Hydrogen Diffusion in the Thermosphere by Monte Carlo Method

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Abstract. The diffusion-limited flow of hydrogen in the thermosphere is obtained by a Monte Carlo calculation. The resulting density distribution has a steeper gradient than current theories predict, even taking omission of gravity into account. The departure is ascribed to increasing nonvalidity of the Chapman-Enskog diffusion coefficient as density changes over a mean free path become significant.

INTRODUCTION

The altitude distribution of minor nonpermanent constituents of the atmosphere and their rate of escape have been treated by several authors, among them Spitzer [1952], Nicolet [1960], and Mange [1961]. In general these authors base their analysis on the hypothesis that a minor constituent exists in hydrostatic equilibrium under its own partial pressure. Mange [1961] and Bates and Patterson [1961] have taken flow into account to arrive at a steeper density gradient for hydrogen than would exist without flow, but hydrostatic equilibrium is still assumed. In an earlier study, not based on this hypothesis, Helge-Petersen [1928] concluded that flowing helium did not attain hydrostatic equilibrium and therefore would decrease in density more rapidly with height than if such equilibrium were attained (see also Mitra [1952, p. 24]).

Recently some indirect evidence has been obtained for the hypothesis that the hydrogen density gradient may be steeper than the current literature predicts. The intensity of night-sky Lyman-α radiation was found in a rocket flight (J. M. Coogan, private communication) to decrease quite rapidly above 200 km, by 30 per cent in 50 km. Such a decrease may reflect a steep density gradient, though the radiative transfer process is a complicated one.

Donahue and Thomas [1963], also, suggest that the Lyman-α radiation appears to originate, in part, by single scattering from an extensive hydrogen envelope which is denser than would be expected on the basis of current estimates of the escaping flux.

These factors motivated the present work, in which the Monte Carlo method is applied to the problem of hydrogen diffusion in the thermosphere, the object being to avoid hypotheses such as that of hydrostatic equilibrium and to simulate actual collisions as closely as possible.

The physical model dealt with here is probably best described by a comparison with the analytic treatments of Nicolet [1960] and others. In such treatments the vertical transport (flow) velocity of a minor constituent of the atmosphere is given by

\[ w = -D \left[ \frac{1}{n_l} \frac{dn_l}{dz} + \frac{m_l g}{kT} \right] + \left( 1 + \alpha_T \right) \frac{dT}{dz} \]  

(1)

where

- \( D \) = diffusion coefficient.
- \( n_l \) = number density of minor constituent.
- \( m_l \) = mass of minor constituent.
- \( \alpha_T \) = thermal diffusion coefficient.

The first term on the right-hand side represents usual diffusion due to concentration gradient. It is the major term and the one essentially calculated here. The second and third represent the effect of gravity, and the fourth, a relatively minor term, represents thermal diffusion.

In the present calculation we do not include the effect of gravity; i.e., between collisions no force is assumed to act on the H atoms. Therefore the flow is limited only by diffusion. The effect of this admittedly severe simplification will be discussed below.

The thermal diffusion term is believed to be automatically present in the calculation, since it results from exchange of momentum between
the H atoms and N₂ molecules during elastic collisions.

**Application of Monte Carlo Method**

We assume for simplicity that the major atmospheric constituent is nitrogen, whose density and temperature depend on altitude as in the Cospar atmosphere. The region dealt with lies between 100 and 500 km. This region is divided into 100 zones; each zone is 4 km thick and has the appropriate Cospar density and temperature.

Hydrogen atoms are introduced at levels of interest and made to undergo collisions with successive nitrogen molecules produced from appropriate Maxwell-Boltzmann (MB) distributions. In each collision, scattering is assumed to be isotropic in the center-of-mass system. The cross section for H-N₂ collisions is not available either experimentally or theoretically, so that an assumption about the interaction potential is necessary. A Lennard-Jones (6-12) potential was assumed here, as will be discussed below. Conveniently, the possibility of formation of an H-N₂ complex may be ruled out because of the high stability of N₂. A detailed description of the process constructed follows.

