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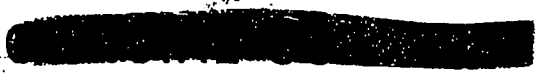
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COLLISION KINETICS IN A SHOCK WAVE

by

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Chief, Declassification Branch

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PHYSICS AND MATHEMATICS



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## Collision Kinetics in a Shock Wave \*

## ABSTRACT

A calculation of the spatial distribution of energetic molecular collisions within a plane shock transition is made, using the Mott-Smith solution of the Boltzmann equation for a shock wave. The results are applied to polytropic gases. If a shock leaves a material with a considerable mass velocity but a moderate temperature, then the transition itself may contain very many high energy collisions. This condition is shown not to obtain for gases; it may possibly occur in liquids or solids.

\* The material in this report was given a limited initial distribution as Report GMX-R-102.

  
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## Collision Kinetics in a Shock Wave


I. Qualitative Discussion

The elementary shock theory requires modification when the material is altered in composition by passage of the shock, that is when the shock produces a chemical transformation or ionization. These processes occur in certain fixed proportions of the collisions between pairs of molecules for which the kinetic energy relative to the center of mass is greater than the chemical activation energy or the ionization potential respectively. A calculation of the rates of such effective collisions has been made in the rigid sphere approximation using the Mott-Smith solution of the Boltzmann equation for a shock wave<sup>1</sup>. The calculation yields information about the possible rates of processes involving inelastic collisions throughout the submicroscopic structure of a shock wave. Certain conclusions and speculations can thus be made about the detailed mechanism of initiation of chemical reaction in steady detonation of homogeneous explosives and about the production of dissociation and ionization by shocks in inert gases.

The qualitative results can be summarized briefly. If the shock produces a moderate mass velocity and considerable temperature change (as in a gas with ratio of specific heats,  $\gamma$ , not

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<sup>1</sup>H. M. Mott-Smith, The Solution of the Boltzmann Equation for a Shock Wave, Phys.Rev. 82, 885(1951)



too near one) then the thin region of the shock transition itself contains a negligible number of especially energetic collisions. One can calculate the state behind the shock in the elementary way and simply estimate the importance of composition changing processes using the equilibrium Boltzmann distribution for this state. But, if the shock produces a fairly high mass velocity and only a moderate temperature change (as in a gas of  $\gamma$  very near one), then it turns out that there can be many-fold more effective collisions within the few mean-free-paths which have been shown to constitute the shock itself<sup>1,2,3</sup> than in an equivalent region behind it. The increase can be estimated using the results of the present calculation.

It is possible to understand this behavior qualitatively. The Mott-Smith molecular velocity distribution for a shock consists of two Maxwell distributions having the temperatures of the shocked and unshocked material respectively and moving in bulk relative to one another with velocity equal to the mass velocity jump across the shock. Within the shock transition region the distribution is an appropriately variable linear combination of these two. Very roughly, a representative collision in the shock between two molecules of mass  $m$ , one from each of the two distributions, takes place at least with relative velocity  $u$ , the mass velocity change

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<sup>2</sup>R. Becker, Z.Physik, 8, 321(1923).

<sup>3</sup>L. H. Thomas, J.Chem.Phys., 12, 449(1944).

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across the shock. The kinetic energy of such a collision relative to the center of mass is  $\frac{1}{4}m\bar{u}^2$ . On the other hand the average relative energy of collision for a Maxwellian gas at temperature  $T$  is<sup>4</sup>  $2kT$ . Thus if  $\frac{1}{4}m\bar{u}^2/2kT$  is appreciably greater than one behind the shock then effective collisions are likely to be significantly numerous within the shock transition region.

## II. Calculation of the Number of Effective Collisions in a Shock.

Assume a one-dimensional stationary shock in a pure gas consisting of smooth rigid spherical molecules of mass  $m$ . The gas is streaming into the shock in the positive  $x$  direction with supersonic velocity  $u_\alpha$  and out of the shock with subsonic velocity  $u_\beta$ . Mott-Smith assumes<sup>1</sup> for the relative frequency of molecular velocity  $\vec{c}$  at position  $x$  the distribution function

$$f(x, \vec{c}) = f_\alpha(x, \vec{c}) + f_\beta(x, \vec{c}) \quad (1)$$

with

$$f_\alpha(x, \vec{c}) = n_\alpha(x) \left[ \frac{m}{2kT_\alpha} \right]^{\frac{3}{2}} \exp \left[ -\frac{m(\vec{c} - \vec{i}u_\alpha)^2}{2kT_\alpha} \right] \quad (2)$$

and similarly for  $f_\beta$  with  $\beta$  replacing  $\alpha$  throughout. Here  $n$  is molecular density,  $T$  temperature,  $k$  the Boltzmann Constant and  $\vec{i}$  is a unit vector in the positive  $x$  direction. Subscripts  $\alpha$  and  $\beta$  denote quantities characteristic of the two simple Maxwell functions from which the total shock distribution is constructed. The density

<sup>4</sup>Chapman and Cowling-The Mathematical Theory of Non-Uniform Gases-p. 93.

