THE PRESENT STATUS OF THE TOWNSEND-HUXLEY EXPERIMENT IN THEORY AND PRACTICE

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Abstract

Many theoretical investigations have been made of the Townsend-Huxley lateral diffusion experiment but only the most recent have included the spatial variation of the energy distribution function and anisotropic diffusion. Nevertheless, almost without exception, self-consistent and apparently accurate experimental results have been obtained using formulae resulting from these earlier analyses even though, as is now known, these analyses were based on inadequate theoretical foundations. This paradoxical situation has been further confused by the fact that some of the analyses have been based on apparently unrealistic boundary conditions while others have contained an error or a fallacy. A review of the development of the theory up to the present time is given here with the aim of clarifying the position and demonstrating the validity of the data published from a number of laboratories.

I. INTRODUCTION

A long-standing problem in the interpretation of the results of the Townsend– Huxley diffusion experiment has been the success of a semi-empirical formula for which no completely satisfactory theoretical basis has been found (Huxley and Crompton 1955; Hurst and Liley 1965). In a recent paper Lowke (1971) has made a significant contribution towards the solution of this problem but, notwithstanding, some confusion is still likely to remain concerning the present status of the theory and the application of earlier theories. In particular there may be reservations about the results that were derived previously either on the basis of the semi-empirical formula or by using other formulae that were apparently successful.

Prior to Lowke's (1971) paper, analyses of the Townsend-Huxley experiment were, almost without exception, based on the assumption of a spatially independent energy distribution function and, as a consequence, it was assumed that the diffusion of the electrons was isotropic. This fundamental assumption has now been shown to be inadequate. Where more complete analyses including spatial dependence were undertaken (Parker 1963; Francey 1969*a*, 1969*b*; Desloge and Mitchell 1970) the simplifications demanded by the complexity of the problem considerably reduced the applicability of the results of the analyses (see Section IV).

There are several reasons for the present confusion. In the first place the remarkably successful semi-empirical formula was the consequence of an algebraic error in a derivation based on the assumption of isotropic diffusion (Huxley 1940). Moreover it proved possible subsequently to provide some theoretical justification for it (Huxley and Crompton 1955; Hurst and Liley 1965), although the validity of the additional assumptions necessary was never shown directly (see Section III).

* Ion Diffusion Unit, Research School of Physical Sciences, Australian National University, P.O. Box 4, Canberra, A.C.T. 2600. More recently the same formula was derived by Lawson and Lucas (1965), also on the basis of isotropic diffusion, but the argument used by these authors appears to be incorrect. To this somewhat confused theoretical picture must be added the failure of experiments in some instances to substantiate the semi-empirical formula in circumstances in which the basis for it now appears to be well understood (Warren and Parker 1962). It is the purpose of this paper to clarify the position in order that the validity of experimental data from this type of experiment can be properly assessed.

II. SOLUTIONS BASED ON ISOTROPIC DIFFUSION

In the Townsend-Huxley experiment (Huxley and Crompton 1962), the proportion R of the total current received by the central portion of the anode (or central strip in some instances) has, until recently, been thought to depend only on the ratio $E/(D/\mu)$ and on the dimensions of the apparatus, in particular its length h and the radius of the central disc (or the half-width of the strip) which will be denoted by b. Here E denotes the electric field, assumed to be parallel to the z axis in what follows, D is the (isotropic) diffusion coefficient, and $\mu = W/E$ is the electron mobility, W being the drift velocity. Until recently the relationship between R and D/μ , b, and h was found by solving the steady state continuity equation

$$D\,\nabla^2 n = W\,\partial n/\partial z\,,\tag{1}$$

where n(x, y, z) is the electron number density (but see also Lucas 1964). Three basically similar solutions of this equation have been proposed and used, each based on somewhat different assumptions concerning the boundary conditions. These solutions will now be described and commented on without stressing the mathematical detail which is straightforward but lengthy (see e.g. Huxley 1940, 1972; Warren and Parker 1962; Lowke 1971).

