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Magnetic Effects on the Diffusion of Ferromagnetic Colloidal Particles Dispersed in a Current-Carrying Fluid Medium

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A theoretical investigation is made of the magnetic effects on the diffusion of ferromagnetic colloidal particles dispersed in a current-carrying fluid medium. Specifically, the following problem is analyzed: Consider an infinitely long circular cylindrical metallic-based ferrofluid confined at its cylindrical surface $r'=a$ by an impermeable wall. The initial distribution of dispersed ferromagnetic particles is cylindrically symmetric but otherwise arbitrary depending only on the radial distance from the axis. At time $t'=0$, a uniform electric current is made to flow through the ferrofluid in the axial direction. In the presence of the magnetic field associated with the current flow, an analytical solution for the subsequent spatial and temporal behavior of the particle distribution function was obtained and then shown to reduce to the corresponding classical diffusion problem in the absence of the magnetic force. The effect of the magnetic force on the relaxation process and on the equilibrium distribution itself was then examined numerically for the specific case where the initial probability distribution function is a delta function located at the axis.

I. INTRODUCTION

MAGNETICALLY responsive fluids called ferrofluids composed of ferromagnetic particles of approximately domain size homogeneously dispersed in a liquid carrier have recently been synthesized in the laboratory.[1] One particular class of ferrofluids consisting of colloidal dispersions of ferrite particles in organic-based liquids are made stable against particle agglomeration by the addition of a surfactant. The mechanism preventing agglomeration is here due to the short-range repulsive force arising from the compression of an absorbed layer of surfactant on the particle surface balancing the attractive London and magnetic forces.[2] A most attractive ferrofluid from the applicational viewpoint would consist of a colloidal dispersion of magnetic particles in a metallic-based fluid. While the synthesis of metallic-based ferrofluids stable against particle agglomeration has so far met with limited success, there is no reason in principle preventing their ultimate production. The discovery of the existence of stable ferrofluids has led to the development of the phenomenology called ferrohydrodynamics[3] defined as the fluid dynamic and heat transfer processes associated with the motion of incompressible magnetically polarizable fluids in the presence of magnetic-field and temperature gradients.

Our particular concern here is to investigate theoretically the magnetic effects on the process of diffusion of ferromagnetic colloidal particles dispersed in a metallic-based current-carrying fluid medium. Interest here lies in the important fact that the usual situation in which the diffusion process is driven exclusively by particle density gradients is no longer applicable, but that the equations describing the diffusion process must be modified to take into account the effect of the external magnetic force acting on the diffusing particle resulting from the interaction of the applied magnetic field and the particle's induced magnetic dipole moment.

We choose a particularly simple geometry which makes the analysis amenable to exact mathematical treatment. Specifically, we examine the following situation: Consider an infinitely long circular cylindrical metallic-based ferrofluid confined at its cylindrical surface $r'=a$ by an impermeable wall (cylindrical coordinates, r', θ', z'). The initial distribution of dispersed ferromagnetic particles is cylindrically symmetric, but otherwise arbitrary, depending only on the radial distance from the axis. At time $t'=0$ a uniform electric current is made to flow through the ferrofluid in the axial direction. In the presence of the magnetic field associated with the current flow, we wish to determine the subsequent spatial and temporal behavior of the particle distribution function with specific emphasis on its long-time behavior, i.e., its behavior near the equilibrium distribution.

II. MATHEMATICAL FORMULATION

A. External Force on Ferromagnetic Particle Partaking of the Conduction Process

The mks system of units is used throughout. We assume that the ferromagnetic particle is itself metallic so that it partakes of the electrical conduction process. The external force on a colloidal particle having a magnetic dipole moment $\mathbf{m}_0$ diffusing through a current-carrying conducting fluid medium and itself participating in the conduction process is

$$\mathbf{F}=-\int_{\substack{\text{particle}\\ \text{surface}}} p\hat{n}\, dS+\int_{\substack{\text{particle}\\ \text{volume}}} \mathbf{j}\times\mathbf{B}\, dv+\mu_0(\mathbf{m}_0\cdot\nabla)\mathbf{H}, \quad (1)$$

where the first term represents the integrated pressure force acting on the surface of the particle, the second term is the Lorentz force, and the last term is the

[1] R. E. Rosensweig, J. W. Nestor, and R. S. Timmins, Chemical Engineering Joint Meeting, London, 1965, Paper 5.14 A.I.Ch.E—I. (unpublished).
[2] E. L. Mackor, J. Colloid Sci. 6, 492 (1951).
[3] J. L. Neuringer and R. E. Rosensweig, Phys. Fluids 7, 1927 (1964).

magnetic force acting on the particle's induced magnetic dipole moment. Here μ_0 represents the free-space permeability.

Now the macroscopic momentum equation for the *mixture* when no gross motions are taking place is

$$-\nabla p+\mathbf{j}\times\mathbf{B}+\mu_0(\mathbf{M}\cdot\nabla)\mathbf{H}=0, \qquad (2)$$

where $\mathbf{M}$ is the magnetization per unit volume of mixture (i.e., dipole moment per unit volume of mixture). The last term of Eq. (2) can be written as $(\epsilon/v_p)\mu_0(\mathbf{m}_0\cdot\nabla)\mathbf{H}$, where ϵ is the volume loading fraction (i.e., the volume occupied by the ferromagnetic particles per unit volume of mixture) and v_p is the volume of a ferromagnetic particle.