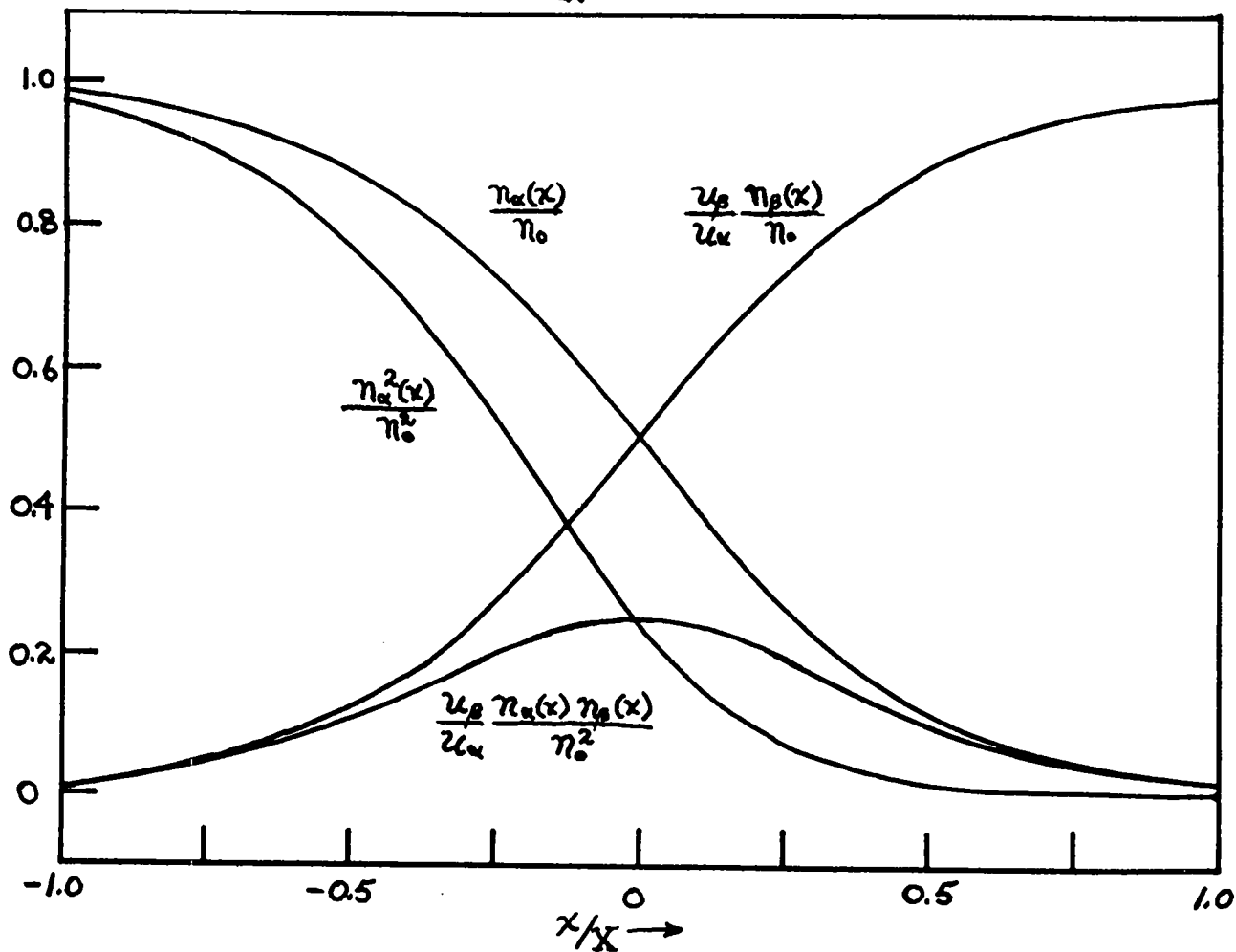


Figure 1 - Spatial dependence of the shock-wave distribution function.

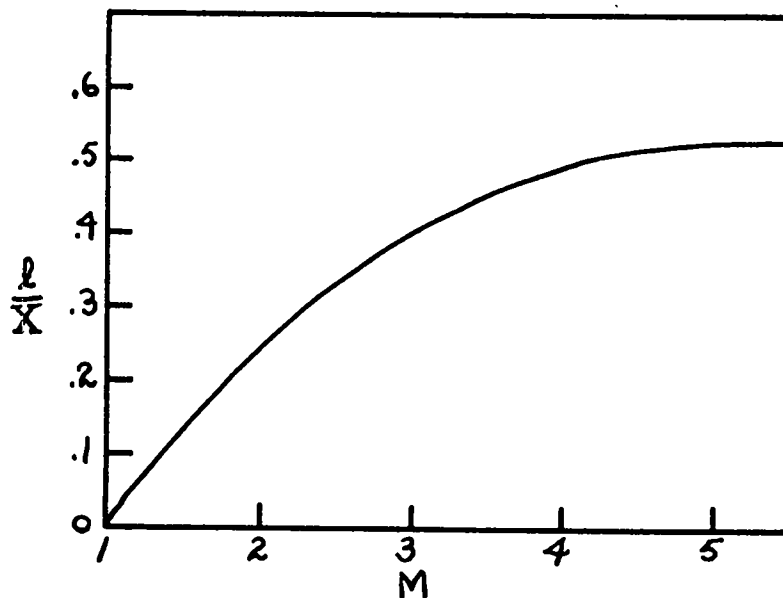


Figure 2 - Ratio of mean free path to shock-wave thickness as a function of Mach number. (Sketched from Ref. 1)