(a) Townsend's Ratio Formula

The first solution was proposed by Townsend (see e.g. Townsend 1915) who took into account not only the finite size of the entrance aperture (electron source) in the cathode but also the effect of the cylindrical boundary of his apparatus. The boundary condition he adopted was n = 0 over a cylindrical surface of radius crepresenting the guard electrodes and over the surface of the cathode except for the entrance aperture, of radius a, over which n was assumed to be constant. Perhaps surprisingly, since the electron density at the cylindrical boundary was small compared with its value at the anode, the same boundary condition was not imposed at the anode, the assumption being made that the distribution of current over the anode was the same as the distribution of number density within an undisturbed stream. The current was therefore calculated simply by finding the flux due to drift (proportional to nW) across a geometrical plane replacing the anode. The flux due to diffusion (proportional to $D \partial n/\partial z$) was omitted. With these assumptions, Townsend obtained the formula

$$R = \frac{b \sum_{k} \{J_1(\mu_k a) J_1(\mu_k b) \exp(-\theta_k h) / x_k^2 J_1^2(\mu_k c)\}}{c \sum_{k} \{J_1(\mu_k a) \exp(-\theta_k h) / x_k^2 J_1(\mu_k c)\}},$$
(2)

where J_1 is a Bessel function of the first kind, $\mu_k = x_k/c$, x_k is the *k*th root of $J_0(x) = 0$, and $\theta_k = (\lambda^2 + \mu_k^2)^{\frac{1}{2}} - \lambda$, with $\lambda = E/2(D/\mu)$. Townsend and his collaborators used this formula with remarkable success in many investigations. As will become apparent, the reason for this success was the geometry of the apparatus to which it was applied.

Although applicable to the form of the apparatus in which a slit source and strip division of the anode are used, the solution developed by Pidduck (1925) is worthy of note because it appears to have been the first to satisfy the boundary condition n = 0 at the anode as well as the other boundary conditions. After he had solved equation (1) for the correct boundary conditions Pidduck calculated the current to the anode by considering the diffusive component only. As applied to the present problem, Pidduck's solution should therefore lead to equation (3) in the following subsection.

(b) Huxley's Formula based on Assumptions of Point Source and n = 0 at Cathode and Anode

The solutions of Townsend (1915) and Pidduck (1925) had two basic shortcomings: in the first place they were inconvenient to use because of the slow convergence of the infinite series and, secondly, they were based on the assumption that n is constant over the entrance aperture. Moreover, as we have seen, Townsend's solution did not satisfy the anode boundary condition. Huxley (1940) therefore proposed a form of the apparatus and the corresponding solution* of equation (1) which avoided these difficulties and also a further problem that arises from the unknown structure of the stream at the source. Realizing from Townsend's work that a small hole acts as a point source and that the diameter of the guard electrodes can be made sufficiently large, without inconvenience, to ensure that their influence is negligible, Huxley developed a solution which was built up of the contributions of an infinite number of dipole-like terms. The first "dipole" represented the source and ensured that n was zero over the cathode except at the origin; a second "dipole" was placed beyond the anode to neutralize the number density (but not the flux) at the anode from the source dipole, thus satisfying the boundary condition at this electrode; a third was placed behind the cathode to neutralize the small perturbation to n at the cathode due to the second dipole, and so on. In practice, apart from exceptional experimental circumstances, contributions from terms higher than the second are negligible, and they will be disregarded in what follows.

