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¹⁷We have calculated the classical kinetic-energy correction by different methods and have always found a leading term of the form (24) and additional terms of higher order in T^{-1} . We are now studying in more detail the relation between the classical kinetic- and potential-energy corrections and will report our results elsewhere. But it should be clear that potential-energy terms which violate Klein's original theorem result from the exclusion of bound electrons, and cannot be obtained from a classical theory which does not explicitly distinguish between free and bound electrons.

Transport Model for Converting Charged Species in Drift Tubes*

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The three-dimensional transport equations of converting charged species are solved, taking into consideration the effect of the following for both primary and secondary species: different electron and/or ion temperatures, different initial radial distributions, an absorbing drift-tube wall, and the distinction between radial and axial diffusion coefficients. The solution bears explicit dependence on all the relevant geometries of the drift-tube assembly, making it applicable to all known types of drift-tube arrangements. Three realistic initial charge distributions are considered. Remaining inadequacies of this transport model are critically examined.

I. INTRODUCTION

In the past decade, various articles¹⁻¹¹ have offered analytical solutions to transport models of converting ions in drift tubes. However, none of the models was treated in a way that is general enough to be applicable to all known arrangements of drift-tube assemblies.^{6, 9, 12-17, 22} Moreover, their models are not three dimensional in a complete sense, in that none of them consider for both parent and daughter species (a) effects of different ion temperatures, (b) effects of different initial radial distributions, (c) distinction between radial and axial diffusion coefficients, and (d) effects of an absorbing drift-tube wall. For accurate interpretation of drift-tube data, which generally have prominent three-dimensional characteristics, such features should be included in the mathematical solutions. For experiments designed to interpret the drift-tube data to 1% accuracy,²⁰⁻²² the advantage of having the solutions of a versatile trans-

port model establishing guidelines is considerable. On the other hand, it should be stated that experiments^{1, 2, 20-22} can be designed to minimize the above listed effects, not properly accounted for owing to the inadequacies of the theoretical model. But the accompanying requirements are often severe. These requirements are usually a combination of the following: (1) the construction of a bulky drift tube, (2) the acceptance of unnecessary limitations on the dynamical ranges of the experiment, and (3) the acceptance of time-consuming internal consistency checks for every simplification assumed in the theoretical model at every extreme of the parametric space.

II. STATEMENT OF THE PROBLEM

The relevant transport equations describing the three-dimensional drift and diffusion phenomena of two charged species 1 and 2, where species 1 converts to species 2 are

$$\frac{\partial \rho_1}{\partial t} = \nabla \cdot (\mathfrak{D}_1 \cdot \nabla \rho_1) - \vec{v}_1 \cdot \nabla \rho_1 - \alpha \rho_1, \quad (1)$$

$$\frac{\partial \rho_2}{\partial t} = \nabla \cdot (\mathfrak{D}_2 \cdot \nabla \rho_2) - \vec{v}_2 \cdot \nabla \rho_2 + \alpha \rho_1, \quad (2)$$

where ρ , \mathfrak{D} , \vec{v} , and α are, respectively, the charge density, diffusion tensor, drift velocity, and conversion frequency.

The region of interest for these equations is a cylindrical cavity of infinite length, and radius, r_0 , along whose surface, the charge density is assumed to be zero. Hence, the boundary conditions are

$$\rho_i(r_0, \theta, z, t) = 0, \quad i = 1, 2; \quad (3)$$

$$\rho_i(r, \theta, \pm \infty, t) = 0, \quad i = 1, 2. \quad (4)$$

The origin of the coordinate system is at the entrance shutter of the drift region. The initial conditions are

$$\rho_i(r, \theta, z, 0) = f_i(r)[H(z + h_i) - H(z)], \quad i = 1, 2, \quad (5)$$

where H and h_i are, respectively, a Heaviside step function and the axial width of the initial slab of the i th species. Hence, Eq. (5) represents an arbitrary radial distribution, with azimuthal symmetry, and a step-function axial distribution of width h_i .

