Multiple Isotropic Scattering*

PAUL I. RICHARDS
Technical Operations, Inc., Arlington, Massachusetts
(Received April 21, 1955)

A formalism is derived for approximately solving problems in the transport of radiation or particles by isotropic scattering with absorption. Although the present theory is very similar to diffusion theory, comparison with rigorous solutions where available shows that the results are more accurate than those of diffusion theory especially in highly absorbing media and in regions close to sources. Moreover certain ambiguities and difficulties in extending diffusion theory are eliminated. The derivation proceeds directly from rigorous transport equations and is based on a series expansion for integrals of the Helmholtz type.

1. INTRODUCTION

1.1 Need for a Simple Theory

THE transport or "diffusion" of various radiations and particles through scattering media is governed by the classical Boltzmann transport equation originally derived in the kinetic theory of gases. Unfortunately, rigorous solution of this equation involves mathematical difficulties of a high order. Only recently[1,2] has the simplest problem been rigorously solved, namely the distribution due to an isotropic point source in an infinite isotropically scattering medium. Recent years have also seen numerical solutions[3,4] for a class of problems involving uniformly illuminated plane slabs.

In the vast majority of problems, however, recourse must generally be made to diffusion theory.[5,6] The latter is simple enough to allow solution of a much wider range of problems but suffers several unfortunate drawbacks. Since the concept of diffusion in response to a concentration-gradient is itself an approximation, the coefficient of diffusivity is poorly defined and several values have been proposed.[1,5,6] Secondly, simple diffusion theory cannot be realistic in regions where the motion is highly organized in direction as is the case near a source or in a highly absorbing medium. The results therefore cannot be trusted in such regions where the "direct beam" may be dominant. Finally, it is difficult to establish realistic boundary conditions where the scattering medium gives way to vacuum or a pure absorber. This is espically true if the boundary is directly illuminated by an external source.

In spite of these disadvantages, diffusion theory has been widely and usefully applied to problems where other theoretical methods are intractable.

1.2 Scope of Present Study

It is the aim of this paper to develop a formalism for isotropic scattering as simple and as widely useful as diffusion theory, but possessing the following advantages:

(a) unequivocal values for constants;
(b) definite boundary conditions even under external illumination;
(c) more accurate results particularly in regions near a source and in media which are highly absorbing.

Basic relations are set up in paragraph 2, the approximate formalism is developed in paragraph 3 and a comparison with other theories is given in paragraph 4.

2. EXACT RELATIONS

2.1 Limitations and Notation

We consider throughout the following only media which can absorb and scatter isotropically. Although the formalism can, like the diffusion theory, be applied to media which scatter anisotropically, the reduction to an equivalent isotropic scattering is outside the scope of the present discussion. For definiteness in terminology, we shall speak of light scattering, although of course the theory covers neutrons and other radiations as well.

Only steady-state distributions will be considered; time dependent problems are outside the scope of this study but can be reduced (reference 2, p. 47) to steady state problems.

Vacuum regions will be replaced by absorbing media in the formal development because absorbers are easier to treat in a unified manner and the two have the same effect on photon distribution within other media; neither returns photons.[7]

With these conventions it will then be assumed that

* This work was sponsored by Air Force Cambridge Research Center, Air Research and Development Command.

[1] C. C. Grosjean, Verhandel Koninke Vlaam. Acad. Wetenschap. Belgie, **13**, No. 36 (1951); also Physica **19**, 29 (1953).

[2] Case, deHoffmann, and Placzek *Introduction to the Theory of Neutron Diffusion*. (U. S. Government Printing Office, Washington, D. C., 1953), Vol. 1, Chap. 4.

[3] S. Chandrasekhar, *Radiative Transfer* (Oxford University Press, Oxford, 1950).

[4] Chandrasekhar, Elbert, and Franklin, Astrophys. J. **115**, 244–268, 269–278 (1952).

[5] S. Glasstone and M. G. Edlund, *Elements of Nuclear Reactor Theory* (D. Van Nostrand Company, Inc., New York, 1952).