The current to the anode now arises simply from the diffusive flux, since the flux due to drift is zero, and the formula for the current ratio is

$$R = 1 - \{(h/d) - (1/\lambda h) + (h/d^2\lambda)\}(h/d) \exp\{-\lambda(d-h)\},$$

$$d = (b^2 + h^2)^{\frac{1}{2}}.$$
(3)

where

An identical ratio formula may be derived from equation (5) of the paper by Warren and Parker (1962) whose solution is based on the same formalism. These authors extended the solution to allow for a finite aperture and for a cylindrical

* Although the formalism of this solution was correct, an algebraic error led to the so-called Huxley empirical formula (Section II(c)) rather than equation (3).

boundary of finite radius, thus solving equation (1), with the correct boundary conditions everywhere, for an apparatus of the form proposed by Townsend. As expected, they found that these refinements were generally unnecessary for analysing the results of their experiments since their apparatus had a relatively small entrance aperture and guard electrodes of reasonably large diameter. Over the greater part of the working range of the experimental parameters, therefore, the ratio curves in Figures 2 and 3 of Warren and Parker's paper may be calculated using the simple formula (3) above.

Since equation (3) was based on a solution that satisfied the boundary conditions everywhere, and since the current to the anode was correctly calculated from the diffusive flux, its application was expected to lead to the satisfactory interpretation of the experimental results. Unfortunately it did not. Crompton and Jory (1962) showed that the ratio formula gave consistent results for a considerable range of the experimental parameters but that it failed elsewhere. This failure was attributed to a breakdown of the assumed boundary conditions. On the other hand, Warren and Parker (1962) found that the formula was generally not satisfactory in interpreting their experimental results, and they were forced to abandon it everywhere in favour of an empirical curve that led to a consistent interpretation. They accounted for the failure in this case in terms of field distortion within the apparatus, an explanation that now seems almost certain to have been correct. No such explanation could be used to account for Crompton and Jory's results, however, since field distortion would be expected to lead to a *universal* failure of the formula, and not simply to a failure over part of the range as observed.

Finally, an anomaly arose at this point to which a good deal of the subsequent confusion can be attributed: in the regime in which equation (3) failed to give a consistent interpretation of Crompton and Jory's (1962) results, the formula given in the following subsection was found to fit the data so well that its validity seemed unquestionable.

(c) Huxley's Semi-empirical Formula

The third solution, the so-called empirical or Huxley formula, had an accidental origin. Owing to an algebraic error to which attention was drawn by Huxley and Crompton (1955),* this formula, namely

$$R = 1 - (h/d) \exp\{-\lambda(d-h)\}, \qquad (4)$$

was derived by Huxley (1940) as the current ratio appropriate to the conditions enumerated in Section II(b) instead of the correct formula given in equation (3).

It is interesting to note that this formula may be derived on the basis of the assumptions adopted by Townsend (Section II(a)) provided $a \to 0$ and $c \to \infty$. Thus, in the terminology of this paper, the semi-empirical formula may also be derived by assuming that the point source behaves as a dipole and that the current distribution at the anode is the same as the distribution of number density at a geometrical plane representing the anode.

* The ratio formula for the dipole solution given by Huxley and Crompton (1955) was itself incorrect, although the error is insignificant.

III. ATTEMPTS TO JUSTIFY SEMI-EMPIRICAL FORMULA

Since equation (4) had led to the successful interpretation of a large body of experimental data (Crompton and Sutton 1952), some justification for it was sought. It was first shown (Huxley and Crompton 1955) that the formula resulted from the use of a pole-like source term and an image to make n = 0 over the anode. However, although the anode boundary condition was met, the cathode boundary condition was not. Since the reflection coefficient for low energy electrons from metal surfaces is known to be high, the suggestion of a breakdown of the electrode boundary conditions was not unreasonable, although a breakdown at the cathode and not at the anode was entirely speculative. A more rigorous test of the empirical formula by Crompton and Jory (1962), who tested their results in the region of significant disagreement between equations (3) and (4) against results taken under conditions where the two ratio formulae converged, led to a further examination of the theory and to two alternative derivations of equation (4).

Hurst and Liley (1965) showed that the formula could be obtained by using generalized boundary conditions at anode and cathode and suggested that the use of equation (4), interpreted in this way, could lead to an estimate of the reflection coefficient at the metal surfaces.