Each zone \( z \) has an MB distribution of N₂ molecules determined by its temperature \( T \). A hydrogen atom is injected into zone \( z = 1 \), with velocity chosen at random from the MB distribution of this zone. An N₂ molecule is produced with velocity also chosen at random from the MB distribution. The collision cross section, which depends on the relative velocities, is computed, and it leads to an actual path traversed. The collision then occurs, and the resulting velocity of the H atom is obtained. A new N₂ molecule is produced, and the process is repeated. Transitions from one zone to the next are handled much as in Cashwell and Everett [1959]. However, in the upper zones, which have low density, a collision-free path may pass through many zones, so that a procedure was incorporated to add traversal times to the intervening zones.

**The Hydrogen-Nitrogen Collision Process**

(a) As was mentioned above, scattering is assumed to be isotropic in the center-of-mass system. The collision formulas of Cashwell and Everett [1959, pp. 56-60] were used in their general form, i.e. as applied to collisions with a moving target particle rather than a fixed target.

(b) A cross section for collisions between H atoms and N₂ molecules was obtained by means of ‘combining rules,’ which are discussed by Hirschfelder et al. [1954, p. 567]. The intermolecular potential assumed was the Lennard-Jones 6-12 potential,

\[
\phi(r) = 4\varepsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^{6} \right]
\]

where

- \( \sigma = \) distance at which potential is zero.
- \( \varepsilon = \) depth of potential minimum.

For two dissimilar molecules, the parameters \( \sigma \) and \( \varepsilon \), according to the combining rules, are to be taken as

\[
\sigma_{12} = \frac{1}{2} (\sigma_1 + \sigma_2)
\]
\[
\varepsilon_{12} = \sqrt{\varepsilon_1 \varepsilon_2}
\]

where \( \sigma_1, \sigma_2, \varepsilon_1, \varepsilon_2 \) are the parameters that hold for like-molecular collisions of molecules 1 and 2, respectively.

The collision cross section for this intermolecular potential is (in the notation of Hirschfelder et al. [1954])

\[
Q^{(1)} = \pi \sigma_{13}^2 Q^{(1)*}
\]

where the reduced cross section \( Q^{(1)*} \) is a function only of the reduced kinetic energy of relative motion \( g^* \):

\[
g^* = \frac{\mu g^2}{\varepsilon_{12}}
\]

\( g \) being the initial relative speed of the colliding molecules and \( \mu \) the reduced mass.

The quantity \( Q^{(1)*} \) is plotted on page 558 of

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**Fig. 1.** Steady-state fluxes in two regions.
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Hirschfelder et al. [1954]; values were taken directly off this curve for the purpose of the calculation.

The parameters \( \sigma_1 = \sigma_H, \ v_1 = v_H, \ v_2 = v_N, \ v_3 = v_N \) are thus needed. \( \sigma_N \) and \( v_N \) are given in Table I-A of Hirschfelder et al. [1954, p. 111] as \( 3.75 \times 10^{-8} \text{ cm} \) and \( 1.10 \times 10^{-14} \text{ erg} \), respectively. For hydrogen the Lennard-Jones parameters are not given, but the \( H_2 \) potential is well known and is given, for example, by Herzberg [1950]. On the assumption that this actual potential closely resembles a Lennard-Jones potential, the effective values of \( \sigma_H \) and \( v_H \) are 0.39 \( \times 10^{-4} \text{ cm} \) and 7.16 \( \times 10^{-14} \text{ erg} \), respectively. The second value is large because \( H_2 \) is a tightly bound molecule, which means that \( v_{12} = (v_H,v_N)^{1/2} \) is correspondingly large, implying a very large cross section indeed for small relative velocities. This is considered reasonable, however, and in any case the above combining rules seem to be the only ones available for obtaining intermolecular potentials for such dissimilar molecules.

(c) The effect of ignoring atomic oxygen may be briefly discussed at this point. The cross section for H-O collisions must be large, since the OH radical can be formed in three-body collisions. Thus the presence of O will hinder diffusion of H, decreasing the density gradient somewhat. It would be feasible to actually include a given proportion of O atoms in the Monte Carlo calculation to verify this qualitative statement, but this was not done here. Bates and Nicolet [1950] estimate that the density of O may be as high as \( 10^{10} \text{ atoms/cm}^3 \) at 95 km, the O/N ratio increasing with altitude, so that such a refinement might be worth while.