[6] R. F. Christy in *Lecture Series in Nuclear Physics* (U. S. Government Printing Office, Washington, D. C., 1947), pp. 115–132.

[7] Obvious modifications in treatment are required if photons may stream through vacuum from one scattering region to another. These modifications will not be explicitly considered here. Although the difficulties so introduced can be great, the present theory is no more vulnerable in this respect than others.

the (total) mean free path λ is constant throughout all space. To simplify expressions we will often employ length units chosen so that $\lambda=1$.

The "albedo" of scattering, $\omega(\mathbf{r})$, however, will be allowed to vary arbitrarily with position. ($\omega=$ scattering cross section/total cross section $=\lambda/\lambda_{scat}=1-$ probability of absorption per encounter.) In particular $\omega=0$ in vacuum regions.

Other notations which will be used below are as follows:

$v=$ velocity of photons,
$\rho=$ density $=$ number of photons/cm^3,
$\mathbf{j}=$ current $=$ *net* vector flux of photons/cm^2 sec,
$q=$ number of photons emitted by sources/cm^3 sec,
$\rho_d=$ density of unscattered photons,
$\mathbf{j}_d=$ *net* flux of unscattered photons.

The latter two quantities refer to "direct beam" contributions. Note that where several such beams cross at a point, $\mathbf{j}_d$ represents their vector sum whereas ρ_d represents the sum of their magnitudes, similarly $q(\mathbf{r})$ is a scalar sum of photons emitted per unit volume per unit time. Note that the sources need not be isotropic.

2.2 Basic Equations

As shown in standard references[2,3] the basic equation for transport by isotropic scattering may be written

$$\rho(\mathbf{r})=\rho_d(\mathbf{r})+\int\int_{-\infty}^{\infty}\int\rho(\mathbf{r}')\omega(\mathbf{r}')\frac{e^{-|\mathbf{r}-\mathbf{r}'|/\lambda}}{4\pi|\mathbf{r}-\mathbf{r}'|^2\lambda}d\tau'. \quad (1)$$

This relation is easily derived by considering the photon density at $\mathbf{r}$ as made up of (a) direct beam contribution plus (b) photons scattered at $\mathbf{r}'$ and traveling without further scattering to position $\mathbf{r}$. The integral in (1) is to be taken over all space but $\omega(\mathbf{r}')$ is understood to vanish by the convention adopted above in regions where no scattering can occur.

It should be noted that Eq. (1) together with given $\rho_d(\mathbf{r})$ and $\omega(\mathbf{r})$ completely determine the solution of the entire problem. Once the density is known [Eq. (1) has a unique solution[2] for given $\rho_d(\mathbf{r})$, $\omega(\mathbf{r})$], the current and other quantities are completely determined. There is no need to use the current in setting boundary conditions on (1).

Nevertheless, since the current is often of interest in the results, we mention here some relations which will be used below.

$$\nabla\cdot\mathbf{j}=q(\mathbf{r})-[1-\omega(\mathbf{r})]v\rho(\mathbf{r})/\lambda, \quad (2)$$

$$\nabla\cdot\mathbf{j}_d=q(\mathbf{r})-v\rho_d(\mathbf{r})/\lambda, \quad (3)$$

$$\nabla x\mathbf{j}=0=\nabla x\mathbf{j}_d. \quad (4)$$

The first two relations are essentially conservation equations. The first states that photons are gained from the sources and lost only by absorbing collisions. Equation (3) states that *unscattered* photons are supplied by the sources and lost be *either* absorbing or scattering encounters.

Equation (4) may be readily proved by setting up integral expressions for $\mathbf{j}$ and $\mathbf{j}_d$ in terms of ρ, ρ_d and q by the method suggested under (1). Taking the curl of these expressions then establishes (4). Note that (4) implies that $\mathbf{j}$ and $\mathbf{j}_d$ are completely determined by the quantities on the right-hand sides of (2) and (3).