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variations along  $x$  are found by Mott-Smith to be

$$n_\alpha(x) = \frac{n_0}{1 + e^{4x/X}} \quad n_\beta(x) = \frac{u_\beta}{u_\beta} \frac{n_0}{1 + e^{4x/X}} \quad (3)$$

where  $n_0 = n_\alpha(-\infty)$  is the density in the supersonic stream far from the shock and  $X$  is the shock thickness defined in the usual way as  $X = [n(+\infty) - n(-\infty)] / |dn/dx|_{\max}$ . The ratio of  $X$  to the mean-free-path in the unshocked gas,  $\ell$ , is found by Mott-Smith to be a complicated function of the shock strength characterized by the Mach number  $M$  and the ratio of specific heats  $\gamma$ , though nearly independent of the latter. The functions  $n_\alpha(x)$  and  $n_\beta(x)$  are sketched in Figure 1, and the dependence of  $X/\ell$  on  $M$  is shown in Figure 2.

The rate of collisions in unit volume at position  $x$  between a pair of molecules 1 and 2, having relative velocity  $g = |\vec{c}_2 - \vec{c}_1|$  equal to or greater than some specified velocity  $g_0$  is given by<sup>5</sup>

$$Z(x, g_0) = \pi \sigma^2 \iint_{g \geq g_0} f(x, \vec{c}_1) \cdot f(x, \vec{c}_2) \cdot g \cdot d\vec{c}_1 d\vec{c}_2 \quad (4)$$

where  $\sigma$  is the effective molecular diameter. Inserting (1) and (2) in (4)

$$Z(x, g_0) = \sigma^2 n_\alpha^2 g_0 \Phi_\alpha + \sigma^2 n_\alpha n_\beta (u_\alpha - u_\beta) \Psi_{\alpha\beta} + \sigma^2 n_\beta n_\alpha (u_\beta - u_\alpha) \Psi_{\beta\alpha} + \sigma^2 n_\beta^2 g_0 \Phi_\beta \quad (5)$$

where

$$\Phi_\alpha = \frac{\pi^2}{8 g_0 c_\alpha^2} \iint_{g \geq g_0} e^{-\frac{(\vec{c}_1 - \vec{u}_\alpha)^2}{2c_\alpha^2} - \frac{(\vec{c}_2 - \vec{u}_\alpha)^2}{2c_\alpha^2}} \cdot g \cdot d\vec{c}_1 d\vec{c}_2 \quad (6)$$

<sup>5</sup> See reference 4, p.90 ff.





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and

$$\Psi_{\alpha\beta} = \frac{\pi^2}{8(u_\alpha - u_\beta) c_\alpha^3 c_\beta^3} \iint_{\mathfrak{g} > \mathfrak{g}_0} e^{-(\vec{c}_1 - \vec{u}_\alpha)^2 / 2c_\alpha^2 - (\vec{c}_2 - \vec{u}_\beta)^2 / 2c_\beta^2} \mathfrak{g} \cdot d\vec{c}_1 d\vec{c}_2 \quad (7)$$

in which  $c_\alpha^2 = kT_\alpha/m$ ,  $c_\beta^2 = kT_\beta/m$ . Similar expressions for  $\Phi_\beta$  and  $\Psi_{\beta\alpha}$  are obtained by interchanging  $\alpha$  and  $\beta$  in (6) and (7). From symmetry considerations,  $\Psi_{\alpha\beta} = -\Psi_{\beta\alpha}$ . To evaluate these integrals we make in (6) the substitution

$$\begin{aligned} \vec{c}_1 &= -\frac{1}{2} \vec{\mathfrak{g}} + \vec{\mathfrak{C}} \\ \vec{c}_2 &= \frac{1}{2} \vec{\mathfrak{g}} + \vec{\mathfrak{C}} \end{aligned} \quad (8)$$

and in (7) the substitution

$$\begin{aligned} \vec{c}_1 &= -\frac{c_\alpha^2}{c_\alpha^2 + c_\beta^2} \vec{\mathfrak{g}} + \vec{\mathfrak{C}} \\ \vec{c}_2 &= \frac{c_\beta^2}{c_\alpha^2 + c_\beta^2} \vec{\mathfrak{g}} + \vec{\mathfrak{C}} \end{aligned} \quad (9)$$