III. SOLUTION

Using the separation of variables approach, where a_{ik} are constants to be determined from the initial conditions:

$$\rho_i(r, z, t) = \sum_{k=1}^{\infty} a_{ik} R_k(r) Z_{ik}(z, t), \quad i = 1, 2, \quad (6)$$

the separated transport equations, which are consistent with the boundary conditions and the assumption of diagonal diffusion tensors, are

$$\frac{d^2 R_k}{dr^2} + \frac{1}{r} \frac{dR_k}{dr} = -\frac{\beta_k^2}{r_0^2} R_k, \quad (7)$$

$$\frac{\partial Z_{1k}}{\partial t} = D_{z1} \frac{\partial^2 Z_{1k}}{\partial z^2} - v_1 \frac{\partial Z_{1k}}{\partial z} - \left(\alpha + \frac{\beta_k^2}{r_0^2} D_{r1} \right) Z_{1k}, \quad (8)$$

$$\frac{\partial Z_{2k}}{\partial t} = D_{z2} \frac{\partial^2 Z_{2k}}{\partial z^2} - v_2 \frac{\partial Z_{2k}}{\partial z} + \alpha Z_{1k} - \frac{\beta_k^2 D_{r2}}{r_0^2} Z_{2k}, \quad (9)$$

where β_k is the k th root of J_0 , which is a Bessel function of the first kind and zeroth order. D_{r1} and D_{z1} are, respectively, radial and axial diffusion coefficients of the i th species.

A solution to Eq. (7) is

$$R_k(r) = J_0(\beta_k r/r_0), \quad k = 1, 2, 3, \dots \infty. \quad (10)$$

These solutions to the radial equation form a complete set, and we incorporate the standard procedure of expanding the initial radial distribution in terms of the eigenfunctions of this complete set. Hence,

$$f_i(r) = \sum_{k=1}^{\infty} a_{ik} J_0(\beta_k r/r_0), \quad (11)$$

and the a_{ik} can be evaluated using the orthogonality of the eigenfunctions,

$$a_{ik} = [2/r_0^2 J_1^2(\beta_k)] \int_0^{r_0} f_i(r) J_0(\beta_k r/r_0) r dr. \quad (12)$$

Equations (8) and (9) are solved by the use of Fourier transforms. The solution for the ion distribution of the daughter species is composed of two infinite sums, in k . The first is due to initial injection of the daughter species. The second sum in k , which shall be referred to as the reaction term, is due to conversion to the daughter species from the parent species. This latter sum is composed of two partial sums, due to the distribution of poles occurring in a contour integration. The first goes from $k=1$ to N , where N is the largest positive integer assumed by k such that $\alpha + D_r \gamma_k > 0$. The second partial sum goes over all other values of k where $\alpha + D_r \gamma_k < 0$. In cases where D_r is always positive, N clearly equals infinity, and the second sum is not taken.

$$\rho_1(r, z, t) = \frac{1}{2} \sum_{k=1}^{\infty} a_{1k} J_0(\beta_k r/r_0) \exp(-\alpha_{1k} t) (\operatorname{erf} \xi_1' - \operatorname{erf} \xi_1), \quad (13)$$

$$\begin{aligned} \rho_2(r, z, t) = & \frac{1}{2} \sum_{k=1}^{\infty} a_{2k} J_0(\beta_k r/r_0) \exp(-\alpha_{2k} t) (\operatorname{erf} \xi_2' - \operatorname{erf} \xi_2) \\ & + \frac{\alpha}{4D_z \lambda} \sum_{i=1}^2 \sum_{j=1}^2 \left(\sum_{k=1}^N \frac{(-1)^{i+j} a_{1k}}{\sigma_j} \exp(-\alpha_{1k} t) J_0(\beta_k r/r_0) [\exp(\eta_{ijk}^2 + 2\eta_{ijk} \xi_{ij})] \right. \\ & \times \operatorname{erfc}(\xi_{ij} + \eta_{ijk}) - \exp(\eta_{ijk}^2 + 2\eta_{ijk} \xi_{ij}'') \operatorname{erfc}(\xi_{ij}'' + \eta_{ijk}) - (\operatorname{erf} \xi_{ij}'' - \operatorname{erf} \xi_{ij}) \\ & + \sum_{k=N+1}^{\infty} \frac{(-1)^{i+j} a_{1k}}{\sigma_j} \exp(-\alpha_{ik} t) J_0(\beta_k r/r_0) [\exp(\eta_{ijk}^2 + 2\eta_{ijk} \xi_{ij})] \\ & \left. \times \operatorname{erfc}(\xi_{ij} + \eta_{ijk}) - \exp(\eta_{ijk}^2 + 2\eta_{ijk} \xi_{ij}'') \operatorname{erfc}(\xi_{ij}'' + \eta_{ijk}) - (\operatorname{erf} \xi_{ij}'' - \operatorname{erf} \xi_{ij}) \right), \quad (14) \end{aligned}$$

