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We study the analytical solution of the time-dependent elastic Boltzmann transport equation in an infinite uniform isotropic medium with an arbitrary phase function. We calculate (1) the exact distribution in angle, (2) the spatial cumulants at any angle, exact up to an arbitrary high order n. At the second order, $n = 2$, an analytical, hence extremely useful combined distribution in position and angle, is obtained as a function of time. This distribution is Gaussian in position, but not in angle. The average center and spread of the half-width are exact. By the central limit theorem the complete distribution approaches this Gaussian distribution as the number of collisions (or time) increases. The center of this distribution advances in time, and an ellipsoidal contour that grows and changes shape provides a clear picture of the time evolution of the particle migration from near ballistic, through snake-like, and into the final diffusive regime. This second-order cumulant approximation also provides the correct ballistic limit. Algebraic expressions for the nth order cumulants are provided. The number of terms grows rapidly with n, but our expressives are recursive and easily automated.

I. Introduction

Search for an analytical solution of the time-dependent elastic Boltzmann transport equation has lasted for many years.[1-3] Besides being considered as a classical problem in fundamental research in statistical dynamics, a novel approach to an analytical solution of this equation may have applications in a broad variety of fields. To our knowledge, an exact solution, even in an infinite uniform medium, is available only for isotropic scattering case, given by E. H. Hauge,[4] in the form of a Fourier transform in space and Laplace transform in time. Based on the angular moment expansion with cut-off to certain order, the Boltzmann transport equation is transferred to a series of moment equations. In the lowest order, a diffusion equation is derived and its analytical solution in an infinite uniform medium is obtained for anisotropic scattering cases. This analytical solution has been broadly applied in many applications. For example, the solution of inverse problems in optical tomography, such as the location of a tumor in a woman's breast from the scattering of light pulses, requires the inversion of a weight matrix[5] obtained by convoluting two Green's functions of the forward scattering problem. The analytical solution of the diffusion equation has provided the needed Green's function. A similar procedure can be applied to other problems, such as using a laser to monitor cloud distributions, to detect objects inside a cloud, or the use of low-frequency sound to detect oil-bearing layers deep under water. The diffusion approximation fails at early times when the particle distribution is still highly anisotropic. The solutions of the diffusion equation or the telegrapher's equation do not produce the correct ballistic limit of particle propagation.[6] Numerical approaches, including the Monte Carlo method, are the main tools in solving the elastic Boltzmann equation; however, detailed solution of a five-dimensional Boltzmann transport equation using a predominately numerical approach leads to prohibitive CPU times.

In this paper, we seek an analytical solution of the elastic Boltzmann transport equation in an infinite uniform medium. We assume that the phase function, $P(\mathbf{s}, \mathbf{s}_0)$, depends only on the scattering angle: $P(\mathbf{s}, \mathbf{s}_0) = P(\mathbf{s}\cdot\mathbf{s}_0)$, where the velocity $\mathbf{v} = v\mathbf{s}$, $\mathbf{s}$ is a unit vector of direction, and v is the (constant) speed in the medium. Under this assumption, we can handle an arbitrary phase function. We obtain the exact angular distribution as a function of time. Based on this solution, we use a cumulant expansion of the particle distribution, $I(\mathbf{r}, \mathbf{s}, t)$, and derive exact spatial cumulants up to an arbitrary high order at any angle and time. A cut-off at second order yields a simple analytical expressions for $I(\mathbf{r}, \mathbf{s}, t)$, as a function of position $\mathbf{r}$, angle $\mathbf{s}$, and time t, and the particle density distribution, $N(\mathbf{r}, t)$, as a function of position $\mathbf{r}$ and time t. These spatial Gaussian distributions have the exact first cumulant (the position of center of the distribution) and the exact second cumulant (the half-width of spread of the distribution). After many scattering events have taken place, the law of large numbers (the central limit theorem) guarantees that the spatial Gaussian distribution that we calculate will become accurate in detail, since the higher cumulants become relatively small. At early times, the spread of the distribution is narrow, hence, the spatial distribution function can be claimed quantitatively accurate for many applications, in the sense that it has the correct mean position and the correct half-width of spread as a function of time. Measurement of the higher order cumulants could require measuring instruments of extreme resolution.

The remainder of this paper is organized as follows. Section II describes the derivation of formula: (1) obtaining an exact solution of the distribution in angle, (2) obtaining an exact formal solution in position and angle, (3) using the cumulant expansion to calculate the exact analytical expressions of cumulants up to an arbitrary high order, (4) describing the

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calculation of the particle distribution function using a spatial Fourier transform. Section III discusses using a cut-off at second order to produce explicit expressions of the distribution function and the density distribution. A brief discussion and summary then follows in Section IV. In the Appendix, we derive analytical formulas for evaluating integrals in eq 12.