These transformations verify  $|\vec{c}_2 - \vec{c}_1| = \mathfrak{g}$ , eliminate cross terms in the variables of integration from the exponential, and have Jacobians equal to unity. The results of the substitutions are

$$\Phi_\alpha = \frac{\pi^2}{8\mathfrak{g}_0 c_\alpha^3} \iint_{\mathfrak{g} > \mathfrak{g}_0} \mathfrak{g} \exp \left[ \frac{\mathfrak{g}^2}{4c_\alpha^2} + \frac{\mathfrak{C}^2}{c_\alpha^2} + \frac{2\mathfrak{g} \cdot \vec{\mathfrak{C}}}{c_\alpha^2} + \frac{2\mathfrak{g}_0 \vec{\mathfrak{C}} \cdot \vec{\mathfrak{I}}}{c_\alpha^2} \right] d\mathfrak{g} d\vec{\mathfrak{C}} \quad (10)$$

and

$$\begin{aligned} \Psi_{\alpha\beta} = \frac{\pi^2}{8(u_\alpha - u_\beta) c_\alpha^3 c_\beta^3} \iint_{\mathfrak{g} > \mathfrak{g}_0} \mathfrak{g} \exp \left[ \frac{\mathfrak{g}^2}{2(c_\alpha^2 + c_\beta^2)} + \frac{1}{2} \left( \frac{1}{c_\alpha^2} + \frac{1}{c_\beta^2} \right) \mathfrak{C}^2 + \frac{\mathfrak{g} \cdot \vec{\mathfrak{C}}}{c_\alpha^2} + \frac{\mathfrak{g} \cdot \vec{\mathfrak{C}}}{c_\beta^2} \right. \\ \left. + \frac{(u_\alpha - u_\beta) \vec{\mathfrak{C}} \cdot \vec{\mathfrak{I}}}{c_\alpha^2 + c_\beta^2} - \left( \frac{u_\alpha}{c_\alpha^2} + \frac{u_\beta}{c_\beta^2} \right) \vec{\mathfrak{C}} \cdot \vec{\mathfrak{I}} \right] d\mathfrak{g} d\vec{\mathfrak{C}} \end{aligned} \quad (11)$$

The expressions for  $\Phi$  and  $\Psi$  are now products of single integrals each of the form

$$J_m(n_0; a, b) = \int_{n_0}^{\infty} \pi^n e^{-a^2 n^2 - b \vec{n} \cdot \vec{\mathfrak{I}}} d\vec{n} = \int_0^{\infty} \int_0^{2\pi} \int_0^{\pi} \pi^n e^{-a^2 n^2 - b n \cos \theta} n^2 \sin \theta d\theta d\phi dn \quad (12)$$





In particular we need the following results which can be obtained by straightforward integration

$$\mathcal{D}_0(a, t) = \left(\frac{\pi t}{a}\right)^3 e^{(t/2a)^2} \tag{13}$$

$$\mathcal{D}_1(\pi_0; a, 0) = \frac{2\pi}{a^2} e^{-a^2 \pi_0^2} [a^2 \pi_0^2 + 1] \tag{14}$$

$$\mathcal{D}_1(\pi_0; a, t) = \frac{\pi}{a^2 t} e^{(t/2a)^2} \left[ \left[1 + \left(\frac{t}{2a}\right)^2\right] \left\{ \mathcal{E}_f\left(a\pi_0 + \frac{t}{2a}\right) - \mathcal{E}_f\left(a\pi_0 - \frac{t}{2a}\right) \right\} + \left(a\pi_0 + \frac{t}{2a}\right) e^{-\left(a\pi_0 + \frac{t}{2a}\right)^2} - \left(a\pi_0 - \frac{t}{2a}\right) e^{-\left(a\pi_0 - \frac{t}{2a}\right)^2} \right] \tag{15}$$

where

$$\mathcal{E}_f x = \int_0^x e^{-s^2} ds \tag{16}$$

Using (12)-(15) in (10) and (11) and letting

$$\eta^2 = \frac{(u_\alpha - u_\beta)^2}{2(c_\alpha^2 + c_\beta^2)} = \frac{m(u_\alpha - u_\beta)^2}{2k(T_\alpha + T_\beta)} \quad ; \quad w^2 = \frac{g_0^2}{2(c_\alpha^2 + c_\beta^2)} = \frac{mg_0^2}{2k(T_\alpha + T_\beta)} \tag{17}$$

$$w_\alpha^2 = \frac{g_0^2}{4c_\alpha^2} = \frac{mg_0^2}{4kT_\alpha} \quad ; \quad w_\beta^2 = \frac{g_0^2}{4c_\beta^2} = \frac{mg_0^2}{4kT_\beta} \tag{18}$$

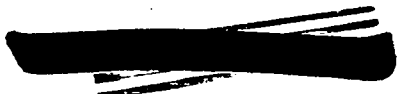
We get after some simplification

$$\begin{aligned} \Phi_\alpha &= \Phi(w_\alpha) \quad ; \quad \Phi_\beta = \Phi(w_\beta) \\ \Phi(w) &= 2\pi^{1/2} e^{-w} \left[ \frac{1}{w} + w \right] \end{aligned} \tag{19}$$

$$\begin{aligned} \Psi_{\alpha\beta} &= -\Psi_{\beta\alpha} = \Psi(w, v) \\ \Psi(w, v) &= \frac{\pi^{1/2}}{2v^2} \left[ \left[1 + v^2\right] \left\{ \mathcal{E}_f(w+v) - \mathcal{E}_f(w-v) \right\} + (w+v) e^{-(w-v)^2} - (w-v) e^{-(w+v)^2} \right] \end{aligned} \tag{20}$$

The functions  $\Phi$  and  $\Psi$  are plotted in Figure 3.