3. SIMPLIFIED THEORY

3.1 Derivation

Since λ has been forced to be constant everywhere we may apply to Eq. (1) the expansion (A.1) derived in the Appendix. Upon evaluating the appropriate integrals defined by (A.2), the result is

$$\rho=\rho_d+\sum_0^\infty(\lambda^2\nabla^2)^n(\omega\rho)/(2n+1), \quad (5)$$

where ∇^{2n} represents an n-fold application of the Laplacian operator.

If terms with $n>1$ are omitted in (5), the result is the standard diffusion equation but with a source term equal to $\rho_d v/\lambda$. However, since the coefficients in (5) fall off only as n^{-1}, neglect of higher order terms is probably not very accurate.

To obtain a more rapidly converging series, operate on both sides of (5) with the operator $(\lambda^2\nabla^2-1)$. Rearranging terms, the result is

$$\lambda^2\nabla^2[(3-2\omega)\rho/3]-(1-\omega)\rho = (\lambda^2\nabla^2-1)\rho_d+2\sum_2^\infty(\lambda^2\nabla^2)^n(\omega\rho)/(4n^2-1). \quad (6)$$

In Eq. (6), the coefficients of high-order terms now fall off very rapidly and we shall make the approximation of dropping the final sum. In this way it is natural to expect that the approximate results so obtained will be more accurate than diffusion theory. Later comparisons will bear out this expectation.

The formalism is considerably simplified if we introduce an auxiliary function $P(\mathbf{r})$ defined by the following equation:

$$\rho(\mathbf{r})=3[P(\mathbf{r})+\rho_d(\mathbf{r})]/[3-2\omega(\mathbf{r})]. \quad (7)$$

Making this substitution in (6) and dropping the infinite sum, one obtains

$$\lambda^2\nabla^2P(\mathbf{r})-K^2P(\mathbf{r})=-(1-K^2)\rho_d(\mathbf{r}), \quad (8)$$

where

$$K(\mathbf{r})=[3(1-\omega)/(3-2\omega)]^{\frac{1}{2}}. \quad (9)$$

Equation (8) with the definitions (7) and (9) forms the basic equation of the present formalism. The functions $\omega(\mathbf{r})$, the albedo for a single encounter, and $\rho_d(\mathbf{r})$, the density due to direct beams, are presumed to be given data for each specific problem.

Before discussing boundary conditions on $P(\mathbf{r})$, it is convenient to derive an expression for the net photon current. From Eq. (4) it follows that both $\mathbf{j}$ and $\mathbf{j}_d$ can be represented as gradients of scalar functions. Consequently we may also set

$$\mathbf{j}-\mathbf{j}_d=\lambda v \nabla f. \tag{10}$$

Subtracting (3) from (2) and making the substitution (10), one obtains

$$\lambda^2\nabla^2 f=\rho_d-(1-\omega)\rho. \tag{11}$$

It then follows from (7), (8), and (9) that

$$\nabla^2(f+P)=0. \tag{12}$$

Both P and f (aside from an undetermined constant) must vanish at infinity since ρ and $\mathbf{j}$ must vanish there. If we envisage any geometrical discontinuities in scattering media (i.e., discontinuities in K and ω) as replaced by very sharp but nevertheless continuous transitions, then on physical grounds all functions must be continuous with their first derivatives and it follows from (12) that f may be taken equal to $(-P)$. Equation (10) then becomes

$$\mathbf{j}=\mathbf{j}_d-\lambda v\nabla P. \tag{13}$$

This relation suffices to determine the net current when (8) has been solved for P.

Note that the derivation has involved no further approximations beyond the use of Eq. (8). Hence $\mathbf{j}$ as determined from (13) will satisfy rigorously the conservation and curl relations (2), (3), and (4) but the density appearing in these will be the approximate density as determined from (7) and (8). In short, Eq. (13) will yield precisely the current implied by the approximate ρ.

3.2 Boundary Conditions

In solving actual problems, it is convenient to consider changes in physical properties between various media as discontinuous. It then becomes necessary to specify the behavior of the fundamental auxiliary function P at such discontinuities.

Since photons do not accumulate at boundaries, it follows from (13) that the normal component of ∇P is continuous. This result may also be derived from (8) by integrating over a "pill box" volume which straddles the interface and letting the height of the box approach zero.