II. Derivation of Cumulants to an Arbitrary High Order

The elastic Boltzmann kinetic equation of particles, with magnitude of velocity v, for the distribution function $I(\mathbf{r}, \mathbf{s}, t)$ as a function of time t, position $\mathbf{r}$, and direction $\mathbf{s}$, in an infinite uniform medium, from a point pulse light source, $\delta(\mathbf{r} - \mathbf{r}_0)\,\delta(\mathbf{s} - \mathbf{s}_0)\,\delta(t - 0)$, is given by[3]

$$\partial I(\mathbf{r}, \mathbf{s}, t)/\partial t + v\mathbf{s}\cdot\nabla_r I(\mathbf{r}, \mathbf{s}, t) + \mu_a I(\mathbf{r}, \mathbf{s}, t) = \mu_s \int P(\mathbf{s}, \mathbf{s}')[I(\mathbf{r}, \mathbf{s}', t) - I(\mathbf{r}, \mathbf{s}, t)]d\mathbf{s}' + \delta(\mathbf{r} - \mathbf{r}_0)\,\delta(\mathbf{s} - \mathbf{s}_0)\,\delta(t - 0) \quad (1)$$

where μ_s is the scattering rate, μ_a is the absorption rate, and $P(\mathbf{s}', \mathbf{s})$ is the phase function, normalized to $\int d\mathbf{s}'\, P(\mathbf{s}', \mathbf{s}) = 1$. When the phase function depends only on the scattering angle in an isotropic medium, we can expand the phase function in Legendre polynomials with constant coefficients,

$$P(\mathbf{s}, \mathbf{s}') = \frac{1}{4\pi}\sum_l a_l P_l(\mathbf{s}\cdot\mathbf{s}') \quad (2)$$

We first study the dynamics of the distribution in direction space, $F(\mathbf{s}, \mathbf{s}_0, t)$, on a spherical surface of radius 1, which is equivalent to the velocity space in the elastic scattering case. The kinetic equation for $F(\mathbf{s}, \mathbf{s}_0, t)$ can be obtained by integrating eq 1 over the whole spatial space, $\mathbf{r}$. The spatial independence of μ_s, μ_a, and $P(\mathbf{s}, \mathbf{s}')$ retains translation invariance. Thus the integral of eq 1 obeys

$$\partial F(\mathbf{s}, \mathbf{s}_0, t)/\partial t + \mu_a F(\mathbf{s}, \mathbf{s}_0, t) + \mu_s[F(\mathbf{s}, \mathbf{s}_0, t) - \int P(\mathbf{s}, \mathbf{s}')\, F(\mathbf{s}', \mathbf{s}_0, t)d\mathbf{s}'] = \delta(\mathbf{s} - \mathbf{s}_0)\,\delta(t - 0) \quad (3)$$

In contrast to eq 1, if we expand $F(\mathbf{s}, \mathbf{s}_0, t)$ in spherical harmonics, its components do not couple with each other. Therefore, it is easy to obtain the exact solution of eq 3:[7]

$$F(\mathbf{s}, \mathbf{s}_0, t) = \exp(-\mu_a t)\sum_l \frac{2l+1}{4\pi}\exp(-g_l t)P_l(\mathbf{s}\cdot\mathbf{s}_0) = \exp(-\mu_a t)\sum_l \frac{2l+1}{4\pi}\exp(-g_l t)\sum_m Y_{lm}(\mathbf{s})Y^*_{lm}(\mathbf{s}_0) \quad (4)$$

where $g_l = \mu_s[1 - a_l/(2l + 1)]$. Two special values of g_l are $g_0 = 0$, which follows from the normalization of $P(\mathbf{s}, \mathbf{s}')$ and $g_1 = v/l_t$, where l_t is the transport mean free path, defined by $l_t = v/[\mu_s(1 - \overline{\cos\theta})]$, where $\overline{\cos\theta}$ is the average of $\mathbf{s}\cdot\mathbf{s}'$ with $P(\mathbf{s}, \mathbf{s}')$ as weight. In eq 4, $Y_{lm}(\mathbf{s})$ are spherical harmonics. Equation 4 serves as the exact Green's function of particle propagation in velocity space. Since in an infinite uniform medium this function is independent of the source position, $\mathbf{r}_0$, requirements for a Green's function are satisfied, especially, a Chapman–Kolmogorov condition is obeyed: $\int d\mathbf{s}' F(\mathbf{s}'', \mathbf{s}', t - t')\, F(\mathbf{s}', \mathbf{s}, t' - t_0) = F(\mathbf{s}'', \mathbf{s}, t - t_0)$. In fact, in an infinite uniform medium, this propagator determines all particle migration behavior, including its spatial distribution, because displacement is an integration of velocity over time. The distribution function $I(\mathbf{r}, \mathbf{s}, t)$ (the source is located at $\mathbf{r}_0 = 0$) is given by

