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Motion of Charged Particles in a Homogeneous Reacting Medium with a One-dimensional Geometry

Russell K. Standish

Department of Theoretical Physics, Research School of Physical Sciences, Australian National University, G.P.O. Box 4, Canberra, A.C.T. 2601, Australia.

Abstract

We study the approach to steady state of a system described by the Klein-Kramers equation with a constant source, to see how the source structure effects disappear away from the source. We link this to some earlier work on non-hydrodynamic effects in time-of-flight swarm experiments to show how this theory may be applied to the analysis of steady-state Townsend swarm experiments.

1. Introduction

The evolution of a distribution function $f(c,z,t)$ of heavy particles reacting with a neutral homogeneous medium is well described by means of the Klein-Kramers equation with a uniform force term. For a one-dimensional geometry, this reads

$$(\partial_t + c\partial_z + \mathcal{M}) \equiv \left(\partial_t + c\partial_z + a\partial_c + \nu - \nu_1 - \nu_1 c\partial_c - \nu_2\partial_c^2\right) f(c,z,t) = S(c,z,t), \qquad (1)$$

where c is the velocity coordinate, z the position coordinate, a the acceleration due to an electric field, and S the source distribution. The reaction rate ν is positive when ionisation occurs, and negative when attachment occurs. The coefficients ν_1 and ν_2 are related to physical quantities by

$$\nu_1 = m\nu_c/M, \quad \nu_2 = k_B T\nu_c/M, \qquad (2)$$

where m is the charged particle mass, M the neutral mass, k_B the Boltzmann constant, T the neutral gas temperature and ν_c the collision frequency.

This equation has received much attention in recent years as an improvement in technique has allowed some progress on the problem of diffusing particles near an absorbing boundary, posed a long time ago by Wang and Uhlenbeck (1945) [see Selinger and Titulaer (1984) for a review]. The interest in this problem comes from modelling chemical reactions near catalysts, and coagulation of colloids. For a good introduction to the Klein-Kramers equation, and to Fokker-Planck equations in general, see Risken (1984).

In this work, we are interested in systems where the source term is constant, and in particular, though not necessarily restricting ourselves it, a source located as a delta function at the origin: $S(c,z,t) = S(c)\delta(z)$. The initial approach

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would be to construct the solutions from a linear combination of solutions that decay away from the origin along the lines of Selinger and Titulaer (1984). However, this approach clearly fails to model the situation of ionisation (see Fig. 3 below) where the particle density grows exponentially away from the origin with a well-defined growth constant. If we are to include solutions that grow away from the origin, then we are faced with the question of which solutions should be included, as there happen to be an infinite number of them forming an unbounded sequence of growth constants. The solution to this problem lies in analysing equation (1) dynamically with a source that is switched on at time $t = 0$. It will be found that some of the modes propagate in the direction of the electric field, and the others in the opposite direction.

2. Eigenfunctions

The operator $ick + \mathcal{M}$ admits a complete set of eigenfunctions,

$$(ick + \mathcal{M})\,\Psi_n(c, ik) = -\omega_n(ik)\Psi_n(c, ik), \tag{3}$$

and the adjoint similarly admits a complete set of eigenfunctions,

$$\left(ick + \tilde{\mathcal{M}}\right)\Phi_n(c, ik) = -\omega_n(ik)\Phi_n(c, ik). \tag{4}$$

The two sets of eigenfunctions may be normalised so as to satisfy the bi-orthonormality relationship

$$\int \Psi_m(c, ik)\Phi_n(c, ik)\mathrm{d}c = \delta_{mn}. \tag{5}$$

In the following, we use dimensionless units, in which $\nu_1 = 1$ and $\nu_2 = \frac{1}{2}$. In these units, the mean free time of the charged particle is m/M and the mean free path is $\frac{1}{2}(m/M)^3$. This model can be solved by finding a similarity transformation, and a variable substitution that transforms equation (3) into the Schrödinger equation for the harmonic oscillator problem (Standish 1987). The solutions to equation (3) and (4) are given by