Formation and dissociation of OH can probably be neglected above 100 km, since, according to Bates and Nicolet [1950], its density is only \( 10^9 \text{ atoms/cm}^3 \) at 90 km and drops rapidly.

**Calculation of Transmissions and Fluxes**

The Monte Carlo method can be used to obtain the density distribution of particles diffusing through an extended dense inhomogeneous medium, but it runs into the difficulty that insufficient numbers of particles penetrate the full extent of the medium. Kahn [1956] and others have devised methods of weighting ('splitting,' etc.) to circumvent this difficulty.

The alternative method used here describes the diffusion problem in terms of a transmission and reflection problem, utilizing the fact that transmission coefficients of finite regions of the medium are easily obtainable by the Monte Carlo method. The transmission relations have been essentially derived (in more complex form) in fields like multilayer optics [Heavens, 1955].

Consider two 'slab' mediums in contact, whose transmission coefficients separately are \( T_1, T_2 \); i.e., out of a flux of \( N_\nu \) particles cm\(^{-2}\) sec\(^{-1}\) injected into the first slab, a flux \( T_1N_\nu \) will be transmitted. Of this, in turn, a fraction \( T_2 \) will be further transmitted through the second slab. There are multiply reflected and transmitted fluxes (Figure 1), and if the system is in a steady state (as much flux leaving the system as entering), we find, when these partial fluxes are totaled, that: (a) the transmitted flux at plane \( i = 3 \) is given by

\[
N_\nu T_1 T_2 / (1 - R_1 R_2)
\]

where \( R_1 = 1 - T_1; R_2 = 1 - T_2 \), so that the over-all transmission is given by \( T_1 T_2 / (1 - R_1 R_2) \); and (b) the forward flux at plane \( i = 2 \) is

\[
N_\nu T_1 / (1 - R_1 R_2)
\]

The backward flux at plane \( i = 2 \) is

\[
N_\nu T_2 / (1 - R_1 R_2)
\]

so that the flux difference at this plane is

\[
N_\nu T_1 T_2 / (1 - R_1 R_2)
\]

Table 1

<table>
<thead>
<tr>
<th>Region, km</th>
<th>Transmission</th>
</tr>
</thead>
<tbody>
<tr>
<td>140-152</td>
<td>0.0040 ± 0.0015</td>
</tr>
<tr>
<td>152-168</td>
<td>0.0074 ± 0.0026</td>
</tr>
<tr>
<td>168-184</td>
<td>0.0116 ± 0.0034</td>
</tr>
<tr>
<td>184-200</td>
<td>0.0183 ± 0.0037</td>
</tr>
<tr>
<td>200-296</td>
<td>0.0095 ± 0.0037</td>
</tr>
<tr>
<td>296-496</td>
<td>0.0231 ± 0.0061</td>
</tr>
</tbody>
</table>

Fig. 2. Medium consisting of \( n \) regions.
Fig. 3. Density distributions of assumed main nitrogen atmosphere (dashed line), Bates-Patterson hydrogen atmosphere (dash-dot line), hydrogen atmosphere derived here (solid line); \( k \) is a different (unspecified) parameter for each curve.

\[
Nv_i T_i T_j/(1 - R_i R_j)
\]

in agreement with \((a)\).

Now consider a slab medium of thickness \( l \) in the \( z \) direction. Divide it by planes into \( n \) regions, as in Figure 2, with the planes labeled by the index \( i \). The transmission coefficients \( T_1, T_2, \ldots, T_n \) of the regions are assumed to be known (by injecting particles into one such region at a time).

The combined transmission of the last two regions is

\[
T_1 = T_{n-1} T_n/(1 - R_{n-1} R_n)
\]

The combined transmission of the last three regions is then

\[
T_2 = T_{n-2} T_1/(1 - R_{n-2} R_1)
\]

where \( R_i = 1 - T_i \).

By continuing this iteration process, the overall transmission is obtained:

\[
T = T_{n-1}
\]

The forward and backward fluxes at any given plane \( i \) can also be obtained by computing the combined transmissions of the regions to the left and to the right of the plane and by applying the two-region formulas above.

