Anisotropic Diffusion
in Electron Swarms

J. L. A. Francey and D. A. Jones

Department of Physics, Monash University, Clayton, Vic. 3168.

Abstract

We show that the distribution functions derived by Parker (1963) in his analysis of the Townsend–Huxley experiment can be used to calculate D_L/D, the ratio of longitudinal to isotropic diffusion coefficients for electron swarms in electrostatic fields in gases. In the case of a constant collision frequency interaction our results agree with previous calculations, whilst for a constant mean free path we find $D_L/D = 0.58$. This result is some 16% higher than previously published values but provides better agreement with experiment for electrons in helium.

1. Introduction

The anisotropic nature of the diffusion coefficient for electrons in electrostatic fields in gases has been known since the work of Wagner *et al.* (1967). The cause of this anisotropy is the position dependence of the electron energy distribution function. In a uniform electron gas this function is independent of position and is given by the normal Davydov expression, whilst in a nonuniform gas the presence of spatial gradients leads to the energy distribution function becoming position dependent, and it is this effect which causes the anisotropy in the diffusion coefficients.

Since the work of Wagner *et al.* (1967) several authors (Parker and Lowke 1969; Skullerud 1969; Huxley 1972) have provided successful descriptions of this effect by allowing for the position dependence of the energy distribution function in their solutions of the Boltzmann equation. Skullerud followed a perturbation procedure originally suggested by Wannier (1953) in some work on ion motion in strong electric fields. By making an assumption regarding the form of the number density he obtained an equation describing the corrections to the distribution function caused by the spatial gradients. Parker and Lowke considered the case of an electron pulse drifting along the field direction and after finding corrections to the pulse shape caused by the spatial gradient terms they then determined the diffusion coefficient from the mean square random displacement of the electrons along the field direction. Huxley assumed that the perturbation on the distribution function could be expanded in the complete set consisting of the number density and all its derivatives, the expansion coefficients being functions of energy which were found by equating to zero successive terms in the Boltzmann equation.

The three methods outlined above are similar in that they each include the effect of spatial gradients as a perturbation on a uniform number density. Indeed, to first order in the correction terms we find that they are identical, the distribution function

in each case being written in the form

$$f_0(v, \mathbf{r}) = n(\mathbf{r}) f_0^*(v) + g(v) \, \partial n(\mathbf{r})/\partial z + \text{h.t.}, \tag{1}$$

h.t. indicating the presence of higher order terms. Here the two-term approximation to the distribution function $f(v, \mathbf{r})$ has been used (Francey and Stewart 1972), $f_0(v, \mathbf{r})$ is the isotropic (in velocity space) part, $f_0^*(v)$ is the Davydov distribution (applicable only in the case of vanishingly small spatial gradients), $n(\mathbf{r})$ is the number density, z is the spatial coordinate in the field direction and $g(v)$ is a function of velocity which is found by requiring that equation (1) satisfy the full Boltzmann equation.

The presence of the additional term in $\partial n/\partial z$ in equation (1) makes the identification of an anisotropic diffusion coefficient very simple. If we substitute equation (1) into the expression for the longitudinal flux Γ_L given by

$$\Gamma_L = -\tfrac{4}{3}\pi \int_0^\infty \frac{v^4}{v_c}\left(\frac{\partial f_0}{\partial z} + \frac{eE}{mv}\frac{\partial f_0}{\partial v}\right) \mathrm{d}v, \tag{2}$$

in which v_c is the collision frequency for momentum transfer, E the strength of the electrostatic field and e and m the charge and mass of an electron, there results

$$\Gamma_L = wn - \left(D + \tfrac{4}{3}\pi \int_0^\infty \frac{v^4 eE}{v_c \, mv}\frac{\partial g}{\partial v}\,\mathrm{d}v\right)\frac{\partial n}{\partial z} + \text{h.t.}, \tag{3}$$

where w and D are the normal (isotropic) expressions for the drift velocity and diffusion coefficient. This gives

$$D_L = D - \tfrac{4}{3}\pi \int_0^\infty g(v)\frac{\mathrm{d}}{\mathrm{d}v}\left(\frac{av^3}{v_c}\right)\mathrm{d}v, \tag{4}$$

where D_L is the coefficient for diffusion along the field direction and a is the acceleration of the electron in the field E. Equation (4) is the expression for D_L found by Parker and Lowke (1969), Skullerud (1969) and Huxley (1972).