Equation (4) was also derived by Lawson and Lucas (1965) using a new approach, but this derivation, which was based on the correct boundary condition n = 0 at anode and cathode, contained a fallacy. In an earlier paper on which this work was based (Lucas 1964), Lucas took as his starting point the time-dependent continuity equation

$$\partial n/\partial t = D \,\nabla^2 n - W \,\partial n/\partial z + \alpha W n \,, \tag{5}$$

which also includes a term αWn to account for ionization, α being the primary ionization coefficient. For the purpose of the present paper, which is restricted for simplicity to situations where ionization is negligible, this term may be omitted. Lucas's solution of equation (5) comprised the sum of a number of terms of the form

$$n(x, y, z, t) = [n_{0s}/(4\pi Dt)^{3/2}]\exp[-\{(z - d_s - Wt)^2 + x^2 + y^2\}/4Dt], \quad (6)$$

which represented drifting and diffusing electron groups released at time t = 0 as point distributions of n_{0s} electrons at positions d_s along the z axis, the source group being released at the origin. The purpose of the higher order terms, released at appropriate positions with appropriate strengths, was to preserve the boundary conditions at cathode and anode. With this formalism Lawson and Lucas (1965) obtained the formula for R

$$R \simeq 1 - \left[(h/d) + \{1 - (h/d)\} \exp(-2\lambda h) \right] \exp\{-\lambda (d-h)\}, \tag{7}$$

in the notation of the present paper. For $h/d \approx 1$ and for all values of λh that are experimentally feasible, equation (7) reduces to the Huxley formula. If correct, this result would be surprising, since it should be possible to build up the steadystate solution as the sum of the electron number densities resulting from an infinite series of travelling groups (Huxley 1972), it being assumed here as in nearly all other derivations that electron-electron interactions are negligible. The formulae for the current ratios derived from each approach, i.e. equations (3) and (7), should therefore be identical. This apparent anomaly arises from an error in Lucas's (1964) original paper whereby some of the travelling groups were given a velocity of -W. Thus although the boundary conditions are satisfied the continuity equation is not (D. S. Burch, personal communication).

IV. Solutions based on Spatially Dependent Energy Distribution Functions and Non-isotropic Diffusion

A further attempt to put the theory on a firmer footing was made by Parker (1963) whose work is the forerunner of present theories which take account of the spatial dependence of the energy distribution function within the diffusion chamber. Parker showed that the spatial dependence could lead to significant errors in the conventional interpretation of the experiment although he concluded that "it would appear ... in most cases the experiments have not been appreciably affected by using the usual interpretation of this experiment* and in the cases where $(b/h)^2$ was large enough for appreciable deviations to exist, such deviations were masked by other effects". Although this statement referred specifically to Warren and Parker's (1962) experiments it is in fact generally true. Unfortunately Parker was unable to formulate his theory to take account of the energy dependence of the momentum transfer cross section, other than in a particular case (constant collision frequency), or to include the effect of electrode boundaries or inelastic collision processes.

Desloge and Mitchell (1970) adopted a different method of solution but again had to restrict their analyses to a case corresponding to apparently unrealistic boundary conditions. Like Parker, they were able to give criteria which had to be met if the effects of the spatial variation of the energy distribution function were to be unimportant, but the ratio formula which they derived, namely

$$R = 1 - \frac{1}{2}(1 + h/d) \exp\{-\lambda(d - h)\},$$

is not the same as any of those yet cited. It corresponds to an isolated point source ("pole"), and is based on the assumption that the current to the anode is the sum of the currents due to drift and diffusion at a geometrical plane replacing the anode. Their solution therefore corresponds to a point source solution with no boundaries of any description.