Let these fluxes at the plane \( i \) be denoted by \( n_i^+ \bar{v}_i \) and \( n_i^- \bar{v}_i \), respectively. The total density at the plane \( i \) is \( (n_i^+ + n_i^-) \); thus the total flux \( \bar{v}_i(n_i^+ + n_i^-) \) must be divided by the mean particle velocity \( \bar{v}_i \), which is not obtainable from the transmission measurements alone. To find \( \bar{v}_i \) we must relate the transmission description of
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TABLE 2

<table>
<thead>
<tr>
<th>Altitude, km</th>
<th>i</th>
<th>$F_i^+$</th>
<th>$F_i^-$</th>
<th>$S_i = (F_i^+ + F_i^-)$</th>
<th>$M_i = (F_i^+ - F_i^-)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>140</td>
<td>1</td>
<td>1.0000</td>
<td>0.9985</td>
<td>1.9985</td>
<td>0.0015</td>
</tr>
<tr>
<td>152</td>
<td>2</td>
<td>0.6281</td>
<td>0.6266</td>
<td>1.255</td>
<td>0.0015</td>
</tr>
<tr>
<td>168</td>
<td>3</td>
<td>0.4278</td>
<td>0.4263</td>
<td>0.854</td>
<td>0.0015</td>
</tr>
<tr>
<td>184</td>
<td>4</td>
<td>0.3005</td>
<td>0.2990</td>
<td>0.600</td>
<td>0.0015</td>
</tr>
<tr>
<td>200</td>
<td>5</td>
<td>0.2204</td>
<td>0.2189</td>
<td>0.439</td>
<td>0.0015</td>
</tr>
<tr>
<td>296</td>
<td>6</td>
<td>0.06466</td>
<td>0.06316</td>
<td>0.128</td>
<td>0.0015</td>
</tr>
<tr>
<td>496</td>
<td>7</td>
<td>0.0015</td>
<td>0.0000</td>
<td>0.0015</td>
<td>0.0015</td>
</tr>
</tbody>
</table>

the system to its description as a diffusing process.

A given atom, moving in a random walk, undergoes a displacement $\xi$ along the $z$ axis in a time $\Delta t$. $\xi^2$, the mean square of such displacements for all atoms originating in a volume $\Delta V$, is related [Kennard, 1938] to the diffusion coefficient $D_i$ by

$$D_i = \lim_{\Delta t \to 0} \frac{\xi^2}{\Delta t} \quad (3)$$

The mean displacement $\bar{\xi}_i$ at the plane $i$ also gives the velocity $\bar{\xi}_i$ required above; i.e.,

$$\bar{v}_i = \lim_{\Delta i \to 0} \frac{\bar{\xi}_i}{\Delta t} \quad (4)$$

The total density at plane $i$ is then given by

$$n_i^+ + n_i^- = \frac{(n_i^+ + n_i^-)\bar{v}_i}{\bar{v}_i} = f_i(Nv_i, T_1, T_2, \ldots, T_n) \quad (5)$$

where $f_i$ is constructed as described above (combining transmissions to right and to left of the given plane, etc., to arrive at the sum of forward and backward fluxes).

The mean time $t_i = 1/\bar{v}_i$ spent per atom in a unit volume at plane $i$ is more directly gotten in practice than $\bar{v}_i$, so that (5) is used in the form

$$n_i = t_i(n_i^+ + n_i^-)\bar{v}_i \quad (5a)$$

In the actual computer program, atoms injected at each plane are always picked from an isotropic and Maxwellian distribution, a distribution that cannot strictly hold in the actual diffusion process. The error resulting from this approximation should be small, however.

Results

(a) Transmissions and fluxes. Several thousand H atoms were successively injected at altitudes of 140, 152, 168, 184, 200, 296, 496 km, corresponding respectively to planes $i = 1, 2, 3, 4, 5, 6, 7$. The resulting transmissions are given in Table 1. These results were used to find values for forward and backward fluxes, which are given, with their sums and differences, in Table 2.

The over-all transmission of the atmosphere from 140 to 500 km is therefore 0.0015, which is also the net upward flux when unit flux is injected at 140 km. (Extra significant figures included in the table entries are retained for calculational purposes and are not intended to represent unwarranted precision).