$$I(\mathbf{r}, \mathbf{s}, t) = \langle \delta(\mathbf{r} - v\int_0^t \mathbf{s}(t')dt')\; \delta(\mathbf{s}(t) - \mathbf{s})\rangle \quad (5)$$

where $\langle ...\rangle$ means the ensemble average in the velocity space. The first δ function imposes that the displacement, $\mathbf{r} - 0$, is given by the path integral. The second δ function assures the correct final value of direction. Equation 5 is an exact formal solution of eq 1, but can not be evaluated directly. We make a Fourier transform for the first δ-function in eq 5, then make a cumulant expansion,[8] and obtain

$$I(\mathbf{r}, \mathbf{s}, t) = F(\mathbf{s}, \mathbf{s}_0, t)\frac{1}{(2\pi)^3}\int d\mathbf{k}\, \exp\left\{i\mathbf{k}\cdot\mathbf{r} + \sum_{n=1}^{\infty}\frac{(-iv)^n}{n!}\sum_{j_n}\cdots\sum_{j_1} k_{j_n}\ldots k_{j_1}\langle \int_0^t dt_n \ldots \int_0^t dt_1\, T[s_{j_n}(t_n)\ldots s_{j_1}(t_1)]\rangle_c\right\} \quad (6)$$

where T denotes time-ordered multiplication.[9] In eq 6, index c denotes cumulant, which is defined in many statistics textbooks,[10] as $\langle A\rangle_c = \langle A\rangle$, $\langle A^2\rangle_c = \langle A^2\rangle - \langle A\rangle\langle A\rangle$, and a general expression relating $\langle A^m\rangle$ and $\langle A^m\rangle_c$, which is given by:

$$\langle A^m\rangle = m! \sum_{m_1, m_2, \ldots} \frac{1}{m_1!}\left(\frac{\langle A\rangle}{1!}\right)^{m_1} \frac{1}{m_2!}\left(\frac{\langle A^2\rangle_c}{2!}\right)^{m_2} \cdots \frac{1}{m_n!}\left(\frac{\langle A^n\rangle_c}{n!}\right)^{m_n} \times \delta(m - m_1 - 2m_2 - \ldots - nm_n - \ldots) \quad (7)$$

Hence, if $\langle A^m\rangle$ $m = 1, 2, \ldots n$ have been calculated, $\langle A^m\rangle_c$ $m = 1, 2, \ldots n$ can be recursively obtained and conversely.[10] In the following, we derive the analytical expression for the ensemble average $\langle \int_0^t dt_n \ldots \int_0^t dt_1\, T[s_{j_n}(t_n) \ldots s_{j_1}(t_1)]\rangle$. Using a standard time-dependent Green's function approach, it is given by

$$\langle \int_0^t dt_n \ldots \int_0^t dt_1\, T[s_{j_n}(t_n)\ldots s_{j_1}(t_1)]\rangle = \frac{1}{F(\mathbf{s}, \mathbf{s}_0, t)}\{\int_0^t dt_n \int_0^{t_n} dt_{n-1} \ldots \int_0^{t_2} dt_1 \int d\mathbf{s}^{(n)} \int d\mathbf{s}^{(n-1)} \ldots \int d\mathbf{s}^{(1)} F(\mathbf{s}, \mathbf{s}^{(n)}, t - t_n)s^{(n)}_{j_n} F(\mathbf{s}^{(n)}, \mathbf{s}^{(n-1)}, t_n - t_{n-1})s^{(n-1)}_{j_n-1} \ldots F(\mathbf{s}^{(2)}, \mathbf{s}^{(1)}, t_2 - t_1)s^{(1)}_{j_1} F(\mathbf{s}^{(1)}, \mathbf{s}_0, t_1 - 0) + \text{perm}\} \quad (8)$$

where the word "perm" means all $n! - 1$ terms obtained by permutation of $\{j_i\}$, $i = 1, \ldots n$, from the first term. In eq 8, $F(\mathbf{s}^{(i)}, \mathbf{s}^{(i-1)}, t_i - t_{i-1})$ is given by eq 4. Since eq 4 is exact, eq 8 provides the exact nth moments of the distribution. In Cartesian coordinates three components of $\mathbf{s}$ are $[s_x, s_y, s_z]$. For convenience in calculation, however, we will use the components of $\mathbf{s}$ on the basis of spherical harmonics:

$$\mathbf{s} = [s_1, s_0, s_{-1}] \equiv [Y_{11}(\mathbf{s}), Y_{10}(\mathbf{s}), Y_{1-1}(\mathbf{s})] = [-2^{-1/2}\sin\theta\, e^{+i\phi}, \cos\theta, +2^{-1/2}\sin\theta\, e^{-i\phi}]$$