$$\omega_n(ik) = \nu - n - aik + \tfrac{1}{2}(ik)^2, \tag{6}$$

$$\Psi_n(c, ik) = \pi^{-\frac{1}{2}}2^{-n}(n!)^{-1}\exp\left[-(c-a)^2/2 - (c-a+ik)^2/2\right]\mathrm{H}_n(c-a+ik), \tag{7}$$

$$\Phi_n(c, ik) = \exp\left[(c-a)^2/2 - (c-a+ik)^2/2\right]\mathrm{H}_n(c-a+ik), \tag{8}$$

where H_n is the nth order Hermite polynomial [Abramowitz and Stegun (1965), 22.2.24].

Firstly, let us consider solving the steady-state problem

$$(\partial_z + \mathcal{M})f(c, z) = 0; \qquad z \neq 0,$$

$$f(c, 0) = S(c). \tag{9}$$

From equation (3), equation (1) is solved by solutions of the form

$$\exp(q_n^{\pm} z)\Psi_n(c, q_n^{\pm}), \tag{10}$$

where the $q_n^{\pm}$ are the roots of $\omega_n(q_n^{\pm}) = 0$:

$$q_n^{\pm} = a \pm \left[a^2 + 2(n - \nu)\right]^{\frac{1}{2}}. \tag{11}$$

A theorem of Protopopescu (1987) tells us that $\Psi_n(c, q_n^{\pm})$ form a complete set in velocity space, so we can look for an expansion of the form

$$f(c,z) = \begin{cases} \displaystyle\sum_{n=0,\, r=\pm}^{\infty} a_n^r \exp(q_n^r z)\Psi_n(c, q_n^r), & z > 0 \\ \displaystyle\sum_{n=0,\, r=\pm}^{\infty} b_n^r \exp(q_n^r z)\Psi_n(c, q_n^r), & z < 0. \end{cases} \tag{12}$$

If the set of functions used to represent f for positive z is disjoint from the set used for negative z, then the completeness theorem assures us that the expansion is unique, which is required if the solution (12) is to be sensible. However, there are an infinite number of ways in which the functions $\Psi_n(c, q_n^{\pm})$ can be divided between the two regions of z. To get some insight into the problem, the number density was computed numerically for the Klein-Kramers model.

Numerical Studies of the Klein-Kramers Model

We numerically computed the number density $\int_{-\infty}^{\infty} f(c,z)\mathrm{d}c$ as a function of z, and the reaction rate ν. In these computations charged particles are injected at a constant rate into the drift region with velocity equal to the drift velocity a, which is set to unity. The computations were carried out using an exact form of the Green function. This can be calculated from equations (6) to (8) by means of the spectral representation

$$G(c,z,t;c',z',t') = \Theta(t-t') \int_{-\infty}^{\infty} \sum_{n=0}^{\infty} \Psi_n(c, ik)\Phi_n(c', ik) \exp[\omega_n(ik)t + ik(z - z')]\mathrm{d}k, \tag{13}$$

where $\Theta(t > 0) = 1, \Theta(t < 0) = 0$ is the Heaviside step function. The sum over n may be performed by using a generating function for Hermite polynomials [Erdélyi *et al.* (1954), 10.13.22] upon which the integration over k becomes a standard Gaussian integral.

The phase space distribution may be found by integrating the Green function over source times $0 \le t' \le t$:

$$\begin{aligned} f(c,z,t) &= \int_0^t \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} G(c,z,t;c',z',t')S(c',z',t')\mathrm{d}c'\mathrm{d}z'\mathrm{d}t' \\ &= \int_0^t G(c,z,t;a,0,t')\mathrm{d}t'. \end{aligned} \tag{14}$$

Firstly, the integration over c to find the number density was performed analytically, and then the time integration was performed numerically using an adaptive integrator.

