial energies, temperatures, and pressures.

I. INTRODUCTION

An understanding of the transport of muonic hydrogen and/or deuterium atoms in a medium of molecular hydrogen and/or deuterium is of interest in the analysis of muon experiments\(^1\) and also in the study of muon-catalyzed fusion.\(^3\) In this paper we develop a method for treating this transport which is basically a modified time-dependent diffusion theory, similar to the Selengut-Goertzel method\(^4\) developed in the 1950s for treating neutron transport in \(\text{H}_2\text{O}\) reactors. The muonic situation is considerably more complicated than the neutron case, because muonic atoms can be exited and de-excited between hyperfine states by colliding with the molecules. The hyperfine splitting, negligibly small in the case of ordinary atomic hydrogen, is around 0.183 eV for muonic hydrogen because it is proportional to the muonic mass squared. Since the kinetic energy of the muonic atom during this diffusion process varies in the range from about 1 eV to thermal energy, we see that the hyperfine splitting cannot be ignored.

Actually, the kinematics for the processes considered here have been developed and published previously.\(^5\) The results of Ref. 9 will be used extensively in the present work.

Since we are using a variant of diffusion theory, our results are not applicable to some of the experimental data, namely, those experiments which were done in optically thin systems. For such systems we suggest a multiple-collision expansion, which we have implemented to low order. Results of these calculations are presented along with the diffusion theory results.

The outline of the remainder of this paper is as follows. In Sec. II we show how the diffusion approximation is obtained from the (linear) transport equation and in particular derive an expression for the diffusion coefficient. The parameters in the diffusion equation are certain scattering kernels and average scattering angles, which were obtained already in Ref. 9. Doppler correction and the effect of molecular rotations were also included in Ref. 9.

In Sec. III, we show how to separate the uncollided flux, which can be computed analytically. Then the diffusion equation is solved for the collided flux, with such cross sections as are available.\(^6\) All calculations are done in slab geometry, which is an excellent model of the experimental setup. For the sake of clarity, the calculations do not take into account the finite lifetime of the muon \(\tau_m = 2 \mu\text{s}\), since it is independent of the scattering process and its effect can be included by multiplying all the results by the factor \(\exp(-t/\tau_m)\).

The diffusion equation has been solved numerically on the Cray XM-P at the Pittsburgh Computer Center. The numerical techniques are standard, and we do not give any details in the paper. In Sec. IV we present results for a number of initial source energies and a number of temperatures for both muonic hydrogen and deuterium atoms diffusing in molecular hydrogen and deuterium, respectively. In Sec. V we present the results of low-order multiple-scattering calculation which can be applied to optically thin media. Our results are seen to compare favorably with the results of a Monte Carlo simulation.\(^7\)

II. DERIVATION OF THE DIFFUSION EQUATION

The muonic atoms can exist in two hyperfine states (singlet and triplet in the case of hydrogen and doublet and quartet in the case of deuterium). We denote the angular flux of muonic atoms as a two component vector

\[
\Phi(t, x, \mu, E) = \begin{bmatrix} \phi_s(t, x, \mu, E) \\ \phi_f(t, x, \mu, E) \end{bmatrix},
\]

where \(t\) is time, \(x\) is the position, \(\mu\) is the cosine of the angle between the velocity and the \(x\) axis, and \(E\) is the energy. We assume the muonic atoms to be diffusing in an infinite slab of thickness \(d\). The notation is standard.\(^8\) \(\Phi\) obeys a linear Boltzmann equation\(^9\)

\[
\frac{1}{v} \frac{\partial \Phi(t, x, \mu, E)}{\partial t} + \mu \frac{\partial \Phi}{\partial x} + \Sigma(E) \Phi = \int \Sigma(E', \mu' \rightarrow E, \mu) \Phi(t, x, \mu', E') d\mu' dE'.
\]