The recurrence relation of the spherical harmonics is given by

$$Y_{lm}(\mathbf{s})Y_{1j}(\mathbf{s}) = \sum_i Y_{l+i\, m+j}(\mathbf{s})\langle l, 1, m, j|l + i, m + j\rangle \times \langle l, 1, 0, 0|l + i, 0\rangle,\ i = \pm 1 \quad (9)$$

where $\langle l_1, l_2, m_1, m_2|l, m\rangle$ is the Clebsch–Gordan coefficients of angular momentum theory,[11] which are

$$\langle l-i,1,m,j|l,m+j\rangle = \begin{bmatrix} \left[\frac{(l-m)(l-m+1)}{(2l-1)2l}\right]^{1/2} & \left[\frac{(l+m)(l-m+1)}{2l(l+1)}\right]^{1/2} & \left[\frac{(l+m)(l+m+1)}{(2l+2)(2l+3)}\right]^{1/2} \\ \left[\frac{(l-m)(l+m)}{(2l-1)l}\right]^{1/2} & \left[\frac{m^2}{l(l+1)}\right]^{1/2} & -\left[\frac{(l+m+1)(l-m+1)}{(l+1)(2l+3)}\right]^{1/2} \\ \left[\frac{(l+m)(l+m+1)}{(2l-1)2l}\right]^{1/2} & -\left[\frac{(l-m)(l+m+1)}{2l(l+1)}\right]^{1/2} & \left[\frac{(l-m)(l-m+1)}{(2l+2)(2l+3)}\right]^{1/2} \end{bmatrix}$$

with the row index (from above) $j = -1, 0, 1$ and the column index (from left) $i = 1, 0, -1$. The orthogonality relation of spherical harmonics is given by

$$\int \mathrm{d}\mathbf{s} Y^*_{l'm'}(\mathbf{s}) Y_{lm}(\mathbf{s}) = \frac{4\pi}{2l+1}\delta_{l,l'}\delta_{m,m'} \quad (10)$$

Using eqs 9 and 10, integrals over $\mathrm{d}\mathbf{s}^{(n)}$ ... $\mathrm{d}\mathbf{s}^{(1)}$ in eq 8 can be analytically performed. We obtain, when $\mathbf{s}_0$ is set along z, that

$$\langle \int_0^t \mathrm{d}t_n \cdots \int_0^t \mathrm{d}t_1 T[s_{j_n}(t_n)\ldots s_{j_1}(t_1)]\rangle = \frac{1}{F(\mathbf{s}, \mathbf{s}_0, t)}\left\{\sum_l Y_{[l\,\sum_{m=1}^n j_m]}(\mathbf{s}) \sum_{i_n}\cdots\sum_{i_1} \frac{2\left(l - \sum_{m=1}^{n} i_m\right)+1}{4\pi} \times D^l_{i_n\ldots i_1}(t) \prod_{k=1}^{n} \langle l - \sum_{m=1}^{n-k+1} i_{n-m+1}, 1, \sum_{m=1}^{k-1} j_m, j_k | l - \sum_{m=1}^{n-k} i_{n-m+1}, \sum_{m=1}^{k} j_m\rangle \langle l - \sum_{m=1}^{n-k+1} i_{n-m+1}, 1, 0, 0 | l - \sum_{m=1}^{n-k} i_{n-m+1}, 0\rangle\right\} + \text{perm} \quad (11)$$

where $i_f = \pm 1, f = 1, 2, \ldots n$, and

$$D^l_{i_n\ldots i_1}(t) = \exp(-\mu_a t)\{\int_0^t \mathrm{d}t_n \int_0^{t_n} \mathrm{d}t_{n-1}\cdots\int_0^{t_2}\mathrm{d}t_1 \exp[-g_l(t-t_n)] \exp[-g_{l-i_n}(t_n - t_{n-1})]\ldots\exp[-g_{l-\sum_{k=1}^{n} i_{n-k+1}}(t_1 - 0)]\} \quad (12)$$

