

## ESTIMATION OF VAPORIZATION HEATS OF SIMPLE MOLECULE LIQUIDS FROM THE HARD BODY EQUATIONS OF STATE

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Vaporization of liquids formed by simple molecules are studied theoretically exploiting *a*) a hard sphere equation of state with the reference hard sphere diameters determined from the parameters of the L-J 12-6 potential by the relation of Barker and Henderson, *b*) a hard convex body equation of state with parameters determined from the structure of the respective molecules.

A hard sphere equation of state derived from the scaled particle theory<sup>1</sup> (this equation is identical with the *c*)-relation of the Percus-Yevick approximation<sup>2-4</sup>) has been proved to give a fair agreement with the Monte Carlo pseudoexperimental data on hard spheres and represents a sound basis for the estimation of thermodynamic functions of real systems. Yosim and Owens<sup>5</sup> applied the mentioned equation of state to the determination of the vaporization heats of simple liquids. They put the diameter of representative hard spheres equal to the  $\sigma$ -parameter of the Lennard-Jones 12-6 potential and obtained (under plausible assumptions) an agreement of calculated and experimental data within 20%. A hard sphere equation of state of Carnahan and Starling<sup>6</sup>, which fits pseudoexperimental data of a hard sphere system at high densities considerably better than the scaled particle equation of state, was applied for the calculation of the vaporization heats under the strictly same conditions as Yosim and Owens did it and a slight improvement resulted<sup>7</sup>.

In this work an attempt is made to improve the determination of the vaporization heats through the use of a better hard sphere diameter estimation, exploiting the relation derived in their perturbation theory by Barker and Henderson<sup>8</sup>. In order to study the effect of the molecular shape, hard convex bodies were used as a representative system instead of hard spheres in the second part of this work. This step was enabled by the knowledge of an approximative hard convex body equation of state derived recently<sup>9,10</sup>.

### THEORETICAL

A convex body  $A$  [defined<sup>11,12</sup> as a set of points, where with any pair of points all points of the connecting line lie inside the set] can be characterized by three functionals: the volume  $V_A$ , surface area  $S_A$  and by the integral of mean curvature  $M_A$ . A parallel body  $C$  to the convex body  $A$  is formed by a set of points forming an

envelope of all spheres with a radius  $\xi$ , the centers of which lie on the surface of the A-body. Functionals of the parallel body with respect to the functionals of the A-body are given by Eqs (1)–(3):

$$V_C = V_A + S_A \xi + 4\pi R_A \xi^2 + \frac{4}{3}\pi \xi^3, \quad (1)$$

$$S_C = S_A + 8\pi R_A \xi + 4\pi \xi^2, \quad (2)$$

$$R_C = R_A + \xi, \quad (3)$$

where  $R = M/4\pi$ .

The derivation of the hard convex body equation of state is based on the extension of the idea of Reiss and coworkers<sup>1</sup> who scaled the size of the given sphere and expressed the effect of this scaling on the configurational energy. Similarly Gibbons<sup>10</sup> scaled the size of the convex body of the given shape by changing the radius of the sphere to which the convex body is inscribed; this step is equivalent to the dilatation of the convex body<sup>11</sup> with the dilatation coefficient  $\lambda$  [for any vector of the dilated body  $\bar{x}$ ,  $\bar{x} = \lambda x$ ]. For the dilatation coefficient  $\lambda \leq 0$ , excluded volumes for the interaction of the scaled particle with ordinary C-particles do not overlap and the work  $W$  required to add a “ $\lambda$ -particle” to the one component system of hard convex bodies is given by Eq. (4)

$$\beta W(\lambda) = -\ln(1 - NV_{C+\lambda C}/V). \quad (4)$$

Here  $\beta = (kT)^{-1}$ ,  $N$  is the number of particles,  $V$  the volume of the system, and the average excluded volume  $V_{C+\lambda C}$  can be determined from Eq. (5)

$$V_{C+\lambda C} = V_C(1 + \lambda^3) + R_C S_C(\lambda + \lambda^2). \quad (5)$$