Since we must have the number density given by

$$n(\mathbf{r}) = \int_0^\infty 4\pi v^2 f_0(v, \mathbf{r})\,\mathrm{d}v \tag{5}$$

and $f_0^*(v)$ is normalized so that

$$\int_0^\infty 4\pi v^2 f_0^*(v)\,\mathrm{d}v = 1, \tag{6}$$

we see that $g(v)$ must satisfy

$$\int_0^\infty 4\pi v^2 g(v)\,\mathrm{d}v = 0. \tag{7}$$

Thus, in the case of a velocity-independent collision frequency, equation (4) implies that $D_L = D$, that is, diffusion is isotropic.

It would seem that a more accurate approach to this problem would be to solve the Boltzmann equation without making the assumption that the spatial gradients

are small. This has already been done by Parker (1963) in his analysis of the Townsend–Huxley experiment. Briefly, his method was to first find a suitable change of variables which allowed the Boltzmann equation to be separated into a set of ordinary eigenvalue equations. A Green's function could then be found by expansion in terms of these eigenfunctions and the solution to the inhomogeneous Boltzmann equation finally obtained by integration over the product of the Green's function and the source term in the usual manner. Parker found that the inclusion of spatial gradients in this way did lead to a position-dependent energy distribution function, but since this work predated the discovery of anisotropic diffusion he merely concluded that the presence of spatial gradients could lead to errors of the order of 20% in the usual interpretation of the Townsend–Huxley experiment.

The purpose of this paper is to show that the distribution functions derived by Parker (1963) can be used to obtain anisotropic diffusion coefficients which are not too different from those obtained by the previous authors. In the next section we briefly review Parker's solution of the Boltzmann equation for the case of a constant electron mean free path and then describe two equivalent methods for calculating the longitudinal diffusion coefficient. The case of a constant collision frequency is described in Section 3, and in Section 4 we compare these results with previously published values.

2. Constant Cross Section

The appropriate form of the Boltzmann equation in the large E/P limit is

$$
\frac{1}{\varepsilon}\frac{\partial}{\partial\varepsilon}\left[\varepsilon\left\{\varepsilon f_0 + \frac{M(eE\lambda)^2}{6m}\left(\frac{\partial f_0}{\partial\varepsilon} + \frac{1}{eE}\frac{\partial f_0}{\partial z}\right)\right\}\right]
$$
$$
+ \frac{MeE\lambda^2}{6m}\frac{\partial^2 f_0}{\partial\varepsilon\,\partial z} + \frac{M\lambda^2}{6m}\nabla^2 f_0 = -\frac{M\lambda S}{2(2m\varepsilon)^{\frac{1}{2}}}. \tag{8a}
$$

Here ε is the electron energy, λ the electron mean free path, M the mass of a gas molecule and P the gas pressure. This equation describes a steady electron stream emitted from a point source at the origin. The system has cylindrical symmetry and is described by the cylindrical coordinates ρ and z. The electrostatic field E points in the direction of the negative z axis. The quantity S is the source term which emits one electron per second with energy ε_0 at the point $\rho = z = 0$ and is given by

$$
S(\varepsilon, z, \rho) = \delta(z)\frac{\delta(\rho)}{2\pi\rho}\frac{\delta(\varepsilon - \varepsilon_0)}{(4\pi/m)(2\varepsilon/m)^{\frac{1}{2}}}.
$$

Defining dimensionless variables y, ξ and η by

$$
y = (A\varepsilon)^2, \qquad \xi = 2A(\varepsilon - eEz), \qquad \eta = 2eEA\rho,
$$

where

$$
A = (eE\lambda)^{-1}(3m/M)^{\frac{1}{2}},
$$

equation (8a) becomes

$$
\frac{\partial}{\partial y}\left\{y\left(f_0 + \frac{\partial f_0}{\partial y}\right)\right\} + y^{\frac{1}{2}}\frac{\partial f_0}{\partial\xi} + \eta^{-1}\frac{\partial}{\partial y}\left(\eta\frac{\partial f_0}{\partial y}\right) = -\left(\frac{A}{2m}\right)^{\frac{1}{2}}\frac{M\lambda S}{4y^{\frac{1}{2}}}. \tag{8b}
$$