(b) Diffusion times and relative densities. At each of the planes $i$, H atoms were successively injected and timed as they diffused through a 4-km distance. The mean times and resulting relative densities are presented in Table 3.

It is to be noted that $t_i > t_o$, which appears to be a real effect, explainable physically as due to freedom of the atoms to travel horizontally for long distances without collisions.

TABLE 3

<table>
<thead>
<tr>
<th>Altitude, km</th>
<th>$i$</th>
<th>Time to Diffuse 4 km, sec</th>
<th>Relative Density $t_iS_i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>140</td>
<td>1</td>
<td>68.1</td>
<td>136</td>
</tr>
<tr>
<td>152</td>
<td>2</td>
<td>25.5</td>
<td>32</td>
</tr>
<tr>
<td>168</td>
<td>3</td>
<td>16.4</td>
<td>14</td>
</tr>
<tr>
<td>184</td>
<td>4</td>
<td>11.5</td>
<td>6.9</td>
</tr>
<tr>
<td>200</td>
<td>5</td>
<td>9.0</td>
<td>3.95</td>
</tr>
<tr>
<td>296</td>
<td>6</td>
<td>2.1</td>
<td>0.27</td>
</tr>
<tr>
<td>496</td>
<td>7</td>
<td>4.4</td>
<td>0.0066</td>
</tr>
</tbody>
</table>
These relative densities are plotted in Figure 3, together with the main nitrogen atmosphere and the Bates-Patterson hydrogen atmosphere [Bates and Patterson, 1961]. The Bates-Patterson atmosphere, constructed for an exosphere temperature of 1500°K, is representative of the hydrogen distributions obtained by means of equation 1. The exponent $k$ of the density scale is not specified because the absolute densities are not necessary here; $k$ takes a different value for each curve.

**DISCUSSION OF RESULTS**

The Monte Carlo curve represents diffusion through a slab medium bounded by vacuum on both sides. The density gradient in vacuum is zero (free particles), so that the density gradient just within the medium must be zero also. This will hold for a distance into the medium that is of the order of a mean free path, i.e. until significant scattering occurs. This distance is only about 0.04 km at the lower boundary (140 km) but about 25 km at the upper boundary (500 km). The MC curve is based on seven widely separated points; therefore it does not show the change of slope that should occur within the border regions, but for accuracy it is terminated at 470 km.

This density curve has a much steeper slope than that of Bates and Patterson. We first ask whether the net flow relation

$$f = D_i \frac{dn(H)}{dz},$$

is satisfied for this curve, where $D_i$ is the diffusion coefficient at plane $i$, computed from the applicable formula of Hirschfelder et al. [1954, p. 539]:

$$D_{12} = 0.00268 \left[ \frac{T^3(M_1 + M_2)/M_1 M_2}{p \sigma_{12}^2 \Omega_{12}^{(1.1)} T_{12}^*} \right]$$

where

- $p = \text{pressure in atmospheres.}$
- $T_{12}^* = kT/\epsilon_{12}$.
- $\sigma_{12}, \epsilon_{12}/k$ are potential parameters in angstroms and degrees Kelvin, respectively.
- $\Omega_{12}^{(1.1)}$ is an integral given in Table 1 [Hirschfelder et al., 1954, p. 1126] as a function of $T$.

The product $D_i \frac{dn(H)}{dz}$, is given in Table 4. It is seen to decrease more and more rapidly with altitude, instead of remaining constant as we would normally expect.

The explanation for this nonconstancy is believed to be the fact that the Chapman-Enskog kinetic theory, from which the diffusion coefficient is derived, becomes invalid when density and temperature changes are appreciable over a mean free path [Hirschfelder et al., 1954, pp. 18-20]. An effective diffusion coefficient in such a case does not appear to have been derived in the literature. The situation has been dealt with to some extent in the field of gas dynamics [Schaaf and Chambre, 1961], which we may qualitatively discuss to show its bearing on the atmospheric problem.