Here \(v\) is the speed of the muonic atom [equal to \((2E/m)^{1/2}\)] and \(\Sigma(E)\) is a diagonal matrix

\[
\Sigma(E) = \begin{bmatrix} \Sigma_s(E) & 0 \\ 0 & \Sigma_f(E) \end{bmatrix},
\]

where \(\Sigma_s(E)\) and \(\Sigma_f(E)\) are the total cross section for

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scattering of singlet (triplet) muonic atoms. The scattering
kernel \( \Sigma(E', \mu' \rightarrow E, \mu) = \Sigma(E' \rightarrow E, \mu_0) \), where \( \mu_0 \) is
the cosine of the scattering angle, can be written as
\[
\Sigma(E', \mu' \rightarrow E, \mu) = \frac{\Sigma_{SS}(E', \mu' \rightarrow E, \mu) \Sigma_{TS}(E', \mu' \rightarrow E, \mu) \Sigma_{TT}(E', \mu' \rightarrow E, \mu)}{\Sigma_{SS}(E', \mu' \rightarrow E, \mu) + \Sigma_{TS}(E', \mu' \rightarrow E, \mu) + \Sigma_{TT}(E', \mu' \rightarrow E, \mu)} .
\]
(4)

Here \( \Sigma_{ab} \) is the scattering cross section for an atom in the
spin state \( a \) to scatter to spin state \( b \). The total cross sections
of Eq. (3) are related to those of Eq. (4) by
\[
\Sigma_a(E') = \int dE' d\mu [\Sigma_{oo}(E', \mu' \rightarrow E, \mu) + \Sigma_{ab}(E', \mu' \rightarrow E, \mu)] .
\]
(5)

To develop a diffusion theory, the standard procedure
is to expand the angular flux in a series of Legendre
polynomials and retain only the first two terms:
\[
\Phi(t, x, \mu, E) = \frac{1}{2} \Phi_0(t, x, E) + \frac{1}{2} \mu \Phi_1(t, x, E) ,
\]
where \( \Phi_0 \) is the total flux (or simply the flux) and \( \Phi_1 \) the
current. We obtain the following equations, known in
neutron transport theory as the P-1 approximation:
\[
\frac{1}{v} \frac{\partial \Phi_0(t, x, E)}{\partial t} + \frac{\partial \Phi_1}{\partial x} + \Sigma(E) \Phi_0 = \int \Sigma_0(E' \rightarrow E) \Phi_0(t, x, E') dE' ,
\]
(7a)
\[
\frac{1}{v} \frac{\partial \Phi_1(t, x, E)}{\partial t} + \frac{1}{3} \frac{\partial \Phi_0}{\partial x} + \Sigma(E) \Phi_1 = \int \Sigma_1(E' \rightarrow E) \Phi_1(t, x, E') dE' .
\]
(7b)

The kernels \( \Sigma_0(E' \rightarrow E) \) and \( \Sigma_1(E' \rightarrow E) \) are the zeroth
and the first moments of the scattering kernel
\( \Sigma(E', \mu' \rightarrow E, \mu) \). \( \Sigma_0(E' \rightarrow E) \) is derived in Ref. 9. In
the spirit of the Selengut-Goertzel approximation we set
\[
\int \Sigma_1(E' \rightarrow E) \Phi_1(E') dE' \approx \left[ \Sigma_{SS}(E) \hat{\mu}_{0SS}(E) \Sigma_{TS}(E) \hat{\mu}_{0TS}(E) \Sigma_{TT}(E) \hat{\mu}_{0TT}(E) \right] \Phi_1(E) ,
\]
(8)
where the average cosine of the scattering angle for
scattering from spin state \( a \) to spin state \( b \), \( \hat{\mu}_{0ab}(E) \), is
also derived in Ref. 9. Also, as is customary in diffusion
theory, the term \( \partial \Phi_1/\partial t \) is neglected.