Note that all ensemble averages have been performed. Equation 12 involves integrals of exponential functions, which can be analytically performed. An explicit expression for evaluating integrals in eq 12 is presented in the Appendix. Equation 12 includes all related scattering and absorption parameters, g_l, $l = 0, 1, \ldots$ and μ_a, and determines the time evolution dynamics. The final particle direction, $\mathbf{s}$, appears as argument of the spherical harmonics $Y_{lm}(\mathbf{s})$ in eq 11. Substituting eq 12 into eq 11, and using a standard cumulant procedure, the cumulants as functions of angle $\mathbf{s}$ and time t up to an arbitrary nth order can be analytically calculated. The final position, $\mathbf{r}$, appears in eq 6, and its component can be expressed as $|\mathbf{r}|\ Y_{1j}(\hat{\mathbf{r}})$, $j = 1, 0, -1$, with $|\mathbf{r}|$ and $\hat{\mathbf{r}}$ are, separately, magnitude and unit direction vector of $\mathbf{r}$. Then, performing a numerical three-dimensional inverse Fourier transform over $\mathbf{k}$, an approximate distribution function, $I(\mathbf{r}, \mathbf{s}, t)$, accurate up to nth cumulant, is obtained.

III. Gaussian Approximation of the Distribution Function

By a cut-off at the second cumulant, the integral over $\mathbf{k}$ in eq 6 can be analytically performed, which directly leads to a Gaussian spatial distribution displayed in eq 13. The exact first cumulant provides the correct center position of the distribution. The exact second cumulant provides the correct half-width of spread of the distribution. Moreover, the central limit theorem claims that as the number of collision events become large enough, the resulting Gaussian distribution approaches detailed accuracy beyond first two exact cumulants. At early time, spread of the spatial distribution is narrow, possibly narrower than the available detection instruments, hence, a spatial distribution with exact first and second cumulants may provide an accurate enough description of particle distribution for many applications.

For the reader's convenience, the expressions below are given in Cartesian coordinates with indices α, $\beta = [x, y, z]$. These expression is obtained by use of an unitary transform $s_\alpha = U_{\alpha j} s_j$ $j = 1, 0, -1$ from eq 11, (up to second order) which is based on $s_j = Y_{1j}(\mathbf{s})$, with

$$U = \begin{bmatrix} -2^{-1/2} & 0 & 2^{-1/2} \\ 2^{-1/2} i & 0 & 2^{-1/2} i \\ 0 & 1 & 0 \end{bmatrix}$$

We set $\mathbf{s}_0$ along the z direction and denote $\mathbf{s}$ as (θ, ϕ). Our cumulant approximation to the particle distribution function is given by

$$I(\mathbf{r}, \mathbf{s}, t) = \frac{F(\mathbf{s}, \mathbf{s}_0, t)}{(4\pi)^{3/2}} \frac{1}{(\det B)^{1/2}} \exp\left[-\frac{1}{4}(B^{-1})_{\alpha\beta}(r - r^c)_\alpha (r - r^c)_\beta\right] \quad (13)$$

with the center of the packet (the first cumulant), denoted by $\mathbf{r}^c$, located at

$$r^c_z = G\sum_l A_l P_l(\cos\theta)[(l+1)f(g_l - g_{l+1}) + l f(g_l - g_{l-1})] \quad (14.1)$$

$$r^c_x = G\sum_l A_l P^{(1)}_l(\cos\theta)\cos\phi[f(g_l - g_{l-1}) - f(g_l - g_{l+1})] \quad (14.2)$$

where $G = v\exp(-\mu_a t)/F(\mathbf{s}, \mathbf{s}_0, t)$, $A_l = (1/4\pi)\exp(-g_l t)$, g_l is defined after eq 4, and

$$f(g) = [\exp(gt) - 1]/g \quad (15)$$

r^c_y is obtained by replacing $\cos\phi$ in eq 14.2 by $\sin\phi$. In eq 14, $P^{(m)}_l(\cos\theta)$ is the associated Legendre function.

The square of the average spread width (the second cumulant) is given by

$$B_{\alpha\beta} = vG\Delta_{\alpha\beta} - r^c_\alpha r^c_\beta/2 \quad (16)$$

where all the coefficients are functions of angle and time:

$$\Delta_{zz} = \sum_l A_l P_l(\cos\theta)\left[\frac{l(l-1)}{2l-1}E^{(1)}_l + \frac{(l+1)(l+2)}{2l+3}E^{(2)}_l + \frac{l^2}{2l-1}E^{(3)}_l + \frac{(l+1)^2}{2l+3}E^{(4)}_l\right] \quad (17.1)$$