If the relative kinetic energy effective for some particular process is  $E$ , then



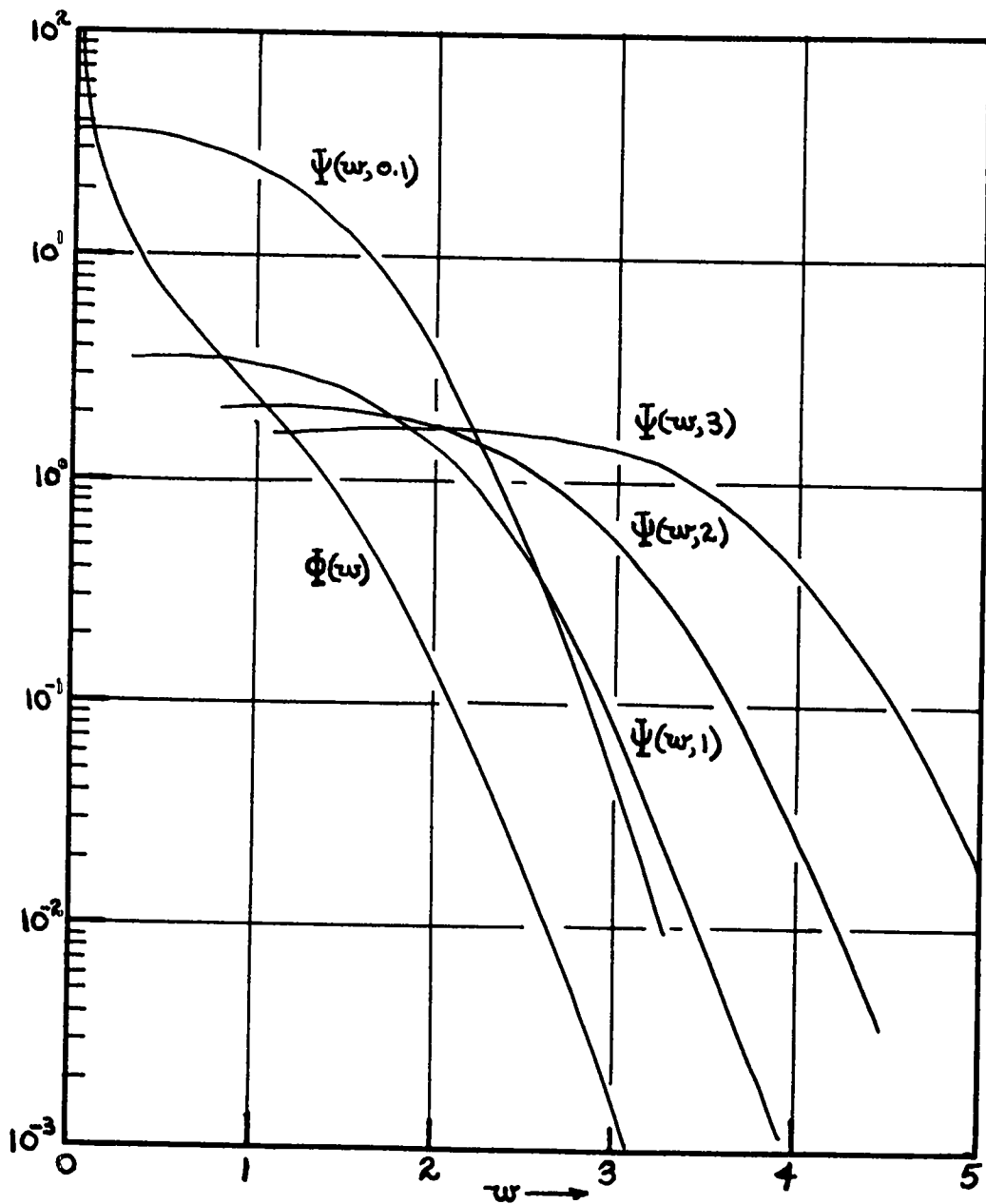


Figure 3 - The functions  $\Phi(w)$  and  $\Psi(w, v)$ .

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$$m g_0 = 4E ; \quad w^2 = \frac{2E}{k(\tau_\alpha + \tau_\beta)} ; \quad w_\alpha^2 = \frac{E}{k\tau_\alpha} ; \quad w_\beta^2 = \frac{E}{k\tau_\beta} \quad (21)$$

We can then calculate the rate of effective collisions in any part of the shock, using (5) together with the relations (17)-(21).

It is convenient to compare the collision rate at an  $x$  of interest with the collision rate far behind the shock. Using (3), (5) and (17)-(20) we find

$$\frac{Z(x, g_0)}{Z(\infty, g_0)} = \frac{n_\alpha^2(-x)}{n_0^2} + \frac{2n_\alpha(x) n_\alpha(-x) u_\beta v \Psi(w, v)}{n_0^2 u_\alpha w \Phi(w_\beta)} + \frac{n_\alpha^2(x) u_\beta^2 \Phi(w_\alpha)}{n_0^2 u_\alpha^2 \Phi(w_\beta)} \quad (22)$$