If we assume a product solution of the homogeneous equation in the form

$$f_0(y, \xi, \eta) = F(y)\, A(\xi)\, B(\eta),$$

we then obtain the two eigenvalue equations

$$\mathrm{d}^2 B/\mathrm{d}\eta^2 + \eta^{-1}\, \mathrm{d}B/\mathrm{d}\eta + K^2 B = 0 \qquad (9a)$$

and

$$y\, \mathrm{d}^2 F/\mathrm{d}y^2 + (1+y)\, \mathrm{d}F/\mathrm{d}y + (1 - K^2 + y^{\frac{1}{2}}\phi)F = 0, \qquad (9b)$$

where K and ϕ are separation constants. The solution to equation (9a) is simply the zero-order Bessel function $J_0(K\eta)$ with eigenvalue K. The solution to equation (9b) does not have a simple form, but the requirement that F become zero as y tends to infinity will provide a set of eigenvalues ϕ_l and eigenfunctions F_l characterized by the label l.

A Green's function for equation (8b) can be found by expansion in the eigenfunctions F_l and has the following form,

$$G(y, \xi, \eta \mid \bar{y}, \bar{\xi}, \bar{\eta}) = \sum_l \sum_K J_0(K\bar{\eta})\, \bar{\eta}\, F_l(K^2, \bar{y})\, \exp\bar{y}$$

$$\times J_0(K\eta)\, F_l(K^2, y)\, \exp\{-(\xi - \bar{\xi})\phi_l\},$$

so that f_0 is given by

$$f_0(y, \xi, \eta) = \int G(y, \xi, \eta \mid \bar{y}, \bar{\xi}, \bar{\eta}) \left\{ \left(\frac{A}{2m} \right)^{\frac{1}{2}} \frac{M\lambda\, S(\bar{y}, \bar{\xi}, \bar{\eta})}{4\bar{y}^{\frac{1}{2}}} \right\} \mathrm{d}\bar{y}\, \mathrm{d}\bar{\xi}\, \mathrm{d}\bar{\eta}$$

$$= \frac{CeEA}{\pi D} \int_0^\infty \mathrm{d}K\; K\, J_0(K\eta) \exp(y_0)\, F_0(K^2, y_0)\, F_0(K^2, y)\, \exp\{-\phi_0(2y_0^{\frac{1}{2}} - \xi)\}. \qquad (10)$$

In this last step the sum over K has been replaced by an integral, since the system is unbounded in the radial direction, and the sum over l has been replaced by just the first term, corresponding to $l = 0$, as the higher order terms decay exponentially with distance from the source. The constant C is the usual normalization constant for the Druyvestyn energy function, $\exp\{-(A\varepsilon)^2\}$, and D is the corresponding diffusion coefficient.

To proceed further we need to know the dependence of F_0 and ϕ_0 on K^2. Parker (1963) makes the following assumptions,

$$F_0(K^2, y) = F_0^{(0)} + K^2 F_0^{(1)} + K^4 F_0^{(2)} + \dots,$$

$$\phi_0(K^2) = \phi_0^{(0)} + K^2 \phi_0^{(1)} + K^4 \phi_0^{(2)} + \dots.$$

Substituting these expansions into equation (10), carrying out the integration and then reverting back to the original variables, we obtain the final expression for f_0 found by Parker, namely

$$f_0 = \frac{C \exp\{-(A\varepsilon)^2\}}{4\pi Dz} \left(1 - \frac{\varepsilon}{eEz}\right)^{-1} \exp\left\{-\frac{w\rho^2}{4Dz}\left(1 - \frac{\varepsilon}{eEz}\right)^{-1}\right\}$$

$$\times \left(1 + \frac{g_1(y)\, L_1(\eta^2/4\theta)}{\theta\, \phi_0^{(1)}} + \frac{g_2(y)\, L_2(\eta^2/4\theta)}{(\theta\, \phi_0^{(1)})^2} + \dots\right), \qquad (11)$$

where $\theta = 2eEAz$, the L_n are the Laguerre polynomials and the g functions are given by

$$g_1 = \frac{F_0^{(1)}(y_0)}{F_0^{(0)}(y_0)} + \frac{F_0^{(1)}(y)}{F_0^{(0)}(y)},$$