Integrating Eq. (2) over the volume of a particle and using Gauss' theorem on the pressure term, we obtain

$$-\int_{\substack{\text{particle}\\ \text{surface}}} p\hat{n}dS+\int_{\substack{\text{particle}\\ \text{volume}}}\mathbf{j}\times\mathbf{B}\,dv+\epsilon\mu_0(\mathbf{m}_0\cdot\nabla)\mathbf{H}=0.$$

Substituting the above equation into Eq. (1), we obtain

$$\mathbf{F}=(1-\epsilon)\mu_0(\mathbf{m}_0\cdot\nabla)\mathbf{H} \qquad (3)$$

for the external force acting on the particle. When $\epsilon=1$, the force is zero, as we should expect, since we are now reduced to a one-component magnetic fluid continuum in which the induced pressure force on the particle surface is balanced by the sum of the Lorentz and magnetic-moment forces acting on the particle. Now the local loading fraction ϵ depends on the local particle concentration and so the diffusion process is essentially nonlinear (see next subsection). Limiting ourselves only to investigations of dilute ferrofluids, i.e., $\epsilon\ll1$, we neglect ϵ in comparison to unity in Eq. (3) resulting in the linearization of the equations governing the diffusion process.

If the induced dipole moment always points in the direction of the local magnetic field,[3] then Eq. (3) becomes (neglecting ϵ in comparison to unity)

$$\mathbf{F}=(\mu_0m_0/H)(\mathbf{H}\cdot\nabla)\mathbf{H}=(\mu_0m_0/H)[\tfrac{1}{2}\nabla H^2-\mathbf{H}\times\nabla\times\mathbf{H}],$$

where we have made use of the vector identity $(\mathbf{H}\cdot\nabla)\mathbf{H}=\frac{1}{2}\nabla(H^2)-\mathbf{H}\times\nabla\times\mathbf{H}$. For the particular configuration under consideration, $\nabla\times\mathbf{H}=j\hat{z}'$ and $\mathbf{H}=\frac{1}{2}jr'\hat{\theta}'$ where $\hat{r}'$, $\hat{\theta}'$, $\hat{z}'$ are the unit vectors along the corresponding cylindrical coordinate curves.

Substituting in the above equation, we obtain

$$\mathbf{F}=-\tfrac{1}{2}\mu_0m_0j\hat{r}', \qquad (4)$$

that is, the external force acting on the particle is constant in magnitude and directed radially toward the axis. It should be emphasized that implicit in the above analysis is the assumption that the particle material and the fluid material have essentially the same electrical conductivity so that the current density remains spatially uniform throughout the cross section.

B. Smoluchowski's Diffusion Equation and Boundary Conditions

The generalization of the ordinary diffusion equation when the diffusing particle is under the influence of outside forces is given by Smoluchowski's equation[4]

$$\partial f/\partial t'=D\nabla^2f-\nabla\cdot((\mathbf{F}/m\beta)f), \qquad (5)$$

where $f(\mathbf{r}',t')d\mathbf{r}'$ is the probability of finding a particle in the interval $d\mathbf{r}'$ between $\mathbf{r}'$ and $\mathbf{r}'+d\mathbf{r}'$ at time t'. The coefficient D is the diffusion coefficient (assumed constant), $\mathbf{F}$ is the outside force acting on dispersed particle, m is the particle mass, and $\beta=6\pi b\eta/m$, where b is the particle (assumed spherical) radius and η is the coefficient of viscosity of the surrounding fluid.

Examining the right-hand side of Eq. (5), one can define a probability flux density $\mathbf{J}$ (i.e., probability of particle crossing unit area per unit time) whose negative divergence equals the right-hand side, i.e.,

$$\mathbf{J}=-D\nabla f+(\mathbf{F}/m\beta)f. \qquad (6)$$

Substituting Eq. (4) for $\mathbf{F}$ into Eqs. (5) and (6) and remembering that the problem at hand involves cylindrically symmetric diffusion in the radial direction only, Eqs. (5) and (6) become

$$\frac{\partial f}{\partial t'}=D\left(\frac{\partial^2 f}{\partial r'^2}+\frac{1}{r'}\frac{\partial f}{\partial r'}\right)+\alpha\frac{\partial f}{\partial r'}+\frac{\alpha}{r'}f, \qquad (7)$$

$$J_r=-(D\partial f/\partial r'+\alpha f), \qquad (8)$$

where $\alpha=\mu_0m_0j/2m\beta$.

We seek a solution of Eq. (7) subject to the following initial and boundary conditions:

$$f(r',t')\rightarrow f_0(r') \quad \text{as} \quad t'\rightarrow0. \qquad (9)$$

The requirement that no particle shall cross the impermeable cylindrical wall at $r'=a$ yields

$$\partial f/\partial r'+(\alpha/D)f=0 \quad \text{at} \quad r'=a \quad \text{for all} \quad t'>0. \qquad (10)$$

Finally, introducing the dimensionless independent variables

$$r=r'/a; \quad t=(D/a^2)t',$$

the system of Eqs. (7), (9), and (10) becomes

$$\frac{\partial f}{\partial t}=\frac{\partial^2 f}{\partial r^2}+\left(\frac{1}{r}+\gamma\right)\frac{\partial f}{\partial r}+\frac{\gamma}{r}f, \qquad (11)$$

subject to the conditions

$$f(r,t)\rightarrow f_0(r) \quad \text{as} \quad t\rightarrow0, \qquad (12)$$

$$\partial f/\partial r+\gamma f=0 \quad \text{at} \quad r=1 \quad \text{for all} \quad t>0, \qquad (13)$$

where the dimensionless parameter $\gamma=\alpha a/D$. Thus, by casting the system in the above nondimensional form,

[4] S. Chandrasekhar, *Selected Papers on Noise and Stochastic Processes*, edited by N. Wax (Dover Publications, Inc., New York, 1954), pp. 42–43; Rev. Mod. Phys. **15**, 1 (1943).

the single parameter γ serves to describe completely the influence of the magnetic force on the diffusion process.