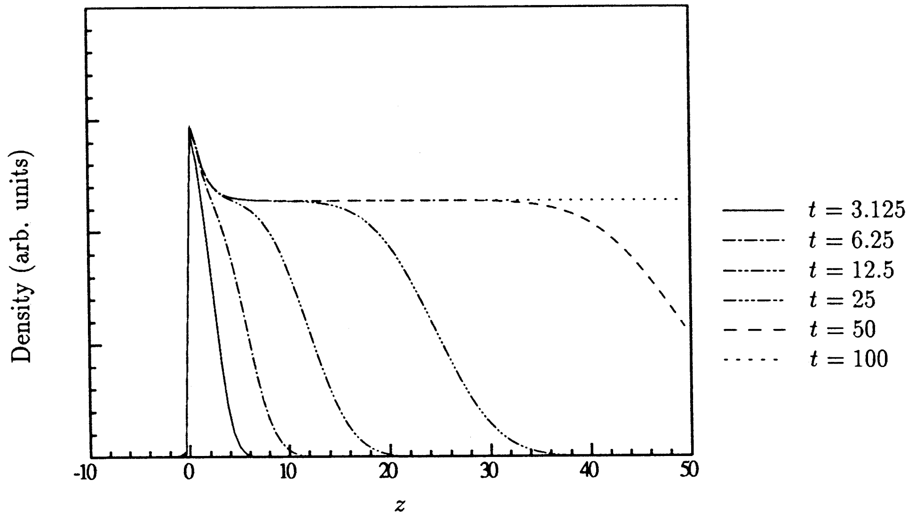

Fig. 1. Density for $\nu = 0$. In Figs 1 to 4 the curves shown are at successively greater times after the source is switched on.

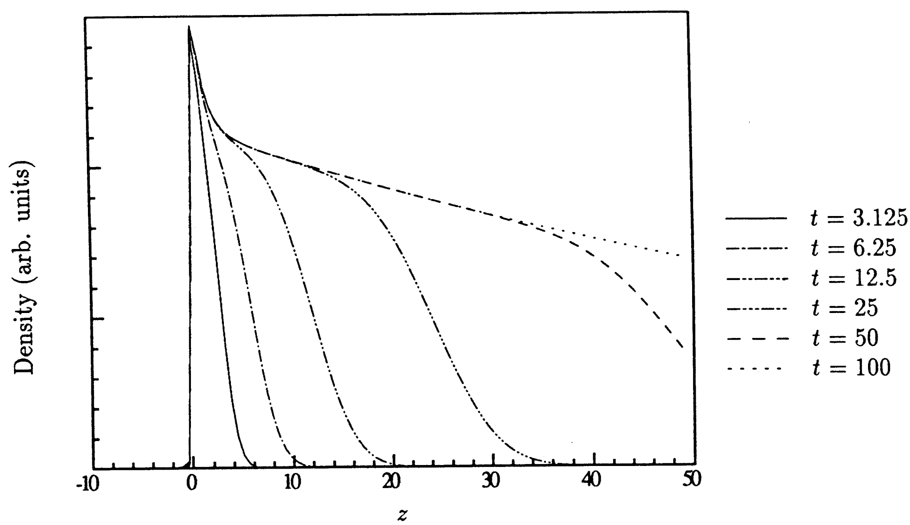

Fig. 2. Density for $\nu = -0.01$.

Figs 1 to 4 show the effect of varying the reaction rate ν. In the cases where $\nu < a^2/2$, the distribution builds up to a steady-state distribution. Non-hydrodynamic effects manifest themselves in a neighbourhood of size $(q_0^- - q_1^-)^{-1}$ (= 1.374, 1.366 and 0.995 for $\nu = -0.01$, 0 and 0.01) around the origin. Outside this region, the non-hydrodynamic modes ($n \neq 0$) are damped exponentially with respect to the hydrodynamic mode ($n = 0$), and it is here that we see exponential behaviour governed by the Townsend ionisation coefficient. In Figs 5 and 6, the steady-state distribution is plotted on a logarithmic plot. It can be seen that the density behaves exponentially in z far from the source. The slopes at