where

$$\begin{aligned} \xi_i &= (z - v_i t) \tau_i, \quad \xi_i' = (z + h_i - v_i t) \tau_i, \quad \xi_i'' = (z + h_1 - v_i t) \tau_i, \quad \alpha_{ik} = \alpha \delta_{i1} + D_{ri} \gamma_k, \quad D_z = D_{z1} - D_{z2}, \\ D_r &= D_{r1} - D_{r2}, \quad v = v_1 - v_2, \quad \gamma_k = \beta_k^2 / r_0^2, \quad \sigma_{jk} = \lambda_k + (-1)^j v / 2D_z, \quad \eta_{ijk} = \sigma_{jk} / 2\tau_i, \\ \lambda_k^2 &= v^2 / 4D_z^2 + \alpha / D_z + \gamma_k D_r / D_z, \quad \eta_{ijk}' = (-1)^k \eta_{ijk}, \quad \xi_{ik} = (-1)^k \xi_i, \quad \tau_i = (4D_{zi} t)^{-1/2}. \end{aligned}$$

While the above solutions are formally composed of infinite sums in k , only the first-few terms are necessary to adequately describe most realistic initial radial distributions.

IV. INITIAL CONDITIONS OF SPECIAL INTEREST

Three types of initial conditions, commonly encountered in experimental situations, are considered below:

(1) The drift-tube assembly is assumed to have a source aperture radius which is the same as that of the drift space. Initially there exists over the source aperture an ion slab of thickness h_i , having uniform density along the axial direction. The radial density distribution of the ion cloud is described by a zeroth-order Bessel function, whose first zero is at the wall of the drift space. Physically, when the time it takes the ions to travel from the source to the collector is long as compared with the characteristic decay time of the higher diffusion modes, such a description is appropriate. Mathematically, such an initial condition can be expressed as

$$\rho_i(\vec{r}, t=0) = C_i J_0\left(\beta_1 \frac{r}{r_0}\right) [H(z+h_i) - H(z)] \quad (15)$$

$$= \sum_{k=1}^{\infty} a_{ik} J_0\left(\beta_k \frac{r}{r_0}\right) [H(z+h_i) - H(z)], \quad (16)$$

where C_i is the charge density of the i th species at the center of the initial ion cloud. Evaluation of the coefficients a_{ik} is trivial. Only the first coefficient is nonzero:

$$a_{ik} = C_i \delta_{1k}, \quad k=1, 2, 3, \dots \infty. \quad (17)$$

(2) The source aperture radius is the same as that of the drift space as in case (1), but the initial ion slab now has uniform density distribution in both the axial and the radial directions. The thickness of the initial slab is still h_i . Mathematically, such a distribution is described by

$$\rho_i(\vec{r}, 0) = C_i [1 - H(r_0)] [H(z+h_i) - H(z)] \quad (18)$$

$$= \sum_{k=1}^{\infty} a_{ik} J_0\left(\beta_k \frac{r}{r_0}\right) [H(z+h_i) - H(z)]. \quad (19)$$

The evaluation of the coefficients a_{ik} by Eq.(12) is straightforward.

$$a_{ik} = 2C_i / [\beta_k J_1(\beta_k)], \quad (20)$$

where J_1 is a Bessel function of the first order. The first six or so terms of the k series suffice

to give a fairly accurate description of the density as a function of \vec{r} and t . For situations, where $(D_i \times \text{total drift time})^{1/2}$ is much larger than r_0 , a few more terms may need to be taken.

(3) The source aperture radius r_s is assumed smaller than that of the drift space. The initial ion slab is cylindrically shaped, having dimensions h_i and r_s . Its distribution is uniform in both the radial and axial direction. Mathematically,

$$\rho_i(\vec{r}, 0) = C_i [1 - H(r_s)] [H(z + h_i) - H(z)] \quad (21)$$

$$= \sum_{k=1}^{\infty} a_{ik} J_0\left(\beta_k \frac{r}{r_0}\right) [H(z + h_i) - H(z)]. \quad (22)$$

The a_{ik} evaluated by Eq. (12) are

$$a_{ik} = \left[2C_i r_s J_1\left(\beta_k \frac{r_s}{r_0}\right) \right] / \left[r_0 \beta_k J_1^2(\beta_k) \right]. \quad (23)$$

The choice of the number of terms is determined by the ratio $(D_i \times \text{total drift time})^{1/2} / r_s$. More than ten terms should be taken, if this ratio is larger than one.