$$g_2 = \frac{F_0^{(2)}(y)}{F_0^{(0)}(y)} + \frac{F_0^{(2)}(y_0)}{F_0^{(0)}(y_0)} + \frac{F_0^{(1)}(y) F_0^{(1)}(y_0)}{F_0^{(0)}(y) F_0^{(0)}(y_0)} - (2y_0^{\frac{1}{2}} - \xi)\phi_0^{(2)},$$

and corresponding higher order expressions. The result (11) should be compared with the function found when the assumption of vanishingly small spatial gradients is made at the start of the solution, namely

$$f_0 = (C/4\pi Dz)\exp\{-(A\varepsilon)^2\}\exp(-w\rho^2/4Dz).$$

We can derive an anisotropic diffusion coefficient from equation (11) in either of two ways. A straightforward method is to calculate the longitudinal flux and number density and then require that

$$\Gamma_L = \bar{w}n - D_L \,\partial n/\partial z, \tag{12}$$

where $\bar{w}$ is the drift velocity in the presence of large spatial gradients. If we expand to order z^{-1} in the correction terms in equation (11), and write

$$n^* = (4\pi Dz)^{-1}\exp(-w\rho^2/4Dz),$$

where n^* is the number density in the case of vanishingly small spatial gradients, then we obtain

$$f_0 = C\exp(-y)n^*\{1 + S(y)(eEAz)^{-1}\},$$

where

$$S(y) = y^{\frac{1}{2}} + \tfrac{1}{4}\pi^{\frac{1}{2}}g_1(y).$$

Use of equations (2) and (5) gives

$$\Gamma_L = wn^*\left\{1 + \frac{D}{w}\left(\frac{1+S_1}{z}\right)\right\} \quad \text{with} \quad S_1 = \int_0^\infty e^{-y}y^{-\frac{1}{2}}S(y)\,dy, \tag{13a}$$

$$n = n^*\left(1 + \frac{S_2}{eEAz\,\Gamma(\frac{3}{4})}\right) \quad \text{with} \quad S_2 = \int_0^\infty e^{-y}y^{-\frac{1}{4}}S(y)\,dy, \tag{13b}$$

and then substitution into equation (12) leads to

$$w + D\left(\frac{1+S_1}{z}\right) = \bar{w} + \frac{\bar{w}D\pi^{\frac{1}{2}}S_2}{w\,\Gamma(\frac{3}{4})z} + \frac{D_L}{z}.$$

Equating terms independent of z here gives $\bar{w} = w$, that is, the drift velocity is not affected by the spatial gradients; while equating terms in z^{-1} gives

$$D_L/D = 1 + S_1 - \pi^{\frac{1}{2}}S_2/\Gamma(\tfrac{3}{4}). \tag{14}$$

Now Parker (1963) has solved the energy eigenvalue equation by a perturbation

calculation using Laguerre functions. This gives the g_1 function the form

$$g_1(y) = -\sum_{n=0}^{\infty} a_n L_n(y).$$

When this expression is used to evaluate the constants S_1 and S_2, the formula for D_L becomes

$$\frac{D_L}{D} = 2 - \frac{\pi^{\frac{1}{2}} \Gamma(\frac{5}{4})}{\Gamma(\frac{3}{4})} - \frac{1}{4}\pi \sum_{n=0}^{\infty} \frac{a_n}{n!} \left(\frac{\Gamma(n+\frac{1}{2})}{\Gamma(\frac{1}{2})} - \frac{\Gamma(n+\frac{1}{4})}{\Gamma(\frac{1}{4})} \right).$$

Using the a_n found in Parker's paper, this leads to

$$D_L/D = 0\cdot584.$$

Since the currently accepted value of the ratio D_L/D is $0\cdot495$, we checked this value by finding a power series solution of the energy eigenvalue equation. In terms of the variable $x = y^2$, equation (9b) is

$$x\, d^2F/dx^2 + (1-2x^2)\,dF/dx + (4\phi x^2 - 4xK^2)F = 0.$$

An acceptable solution to this equation has the form

$$F(x) = 1 + K^2 \sum_{n=2}^{\infty} a_n x^n + O(K^4),$$

where

$$a_n = \frac{2^{n/2}}{n} \frac{2}{n!!} \qquad \text{for } n \text{ even and } \geqslant 2,$$

$$= -\frac{2^{(n+1)/2}}{n\,\pi^{\frac{1}{2}}} \frac{2}{n!!} \qquad \text{for } n \text{ odd and } \geqslant 3.$$