To derive the second boundary condition, it is convenient to consider the limiting case of a continuous transition between media as the distance through the transition is made to approach zero. Assume first that P becomes discontinuous in the limit. Then as the distance of transition is made small, the normal component of the current $\mathbf{j}$ will, by (13) attain an arbitrarily large value within the transition while remaining finite on either side of the boundary. This situation can only represent a "double layer" of sources and sinks distributed over the boundary, and must be ruled out of any physical problem not involving such double layers. Hence the remaining boundary condition is that P itself is continuous.

It may be noted that ρ as given by (7) will then be discontinuous at boundaries. This is not in general true of a rigorous solution,[2] but the latter often has a sudden drop very near an absorbing boundary, and the discontinuity given by the present formalism is presumably an approximation to this sudden drop.

It is of interest to note that the simple formalism presented here also automatically gives an extrapolated endpoint.[6] Specifically, if we assume a plane boundary located in a region where $\rho_d=0$ and separating a vacuum or an absorbing medium from a medium with finite ω, then the solution of (8) on the absorbing side is of the form $\exp(-|Z|/\lambda)$ where Z is distance measured normally from the boundary. By continuity of P and its normal derivative, the value and derivative of ρ will then have the ratio λ just inside the interface, and a linear extrapolation of ρ into the absorbing medium will then vanish at a distance λ from the boundary. Thus the formalism gives an extrapolated end point of λ rather than the rigorous value,[2,6] 0.7104λ. However, the present formalism will automatically alter this value to take account of various specific local conditions. There is no need, as in diffusion theory, to use separate arguments to establish its appropriateness in any given situation.

3.3 Summary

Here we collect the above results in a form which would be used to solve a specific problem. For simplicity it is assumed that there are no reflecting boundaries and that the geometry is such that photons cannot stream across a vacuum from one scattering medium to enter another. While the complications thus excluded can be very great, they are not peculiar to the present formalism.

The function, $\omega(\mathbf{r})$ = the probability that an encounter at $\mathbf{r}$ results in (isotropic) scattering rather than absorption, is specified by the properties of the various scattering media. It is assumed that the total mean path λ is constant in all media.

The function, $\rho_d(\mathbf{r})$ = density of unscattered photons, is readily computed from the given distribution of sources. Note that the sources need not themselves be isotropic and that they can be located either inside scattering media or in vacuum. Values of $\rho_d(\mathbf{r})$ will be needed only in regions where $\omega\neq 0$.

After $\rho_d(\mathbf{r})$ has been computed, vacuum regions are replaced by complete absorbers ($\omega=0$). The function $K(\mathbf{r})$ is then computed from

$$K=[(1-\omega)/(1-2\omega/3)]^{\frac{1}{2}}. \tag{14}$$

This completes the setup of the problem.

Solution of the problem is then effected by solving the following equation for the auxiliary function $P(\mathbf{r})$

$$\lambda^2\nabla^2 P-K^2P=-(1-K^2)\rho_d, \tag{15}$$

TABLE I. Comparison of $K(\omega)$ for various theories.

ω	By (14)	$K(\omega)$ Rigorous[a]	Diffusion theory
0	1	1	1.73
0.1	0.982	1.0000	1.64
0.2	0.960	0.9999	1.55
0.3	0.935	0.997	1.45
0.4	0.904	0.986	1.34
0.5	0.867	0.957	1.22
0.6	0.817	0.907	1.09
0.7	0.750	0.829	0.949
0.8	0.655	0.710	0.775
0.9	0.500	0.525	0.548
0.94	0.402	0.414	0.425
0.98	0.240	0.243	0.245
0.99	0.172	0.1725	0.173
1	0	0	0

[a] See reference 2.

subject to the boundary conditions

$$P \to 0 \text{ as } \mathbf{r} \to \infty$$

$$P \text{ and } \mathbf{n}\cdot\nabla P \text{ continuous at interfaces.} \quad (16)$$

The density ρ of photons and the *net* flux $\mathbf{j}$ are then given by

$$\rho = (P+\rho_d)/(1-2\omega/3), \quad (17)$$

$$\mathbf{j} = \mathbf{j}_d - \lambda v \nabla P. \quad (18)$$

In (18) $\mathbf{j}_d$ is the *net* flux of unscattered photons, which is easily computed from the given source distribution (with, of course, vacuum not replaced by absorbers).