$$\Delta_{xx,yy} = \sum_l \frac{1}{2} A_l P_l(\cos\theta)\left[-\frac{l(l-1)}{2l-1}E_l^{(1)} - \frac{(l+1)(l+2)}{2l+3}E_l^{(2)} + \frac{l(l-1)}{2l-1}E_l^{(3)} + \frac{(l+1)(l+2)}{2l+3}E_l^{(4)}\right] \pm \sum_l \frac{1}{2} A_l P_l^{(2)}(\cos\theta)\cos(2\phi)\left[\frac{1}{2l-1}E_l^{(1)} + \frac{1}{2l+3}E_l^{(2)} - \frac{1}{2l-1}E_l^{(3)} - \frac{1}{2l+3}E_l^{(4)}\right] \quad (17.2)$$

where (+) corresponds to Δ_{xx} and (−) corresponds to Δ_{yy}

$$\Delta_{xy} = \Delta_{yx} = \sum_l \frac{1}{2} A_l P_l^{(2)}(\cos\theta)\sin(2\phi)\left[\frac{1}{2l-1}E_l^{(1)} + \frac{1}{2l+3}E_l^{(2)} - \frac{1}{2l-1}E_l^{(3)} - \frac{1}{2l+3}E_l^{(4)}\right] \quad (17.3)$$

$$\Delta_{xz} = \Delta_{zx} = \sum_l \frac{1}{2} A_l P_l^{(1)}(\cos\theta)\cos\phi\left[\frac{2(l-1)}{2l-1}E_l^{(1)} - \frac{2(l+2)}{2l+3}E_l^{(2)} + \frac{1}{2l-1}E_l^{(3)} + \frac{1}{2l+3}E_l^{(4)}\right] \quad (17.4)$$

Δ_{yz} is obtained by replacing $\cos\phi$ in eq 17.4 by $\sin\phi$. In eq 17.1−17.4

$$E_l^{(1)} = [f(g_l - g_{l-2}) - f(g_l - g_{l-1})]/(g_{l-1} - g_{l-2}) \quad (18.1)$$

$$E_l^{(2)} = [f(g_l - g_{l+2}) - f(g_l - g_{l+1})]/(g_{l+1} - g_{l+2}) \quad (18.2)$$

$$E_l^{(3)} = [f(g_l - g_{l-1}) - t]/(g_l - g_{l-1}) \quad (18.3)$$

$$E_l^{(4)} = [f(g_l - g_{l+1}) - t]/(g_l - g_{l+1}) \quad (18.4)$$

A cumulant approximation for the particle density distribution is obtained from the exact expression $N(\mathbf{r}, t) = \langle\delta(\mathbf{r} - v\int_0^t \mathbf{s}(t')\,dt')\rangle$. Using $\int ds\, F(\mathbf{s}, \mathbf{s}', t) = \exp(-\mu_a t)$, we have a Gaussian shape

$$N(\mathbf{r}, t) = \frac{1}{(4\pi D_{zz}vt)^{1/2}}\frac{1}{4\pi D_{xx}vt}\exp\left[-\frac{(z-R_z)^2}{4D_{zz}vt}\right] \times \exp\left[-\frac{(x^2+y^2)}{4D_{xx}vt}\right]\exp(-\mu_a t) \quad (19)$$

with a moving center located at

$$R_z = v[1 - \exp(-g_1 t)]/g_1 \quad (20)$$

and the corresponding diffusion coefficients are given by

$$D_{zz} = \frac{v}{3t}\left\{\frac{t}{g_1} - \frac{3g_1 - g_2}{g_1^2(g_1 - g_2)}[1 - \exp(-g_1 t)] + \frac{2}{g_2(g_1 - g_2)}[1 - \exp(-g_2 t)] - \frac{3}{2g_1^2}[1 - \exp(-g_1 t)]^2\right\} \quad (21.1)$$

$$D_{xx} = D_{yy} = \frac{v}{3t}\left\{\frac{t}{g_1} + \frac{g_2}{g_1^2(g_1 - g_2)}[1 - \exp(-g_1 t)] - \frac{1}{g_2(g_1 - g_2)}[1 - \exp(-g_2 t)]\right\} \quad (21.2)$$

In contrast to eqs 14 and 17, these results are independent of g_l for $l > 2$. Figure 1 shows the moving center of particles, R_z (eq 20), and the diffusion coefficients, D_{zz} and D_{xx} (eqs 21), as a function of time, where g_l is calculated by Mie theory of light scattering[12] assuming (for this figure) that the "uniform" scattering medium consists of water droplets with $r/\lambda = 1$ are uniformly distributed in air, with r the radius of the droplet, λ the wavelength of light, and the index of refraction $m = 1.33$.