The first and last terms in this expression are the contribution of collisions with relative velocity greater than  $g_0$ , due to the intrinsic thermal motion of the shocked and unshocked molecules respectively. The central term is the contribution of high velocity contacts that arises in the complex collision process through which one Maxwell distribution is transformed into another by means of a shock. If we integrate this central term only, over all  $x$ , we get a measure of the total effectiveness of the shock transition in producing high velocity collisions

$$\tau = \frac{2 u_\beta v \Psi(w, v)}{u_\alpha w \Phi(w_\beta)} \int_{-\infty}^{+\infty} \frac{n_\alpha(x) n_\alpha(-x)}{n_0^2} dx \quad (23)$$

Inserting (3) in (23) and performing the integration we find

$$\tau = \frac{\int x u_\beta v \Psi(w, v)}{2 u_\alpha w \Phi(w_\beta)} \quad (24)$$

The quantity  $\tau$  can be interpreted as the thickness of material far behind the shock which produces collisions of relative velocity greater than  $g_0$  at the same rate as produced by the mass velocity jump across the shock.

### III. Application to Polytropic Gases

If a plane, steady shock of Mach number  $M$  is propagating in a polytropic gas having sound velocity  $a_0$ , defining

$$M = u_\alpha / a_0 \quad (25)$$

$$\mu^2 = \frac{\gamma - 1}{\gamma + 1} \quad (26)$$

It can be shown that<sup>6</sup>

$$a_0^2 = \frac{\gamma k T_\alpha}{m} \quad ; \quad u_\alpha - u_\beta = a_0 (1 - \mu^2) \frac{M^2 - 1}{M} \quad (27)$$

$$\frac{p_\beta}{p_\alpha} = (1 + \mu^2) M^2 - \mu^2 \quad ; \quad \frac{u_\beta}{u_\alpha} = 1 - (1 - \mu^2) \frac{M^2 - 1}{M^2} \quad (28)$$

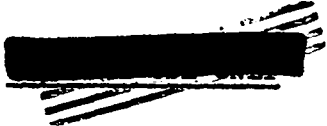
$$\frac{T_\beta}{T_\alpha} = \frac{p_\beta}{p_\alpha} \frac{u_\beta}{u_\alpha} = [(1 + \mu^2) M^2 - \mu^2] \left[ 1 - (1 - \mu^2) \frac{M^2 - 1}{M^2} \right] \quad (29)$$

$$\nu^2 = \frac{m(u_\alpha - u_\beta)^2}{2k(T_\alpha + T_\beta)} = \frac{(1 - \mu^2)(M^2 - 1)^2}{2M^2 + 2[(1 + \mu^2)M^2 - \mu^2][M^2 - (1 - \mu^2)(M^2 - 1)]} \quad (30)$$

where  $p_\beta/p_\alpha \geq 1$  is the pressure ratio across the shock, and all other symbols are as previously defined. These formulae are certainly inaccurate for real gases for the stronger shocks which will be discussed.

The magnitude of  $\nu^2$ , which is almost the quantity  $\frac{1}{4} m u^2 / 2kT$  of Section I, largely determines the number of high energy collisions produced in the shock transition. In Figure 4,  $\nu^2$  is plotted as a function of  $M$  for several values of  $\gamma$ , as given by (30). Clearly  $\nu^2$  is appreciably greater than one only for fairly strong shocks in gases having  $\gamma$  not too different from unity.

<sup>6</sup>Courant and Friedrichs-Supersonic Flow and Shock Waves-Chapter IIIC.



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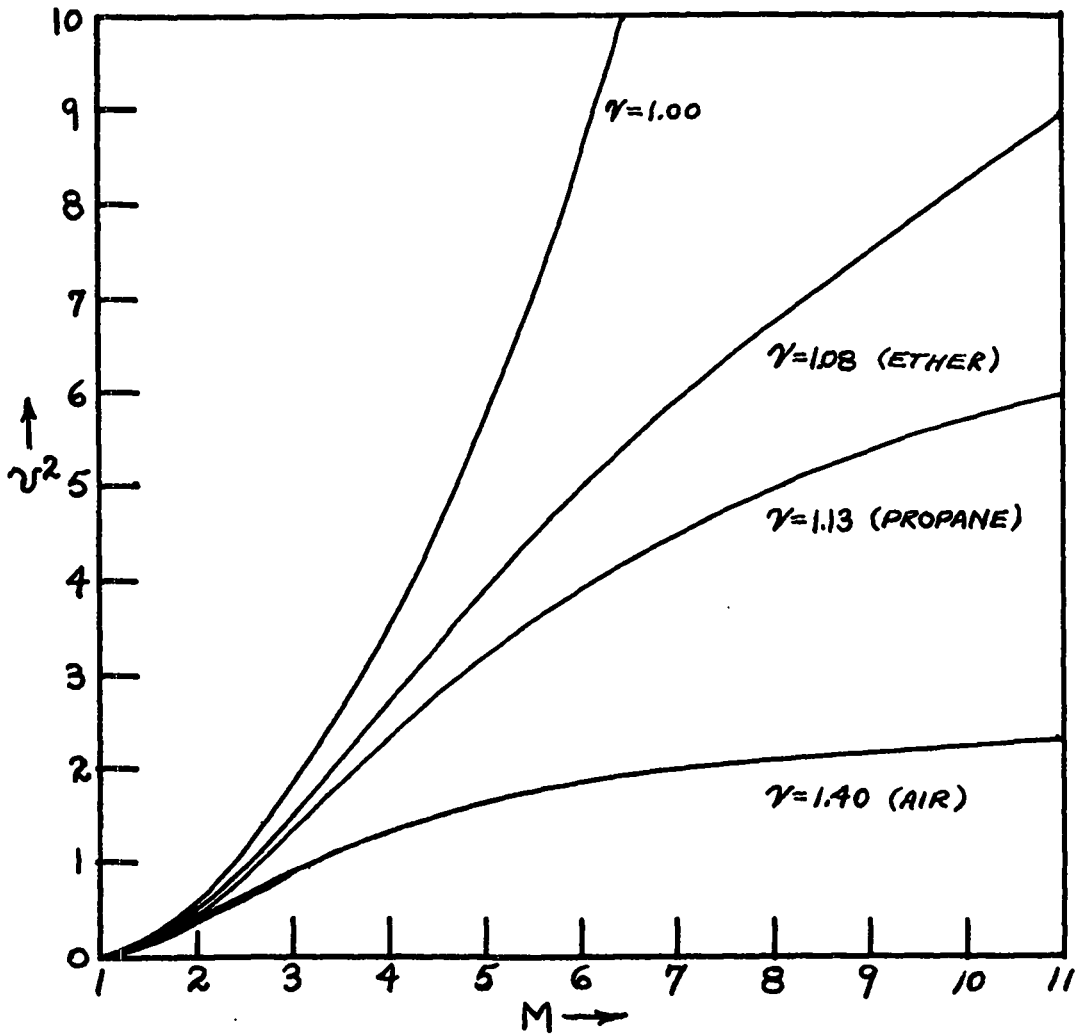