Equations (17) and (18) give ρ and $\mathbf{j}$ assuming vacuum to act as a perfect absorber. Under the limitations stated above, they are thus correct (subject to the approximation of the whole theory) within scattering media, but of course do not represent the actual situation in vacuum regions. If needed, the latter must be separately computed from the former.

The value (18) for $\mathbf{j}$ will satisfy rigorously the conservation relations implied by the approximate value of $\rho(\mathbf{r})$.

It will be seen that the effort of solving a problem is about the same as that required by diffusion theory. Nevertheless, the results are shown below to be more accurate in cases where comparison with rigorous solution is possible.

TABLE II. Values of $4\pi r^2\rho v/S$ from various theories. Point source in an infinite medium, $\lambda=1$.

ω	r	By (19)	Rigorous[a]	Simple diffusion theory	Diffusion theory with rigorous K
	0	2.5	1	0	0
	0.5	1.96	1.52	1.14	1.15
	1	1.84	1.80	1.73	1.77
	1.5	1.77	1.90	1.97	2.05
0.9	2	1.68	1.87	2.01	2.10
	3	1.43	1.60	1.59	1.86
	4	1.13	1.24	1.33	1.47
	5	0.85	0.92	0.97	1.08
	6	0.62	0.65	0.66	0.77
	7	0.44	0.45	0.46	0.53
	0	1.25	1	0	0
	0.5	0.85	0.77	0.73	0.91
	1	0.58	0.54	0.70	1.11
	1.5	0.40	0.37	0.51	1.01
0.3	2	0.28	0.24	0.33	0.82
	3	0.13	0.10	0.12	0.45
	4	0.060	0.043	0.036	0.22
	5	0.028	0.017	0.010	0.10
	6	0.012	0.007	0.003	0.045
	7	0.005	0.003	0.0008	0.019

[a] See reference 2.

4. COMPARISON WITH OTHER THEORIES

4.1 Point Source in Infinite Medium

The present formalism is easily applied to the problem of an isotropic point source in an infinite homogeneous (isotropically) scattering and absorbing medium. In this case we have immediately $\rho_d = S\exp(-r)/4\pi r^2 v$ where S is the source strength in photons/sec and the unit of length has been chosen to make $\lambda=1$. The parameter K is a constant and Eq. (15) is readily solved by the method of Green's function. The final result is

$$\rho = [S/4\pi v(1-2\omega/3)]\{e^{-r}/r^2 + [(1-K^2)/2Kr] \times [e^{-Kr}2\tanh^{-1}K + e^{Kr}E_1((1+K)r) - e^{-Kr}E_1((1-K)r)]\}, \quad (19)$$

where

$$E_1(X) = -\mathrm{Ei}(-X) = \int_X^\infty (e^{-u}/u)du. \quad (20)$$

It may be noted that when ω vanishes (19) becomes the rigorous solution for a perfectly absorbing medium. It may also be shown that for all ω the total number of photons in the distribution (19) has the rigorous[2] value $S/(1-\omega)v$.

In comparing (19) with the corresponding result for diffusion theory, we run across one of the disquieting features of the latter. Simple diffusion theory gives the value $[3(1-\omega)]^{\frac{1}{2}}$ for K. This, of course, is greatly in error for small ω as may be seen in Table I where it also is shown that the value (14) for K is much closer to the rigorous[2] value. In view of this discrepancy, various corrected expressions are often used[6] in diffusion theory. Whatever the value of K employed, diffusion theory with a δ-function source term gives the result $\rho = (3S/4\pi v)(e^{-Kr}/r)$.