V. APPLICATION TO DRIFT EXPERIMENTS

In drift-tube experiments, one is usually interested in the current at the collector, which is

$$I_i(z_0, t) = 2\pi \int_0^{r_c} \left(v_i \rho_i(z_0, r, t) - D_{zi} \frac{\partial \rho_i(z_0, r, t)}{\partial z} \right) r dr, \quad (24)$$

where r_c is the radius of the collector. The differentiation of ρ with respect to z is straightforward, and the integration with respect to r needs only the use of the relationship

$$\int_0^{r_c} J_0\left(\beta_k \frac{r}{r_0}\right) r dr = r_0 r_c J_1\left(\beta_k \frac{r_c}{r_0}\right) / \beta_k. \quad (25)$$

Hence the substitution of Eqs. (13) and (14) into (24) yields an analytical expression for the experimentally measured value $I(z_0, t)$, in terms of t , α , v_1 , v_2 , D_{z1} , D_{r1} , D_{z2} , D_{r2} , C_1 , C_2 , z_0 , r_s , r_0 , r_c . The explicit dependence of I on the last-four parameters, z_0 , r_s , r_0 , and r_c makes the solution applicable to all drift-tube arrangements^{6,9,12-19,22} of cylindrical symmetry. In experiments where the measured quantity is charge collected at z_0 during time interval τ , instead of the instantaneous current $I(z_0, t)$, one needs to compare the data with the expression,

$$q_i(t) = \int_t^{t+\tau} I_i(z_0, t) dt. \quad (26)$$

These solutions are also applicable to drift-tube experiments operated under steady-state condition,^{5,16} provided that one lets $h_i \gg z_0$ and $t = z_0/v_i$ in Eqs. (13) and (14). It being understood that such conditions demand that the solution is valid at the time range, $z_0/v_i \ll t \ll h_i/v_i$, only. The generality of these solutions is shown by the fact that the solutions offered for converting ions in Refs. 1 to 11 are special cases of our treatment.

VI. CRITICISM

Five assumptions made in this transport model are: (1) the wall surrounding the drift space is a perfect absorber, (2) no absorption by the source shutter, (3) no absorption by the collector (collector shutter), (4) the charge distribution is azimuthally symmetric, and (5) the diffusion equations adequately describe the motion of the charge distribution. The validity of the first assumption must be examined separately depending upon the nature of the charged particles involved. In the case of ion transport, the metal guard rings defining the drift space can be considered as genuine perfect absorbers. The only approximation made is the discrete spacing of the rings, instead of a continuous wall. This approximation can be examined with the help of a numerical example. Consider the case, where $v_1 = 50000$ cm/sec, $D_{z1} = D_{r1} = 900$ cm²/sec, $r_0 = r_s = 1$ cm, $h_i = 0.5$ cm, $z_0 = 10$ cm, $\alpha = 0$ sec⁻¹ and the initial radial distribution is uniform. The fractional loss due to radial diffusion, with the assumption of a perfectly absorbing wall, is 0.64; while that for six equally spaced guard rings is found to be 0.59. For drift tubes having guard rings more densely spaced than above, the approximation should be even better. In cases, where fast electrons are involved, the surrounding wall becomes somewhat reflective. Therefore, another approximation is involved. It is found, however, from simple consideration of net radial diffusion losses, that this assumption can be corrected to first order by the introduction of an effective drift-tube radius, r_0' . When the reflectivity R is small, r_0' can be related to the physical drift-tube radius r_0 , through the relationship $r_0' = r_0 / (1 - R)^{1/2}$.

The absorption of charged particles by the source shutter must be examined separately depending upon the size of the source aperture. If the source aperture is of the same size as the drift tube, only the absorption of back diffusion of the admitted charged particles needs to be considered after the shutter is closed. This problem was investigated by Lowke,¹¹ assuming an infinitely thin plane initial charge distribution. Taking his results and modifying it slightly to fit an initial step distribution of axial width h , one obtains

$$E = h / \{ z_0 [\exp(hv/2D) - 1] \}, \quad (27)$$

where E is the relative error in position (as compared to z_0) of the peak density at $t = (z_0 + h/2)/v$. When the diffusion coefficient is small, E can be taken as the relative error in the arrival time of the maximum current. This error is less than 1% for most ion-drift experiments. For electron-drift experiments the error can be significant. If the source aperture is smaller than that of the drift tube, and if the initial charge distribution has a finite width h , then one may consider the additional radial absorption effect by the shutter. The trailing edge of the initial charge distribution is always more severely distorted, because this portion of the distribution is limited by the physical presence of the source shutter to a cross section of radius r_s , while the mathematical representation allows it to diffuse in the radial direction as freely as the leading edge. If the time h/v is very small as compared with the total drift z_0/v , the effect due to this mathematical inadequacy is believed to be insignificant.