Each distribution in eqs 13 and 19 describes a particle "cloud" anisotropically spreading from a moving center, with time-dependent diffusion coefficients. At early time $t \to 0$, $f(g) \approx t + O(t^2)$ in eq 15, and $E_l^{(j)} \approx t^2/2 + O(t^3)$ for $j = 1, 2, 3, 4$ in eqs 18. From eqs 14, 17, and 20−21, we see that for the density distribution, $N(\mathbf{r}, t)$, and the dominant distribution function, that is $I(\mathbf{r}, \mathbf{s}, t)$ along $\mathbf{s} = \mathbf{s}_0$, the center moves as $vt\,\mathbf{s}_0$ and the $B_{\alpha\beta}$ in eq 16 are proportional to t^3 at $t \to 0$. A distribution function $I(\mathbf{r}, \mathbf{s}, t)$ for $\mathbf{s}$ not close to $\mathbf{s}_0$ is small since $F(\mathbf{s}, \mathbf{s}_0, t) \sim t$, for small t. The center moves at a certain direction with displacement proportional to vt, and the $B_{\alpha\beta}$ in eq 16 are proportional to t^2 at $t \to 0$. These results present a clear picture of nearly ballistic motion at $t \to 0$. With increase of time, the motion of the center slows down, and the diffusion coefficients increase from zero. This stage of particle migration is often called a "snake-like mode".

With further increase in time, the lth Legendre component in eqs 4, 14, and 17, exponentially decays with a rate related to g_l. The detailed decay rate, g_l, is determined by the shape of the phase function. Generally speaking, the very high lth components decay in a rate of order of μ_s, as long as its Legendre coefficient a_l distinctly smaller than $2l + 1$. Even in the case that the phase function has a very sharp forward peak, in which there are non-zero a_l for very high lth rank, the a_l are, usually, much smaller than $2l + 1$. Therefore, for the distribution function at time t after the ballistic stage is over, a truncation in summation of l is available.

At large times, the distribution function tends to become isotropic. From eqs 19−21, the particle density, at $t \gg l_t/v$ and $r \gg l_t$, tends towards the conventional diffusion solution with the diffusive coefficient $l_t/3$. Therefore, our solution quantitatively describes how particles migrate from nearly ballistic motion to diffusive motion.

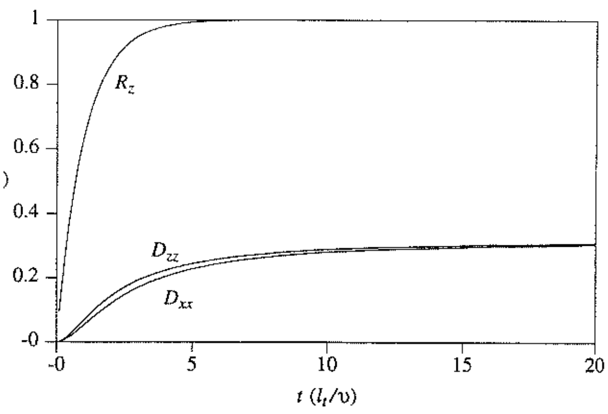

Figure 1. shows the moving center of a particle's density function, R_z (eq 20), and the diffusion coefficients, D_{zz} and D_{xx} (eqs 21), as functions of time, t.

IV. Discussion

The cumulant expansion terminating at the second order is a standard method in statistical mechanics,[10] which neglects cumulants higher than second order, and leads to a Gaussian distribution. If we examine the spatial displacement after each collision event as an independent random variable, $\Delta\mathbf{r}_i$, the total displacement is $\sum\Delta\mathbf{r}_i$ ($i = 1,...N$), with N the number of collision events, which can be estimated by t/μ_s. If we define $\mathbf{Y} = (N)^{-1/2}\sum\Delta\mathbf{r}_i$, the central limit theorem claims that if N is a large number, then $\langle\mathbf{Y}^n\rangle_c/\langle\mathbf{Y}^2\rangle_c \sim N^{1-n/2}$, $n \geq 3$. Therefore, the sum of N variables will have an essentially Gaussian distribution. Therefore, after enough collision events happened, the distributions we have calculated are accurate in detail, not just having the correct center and spread. At early time, the particle's spread is narrow, hence, in many applications the detailed shape is less important than the correct position and correct narrow width of the beam, because of the finite resolution of detection devices.

In case a more accurate distribution at early time is needed, by use of eqs 11 and 12 with its expression in Appendix, and a standard cumulant procedure, the exact higher (up to arbitrary nth) order cumulants can be analytically calculated. Then, performing a numerical three-dimensional Fourier transform, the particle distribution function accurate up to nth order cumulant approximation can be obtained.