Now this expression gives the correct total number of photons only if the *less* accurate value of K is used. Moreover, a similar feature arises in comparing detailed values of ρ. In Table II are shown values of ρ given by various theories for $\omega=0.9$ and $\omega=0.3$. For values of ω near unity, there seems to be little choice between the various approximate theories (in this example at least). For $\omega=0.3$, we note again that the "corrected" diffusion theory is rather poor whereas the simple diffusion theory is surprisingly good in spite of the fact that K is 45% too

TABLE III. Transmission of nonabsorbing plane slab. ($\lambda=1$, $\omega=1$)

cos θ	t	Transmission This paper	Transmission Rigorous[a]	Diffusion theory	cos θ	t	Transmission This paper	Transmission Rigorous[a]	Diffusion theory
	0.05	0.975	0.975	0.98		0.05	0.683	0.680	0.632
	0.1	0.953	0.953	0.95		0.1	0.561	0.549	0.432
1	0.25	0.889	0.890	0.885	0.05	0.25	0.469	0.430	0.200
	0.5	0.802	0.797	0.786		0.5	0.420	0.356	0.100
	1	0.667	0.660	0.632		1	0.350	0.275	0.050
	0.05	0.953	0.954	0.950		0.05	0.488	0.456	0
	0.1	0.910	0.910	0.905		0.1	0.477	0.429	0
0.5	0.25	0.802	0.800	0.786	0	0.25	0.445	0.372	0
	0.5	0.674	0.666	0.632		0.5	0.400	0.313	0
	1	0.522	0.502	0.432		1	0.333	0.242	0
	0.05	0.890	0.890	0.885					
	0.1	0.803	0.801	0.786					
0.2	0.25	0.637	0.627	0.570					
	0.5	0.507	0.480	0.368					
	1	0.402	0.354	0.200					

[a] See reference 4.

large. At very great distances r, however, the reverse must be true since the exponential function will eventually dominate the behavior. Thus, in all, it appears that the present formalism, with no additional corrections, produces a single set of formulas which are much better approximations over large ranges of the parameters.

4.2 Plane Slab Problems

Consider a homogeneous plane slab of thickness t, bounded on both sides by vacuum. Let the slab be uniformly illuminated by parallel radiation making an angle θ with the normal. This problem also has been rigorously solved[3,4] for isotropic scattering.

In Table III, we show the transmission (=power transmitted/power incident) of a nonabsorbing slab as obtained from the various theories. It will be seen that the present formalism in all cases gives results which are better than those of diffusion theory and that in particular it predicts about the right transmission for grazing incidence ($\theta=\pi/2$) while diffusion theory gives a limiting value of zero. In Table III moreover we also have another example of the uncertainty entailed in any simple attempt to correct diffusion theory. The values shown are for the boundary condition $\rho=0$; in this example, use of the extrapolated endpoint (equivalent to using $t+1.4\lambda$ as the thickness) would make the diffusion approximation worse.

In Table IV are shown values of the albedo (=power reflected/power incident) for infinite thickness and various ω, θ. Here again it is seen that the present formalism is a better approximation over the full range of the parameters. Again, the results shown for diffusion theory are those of the simple version; use of diffusion formulas with the rigorous value of K makes the albedo approximation worse although the detailed variation of ρ is presumably better.

TABLE IV. Albedo of half-space.

ω	cos θ	Albedo This paper	Albedo Rigorous[a]	Diffusion theory	ω	cos θ	Albedo This paper	Albedo Rigorous[a]	Diffusion theory
	0	0	0	1		0	0.250	0.452	1
0	0.5	0	0	0.536	0.7	0.5	0.182	0.278	0.678
	1	0	0	0.366		1	0.143	0.208	0.513
	0	0.018	0.052	1		0	0.500	0.694	1
0.1	0.5	0.012	0.024	0.550	0.9	0.5	0.400	0.508	0.785
	1	0.009	0.016	0.378		1	0.333	0.415	0.645
	0	0.065	0.163	1		0	0.719	0.776	1
0.3	0.5	0.044	0.082	0.580	0.95	0.5	0.631	0.626	0.837
	1	0.034	0.058	0.408		1	0.561	0.536	0.720
	0	0.133	0.293	1		0	1	1	1
0.5	0.5	0.093	0.161	0.620	1	0.5	1	1	1
	1	0.071	0.115	0.448		1	1	1	1

[a] See reference 3.