III. SOLUTION

Seeking a separable solution of the form $e^{-\lambda t}R(r)$, with λ representing the separation constant, and substituting into Eq. (11), we obtain the following ordinary differential equation for $R(r)$:

$$r(d^2R/dr^2)+(1+\gamma r)dR/dr+(\lambda r+\gamma)R=0. \quad (14)$$

Let

$$R(r)=r^{-1/2}e^{-\gamma r/2}u(r). \quad (15)$$

Substituting into Eq. (14), we obtain the following differential equation for $u(r)$:

$$4r^2(d^2u/dr^2)=[(\gamma^2-4\lambda)r^2-2\gamma r-1]u. \quad (16)$$

Making the following change of independent variable

$$z=(\gamma^2-4\lambda)^{1/2}r, \quad (17)$$

Eq. (16) becomes

$$4z^2(d^2u/dz^2)=[z^2-2\gamma/(\gamma^2-4\lambda)^{1/2}z-1]u, \quad (18)$$

which is recognized as Whittaker's equation[5]

$$4z^2(d^2u/dz^2)=(z^2-4kz+4m^2-1)u, \quad (19)$$

with $m=0$ and $k=\frac{1}{2}\gamma(\gamma^2-4\lambda)^{-1/2}$.

The solution of Whittaker's equation, regular at $z=0$, is[5]

$$u(z)=M_{k,0}(z)=M_{\gamma/2(\gamma^2-4\lambda)^{1/2},0}(z)$$
$$=z^{1/2}e^{-z/2}{}_1F_1(\tfrac{1}{2}[1-\gamma/(\gamma^2-4\lambda)^{1/2}];1;z), \quad (20)$$

where ${}_1F_1(a;1;z)$ is the confluent hypergeometric function defined by

$${}_1F_1(a;1;z)=1+\sum_{n=1}^{\infty}a(a+1)\cdots(a+n-1)\frac{z^n}{(n!)^2}. \quad (21)$$

Returning to the original variable r, using Eq. (20) in Eq. (15) and suppressing the constant multiplier $(\gamma^2-4\lambda)^{1/4}$, the solution of Eq. (14), regular at the origin, is

$$R(\lambda,\gamma,r)=\exp\{-\tfrac{1}{2}[\gamma+(\gamma^2-4\lambda)^{1/2}]r\}$$
$$\times{}_1F_1(\tfrac{1}{2}[1-\gamma/(\gamma^2-4\lambda)^{1/2}];1;(\gamma^2-4\lambda)^{1/2}r). \quad (22)$$

Substituting Eq. (22) into the boundary condition Eq. (13), i.e.,

$$dR/dr+\gamma R=0 \quad \text{at} \quad r=1, \quad (23)$$

and making use of the recurrence relation[6]

$$(b-a){}_1F_1(a-1;b;z)$$
$$=(b-a-z){}_1F_1(a;b;z)+zd{}_1F_1(a;b;z)/dz$$

leads to the following characteristic equation:

$$G(\lambda,\gamma)=[\tfrac{1}{2}+\gamma/2(\gamma^2-4\lambda)^{1/2}]$$
$$\times\exp\{-\tfrac{1}{2}[\gamma+(\gamma^2-4\lambda)^{1/2}]\}\{[(\gamma^2-4\lambda)^{1/2}-1]$$
$$\times{}_1F_1(\tfrac{1}{2}[1-\gamma/(\gamma^2-4\lambda)^{1/2}];1;(\gamma^2-4\lambda)^{1/2})$$
$$+{}_1F_1(\tfrac{1}{2}[-1-\gamma/(\gamma^2-4\lambda)^{1/2}];1;(\gamma^2-4\lambda)^{1/2})\}=0, \quad (24)$$

whose roots yield the eigenvalues λ_k. It is shown in Appendix A that Eq. (14) can be cast in self-adjoint form having the weight factor $re^{\gamma r}$ which remains of one sign in the region under consideration. Thus, the eigenvalues of the Sturm-Liouville system, Eqs. (14) and (23) [and hence the roots of Eq. (24)] are all real.[7] Further, it is shown in Appendix A that the roots of the characteristic equation are nonrepetitive and in Appendix B that the eigenvalues are all nonnegative. Using the fact that ${}_1F_1(0;1;\gamma)=1$ and ${}_1F_1(-1;1;\gamma)=1-\gamma$, it is seen that $\lambda=\lambda_0=0$ is a root of the characteristic equation independent of γ. For each value of γ, the roots of Eq. (24) generate an infinite set of discreet nonnegative eigenvalues λ_k ($\lambda_0=0$) lying in the interval $0\leq\lambda_k<\infty$. By examining Eq. (22) one would suspect that the eigenfunctions $R(\lambda_k,\gamma,r)$, belonging to those eigenvalues λ_k for which $\lambda_k>\gamma^2/4$, are complex functions since for $\lambda_k>\gamma^2/4$ the exponential as well as the confluent hypergeometric terms involve complex parameters and imaginary arguments. We know, of course, that the functions $R(\lambda_k,\gamma,r)$ must be real since they are the solutions of the Sturm-Liouville system Eqs. (14) and (23) involving real coefficients and real eigenvalues. However, an independent proof that the eigenfunctions $R(\lambda_k,\gamma,r)$ given in Eq. (22) (with λ replaced by λ_k) are real is given in Appendix C.