The three main flow regimes of gas dynamics are the continuum flow regime, the free molecular flow regime, and the transition regime lying between them. The regimes are based on the range of values of a Knudsen number $K$, which is defined as the ratio of mean free path to some significant dimension of the flow field. Very roughly, the continuum regime corresponds to $K \ll 1$, the free molecule regime to $K \geq 1$, and the transition regime to intermediate values of

**TABLE 5**

<table>
<thead>
<tr>
<th>$i$</th>
<th>$l_i, \text{km}$</th>
<th>$h_i, \text{km}$</th>
<th>$K_i = l_i/h_i$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.036</td>
<td>25</td>
<td>0.0014</td>
</tr>
<tr>
<td>2</td>
<td>0.10</td>
<td>33</td>
<td>0.0030</td>
</tr>
<tr>
<td>3</td>
<td>0.12</td>
<td>38</td>
<td>0.0031</td>
</tr>
<tr>
<td>4</td>
<td>0.21</td>
<td>39</td>
<td>0.0054</td>
</tr>
<tr>
<td>5</td>
<td>0.35</td>
<td>41</td>
<td>0.0085</td>
</tr>
<tr>
<td>6</td>
<td>2.0</td>
<td>53</td>
<td>0.038</td>
</tr>
<tr>
<td>7</td>
<td>25.5</td>
<td>83</td>
<td>0.31</td>
</tr>
</tbody>
</table>
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K. Though this division of gas dynamics pertains actually to flow past solid surfaces, it appears relevant in the present study to consider density scale height as a significant dimension. Knudsen numbers $K_i = l_i/h$ are listed in Table 5, using mean free paths $l_i$ taken from the computer calculation. $K$ becomes appreciable compared with unity as altitude increases, so that conditions depart from those of the continuum regime and become those of the transition regime. The diffusion coefficient of the continuum regime will not then be valid.

These observations do not constitute proof of the suggested explanation, of course, but only an argument for its plausibility.

A direct measure of the departure of the diffusion coefficient formula from validity is afforded by the entries of Table 4; since the product $D \cdot \frac{dn(H)}{dz}$ decreases by a factor of 11.5 between the altitudes of 140 and 296 km, $D$ at 296 km is too small by this factor.

As was noted earlier, gravity has been neglected. Its inclusion would decrease net flow, because molecular escape from the critical level near 500 km would then be an important factor in controlling the over-all escape rate. This factor is taken into account by Bates and Patterson, who assume a particular value for the molecularly limited escape flux. The slope of their curve is therefore correctly less than that of the MC curve at lower altitudes, where the effective diffusion coefficient is (approximately) the same for both. At higher altitudes, however, inclusion of this factor would not decrease the slope of the MC curve to that of Bates and Patterson, because of the increasing difference in effective diffusion coefficient. The true curve should then have a slope intermediate between that of Bates and Patterson and of the present curve.

The assumption adopted at the start, that the diffusing hydrogen is not in hydrostatic equilibrium, appears to play a lesser role in steepening the density curve than does the non-validity of the diffusion coefficient, though it is difficult to particularize its effect on the final result.

An approximation was made in setting density of the main atmosphere equal to zero above 500 km. Collisions above this altitude would probably have a negligible effect on the diffusion process.

CONCLUSIONS

The Monte Carlo calculation of diffusion of hydrogen through a nitrogen atmosphere results in a higher gradient of hydrogen density than current theories predict, even allowing for the fact that gravity was not included. The departure is ascribed to increasing nonvalidity of the usual diffusion coefficient with altitude, because the mean free path becomes significantly large compared with density scale height. The usual (calculated) diffusion coefficient is estimated to be an order of magnitude too low at 300-km altitude.

At low altitudes the true curve should have the slope of hydrogen distributions calculated in the usual way, e.g. that of Bates and Patterson. At higher altitudes, however, the true curve's slope will be intermediate between that of the present curve and that of Bates and Patterson.

Since the diffusion-limited flow is found to be higher than in other calculations, the over-all escape flux of hydrogen will also be higher. This could account for the denser hydrogen envelope hypothesized by Donahue and Thomas [1963].

Acknowledgments. I thank J. E. Kupperian for suggesting the problem and for valuable discussions in the course of the work. C. Wade programmed the calculation and made the many necessary runs. Talks with C. L. Wolff were also appreciated.

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(Manuscript received July 11, 1963; revised October 3, 1963.)