4.3 Conclusions

In summary, an approximate formalism has been derived directly from the rigorous isotropic transport equations. Although the labor involved in the solution of specific problems is about the same as that required in diffusion theory, the results appear to be a better approximation particularly for absorbing media. The results are, of course, approximate but if further improvement is required, it is clear from the derivation itself what further steps should be taken to increase the accuracy. This is in contrast to diffusion theory where, as noted above, apparently reasonable attempts to improve accuracy may in fact have the opposite effect. It appears that the present formalism should be widely applicable to problems too complex to be handled by rigorous methods, and we expect to make a number of such applications in the near future.

APPENDIX

The purpose of this appendix is to indicate the derivation of the following expansion.

$$(4\pi)^{-1}\iiint_{-\infty}^{\infty} F(\mathbf{r}')K(|\mathbf{r}'-\mathbf{r}|)d\tau' = \sum_0^\infty a_n \nabla^{2n}F(\mathbf{r})/(2n+1)! \quad \text{(A.1)}$$

where

$$a_n = \int_0^\infty u^{2n+2}K(u)du. \quad \text{(A.2)}$$

The notation ∇^{2n} denotes an n-fold repeated application of the Laplacian operator. No detailed examination of restrictions will be made, but obviously necessary conditions are the convergence of (A.2) for all n and the validity of the Taylor series expansions for F.

To evaluate the integral in (A.1), change the integration variable to $\mathbf{R}=\mathbf{r}'-\mathbf{r}$ and expand $F(\mathbf{R}+\mathbf{r})$ in a three-dimensional Taylor series about $\mathbf{r}$.

$$\iiint_{-\infty}^{\infty} [e^{\mathbf{R}\cdot\nabla}F(\mathbf{r})]K(R)d\tau_R/4\pi,$$

where the exponential operator is defined by its power series. The mth term in this series is itself a sum of 3^m terms which arise from the scalar product in the exponent. Collecting like powers of these ultimate terms, the mth term of the main series can be written

$$\frac{1}{m!}\int_0^\infty K(R)R^{m+2}dR\cdot \sum_{(i+j+k=m)} \binom{m}{i}\binom{m-i}{j} \cdot\left\{\left(\frac{\partial}{\partial x}\right)^i\left(\frac{\partial}{\partial y}\right)^j\left(\frac{\partial}{\partial z}\right)^k F(r)\right\} \oint\oint \frac{X^iY^jZ^k}{4\pi R^m}d\Omega_R. \quad \text{(A.3)}$$

Now by spherical symmetry, the surface integrals in this expression will vanish unless i, j, and k are all even. Consequently, the entire expression (A.3) vanishes unless m is even. The surface integrals in the remaining cases can be evaluated from Dirichlet's integral,[8] a special case of which is (for i, j, k even)

$$\iiint_{r\leq a} x^iy^jz^k d\tau = a^{m+3}\Gamma\left(\frac{i+1}{2}\right)\Gamma\left(\frac{j+1}{2}\right)\Gamma\left(\frac{k+1}{2}\right)\Big/\Gamma\left(\frac{m+5}{2}\right).$$

Differentiating this with respect to a and then setting $a=1$, the integral required in (A.3) is obtained. Substituting the result in (A.3), combining numerical factors, and recalling that i, j, and k are all even, it is found that (A.3) reduces to the term $n=m/2$ in (A.1) and the result follows.

It is perhaps of interest to note that the case $K(x) = -e^{-x}/x$ yields a series (A.1) formally equivalent to the operator $(\nabla^2-1)^{-1}$ thus indicating the well-known fact that this particular K is the Green's function of the operator (∇^2-1).

[8] See, for example, R. C. Tolman, *Principles of Statistical Mechanics* (Oxford University Press, Oxford, 1938), p. 656.