We thus arrive at the diffusion equation
\[
\frac{1}{v} \frac{\partial \Phi_0}{\partial t} - D \frac{\partial^2 \Phi_0}{\partial x^2} + \Sigma(E) \Phi_0 = \int \Sigma_0(E' \rightarrow E) \Phi_0(t, x, E') dE' .
\]
(9a)
and the diffusion matrix \( D \) is given by
\[
D = \frac{M}{3 \det M} ,
\]
(9b)
\[
M = \begin{bmatrix}
\Sigma_T & -\Sigma_{TT} \hat{\mu}_{0TT} & \Sigma_{TS} \hat{\mu}_{0TS} \\
-\Sigma_{TT} \hat{\mu}_{0TT} & \Sigma_{SS} & \Sigma_{TS} \hat{\mu}_{0TS} \\
\Sigma_{TS} \hat{\mu}_{0TS} & \Sigma_{TS} \hat{\mu}_{0TS} & \Sigma - \Sigma_{SS} \hat{\mu}_{0SS}
\end{bmatrix} .
\]
(9c)

The boundary condition for the angular flux is, of
course, the normal nonreentrant condition at \( x = \pm d/2 \).
For the diffusion approximation, we replace this with
the usual extrapolated-endpoint condition:
\[
\Phi_0(\pm d') = 0 ,
\]
(10a)
where the extrapolated endpoint \( d' \) is defined by
\[
d' = \frac{1}{2} d + 0.7 \frac{1}{\Sigma_a(E)(1 - \hat{\mu}_{0aa})} , \quad a = S, T .
\]
(10b)

We see that there is some ambiguity because the extrapo-
lated endpoint is a function of energy and furthermore it
is different for the singlet and triplet case. We use an
average which should be a good approximation for thick
slabs, where the second term in Eq. (10b) is a small correction.

The x dependence is treated by Fourier expansion
\[
\Phi_0(t, x, E) = \sum_{n=1} \Phi_{0n}(t, E) \cos(\alpha_n x) ,
\]
(11)
where
\[
B_n = \frac{(2n - 1) \pi}{2d'} .
\]
(12)
Equation (9) then yields
\[
\frac{1}{v} \frac{\partial \Phi_{0n}(t, E)}{\partial t} + \left[ D \left(E \right) B_n^2 + \Sigma(E) \right] \Phi_{0n} = \int \Sigma_0(E' \rightarrow E) \Phi_{0n}(t, x, E') dE' .
\]
(13)

III. THE UNCOLLIDED FLUX

The accuracy of the diffusion approximation can be
improved considerably by separating the uncollided flux \( \Phi^u \)
and using the first collisions as the source for the
diffusion equation. Clearly \( \Phi^u \) can be defined analyti-
cally, as follows.

The initial source of muonic atoms is isotropic and
independent of \( x \), so \( \Phi^u \) satisfies
\[
\frac{1}{v_0} \frac{\partial \Phi^u(t, x, \mu)}{\partial t} + \mu \frac{\partial \Phi^u}{\partial x} + \Sigma(E_0) \Phi^u = S_0 \delta(t) \delta(E - E_0) ,
\]
(14)
where \( S_0 \) is the source strength (atoms/cm). The Laplace
transform of Eq. (14) gives
\[
\mathcal{L} \Phi^u(p, x, \mu) + \mu \mathcal{L} \Phi^u + \mathcal{L} \Sigma \Phi^u = S_0 \delta(E - E_0) .
\]
(15)

Here \( p \) is the transform variable and \( \mathcal{L} \Phi^u(p, x, \mu) \) the
transformed flux. Since we are only interested in the
solution inside the slab (if \( |x| < d/2 \)), we may consider \( \Sigma \)
and \( S_0 \) as constants, independent of \( x \). The equations for
singlet and triplet are decoupled, so it suffices to solve for
one species:
we can write it as a sum of the uncollided part \( X_u \) and the collided part \( X_c \), where

\[
X_u(t) = 2\phi_u(t, d/2)
\]

\[
= S_0 v_0 \exp[-\Sigma_a(E_0) v_0 t] \begin{cases} 1, & v_0 t < d \\ \left( \frac{d}{v_0 t} \right)^2, & v_0 t > d \end{cases}
\]