It is shown in Appendix A that any two eigenfunctions $R(\lambda_k,\gamma,r)$, $R(\lambda_l,\gamma,r)$ belonging to different eigenvalues $\lambda_k\neq\lambda_l$ are orthogonal to each other with respect to the weight factor $re^{\gamma r}$. Mathematically, from Appendix A,

$$\int_0^1 re^{\gamma r}R(\lambda_k,\gamma,r)R(\lambda_l,\gamma,r)dr=0, \quad (25)$$

and

$$\int_0^1 re^{\gamma r}R^2(\lambda_k,\gamma,r)dr=\int_0^1 re^{-(\gamma^2-4\lambda_k)^{1/2}r}{}_1F_1{}^2(\tfrac{1}{2}[1-\gamma/(\gamma^2-4\lambda_k)^{1/2}];1;(\gamma^2-4\lambda_k)^{1/2}r)dr$$
$$=\frac{e^\gamma}{\gamma}\frac{dR(\lambda_k,\gamma,r)}{dr}\bigg|_{r=1}\frac{dG(\lambda,\gamma)}{d\lambda}\bigg|_{\lambda=\lambda_k}, \quad (26)$$

where $G(\lambda,\gamma)$ is given by Eq. (24).

[5] E. D. Rainville, *Intermediate Differential Equations* (The Macmillan Company, New York, 1964), Chap. 11.

[6] *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (U. S. Department of Commerce, National Bureau of Standards, Washington, D. C., 1964), Appl. Math. Ser. 55, Chap. 13, p. 507, formula (13.4.11).

[7] E. L. Ince, *Ordinary Differential Equations* (Dover Publications, Inc., New York, 1956), Sec. 10.7, pp. 237–238.

On the assumption that our Sturm-Liouville system generates a complete set of eigenfunctions, the results, Eqs. (25) and (26), form the basis by which the initial condition can be satisfied. Thus, if the initial probability distribution function $f_0(r)$ is quadratically integrable with respect to the weight factor $re^{\gamma r}$ in the interval $0 \leq r \leq 1$, i.e.,

$$\int_0^1 re^{\gamma r} f_0^2(r)dr \quad \text{finite},$$

then

$$f_0(r) = \sum_{k=0}^{\infty} A_k R(\lambda_k,\gamma,r),$$

where, using Eqs. (25) and (26),

$$A_k = \int_0^1 f_0(r) re^{\gamma r} R(\lambda_k,\gamma,r)dr \bigg/ \int_0^1 re^{-(\gamma^2-4\lambda_k)^{1/2} r} {}_1F_1{}^2(\tfrac{1}{2}[1-\gamma/(\gamma^2-4\lambda_k)^{1/2}];\, 1;\, (\gamma^2-4\lambda_k)^{1/2} r)dr.$$

Finally, using the above result, the formal solution to the diffusion problem is

$$f(r,t) = \sum_{k=0}^{\infty} \frac{\int_0^1 f_0(r) re^{\gamma r} R(\lambda_k,\gamma,r)dr}{\int_0^1 re^{-(\gamma^2-4\lambda_k)^{1/2} r} {}_1F_1{}^2(\tfrac{1}{2}[1-\gamma/(\gamma^2-4\lambda_k)^{1/2}];\, 1;\, (\gamma^2-4\lambda_k)^{1/2} r)dr} R(\lambda_k,\gamma,r) e^{-\lambda_k t}, \tag{27}$$

where $R(\lambda_k,\gamma,r)$ is given by Eq. (22) with λ_k replacing λ, and λ_k ($\lambda_0 = 0$) are the roots of the characteristic Eq. (24).

IV. SOME SPECIAL SOLUTIONS AND THE EQUILIBRIUM DISTRIBUTION

A. External Force Absent, i.e., $\gamma = 0$

It is at first of interest to determine what the general solution Eq. (27) reduces to for the case when the external magnetic force is absent, i.e., $\gamma = 0$. When $\gamma = 0$, the eigenfunctions Eq. (22) reduce to

$$R(\lambda_k,0,r) = e^{-i(\lambda_k)^{1/2} r} {}_1F_1(\tfrac{1}{2};\, 1;\, 2i(\lambda_k)^{1/2} r). \tag{28}$$

Making use of the identity[8]

$${}_1F_1(\nu+\tfrac{1}{2};\, 2\nu+1;\, 2iz) = \Gamma(1+\nu) e^{iz} \left(\frac{1}{2z}\right)^{-\nu} J_\nu(z),$$

where $\Gamma(1+\nu)$ is the gamma function of argument $(1+\nu)$ and $J_\nu(z)$ the Bessel function of the first kind of order ν, Eq. (28) reduces to, with $\nu = 0$,

$$R(\lambda_k,0,r) = J_0((\lambda_k)^{1/2} r). \tag{29}$$

That is, the eigenfunctions reduce to the Bessel functions of the first kind of order zero.

Substituting Eq. (29) into Eqs. (27) and (24), the solution Eq. (27) takes the form

$$f(r,t) = 2 \sum_{k=0}^{\infty} \frac{\int_0^1 r f_0(r) J_0((\lambda_k)^{1/2} r)dr}{J_0^2((\lambda_k)^{1/2})} J_0((\lambda_k)^{1/2} r) e^{-\lambda_k t} \tag{30}$$

and the characteristic Eq. (24) reduces to

$$J_1(\sqrt{\lambda}) = 0. \tag{31}$$

The system of Eqs. (30) and (31) is recognized as the solution, given by Carslaw and Jaeger,[9] to the classical initial value problem of radial diffusion (heat conduction) in an infinitely long circular cylinder with an impermeable (insulated) wall located at $r = 1$. That Eqs. (27) and (24) should reduce to the solution (30) and (31) is made obvious by the fact that when $\gamma = 0$ our original system of Eqs. (11) through (13) reduces to the ordinary diffusion equation with the usual insulated flux condition at the wall, i.e., $\partial f/\partial r = 0$ at $r = 1$.