The answer to the long-standing paradox referred to at the outset of this paper has now apparently been found in the application of the theory of anomalous longitudinal diffusion (Parker and Lowke 1969; Lowke and Parker 1969) to this problem (Lowke 1971; Huxley 1972). Lowke (1971) assumed that the electron number density within the diffusion chamber could be found by solving a continuity equation that was modified to allow for anisotropic diffusion, namely

$$D_{\rm L}\frac{\partial^2 n}{\partial z^2} + D_{\rm T}\left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2}\right) = W\frac{\partial n}{\partial z}.$$
(8)

This equation embodies the assumption that the transport coefficients W, $D_{\rm L}$, and $D_{\rm T}$ are independent of position even though the phenomenon of anisotropic diffusion arises from the spatial dependence of the velocity distribution function.

* That is, an interpretation based on a spatially independent energy distribution function.

A formal justification of equation (8) was given by Huxley (1972) on the assumption that the velocity distribution function can be expressed as a function of position through the spatial derivatives of n. With this assumption he has shown that the Maxwell-Boltzmann equation including all spatial gradient terms can be satisfied provided n satisfies a continuity equation of this form.

The application of equation (8) to the Townsend-Huxley experiment has at last led to a satisfactory explanation of the success of equation (4). The current ratio formula derived on the basis of equation (8) and the "correct" boundary conditions, that is, n = 0 at cathode and anode, is

$$R = 1 - \{(h/d') - (1/\lambda_{\rm L}h) + (h/\lambda_{\rm L}d'^2)\}(h/d')\exp\{-\lambda_{\rm L}(d'-h)\}, \qquad (9)$$

where $\lambda_{\rm L} = W/2D_{\rm L}$, $d'^2 = h^2 + b'^2$, and $b' = (D_{\rm L}/D_{\rm T})^{\frac{1}{2}}b$. It has been shown (Lowke 1971; Huxley 1972) that provided b/h is small equation (9) leads to experimental results for $D_{\rm T}/\mu$ that are insignificantly different from those calculated using equation (4) provided $D_{\rm L}/D_{\rm T} \approx 0.5$, that is, the momentum transfer cross section is practically independent of energy (Parker and Lowke 1969; Lowke and Parker 1969). Furthermore, for sufficiently large values of $\lambda_{\rm L} h$ (again provided b/h is small) equation (9) converges to equation (4) regardless of the value of $D_{\rm L}/D_{\rm T}$. These conditions have more often than not been met experimentally, and it is for this reason that the flaws in the earlier theories remained undetected for so long.

We finally consider a simple physical argument which indicates why the current ratio R is independent of $D_{\rm L}$ for large values of $\lambda_{\rm L} h$ and why, with this condition, equations (4) and (9) converge provided b/h is small. We first note that equation (4) can be rewritten as

$$R = 1 - (h/d) \exp\{-\lambda b^2/(d+h)\},$$

since $d^2 = h^2 + b^2$. Thus, provided b remains fixed, the ratio formula approaches the form

$$R_{
m lim} = 1 - \exp(-\lambda b^2/2h) \qquad {
m as} \qquad h o \infty$$
 .

The time-dependent continuity equation which allows for anisotropic diffusion (but not ionization) is

$$\frac{\partial n}{\partial t} = D_{\rm L} \frac{\partial^2 n}{\partial z^2} + D_{\rm T} \left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2} \right) - W \frac{\partial n}{\partial z}, \tag{10}$$

where the field is parallel to the z direction. The solution of this equation which corresponds to the release of n_0 electrons at the origin of coordinates at time t = 0 is

$$n = \frac{n_0 \exp(-\rho^2/4D_{\rm T}t) \exp\{-(z-Wt)^2/4D_{\rm L}t\}}{4\pi D_{\rm T}t(4\pi D_{\rm L}t)^{\frac{1}{2}}} = n_0 p_1(\rho, t) p_2(z, t), \qquad (11)$$

where

$$p_1(
ho,t) = rac{\exp(-
ho^2/4D_{
m T}t)}{4\pi D_{
m T}t}, \qquad p_2(z,t) = rac{\exp\{-(z-Wt)^2/4D_{
m L}t\}}{(4\pi D_{
m L}t)^{rac{1}{2}}}.$$

We note that

$$2\pi\int_0^\infty p_1(
ho,t)\,
ho\,\mathrm{d}
ho = \int_{-\infty}^\infty p_2(z,t)\,\mathrm{d}z = 1\,,$$

that is, p_1 and p_2 are probability functions such that, at time t, $2\pi p_1(\rho, t) \rho d\rho$ is the probability of finding an electron in the cylindrical volume bounded by ρ and $\rho+d\rho$, and that $p_2(z,t) dz$ is the probability of finding an electron between the planes z and z+dz.