This gives

$$\frac{D_L}{D} = 2 - \frac{\pi^{\frac{1}{2}} \Gamma(\frac{5}{4})}{\Gamma(\frac{3}{4})} - \frac{1}{4}\pi \sum_{n=2}^{\infty} a_n \left(\frac{\Gamma(\frac{1}{2}n+\frac{3}{4})}{\Gamma(\frac{3}{4})} - \frac{\Gamma(\frac{1}{2}n+\frac{1}{2})}{\Gamma(\frac{1}{2})} \right) = 0\cdot577.$$

To point out the similarity here with the work of Parker and Lowke (1969), Skullerud (1969) and Huxley (1972), we re-derive equation (14) by another method. If we keep terms of order z^{-1} and $(\rho/z)^2$ in equation (11), we can write the distribution in the form

$$f_0 = f_0^* n^* \left[1 + S(y) \left\{ \frac{1}{eEAz} - \left(\frac{\rho}{z}\right)^2 \right\} \right], \qquad (15)$$

where

$$f_0^* = C\exp(-y).$$

From this we find

$$n = n^* \left[1 + \frac{S_2}{\Gamma(\frac{3}{4})} \left\{ \frac{1}{eEAz} - \frac{\pi^{\frac{1}{2}}}{4}\left(\frac{\rho}{z}\right)^2 \right\} \right], \qquad \frac{\partial n}{\partial z} = n^* \left\{ \frac{w}{4D}\left(\frac{\rho}{z}\right)^2 - \frac{1}{z} \right\},$$

where S_2 is given by the definition (13b) as before. If we now arbitrarily add and

substract

$$\frac{S_2}{\Gamma(\frac{3}{4})}\left\{\frac{1}{eEAz} - \frac{\pi^{\frac{1}{2}}}{4}\left(\frac{\rho}{z}\right)^2\right\}$$

in equation (15), f_0 becomes

$$f_0 = f_0^* n + f_0^* \frac{\partial n}{\partial z} \frac{1}{eEA}\left(\frac{S_2}{\Gamma(\frac{3}{4})} - S(y)\right),$$

which has the form of equation (1) with

$$g(y) = \frac{1}{eEA}\left(\frac{S_2}{\Gamma(\frac{3}{4})} - S(y)\right)f_0^*(y). \tag{16}$$

When this expression for $g(y)$ is substituted into equation (4) we again obtain the result (14) for the ratio D_L/D.

3. Constant Collision Frequency

In the case of a constant collision frequency, by a similar procedure to the above Parker (1963) obtained

$$f_0 = \frac{C\exp(-B\varepsilon)}{4\pi Dz}\left(1 - \frac{\varepsilon}{eEz}\right)^{-1}\exp\left\{-\frac{w\rho^2}{4Dz}\left(1 - \frac{\varepsilon}{eEz}\right)^{-1}\right\}, \tag{17}$$

where C is now the normalization constant for the energy function $\exp(-B\varepsilon)$, with $B = 3M^{-1}(Mv_0/eE)^2$, and w and D are the drift velocity and diffusion coefficient appropriate to a constant collision frequency gas. Equation (17) has the advantage of being exact, no expansion in the parameter K^2 being required in this case. Again keeping only terms of order z^{-1} and $(\rho/z)^2$, we find that (17) can be written

$$f_0 = f_0^* n^*\left[1 + \frac{x}{eEB}\left\{\frac{w}{4D}\left(\frac{\rho}{z}\right)^2 - \frac{1}{z}\right\}\right], \tag{18}$$

with

$$x = B\varepsilon \quad \text{and} \quad f_0^* = C\exp(-B\varepsilon).$$

The form (18) gives

$$n = n^*\left[1 + \frac{3}{2eEB}\left\{\frac{w}{4D}\left(\frac{\rho}{z}\right)^2 - \frac{1}{z}\right\}\right], \qquad \frac{\partial n}{\partial z} = n^*\left\{\frac{w}{4D}\left(\frac{\rho}{z}\right)^2 - \frac{1}{z}\right\}.$$