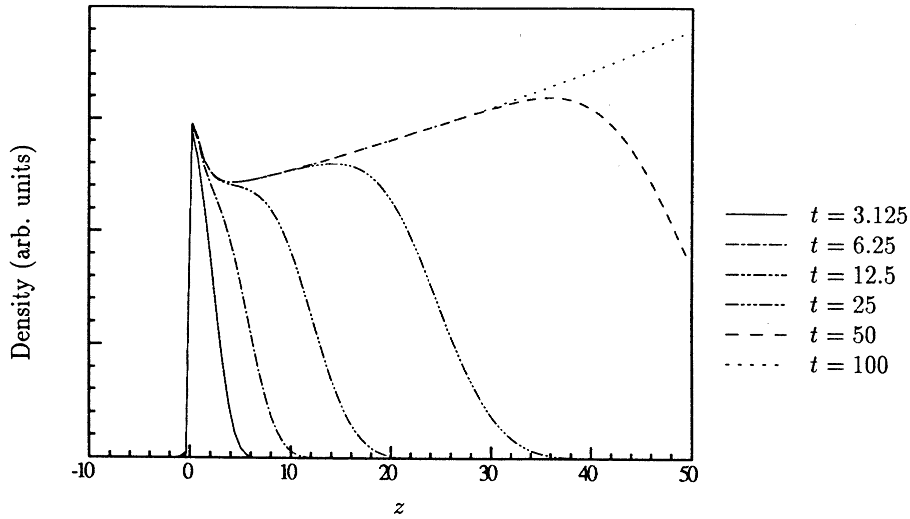

Fig. 3. Density for $\nu = 0.01$.

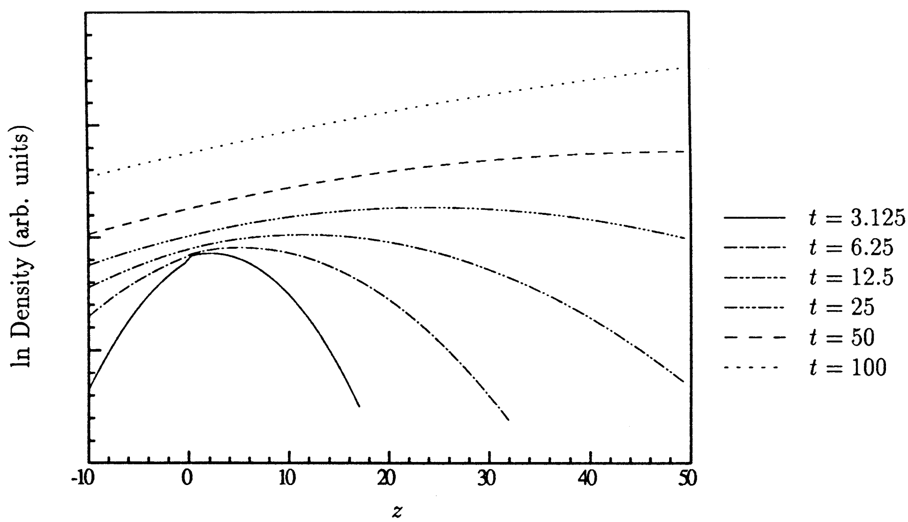

Fig. 4. Logarithmic plot of the density for $\nu = 1$.

either extremity give q_0^- downstream of the source, and q_0^+ upstream. This is evidence that the positive branch roots control the spatial decay of particles diffusing against the electric field, and the negative branch roots control the spatial decay (or growth) of those diffusing with the field.

In Fig. 4, the reaction rate has been increased to larger than $a^2/2$. In this case, no steady state is seen to occur. Rather, the density of charged particles increases exponentially with time. Physically, this can be understood as the electric field not being strong enough to remove at a sufficiently rapid rate the charged particles created by ionisation. This effect will be seen to arise out of the analysis in the next section.