Integration over all times and setting \( S_0 = (2d)^{-1} \) (corresponding to a single atom inside the slab at \( t = 0 \)) yields the ratio of atoms that eventually escape from the slab without suffering a single collision (escape probability\(^{13}\))

\[
U(\delta_a) = \frac{1}{2} \left\{ \frac{1}{\delta_a} (1 - e^{-\delta_a}) + E_2(\delta_a) \right\},
\]

where \( \delta_a = \Sigma_a d \) is the optical width for the atoms in state \( a \) and \( E_2(x) \) is the exponential integral

\[
E_2(x) = \int_1^\infty e^{-xt} t^{-2} dt.
\]

This quantity can give insight into the applicability of the diffusion approximation and/or multiple collision expansion.

We will also need the \( n \)th Fourier component of the total uncollided flux

\[
output_\nu(t) = \frac{8 S_0 d}{(2n - 1)^2 \pi^2} \exp[-v_0 \Sigma_a(E_0) t] \times \begin{cases} 1 + \frac{\sin(2n - 1)\pi \tau}{(2n - 1)\pi \tau}, & \tau - 1 \\ \frac{1}{\tau} + \frac{\sin(2n - 1)\pi \tau}{2n - 1\pi \tau} H(\tau - 1) \end{cases}
\]

where

\[
\tau = \frac{v_0 t}{d}.
\]

**IV. RESULTS**

As we mentioned in Sec. III, the accuracy of the diffusion approximation can be improved by separating the uncollided flux. The \( n \)th Fourier component of the collided flux \( \Phi_c^\nu(t, E) \) then obeys the zero initial condition and satisfies Eq. (13) with the right-hand side supplemented by the source:

\[
S_n(t) = \begin{bmatrix} \Sigma_{SS}(E_0) & \Sigma_{TS}(E_0) \\ \Sigma_{ST}(E_0) & \Sigma_{TT}(E_0) \end{bmatrix} \Phi_c^\nu(t),
\]

where \( \Phi_c^\nu \) was defined by Eq. (23).

The modified Eq. (13) can be solved numerically. We used the multigroup method to discretize the energy variable. The fluxes, the cross sections, and the diffusion matrix were taken to be independent of energy in each group. The trapezoidal formula was used for numerical
integration. By approximating the time derivative by the forward difference we obtained an explicit numerical scheme, where the knowledge of $\Phi(t)$ suffices for the calculation of $\Phi(t + \Delta t)$. The first three terms in the Fourier expansion were retained and 20 energy groups were used.

We present the results of several calculations for the case of deuterium (Figs. 1–3) and hydrogen (Fig. 4). The atomic cross sections (Ref. 10 for hydrogen and Ref. 11 for deuterium) were used and converted to molecular cross sections, as explained in Ref. 9. In the case of hydrogen, the effect of rotations of the scatterers was taken into account by replacing the hyperfine splitting constant ($Q = 0.183$ eV) with the effective inelastic energy transfer $Q_{\text{eff}}(E)$, as defined in Ref. 9. For deuterium, however, the hyperfine splitting is small ($Q = 0.049$), so this correction was ignored. Only the emerging flux $X'(t) = X_s + X_T$ is plotted, since present experiments cannot distinguish between the atoms in the two hyperfine states.

Figure 1 shows the effect of varying the initial energy $E_0$ ($E_0 = 1$, 0.5, and 0.1 eV) in the case of deuterium diffusing in a slab of width $d = 1$ cm, at $p = 1$ bar and $T = 300$ K. Diffusion at two different temperatures $T = 300$ K and $T = 200$ K is shown in Fig. 2 for $d = 1$ cm, $p = 1$ bar, $E_0 = 1$ eV. Finally, pressure has been varied on Fig. 3 for $d = 1$ cm, $T = 300$ K, $E_0 = 1$ eV.