B. Initial Distribution a Normalized Delta Function Located at the Axis

For numerical purposes, in Sec. V, we shall consider the special case where the initial probability distribution function is a normalized delta function located at the axis. Symbolically,

$$f_0(r) = \frac{\delta(r)}{2\pi r} \quad \text{so that} \quad 2\pi \int_0^1 f_0(r) r\, dr = 1. \tag{32}$$

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[8] Reference 6, Chap. 13, p. 509, formula (13.6.1).

[9] H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids* (Clarendon Press, Oxford, England, 1947), Chap. VII, p. 178, formula 11.

Substituting Eq. (32) for $f_0(r)$ into Eq. (27) and remembering that $R(\lambda_k,\gamma,0)=1$, Eq. (27) becomes

$$f(r,t)=\frac{1}{2\pi}\sum_{k=0}^{\infty}\frac{\exp\{-\frac{1}{2}[\gamma+(\gamma^2-4\lambda_k)^{1/2}]r\}\,{}_1F_1(\frac{1}{2}[1-\gamma/(\gamma^2-4\lambda_k)^{1/2}];1;(\gamma^2-4\lambda_k)^{1/2}r)e^{-\lambda_k t}}{\int_0^1 re^{-(\gamma^2-4\lambda_k)^{\frac{1}{2}}r}{}_1F_1{}^2(\frac{1}{2}[1-\gamma/(\gamma^2-4\lambda_k)^{1/2}];1;(\gamma^2-4\lambda_k)^{1/2}r)dr}. \quad (33)$$

C. The Equilibrium Distribution

The equilibrium distribution $f_{\text{eq}}(r)$ is defined as the limit of $f(r,t)$ as $t\rightarrow\infty$. Thus the equilibrium distribution, remembering that $\lambda_0=0$, is given by the first term of Eq. (27). Using the fact that $R(0,\gamma,r)=e^{-\gamma r}$ since ${}_1F_1(0;1;z)=1$, and considering only initial distributions which are normalized, i.e., $2\pi\int_0^1 f_0(r)rdr=1$, we obtain from the first term of Eq. (27)

$$f_{\text{eq}}(r)=\frac{1}{2\pi}\left(e^{-\gamma r}\Big/\int_0^1 re^{-\gamma r}dr\right)$$
$$=\frac{1}{2\pi}\frac{\gamma^2}{1-(\gamma+1)e^{-\gamma}}e^{-\gamma r}. \quad (34)$$

Incidentally, the equilibrium distribution can be obtained without resorting to the solution of the initial value problem. That is, at equilibrium, the probability flux density J_r is everywhere zero. Thus, the equilibrium distribution is simply the solution of $df_{\text{eq}}/dr+\gamma f_{\text{eq}}=0$ subject to the condition that f_{eq} is normalized. The result yields Eq. (34).

The average position of the particle at equilibrium is

$$r_{\text{av}}=\frac{2-(\gamma^2+2\gamma+2)e^{-\gamma}}{\gamma[1-(1+\gamma)e^{-\gamma}]}, \quad (35)$$

and so $0\leq r_{\text{av}}\leq\frac{2}{3}$, when $\infty\geq\gamma\geq 0$.

V. NUMERICAL RESULTS

A tabulation of the first six roots of the characteristic equation for the range of γ lying between zero and 3.0 in increments of 0.5 is shown in Table I. Referring to the analysis of Sec. IV A, it is seen that the row headed by $\gamma=0$ corresponds to the roots[10] of $J_1(\sqrt{\lambda})=0$. While the magnitude of λ_k does not change appreciably with increasing γ, the physical interpretation, however, of the decrease in magnitude of λ_k (particularly λ_1) with increasing γ up to $\gamma=2.0$ and the subsequent increase for $\gamma>2$ will be discussed presently.

TABLE I. Numerical values of the first six roots of the characteristic equation for the range of γ lying between zero and 3.0 in increments of 0.5.

$\gamma \backslash \lambda$	λ_0	λ_1	λ_2	λ_3	λ_4	λ_5
0	0	14.68197	49.21846	103.49945	177.52077	271.28165
0.5	0	14.22966	48.62367	102.81441	176.76947	270.47799
1.0	0	13.89971	48.15217	102.25317	176.14228	269.79864
1.5	0	13.69205	47.80395	101.81573	175.63921	269.24360
2.0	0	13.60661	47.57901	101.50211	175.26026	268.81287
2.5	0	13.64331	47.47737	101.31232	175.00544	268.50646
3.0	0	13.80209	47.49902	101.24636	174.87475	268.32438

[10] Reference 6, Chap. 9, p. 414, Table 9.7.