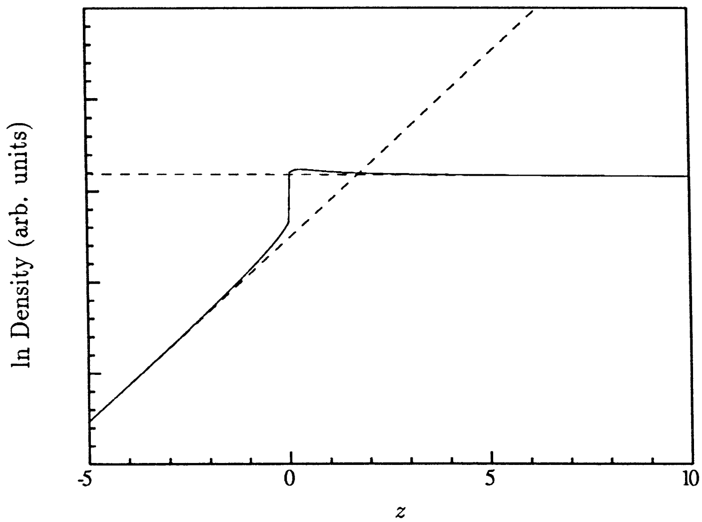

Fig. 5. Logarithmic plot of the steady-state density for the case $\nu = -0.01$. Lines fitted to the tails of the distribution have slopes corresponding to $q_0^+ = 2.01$ and $q_0^- = -0.01$ respectively.

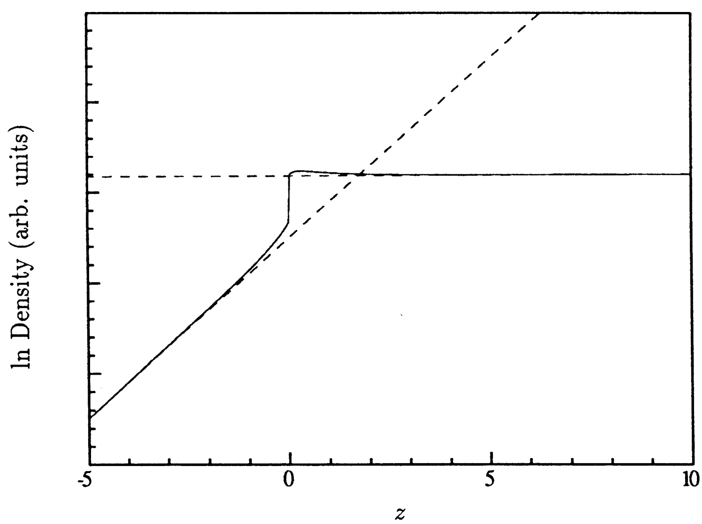

Fig. 6. Logarithmic plot of the steady-state density for the case $\nu = 0.01$. Lines fitted to the tails of the distribution have slopes corresponding to $q_0^+ = 1.98$ and $q_0^- = 0.01$ respectively.

3. Asymptotic Behaviour for Large Times and Distances

In the previous section, we examined a model in which there are an infinite number of roots q_n^r of either sign. The numerical work indicates that the root q_0^- controls the asymptotic exponential behaviour downstream from the source, and that q_0^+ controls the behaviour upstream. In this section, we discuss the time dependence analytically, and show how the steady-state solution is established. It will be seen that the positive and negative branches of the roots control the swarm behaviour upstream and downstream of the source respectively.

We start with the one-dimensional time dependent Klein-Kramers equation for a steady source $S(c,z)$ switched on at $t=0$:

$$(\partial_t + c_z\partial_z + \mathcal{M}) f(c,z,t) = S(c,z)\Theta(t). \tag{15}$$

This may be formally solved by means of the eigenfunctions (3) and (4),

$$f(c,z,t) = \sum_n f_n(c,z,t) \equiv \sum_n \int_{-\infty}^{\infty} \frac{s_n(c,ik)}{\omega_n(ik)} \exp(ikz)\{1 - \exp(\omega_n(ik)t)\}\,\mathrm{d}k, \tag{16}$$

where

$$s_n(c,ik) = \frac{1}{2\pi}\Psi(c,ik)\int\int \exp(-ikz')\Phi_n(c',ik)S(c',z')\mathrm{d}c'\mathrm{d}z'. \tag{17}$$