Figure 4 - Ratio of mass motion and thermal kinetic energies as a function of shock Mach number for several gases.



In Figure 5 the relative number of energetic collisions in two shocks, at three relative velocities each, are plotted as a function of position in the shock for a gas of  $\gamma=1$ . These curves were calculated using (25)-(30) in (22). The pronounced maxima of energetic collisions within the shock transition displayed in this case disappears entirely for  $\gamma>1.1$ .

Tabulated below are some values of  $\tau/x$  for  $\gamma=1$  and  $\gamma=1.08$  (ethyl ether), calculated from (24) and (25)-(30). Even for the low  $\gamma$  of ether the shock transition region contains few energetic collisions compared to the material behind the shock. However, when  $\gamma$  is one, or very nearly so, and  $M$  is sizeable, then the shock transition is seen to be equivalent to a considerable thickness of shocked material.

Length of shocked polytropic gas, in units of shock thickness, which produces collisions of relative velocity  $\geq g_0$  at the same rate as the total number resulting from the mass velocity jump across the shock.

| $\gamma$ | $M$  | $\nu$ | $\frac{g_0}{u_0 - u_1}$ | $\frac{\tau}{x}$  |
|----------|------|-------|-------------------------|-------------------|
| 1.00     | 2.4  | 1     | 1                       | 0.11              |
|          | 2.4  | 1     | 2                       | 0.32              |
|          | 2.4  | 1     | 3                       | 1.42              |
|          | 3.7  | 2     | 2                       | 0.41              |
|          | 3.7  | 2     | 3                       | 9.2               |
|          | 3.7  | 2     | 4                       | $3.0 \times 10^2$ |
|          | 5.0  | 3     | 3                       | $2.0 \times 10^1$ |
|          | 5.0  | 3     | 4                       | $3.4 \times 10^3$ |
|          | 5.0  | 3     | 5                       | $5.0 \times 10^6$ |
|          | 1.08 | 2.5   | 1                       | 1                 |
| 2.5      |      | 1     | 2                       | 0.17              |
| 2.5      |      | 1     | 3                       | 0.31              |
| 5.1      |      | 2     | 2                       | 0.09              |
| 5.1      |      | 2     | 3                       | 0.23              |
| 5.1      |      | 2     | 3                       | 0.37              |
| 11.4     |      | 3     | 3                       | 0.08              |
| 11.4     |      | 3     | 4                       | 0.14              |
| 11.4     |      | 3     | 5                       | 0.10              |