In order to illustrate numerically the manner in which an initial probability distribution relaxes to its equilibrium distribution, we chose to consider the particular case where the initial probability distribution function was a normalized delta function located at the axis. The solution for this case is given by Eq. (33). For the special case $\gamma=1$ and using the first six terms of the series, the variation of the probability distribution function $f(r,t)$ with dimensionless radial distance for the four different dimensionless times $t=1/\lambda_1, 2/\lambda_1, 3/\lambda_1$, and $t=\infty$ is shown in Fig. 1. At least for large times, the

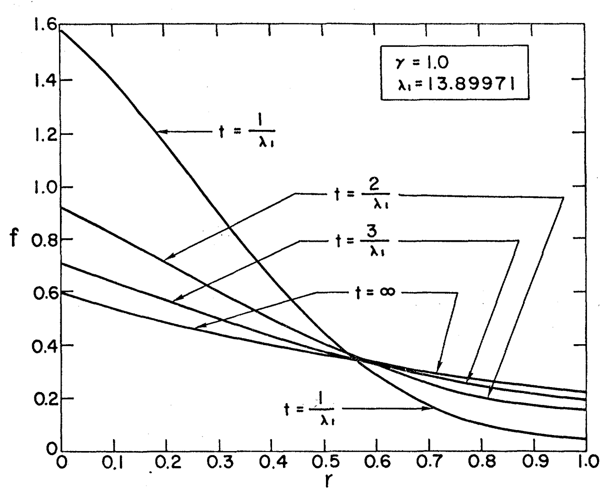

FIG. 1. Variation of the probability distribution function with dimensionless radial distance at various dimensionless times for the case $\gamma=1.0$.

temporal behavior of the distribution function is governed essentially by the second term of Eq. (33). Thus, the reciprocal of the first eigenvalue, i.e., $1/\lambda_1$ is essentially a measure of the relaxation time. Now, at least as far as the equilibrium distribution is concerned, it was shown in Sec. IV C that the mean particle position decreases with increasing γ. Thus, on the average, a particle need not travel as far in order to reach its equilibrium position. At the same time, the mean diffusion speed radially outward decreases with increasing γ since the particle must buck a stronger magnetic force. Since the relaxation time is a measure of the ratio of these two quantities, the results of the table indicate that as γ increases from zero to 2.0 the mean diffusion distance decreases less rapidly with γ than the

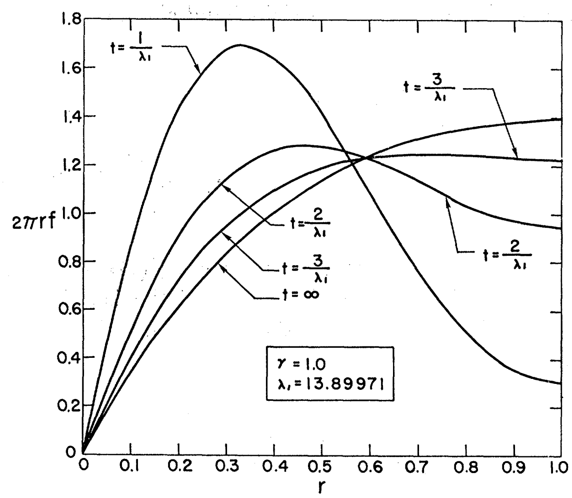

FIG. 2. Variation of the distribution function $2\pi rf$ with dimensionless radial distance at various dimensionless times for the case $\gamma = 1.0$.

mean diffusion speed and that the reverse is true when $\gamma > 2.0$.

For the particular case considered here, the numerical results further indicate that the equilibrium distribution is reached to within 5% in a dimensionless time of the order of four times the relaxation time, i.e., $t = 4/\lambda_1 = 0.2878$. In terms of dimensional time, however, the time taken to practically reach the equilibrium distribution depends on the dimension of the diffusion region and the diffusion constant through the relation $t' = (a^2/D)t$.

The variation of the probability of finding the particle anywhere in the annulus located at r, i.e., $2\pi rf(r,t)$ with dimensionless radial distance for the same dimensionless times as above is shown in Fig. 2. Of particular interest is the existence of a maximum which is continuously displaced towards the surface; the maximum becoming flatter on account of the random motions experienced by the particles. Also to be noted is the manner in which the function continuously adjusts itself with time so that the integral under the curve always remains equal to 1, as it must.

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APPENDIX A

The integrals

$$\int_0^1 re^{\gamma r}R(\lambda_k,\gamma,r)R(\lambda_l,\gamma,r)dr; \quad \int_0^1 re^{\gamma r}R^2(\lambda_k,\gamma,r)dr.$$

We use the notation $R_k = R(\lambda_k,\gamma,r)$, $R_l = R(\lambda_l,\gamma,r)$ and the prime to denote differentiation with respect to r. Consider the differential Eq. (14) for the two eigenfunctions R_k and R_l belonging, respectively, to the eigenvalues λ_k and λ_l, $\lambda_k \neq \lambda_l$, i.e.,

$$rR_k'' + (1+\gamma r)R_k' + (\lambda_k r + \gamma)R_k = 0, \qquad \text{(A1)}$$

$$rR_l'' + (1+\gamma r)R_l' + (\lambda_l r + \gamma)R_l = 0. \qquad \text{(A2)}$$

Casting Eqs. (A1) and (A2) in self-adjoint form by multiplication by $e^{\gamma r}$, we obtain

$$(re^{\gamma r}R_k')' + (\lambda_k r + \gamma)e^{\gamma r}R_k = 0, \qquad \text{(A3)}$$

$$(re^{\gamma r}R_l')' + (\lambda_l r + \gamma)e^{\gamma r}R_l = 0. \qquad \text{(A4)}$$

Multiplying Eq. (A3) by R_l, Eq. (A4) by R_k, subtracting and integrating between the limits 0 and 1, yields

$$(\lambda_k - \lambda_l)\int_0^1 re^{\gamma r}R_kR_l\,dr = \int_0^1 [R_k(re^{\gamma r}R_l')' - R_l(re^{\gamma r}R_k')']dr.$$