We assume that the source has been chosen in such a way that the integral over k in (16) is well defined. For example, we may choose a Gaussian source located at $z=0$:

$$S(c,z) = \exp(-\sigma c^2)\delta(z). \tag{18}$$

Upon substituting (7), (8) and (18) into (17), we find that

$$s_n(c,ik) = \frac{(1-1/\sigma)^{\frac{1}{2}n}}{\sigma^{\frac{1}{2}} 2^n n!} \exp\left[(1-1/4\sigma)k^2 - (c-2a)ik - (c-a)^2\right]$$
$$\times \mathrm{H}_n\left(\frac{ik(1-1/2\sigma)-a}{(1-1/\sigma)^{\frac{1}{2}}}\right)\mathrm{H}_n(c-a+ik). \tag{19}$$

To get s_n to vanish fast enough as $k \to \pm\infty$ for (16) to be convergent, we must choose $\sigma < \frac{1}{4}$.

Since s_n and ω_n are analytic functions of k, the integrand in (16) is analytic. Also, $s_n(c,ik) \to 0$ as $\mathrm{Re}(k) \to \pm\infty$, and so the contour of integration in (16) may be translated by an arbitrary amount. In particular, we may move the contour so that it passes through the saddle point $-iQ_n$ of $\omega_n(ik)$, which is $-ai$. We may then use the method of steepest descent (Jeffreys 1961) to evaluate the time dependent portion of the integral at large times:

$$f_n(c,z,t) \sim \int_{-\infty+Q_n i}^{\infty+Q_n i} \frac{s_n(c,ik)\exp(ikz)}{\omega_n(ik)}\mathrm{d}k$$
$$- \exp\left(\omega_n(Q_n)t\right)\left(\frac{2\pi}{t}\right)^{\frac{1}{2}} \frac{s_n(c,Q_n)\exp(Q_n z)}{\omega_n(Q_n)\left[-\omega_n''(Q_n)\right]}. \tag{20}$$

The behaviour of f in time will depend critically upon the sign of $\omega_n(Q_n)$. If $\omega_n(Q_n)$ is positive for any n, then the time dependent part will grow exponentially, and the system will not approach a steady state. On the other hand, if $\omega_n(Q_n)$ is negative for all n, then the time dependent term is

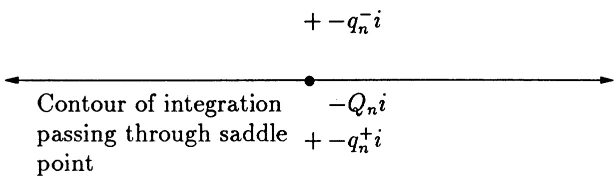

Fig. 7. Singularity structure of $s_n(c, ik)/\omega_n(ik)$ for the Klein-Kramers model. There are two simple poles at $-q_n^{\pm} i$, and a saddle point at $-Q_n i = -(q_n^+ + q_n^-)i$. The fact that the contour of integration must lie between the poles determines that the negative branch controls downstream behaviour, and that the positive branch determines upstream behaviour.

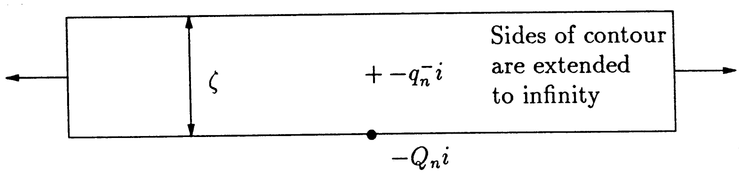

Fig. 8. Contour used for the large z asymptotic argument.

exponentially damped, and a steady state is reached. Now, we get

$$\omega_n(Q_n) = \nu - n - a^2/2. \tag{21}$$

If $\nu > a^2/2$, then there is no steady state approached (Fig. 4), otherwise the system does approach a steady state (Figs 1 to 3).

Let us consider a system satisfying $\omega_n(Q_n) < 0$ for all n. The steady-state term is given by the integral in (20). Since s_n and ω_n are analytic, the only singularities of the integrand occur at the zeros of ω_n. There are only two singularities as shown in Fig. 7. The contour of integration must lie between the poles q_n^+ and q_n^- for the time dependent term to approach zero according to (20). As we shall see, this leads to the term proportional to $\exp(q_n^+ z)$ not contributing to the distribution at positive z and similarly the $\exp(q_n^- z)$ term not contributing to the distribution at negative z.