Diffusion in hydrogen is more interesting because the cross sections vary as a function of energy over two orders of magnitude. The initial statistical mixture (75%
triplet, 25% singlet) changes into predominantly singlet during the first few average collision times. Atoms in the singlet state have a smaller cross section and hence diffuse faster. This results in a bump on the emerging flux curve (the case \( d = 5 \text{ cm}, p = 1 \text{ bar}, T = 300 \text{ K}, E_0 = 1 \text{ eV} \) is shown in Fig. 4). The prediction of the bump, though surprising when it was first obtained, was confirmed by a very simple model, which is presented in the Appendix.

V. MULTIPLE-SCATTERING EXPANSION

In optically thin media, the diffusion approximation is not valid. Most atoms escape from the medium without suffering a single collision. In this case it is convenient to use the multiple collision expansion for the angular flux \( \Phi \)

\[
\Phi = \Phi^* + \sum_{n=1}^{\infty} \Phi^{(n)},
\]

where \( \Phi^{(n)} \) is the \( n \) times collided flux.

Here we present a low-order implementation of Eq. (25), in which we retain only \( \Phi^* \) and \( \Phi^{(1)} \), and neglect the rest. \( \Phi^* \) is defined analytically by Eq. (17). Clearly, the source for \( \Phi^{(1)} \) are first collisions. We take this source to be isotropic, which is a good approximation. Furthermore, we assume that all atoms contributing to \( \Phi^{(1)} \) travel with the average speed \( v_1 \), i.e., we model the continuous speed distribution of \( \Phi^{(1)} \) by \( \delta(v-v_1) \). In the case of elastic scattering,

\[
v_1 = \frac{3A^2 + 1}{3A(A+1)}v_0,
\]

where \( A \) is the ratio of the masses of the target molecule and the muonic atom.

Then \( \Phi^{(1)} \) obeys the equation

\[
\frac{1}{v_1} \frac{\partial \Phi^{(1)}(t,x,\mu)}{\partial t} + \mu \frac{\partial \Phi^{(1)}}{\partial x} + \Sigma(E_1) \Phi^{(1)} = S^{(1)}(t,x),
\]

where \( E_1 = mv_1^2/2 \) and the source \( S^{(1)} \) equals

\[
S^{(1)}(t,x) = \frac{1}{2} \left[ \Sigma_{SS}(E_0) \Sigma_{TS}(E_0) \right] \Phi^0(t,x).
\]

Equation (27) can be readily solved by integration along the characteristics\(^1\) and yields

\[
\Phi^{(1)}(t,x,\mu) = \int_0^t \exp \left[ -\Sigma_a(E_1) \tau \right] \times \sigma_a[\tau,x-\mu v_1(t-\tau)] d\tau.
\]

The once collided emerging flux \( X^{(1)}(t) \),

\[
X^{(1)}(t) = 2 \int_0^1 \mu \Phi^{(1)}(t, d/2, \mu) d\mu,
\]

can be found by numerical quadrature. Trapezoidal rule was used for numerical evaluation of formulas (29) and (30).

We employed the above calculation in the case of deuterium muonic atoms in a slab of width \( d = 0.23 \text{ cm} \), filled with gaseous deuterium at pressure \( p = 0.188 \text{ bar} \) and \( T = 300 \text{ K} \), which corresponds to optical thickness \( \delta = 0.52 \) (Fig. 5). These parameters were used in an experiment done by Siegel.\(^12\) From Eq. (21) we see that 55% of the atoms contribute to the uncollided emerging flux, while numerical integration shows that 26% contribute to the first collided emerging flux. Thus we are ignoring 19% of the atoms, namely, those that suffer more than one collision on their way out of the slab.