Let us assume that $D_{\rm L} = 0$. Then $p_2(z)$ becomes the delta function $\delta(z - Wt)$ and equation (11) becomes

$$n = (n_0/4\pi D_{\rm T} t) \exp(-\rho^2/4D_{\rm T} t) \,\delta(z - W t)\,,\tag{12}$$

which describes the number density within a two-dimensional distribution of charge confined to the plane z = Wt, that is, an axially symmetric, infinitesimally thin distribution moving parallel to the z axis at velocity W. It may be easily shown that the current ratio formula, found by evaluating equation (12) at t = h/W, is given by

$$R = 1 - \exp(-\lambda_{\rm T} b^2/2h) = R_{\rm lim}\,, \tag{13}$$

where $\lambda_{\rm T} \equiv W/2D_{\rm T} \equiv W/2D \equiv \lambda$. This then is the ratio formula which corresponds to the case when every electron released from the source arrives at the anode at the same time. Equation (13) also follows immediately from equation (9) on putting $D_{\rm L} = 0$.

From the preceding argument it is obvious that, even when $D_{\rm L} \neq 0$, the ratio formula must approach $R_{\rm lim}$ provided the spread of arrival times at the anode is small compared with t = h/W. Stated alternatively, one would expect to find a current distribution accurately predicted by $R_{\rm lim}$ provided the half-width of $p_2(z)$ were small compared with h. Now it may be shown without difficulty (see e.g. Wagner, Davis, and Hurst 1967) that if δt is the full width at half maximum of the distribution of arrival times, which is approximately Gaussian, then $\delta t/t$ is proportional to $(\lambda_{\rm L} h)^{-\frac{1}{2}}$, that is, for fixed values of E/N and N, the ratio $\delta t/t \to 0$ as $h \to \infty$. Furthermore, since the effects of the cathode and anode boundaries are localized, their influence on the radial distribution of the travelling group also diminishes as h increases. It follows that, provided h is sufficiently large, the current ratio should be predicted by $R_{\rm lim}$, that is, the ratio should depend only on the lateral diffusion coefficient.

V. Conclusions

In this paper it has been shown that there are two basic reasons why analyses of the Townsend-Huxley lateral diffusion experiment that are based on the assumption of isotropic diffusion have led to consistent results even though diffusion is usually far from isotropic. The fact which accounts for the self-consistence and intrinsic accuracy of the majority of the earlier results is simply that the structure of the diffusing stream was usually determined at a considerable distance from the source. In this case, as we have seen, the lateral spread of the stream is little affected by longitudinal diffusion. On the other hand, a remarkable coincidence was responsible for the success of the original Huxley formula (equation (4)) even when the stream was analysed close to the source, namely that equation (9) is closely resembled by equation (4) when $D_{\rm L}/D_{\rm T} = 0.5$, and that the testing of the formula was carried out using gases for which the momentum transfer cross section is a slowly varying function of energy and for which, as a consequence, $D_{\rm L}/D_{\rm T} \simeq 0.5$. For the above two reasons the analysis of much of the earlier experimental work that was concerned simply with the determination of the ratio $D_{\rm T}/\mu$ is still valid. On the other hand, in situations where attachment and ionization processes play a significant part in determining the spatial variation of the electron number density, the results obtained from analyses based on isotropic diffusion are not valid. The existing data derived from lateral diffusion experiments in these circumstances therefore require re-analysis.

VI. ACKNOWLEDGMENTS

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