Addition and substraction of

$$\frac{3}{2eEB}\left\{\frac{w}{4D}\left(\frac{\rho}{z}\right)^2 - \frac{1}{z}\right\}$$

in equation (18) then leads to

$$f_0 = f_0^* n + f_0^*(\partial n/\partial z)(eEB)^{-1}(\tfrac{3}{2} - x),$$

which again is of the form of equation (1) with

$$g = (eEB)^{-1}(\tfrac{3}{2} - x)f_0^*. \tag{19}$$

Substituting the expression (19) into equation (4) we find

$$D_L/D = 1 - 2\pi^{-\frac{1}{2}} \int_0^\infty (\tfrac{3}{2} - x) e^{-x} x^{\frac{1}{2}}\, dx = 1,$$

that is, diffusion is isotropic for a constant collision frequency gas. This is the result found by each of the previously mentioned authors.

4. Discussion

Whilst our value for the ratio D_L/D in the case of a constant cross section (Section 3) is some 16% higher than the currently accepted value (see also Robson 1972; McIntosh 1974; Skullerud 1974), it does have the advantage of providing better agreement with experiment. Lowke and Parker (1969) have used their results to calculate D_L/μ values for several real gases (μ is the electron mobility). For helium, which is a very good approximation to a constant mean free path and for which no inelastic collisions have to be taken into account, they found that their value of 0.495 successfully explained the major part of the difference between D_L and D, but that their value was still too low by the order of 15%. This point was noted by Elford (1974), who measured D_L/μ in helium with a much better accuracy than the original experiments of Wagner et al. (1967), but unfortunately he was not able to go to high enough values of E/P where the difference becomes pronounced.
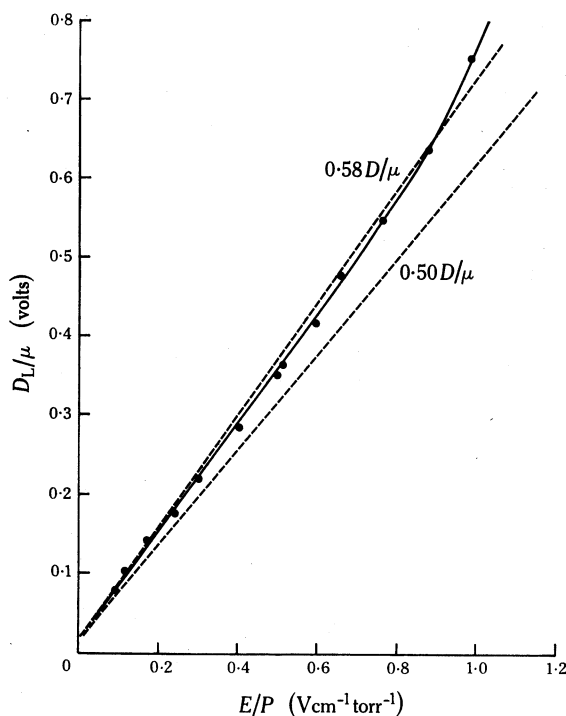

Fig. 1. Comparison of the D_L/μ values of Wagner et al. (1967), as a function of E/P for electrons in helium (solid curve), with the relations $0.50\,D/\mu$ and $0.58\,D/\mu$ (dashed curves) which represent Parker and Lowke's (1969) result and the present work respectively. The D/μ values are those of Crompton et al. (1967).

In Fig. 1 we have plotted, as a function of E/P, the D_L/μ values of Wagner et al. (1967) for electrons in helium and have compared these with the curves for $0.50\,D/\mu$ and $0.58\,D/\mu$, where the D/μ values are those of Crompton et al. (1967). Our ratio gives a better agreement with experiment, although the fit is still not perfect.

The discrepancy between our results and those of Parker and Lowke (1969), Skullerud (1969) and Huxley (1972) is still a little disconcerting, however, despite the better agreement with experiment. Recently Skullerud (1974) has used a generalized Hermite-polynomial expansion of f_0 in the spatial coordinates and claims that this method should be accurate even in the case of large spatial gradients. The value he finds for D_L/D is $0 \cdot 491$. It could be thought that we have not included enough terms in the distribution function. To check this we extended Parker's (1963) analysis to include the term $g_4 L_4/(\theta \phi_0^{(1)})^4$ in equation (11), but found that the additional terms could not be used to change our results; instead they seemed to belong with the higher order derivatives of n. A possible check on this work is to extend it to include a general variation of cross section with energy, and also to include inelastic effects. Such work is proceeding.

Acknowledgments

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