Integrating by parts, yields

$$(\lambda_k - \lambda_l)\int_0^1 re^{\gamma r}R_kR_l\,dr = \{re^{\gamma r}[R_kR_l' - R_lR_k']\}_0^1. \qquad \text{(A5)}$$

Using the boundary condition Eq. (23) in the right-hand side of Eq. (A5), i.e.,

$$R_k'(1) + \gamma R_k(1) = 0,$$

$$R_l'(1) + \gamma R_l(1) = 0,$$

and the fact that the right-hand side of (A5) vanishes at the lower limit, yields

$$\int_0^1 re^{\gamma r}R_k(\lambda_k,\gamma,r)R_l(\lambda_l,\gamma,r)dr = 0. \qquad \text{(A6)}$$

To evaluate $\int_0^1 re^{\gamma r}R^2(\lambda_k,\gamma,r)dr$ we first replace Eq. (A1) by $rR'' + (1+\gamma r)R' + (\lambda r + \gamma)R = 0$, where λ now plays the role of a continuously running parameter having a corresponding solution denoted simply by R. We now proceed in the same exact fashion as above obtaining the following expression equivalent to Eq. (A5):

$$(\lambda - \lambda_k)\int_0^1 re^{\gamma r}RR_k\,dr = \{re^{\gamma r}[RR_k' - R_kR']\}_0^1 = e^{\gamma}[R(\lambda,\gamma,1)R'(\lambda_k,\gamma,1) - R(\lambda_k,\gamma,1)R'(\lambda,\gamma,1)].$$

Using the boundary condition Eq. (23), i.e., $R(\lambda_k,\gamma,1) = -(1/\gamma)R'(\lambda_k,\gamma,1)$, to replace $R(\lambda_k,\gamma,1)$ in the above expression, yields

$$\int_0^1 re^{\gamma r}RR_k\,dr=\frac{e^\gamma}{\gamma}R'(\lambda_k,\gamma,1)\left[\frac{\gamma R(\lambda,\gamma,1)+R'(\lambda,\gamma,1)}{\lambda-\lambda_k}\right].$$

Taking the limit of the above expression as $\lambda\rightarrow\lambda_k$ and remembering that

$$\lim_{\lambda\rightarrow\lambda_k}\left[\frac{\gamma R(\lambda,\gamma,1)+R'(\lambda,\gamma,1)}{\lambda-\lambda_k}\right]=\left.\frac{dG(\lambda,\gamma)}{d\lambda}\right|_{\lambda=\lambda_k},$$

$$\text{since}\quad \gamma R(\lambda_k,\gamma,1)+R'(\lambda_k,\gamma,1)=0,$$

we obtain

$$\int_0^1 re^{\gamma r}R_k^2(\lambda_k,\gamma,r)dr=\frac{e^\gamma}{\gamma}\left.\frac{dR(\lambda_k,\gamma,r)}{dr}\right|_{r=1}\left.\frac{dG(\lambda,\gamma)}{d\lambda}\right|_{\lambda=\lambda_k},\quad \text{(A7)}$$

where G is given by Eq. (24).

Incidentally, the result of Eq. (A7) can be used to prove that the roots of the characteristic equation are nonrepetitive. For, if λ_k were a multiple root, then $(dG/d\lambda)_{\lambda=\lambda_k}=0$. This result forces the left-hand side of Eq. (A7) to equal zero, which is impossible unless $R(\lambda_k,\gamma,r)\equiv 0$.

APPENDIX B:

EIGENVALUES ARE NON-NEGATIVE

Casting the differential Eq. (14) for the eigenfunction $R(\lambda_k,\gamma,r)$ belonging to the eigenvalue λ_k into self-adjoint form by multiplying through by $e^{\gamma r}$ yields, using the same notation as in Appendix A,

$$\lambda_k re^{\gamma r}R_k=-(re^{\gamma r}R_k')'-\gamma e^{\gamma r}R_k. \quad \text{(B1)}$$

Multiplying through by R_k, and integrating between the limits 0 to 1, yields

$$\lambda_k\int_0^1 re^{\gamma r}R_k^2\,dr=-\int_0^1\{R_k[(re^{\gamma r}R_k')'+\gamma e^{\gamma r}R_k]\}dr. \quad \text{(B2)}$$

Integrating by parts, the right-hand side becomes

$$-re^{\gamma r}R_kR_k'\Big|_0^1+\int_0^1 re^{\gamma r}R_k'^2\,dr-\gamma re^{\gamma r}R_k^2\Big|_0^1+\gamma\int_0^1 r(e^{\gamma r}R_k^2)'dr.$$

The integrated terms cancel since $R_k'(1)=-\gamma R_k(1)$. Differentiating out the integrand of the last integral and combining integrals, the right-hand side of Eq. (B2) reduces to

$$\int_0^1 re^{\gamma r}\{R_k'+\gamma R_k\}^2\,dr$$

and hence from Eq. (B2)

$$\lambda_k=\int_0^1 re^{\gamma r}\{R_k'+\gamma R_k\}^2\,dr\bigg/\int_0^1 re^{\gamma r}R_k^2\,dr. \quad \text{(B3)}$$

Thus, since the integrands in both numerator and denominator of Eq. (B3) are positive, λ_k is positive.

APPENDIX C

The eigenfunctions given by Eq. (22) (with λ_k replacing λ) are real.

We need only concern ourselves with those eigenfunctions whose eigenvalues $\lambda_k>\gamma^2/4$.