Since s_n grows much faster than any exponential as $k \to \pm i\infty$, it is not possible to evaluate the integral in (20) by completing the contour around the positive imaginary half-plane for positive z, and around the negative half-plane for negative z. Instead, we must use a large asymptotic argument that is similar to the method described in section 2.6 of Jeffreys (1961). In this, we complete the contour in the fashion shown in Fig. 8, with ζ an arbitrarily large positive but finite value. We may now apply Cauchy's residue theorem to obtain

$$\begin{aligned} f_n(c, z, \infty) &= \int_{-\infty+Q_n i}^{\infty+Q_n i} \frac{s_n(c, ik)\exp(ikz)}{\omega_n(ik)}\,\mathrm{d}k \\ &= 2\pi i \exp(q_n^- z)\,\mathrm{Res}\left(\frac{s_n(c, ik)}{\omega_n(ik)}, iq_n^-\right) + \int_{-\infty+(Q_n+\zeta)i}^{\infty+(Q_n+\zeta)i} \frac{s_n(c, ik)\exp(ikz)}{\omega_n(ik)}\,\mathrm{d}k. \end{aligned}$$

But the absolute value of the second term is

$$\left|\int_{-\infty+(Q_n+\zeta)i}^{\infty+(Q_n+\zeta)i} \frac{s_n(c,ik)\exp(ikz)}{\omega_n(ik)}\mathrm{d}k\right| \leq \exp(-[\zeta+Q_n]z)\int_{-\infty+(Q_n+\zeta)i}^{\infty+(Q_n+\zeta)i}\left|\frac{s_n(c,ik)}{\omega_n(ik)}\right|\mathrm{d}k.$$

Since ζ may be chosen arbitrarily large, the second term must vanish faster than any exponential as a function of z, and so

$$f_n(c,z,\infty) \sim 2\pi i \exp(q_n^- z)\mathrm{Res}\left(\frac{s_n(c,ik)}{\omega_n(ik)}, iq_n^-\right) \quad \text{as} \quad z \to \infty. \tag{22}$$

By taking ζ negative, one can similarly show that

$$f_n(c,z,\infty) \sim 2\pi i \exp(q_n^+ z)\mathrm{Res}\left(\frac{s_n(c,ik)}{\omega_n(ik)}, iq_n^+\right) \quad \text{as} \quad z \to -\infty. \tag{23}$$

In general, we may state the selection principle thus: the contour passing through the saddle point of $\mathrm{Re}[\omega_n(ik)]$ divides the complex plane; those roots of ω_n that lie above this contour contribute to the asymptotic behaviour of f_n downstream of the source, and those that lie below contribute to the asymptotic behaviour upstream of the source.

4. Discussion

In this paper, a theory is developed relating the asymptotic properties of the steady-state solution of the Klein-Kramers equation to the distribution of zeros of the eigenvalues of the inhomogeneous operator $ick+\mathcal{M}$. It was found that the zeros of the lowest eigenvalue give the growth constant far away from the source, whereas the other modes are clustered around the source and decay exponentially way from it. In the non-hydrodynamic theory of time-of-flight swarm experiments (Kumar 1981; Standish 1987) it is assumed that the general inhomogeneous Boltzmann operator $ick+\mathcal{M} \equiv ick + a\partial_c + J$, where J is the collision operator, satisfies the same eigenvalue equations (3) to (5) as does the Klein-Kramers operator. If we further assume that the appropriate functions are analytic, then the theory given in this paper can be used directly to analyse steady-state swarm experiments of a one-dimensional nature such as the steady-state Townsend experiment. At the expense of additional complication, the assumptions on the spectrum of $ick+\mathcal{M}$ and the analyticity assumptions may be relaxed, introducing additional terms in the solution that are generally not exponential in z, but are bounded by an exponential of z, and so will not contribute to the behaviour at large z.

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