Unfortunately, the analysis of the experiments has not been completed yet, so we can only compare our calculation with a Monte Carlo simulation.\(^12\) Although the agreement is apparently good, the two should not be compared directly, because in the experiment the source of muonic atoms is not a \( \delta \) function in energy, but rather a continuous distribution. In the Monte Carlo simulation this distribution is taken to be a Maxwellian. The calculation gives Green's function of the problem, while the experiment yields its convolution with the initial source. We hope that comparison of our calculation with the experiment will yield some information about the energy distribution of the source. We plan to improve the calculation by taking into account more terms in the multiple-collision expansion.

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DIFFUSION OF MUONIC ATOMS

APPENDIX

Here we present the analytically solvable one-speed model, in which all the atoms are assumed to be moving with the same speed \( v \). Simple as it is, the model reasonably describes the diffusion process; in particular, it predicts the bump on the emerging flux curve in the case of hydrogen.

Under the one-speed assumption and neglecting the off-diagonal elements in the diffusion matrix \( D \), the diffusion equation (13) yields

\[
\frac{\partial \Phi_{0n}(t)}{\partial t} = v \left[ -\Sigma_{ST} - B_0^2 D_{SS} \begin{bmatrix} \Sigma_{TS} & -\Sigma_{TS} \end{bmatrix} \right] \Phi_{0n}(t)
\]

where the matrix is constant. The main contribution comes from the first Fourier mode, so we set \( n = 1 \). To simplify the notation we rewrite Eq. (31) as

\[
\frac{\partial \Psi(t)}{\partial t} = \begin{bmatrix} -\sigma_s - \lambda_s & \sigma_T \\ \sigma_s & -\sigma_T - \lambda_T \end{bmatrix} \Psi(t) .
\]

(32)

In the case of hydrogen, the initial condition is

\[
\Psi(t=0) = \frac{1}{4} \begin{bmatrix} 1 \\ 3 \end{bmatrix} .
\]

(33)

The eigenvalues and eigenvectors are

\[
\omega_{\pm} = \frac{1}{2} \left[ -\lambda_s - \lambda_T - \sigma_s - \sigma_T \pm \sqrt{\left( -\lambda_s - \lambda_T + \sigma_s + \sigma_T \right)^2 + 4\sigma_s \sigma_T} \right]^{1/2}
\]

and

\[
v_{\pm} = \begin{bmatrix} 1 \\ u_{\pm} \end{bmatrix}, \quad u_{\pm} = \frac{1}{\sigma_T} (\omega_{\pm} + \lambda_s + \sigma_s)
\]

(34a)

(34b)

and the solution of Eq. (33) is a sum of two exponentials

\[
\Psi(t) = C_+ v_+ \exp(\omega_+ t) + C_- v_- \exp(\omega_- t) ,
\]

(35a)

with

\[
C_+ = \frac{3 - u_+}{4(u_+ - u_-)} , \quad C_- = \frac{1}{4} - C_+ .
\]

(35b)

In the thermal region \( \sigma_S = 0 \) and the solution (35) simplifies into

\[
\Psi(t) = \frac{3\sigma_T}{4} \left( e^{-\lambda_T t} + \frac{3\sigma_T}{4(\lambda_T - \sigma_T - \lambda_s)} e^{-\lambda_s t} \right)
\]

(36)

We are interested in seeing when the emerging flux

\[
X = \lambda_s \Psi + \lambda_T \Psi_T ,
\]

exhibits a bump. Since \( \lambda_T \) is much smaller than \( \lambda_s \), we ignore it. Then \( X \) has a maximum at

\[
t = \frac{1}{\sigma_T - \lambda_s} \ln \left( \frac{3\lambda_T^2}{\lambda_s (4\sigma_T - \lambda_s)} \right)
\]

(37)

subject to the condition

\[
3\sigma_T > \lambda_s ,
\]

(38)

which is the requirement of an optically thick slab, needed for the validity of the diffusion approximation in the first place.

The one-speed model of course cannot predict the initial decay in the emerging flux, which is due to slowing down.

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