Let $p=(4\lambda_k-\gamma^2)^{1/2}$. Then the eigenfunction Eq. (22) becomes

$$R(\lambda_k,\gamma,r)=e^{-\gamma r/2}e^{-ipr/2}{}_1F_1(\tfrac{1}{2}[1+i\gamma/p];1;ipr). \quad \text{(C1)}$$

Since the first factor $e^{-\gamma r/2}$ is real, we need only concern ourselves with the character of the product of the last two factors.

We now make use of the following integral representation for the confluent hypergeometric function[11]:

$${}_1F_1(a;1;z)=\frac{1}{\Gamma(a)\Gamma(1-a)}\int_0^1 e^{zt}t^{a-1}(1-t)^{-a}dt, \quad \text{(C2)}$$

valid for $R_ea<1$. In our case, $R_ea=\frac{1}{2}$ so the restriction on a is satisfied. For our case, Eq. (C2) becomes

$${}_1F_1(\tfrac{1}{2}+i\gamma/2p;1;ipr)=\frac{\sin\pi(\frac{1}{2}+i\gamma/2p)}{\pi}\int_0^1\frac{e^{iprt}}{[t(1-t)]^{1/2}}\left(\frac{t}{1-t}\right)^{i\gamma/2p}dt,$$

[11] Reference 6, Chap. 13, p. 505, formula (13.2.1).

where we have made use of the identity[12]

$$\Gamma(a)\Gamma(1-a)=\pi/\sin\pi a.$$

Now

$$\sin(\pi/2+i\gamma\pi/2p)=\cosh(\gamma\pi/2p)$$

so

$$e^{-ipr/2}{}_1F_1(\tfrac{1}{2}+i\gamma/2p;1;ipr)$$

$$=\frac{\cosh(\pi\gamma/2p)}{\pi}\int_0^1\frac{e^{ipr(t-\frac{1}{2})}}{[t(1-t)]^{1/2}}\left(\frac{t}{1-t}\right)^{i\gamma/2p}dt. \quad \text{(C3)}$$

[12] Reference 6, Chap. 6, p. 256, formula (6.1.17).

Letting $u=1-t$; $du=-dt$; $t-\frac{1}{2}=\frac{1}{2}-u$ (C3) becomes

$$e^{-ipr/2}{}_1F_1(\tfrac{1}{2}+i\gamma/2p;1;ipr)$$

$$=\frac{\cosh(\pi\gamma/2p)}{\pi}\int_0^1\frac{e^{ipr(\frac{1}{2}-u)}}{[u(1-u)]^{1/2}}\left(\frac{1-u}{u}\right)^{i\gamma/2p}du$$

$$=\frac{\cosh(\gamma\pi/2p)}{\pi}\int_0^1\frac{e^{-ipr(u-\frac{1}{2})}}{[u(1-u)]^{1/2}}\left(\frac{u}{1-u}\right)^{-i\gamma/2p}du. \quad \text{(C4)}$$

Comparing the right-hand sides of Eqs. (C3) and (C4), we see that $e^{-ipr/2}{}_1F_1(\frac{1}{2}+i\gamma/2p;1;ipr)$ is also equal to its complex conjugate, hence it is real. Thus, the eigenfunctions $R(\lambda_k,\gamma,r)$ are real.

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Excitation Spectrum of the Bose Liquid

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The assumptions and predictions of the Brueckner-Sawada method (including recent refinements) for the derivation of the excitation spectrum of the high-density boson system are examined using Green's-function techniques and a new method of solving the scattering-matrix equation. In the case of the real interaction potential of He^4 atoms, the spectrum obtained in our approximation has the correct form but the depletion has a meaningless value. It is pointed out that a probable cause of discrepancies is inconsistent omission of certain self-energy terms.

INTRODUCTION

THERE are several microscopic approaches to the derivation of the excitation spectrum of the zero-temperature boson system,[1] which have been used to derive the phonon part of the spectrum. To obtain the roton minimum, methods for a nondilute gas must be used. Some clarification of the reason for the appearance of the roton dip was already given by the argument of Feynman[2] showing connection between quasiparticle energy $\epsilon(k)$ and the liquid-structure function $S(k,\omega)$. The purely microscopic derivations have concentrated almost exclusively on the case of hard-sphere bosons. Brueckner and Sawada[3] (BS) treated the hard core as a screened delta-function potential, and found qualitative agreement with the Landau curve. Parry and ter Haar[4] found that the roton minimum disappears if the depletion effect is included in these calculations. Even poorer agreement was found when an attractive tail was added to the hard core, and they concluded that the hard-sphere boson gas is not as good a model for liquid helium as has been assumed. Liu, Liu, and Wong[5] showed, however, that a qualitatively correct excitation spectrum is found if, instead of this treatment, the hard-sphere potential is replaced by the two-body pseudopotential earlier considered by Lee, Huang, and Yang.[6]

The approach used in the quoted papers is called the Brueckner-Sawada method. Because it still forms one of the main efforts to microscopic derivation of the excitation spectrum of liquid helium II, we consider it worthwhile to study some aspects of the approximations and predictions of the theory. In the next two sections the quasiparticle spectrum, the depletion, and the reaction matrix equation are derived using Green's-function techniques at zero temperature. The BS method is then shown to have the following properties: [1] The quasiparticle energy is assumed to be given by the poles of the single-particle Green's function. [2] In this, only the self-energy resulting from first-order terms of the effective interaction with the particles in the condensate is included. [3] The effective interaction is given by the BS equation for the scattering matrix, in which the propagator between successive

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