In summary, we present an analytical solution of the elastic Boltzmann transport equation in an infinite uniform isotropic medium. Using a cumulant expansion we can analytically calculate cumulants up to an arbitrary high order. By terminating at the second order, we have derived an analytical solution of the distribution function, eq 13, and the density distribution, eq 19, with exact first cumulant (center of the distribution) and exact second cumulant (the half-width of spread of the distribution). These expressions show a clear picture of time evolution of particle migration from ballistic to snake-like, then to diffusion regime.

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Appendix

In this Appendix, we derive an analytical expression of eq 12 to nth order. By defining

$$b_m = g_{[l-\sum_{k=1}^{n-m} i_{n-k+1}]} - g_{[l-\sum_{k=1}^{n-m+1} i_{n-k+1}]} \quad m = 1,...n \quad \text{(A1)}$$

eq 12 can be written as

$$D^l_{i_n...i_1}(t) = \exp(-\mu_a t)\exp(-g_l t)F^{(n)}(t) \quad \text{(A2)}$$

with

$$F^{(n)}(t) = \int_0^t \mathrm{d}t_n e^{b_n t_n} \int_0^{t_n} \mathrm{d}t_{n-1} e^{b_{n-1}t_{n-1}} \ldots \int_0^{t_2} \mathrm{d}t_1 e^{b_1 t_1} \quad \text{(A3)}$$

It is easy to directly calculate eq A3 for low n orders:

$$F^{(1)}(t) = \frac{e^{b_1 t}}{b_1} - \frac{1}{b_1} \quad \text{(A4.1)}$$

$$F^{(2)}(t) = \frac{e^{(b_1+b_2)t}}{b_1(b_1+b_2)} - \frac{e^{b_2 t}}{b_1 b_2} + \frac{1}{(b_1+b_2)b_2} \quad \text{(A4.2)}$$

$$F^{(3)}(t) = \frac{e^{(b_1+b_2+b_3)t}}{b_1(b_1+b_2)(b_1+b_2+b_3)} - \frac{e^{(b_2+b_3)t}}{b_1 b_2(b_2+b_3)} + \frac{e^{b_3 t}}{(b_1+b_2)b_2 b_3} - \frac{1}{(b_1+b_2+b_3)(b_2+b_3)b_3} \quad \text{(A4.3)}$$

In each step of integration, the difficulty is in determining the constant term. In the following we prove that this term is given by $(-1)^n/[b_n(b_n + b_{n-1})...(b_n + b_{n-1} + ... + b_1)]$. Equation A3 can be written as

$$F^{(n)}(t) = \int_0^t \mathrm{d}t' e^{b_n t'} F^{n-1}(t') \quad \text{(A5)}$$

Using integration by parts to eq A5, we obtain

$$F^{(n)}(t) = \frac{1}{b_n}[e^{b_n t}F^{(n-1)}(t) - \int_0^t \mathrm{d}t'\, e^{(b_n+b_{n-1})t'}F^{(n-2)}(t')] \quad \text{(A6)}$$

Recursively applying eq A6, we obtain

$$F^{(n)}(t) = \frac{e^{b_n t}}{b_n}F^{(n-1)}(t) - \frac{e^{(b_n+b_{n-1})t}}{b_n(b_n+b_{n-1})}F^{(n-2)}(t) + ... + (-1)^k \frac{e^{(b_n+b_{n-1}+...+b_{n-k})t}}{b_n(b_n+b_{n-1})...(b_n+b_{n-1}+...+b_{n-k})}F^{n-k-1}(t) + ... + (-1)^{n-1}\frac{e^{(b_n+b_{n-1}+...+b_1)t} - 1}{b_n(b_n+b_{n-1})...(b_n+b_{n-1}+...+b_1)} \quad \text{(A7)}$$

Equation A7 provides formulas to recursively evaluate eq 12 up to nth order. Also, eq A7 produces the above mentioned constant term. An explicit expression of eq 12 can then be written as

$$D^l_{i_n...i_1}(t) = \exp(-\mu_a t)\exp(-g_l t)\sum_{m=0}^{n} \frac{(-1)^m \exp\left(\sum_{k=0}^{n-m} b_{n-k+1}t\right)}{\prod_{j=1}^{n} L_j^{(m)}} \quad \text{(A8)}$$

with $b_{n+1} \equiv 0$, and

$$L_j^{(m)} = \sum_{k=j}^{m} b_k,\ j \leq m \text{ or } L_j^{(m)} = \sum_{k=m+1}^{j} b_k,\ j > m \quad \text{(A9)}$$

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