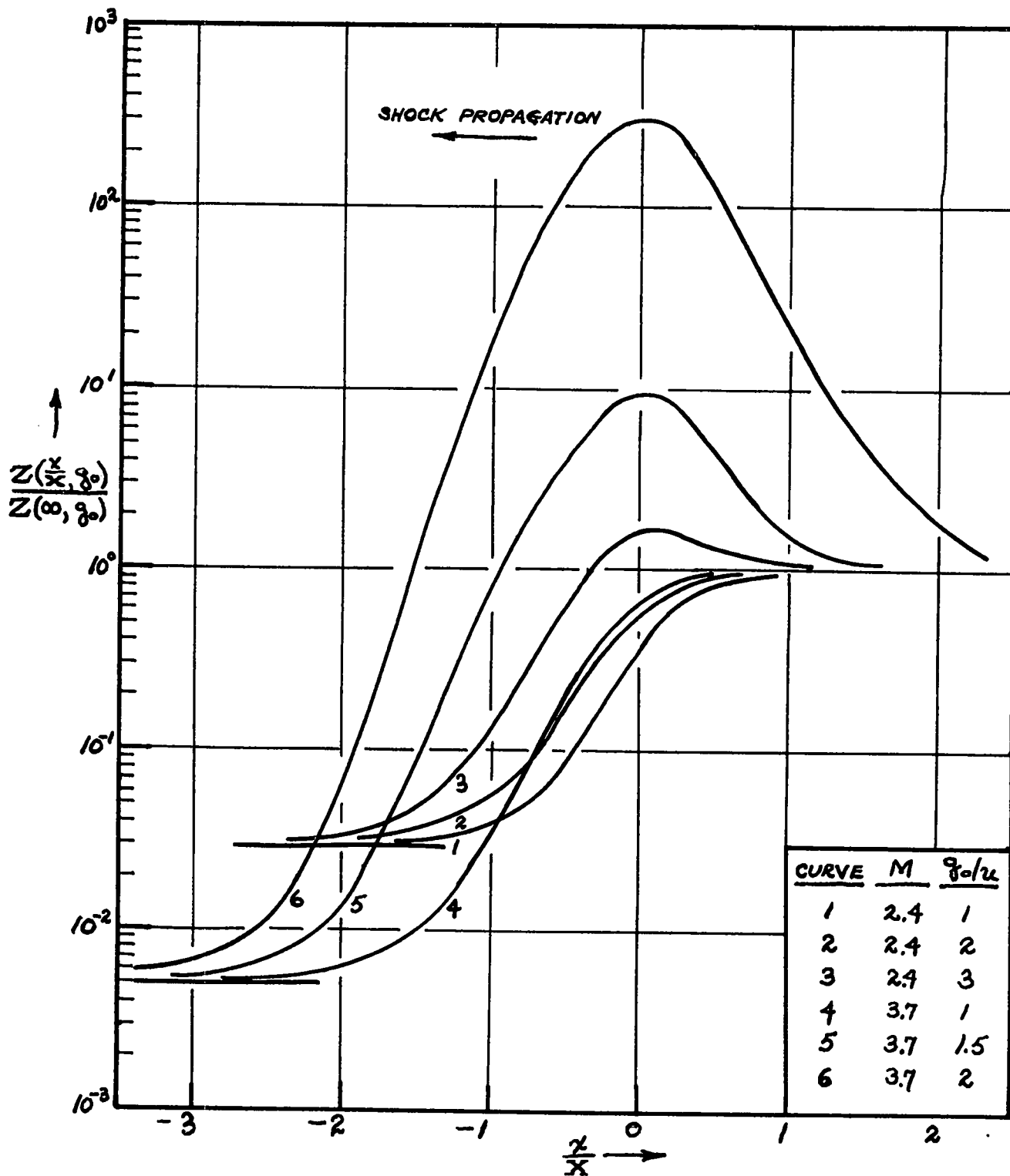


Figure 5 - Relative number of energetic collisions in shocks in a gas with specific heat ratio equal to one.



#### IV. Conclusions

The Mott-Smith solution of the Boltzmann equation, which forsakes the classical Enskog-Chapman method of series expansion for the distribution function and starts instead with an intuitively constructed first approximation peculiarly appropriate to the case of a shock, has in consequence the distinct advantage of providing a strikingly clear and useful qualitative picture of the molecular dynamics in the shock process. The treatment of collision kinetics given here inherits this feature.

Quantitatively the results of the present calculation are useful mostly in a negative sense. The interesting new detail, namely the high concentration of energetic collisions within the shock, is shown not to occur for any real gases of current practical interest. Thus, when any of the common gases are strongly shocked we need not consider the shock per se as a direct origin of ionization, dissociation or chemical reaction, but rather to look for these effects in the state resulting from the shock.

The treatment of collision kinetics has an important bearing on the problem of initiation of chemical reaction in the steady detonation of a homogeneous liquid or solid explosive. The Zeldovich-von Neuman picture of detonation as a shock followed by a steady reaction zone is fully established. There is, however, a lively controversy over how the shock initiates the reaction. von Neuman<sup>7</sup> and others hold that the vehement blow delivered by the

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<sup>7</sup> von Neuman - OSRD 549 (1942)

material velocity in the wave head provides the mechanism needed to start the reaction. Eyring<sup>8</sup> and collaborators are the chief proponents of the theory that the temperature behind the shock front is sufficient to bring the initiation, as well as the subsequent reaction, into the province of ordinary chemical-kinetic theory.

With absolutely no claim to quantitative validity, we can estimate  $\tau/\lambda$  according to (24) for the reaction shock in a condensed explosive. Its value is extraordinarily sensitive to the assumed activation energy and the temperature behind the shock. Over the range of conceivable explosive properties,  $\tau/\lambda$  varies between 1 and  $10^{20}$ , with the most plausible values lying in  $10^5$  to  $10^{15}$ . The view resulting from this consideration definitely leans toward that of von Neuman. In fact it seems likely that a more complicated situation occurs, namely that there are some explosives in which the initiation can be considered of thermal character and others in which the velocity jump causes initiation, a significant fraction of chemical reaction actually occurring within the shock transition.

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<sup>8</sup>Eyring, Powell, Duffey, Parlin - Chem.Rev. 45,69 (1949)

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