The effect of absorption by collector or collector shutter has been investigated by a number of persons.^{11, 23} Whealton and Woo²³ showed that the arrival-time spectrum is not significantly affected (less than 1%) by the assumption of either a completely transmitting or absorbing collector, for conditions where $(vz_0/D > 200)$. When the above condition is severely violated, as it is in some electron transport cases, one would expect that the assumption of a completely absorbing collector is more appropriate.^{11, 22} An analytical solution satisfying such a boundary condition can always be constructed from solutions to a perfectly transmitting collector transport model. The introduction of a negative image term, having appropriate strength for each diffusion mode, starting at $z = 2z_0$ and traveling with the same velocity, shall satisfy the $\rho(z_0, r, t) = 0$ boundary condition for all t . For example, a substitute expression

for Eq. (13), satisfying the absorbing collector boundary condition is

$$\rho_1(z, r, t) = \frac{1}{2} \sum_{k=1}^{\infty} a_{1k} Z_{1k}(z, r, t) + \frac{1}{2} \sum_{k=1}^{\infty} a_{1k} b_{1k} Z_{1k}(z - 2z_0, r, t), \quad (28)$$

where

$$Z_{1k} = J_0\left(\beta_k \frac{r}{r_0}\right) e^{-\alpha_{1k} t} (\operatorname{erf} \xi_{1k}' - \operatorname{erf} \xi_{1k}) \quad (29)$$

and where b_{1k} , the strength factor for k th diffusion mode, is determined by the condition:

$$Z_{1k}(z, r, t) \Big|_{z=z_0} = b_{1k} Z_{1k}(z - 2z_0, r, t) \Big|_{z=z_0}. \quad (30)$$

No experimental or theoretical evaluation on the validity of the fourth assumption was made. However, the azimuthal symmetry of the entire drift structure together with the effect of diffusion, gave reasonable grounds for having confidence in the validity of such an assumption. The restrictions to be observed, whenever the diffusion equations of (1) and (2) are used to describe the motion of converting charged particles in drift tubes, are already discussed elsewhere.^{9, 24}

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Some Properties of Correlation Functions of Irreducible Tensor Operators*

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Properties of quantum-mechanical time-dependent correlation functions of pairs of irreducible tensor operators are derived from the transformation properties of the tensor operators under rotations, the assumed rotational invariance of the Hamiltonian of the system, and the Wigner-Eckart theorem. It is shown that many of the possible correlation functions are zero, and that the nonzero correlation functions can be expressed in terms of reduced matrix elements. Some examples of irreducible tensor operators which occur in applications are given, and their reduced matrix elements are calculated.

I. INTRODUCTION

Quantum-mechanical time-dependent correlation functions of pairs of operators occur in the theory of magnetic resonance and relaxation,¹ and also in the theories of other phenomena, such as microwave resonance, Raman light scattering, and neutron scattering.² The operators whose correlation functions are to be calculated are irreducible tensor operators, or they can be expressed as linear combinations of irreducible tensor operators. However, the properties of irreducible tensor operators do not seem to have been exploited fully in previous calculations of correlation functions.

Some general properties of correlation functions of irreducible tensor operators are derived in Sec. II, and it is shown how such correlation functions can be expressed in terms of reduced matrix elements. In Sec. III correlation functions of tensor operators pertaining to a single molecule in the system are considered. Section IV is concerned with symmetry properties, symmetrized correlation functions, and their Fourier transforms. In Sec. V some examples of irre-

ducible tensor operators are given, and their reduced matrix elements calculated.

II. GENERAL PROPERTIES

An irreducible tensor operator of rank k can be defined³ as a set of $2k+1$ operators T_{km} , $m = -k, -k+1, \dots, k$, which transform under rotations of the coordinate system in the following manner. The operator T_{km} in a coordinate system S' is equal to the operator

$$RT_{km}R^{-1} = \sum_{m'=-k}^k D_{m'm}^k(\alpha\beta\gamma)T_{km'} \quad (1)$$

in a coordinate system S , where $\alpha\beta\gamma$ are the Euler angles specifying the orientation of S' with respect to S . R is the unitary rotation operator

$$R = \exp(-i\alpha L_z) \exp(-i\beta L_y) \exp(-i\gamma L_z), \quad (2)$$

where $\hbar L_y$ and $\hbar L_z$ are, respectively, the y and z components in S of the total angular momentum $\hbar\vec{L}$ of the system. The unitary matrix $\underline{D}^L(\alpha\beta\gamma)$ has elements