For  $\lambda > 0$ ,  $\beta W(\lambda)$  is expressed by a Taylor series expansion in  $\lambda$  (up to  $\lambda^2$  plus a term  $\lambda^3 \beta P V_C$ ); from this expansion  $\beta W(\lambda = 1)$  can be evaluated. Exploiting the Gibbs–Duhem relation and Eq. (6) for the chemical potential  $\mu^{13}$ ,

$$\beta \mu = \ln \varrho \Lambda^3 + \beta W(\lambda = 1) \quad (6)$$

the hard convex body equation of state can be derived:

$$\frac{\beta P}{\varrho} = \frac{1 + y(\gamma - 2) + y^2(\gamma^2/3 - \gamma + 1)}{(1 - y)^3}. \quad (7)$$

Here  $\varrho$  is the number density ( $N/V$ ),  $P$  the pressure of the hard convex body system,

$$y = NV_C/V, \quad \gamma = R_C S_C/V_C, \quad (8, 9)$$

and  $\Lambda^{-3}$  is the translational contribution to the partition function<sup>13</sup>,

$$\Lambda = (h^2/2\pi mkT)^{1/2}. \quad (10)$$

For the hard sphere system [ $R_C = R$ ,  $S_C = 4\pi R^2$ , and  $V_C = 4\pi R^3/3$ ]  $\gamma = 3$  and Eq. (7) reduces to the hard sphere equation of state

$$\beta P/\varrho = (1 + \gamma + \gamma^2)/(1 - \gamma)^3. \quad (11)$$

A formal addition of the term  $-\gamma^3$  in the numerator of Eq. (11) gives the equation of state of Carnahan and Starling. Eq. (7) also yields a correct expression for the second virial coefficient of the hard convex body system (Kihara<sup>12</sup>, Rigby<sup>14</sup>); it represents itself a fair approximation of the hard convex body equation of state and can be used for the estimation of thermodynamic functions of real systems. (The situation is different, however, with mixtures of hard convex bodies of different shapes, where Cauchy's relations are not fulfilled<sup>10</sup> and the equation is not reliable.)

In order to calculate the vaporization heat on the basis of the hard convex body equation of state the following isothermal thermodynamic cycle is assumed: a) One mol of liquid is evaporated at a given temperature and pressure (the liquid volume is  $V_1$ ) to the equilibrium vapour with the volume  $V_g$ . b) The vapour is expanded from the pressure  $P$  to  $P^* \rightarrow 0$  ( $V^* \rightarrow \infty$ ,  $P^*V^* = RT$ ). c) Molecules of the vapour are "discharged" at the pressure  $P^* \rightarrow 0$  so that a representative convex body system results. d) The hard convex body system is compressed from  $V^*$  to the volume equal to  $V_1$  and the pressure  $P^0$  of hard convex bodies. e) Convex particles are "charged" at the constant volume  $V_1$  to the given realistic pair potential so that the original liquid is recovered.

Entropy changes of the respective steps can be expressed by Eqs (12)–(16):

$$\Delta S_a = \Delta H^{vap}/T, \quad (12)$$

$$\Delta S_b = \int_{P^*}^P (\partial V/\partial T)_P dP = RT \ln P - RT \ln P^* + \int_{P^* \rightarrow 0}^P (\partial B/\partial T) dP, \quad (13)$$

$$\Delta S_c = 0, \quad (14)$$

$$\Delta S_d = - \int_{V_1}^{V^*} (\partial P^0/\partial T)_V dV = -1/T \int_{V_1}^{V^*} P^0 dV, \quad (15)$$

and

$$S_e = S_1 - S^0. \quad (16)$$

In Eqs (12)–(16)  $B$  is the residual volume (approximated by the second virial coefficient),  $R$  the gas constant,  $\Delta H^{vap}$  the vaporization heat, the superscript zero denotes the hard convex body system, the asterisk the perfect gas system, and the

superscript 1 the liquid state. The difference  $S_1 - S^0$  equals zero in the first order perturbation treatment with hard sphere diameters independent of temperature (a treatment similar to that of Barker and Henderson); this equality is here assumed. After the substitution of Eq. (7) in Eq. (15) the following relation for the vaporization heat holds

$$\frac{\Delta H^{\text{vap}}}{RT} = \ln \frac{RT}{PV_1} - \left( \frac{\partial B}{\partial T} \right) \frac{P}{R} - \ln(1-y) + \frac{\gamma y}{(1-y)} + \frac{\gamma^2 y^2}{6(1-y)^2}. \quad (17)$$

The second term of the right hand side of the last equation is comparatively small (less than 1%); because of this fact and because second virial coefficients for some of the studied compounds are not known, this term was neglected completely.

## RESULTS AND DISCUSSION

As a first step, the vaporization heats were calculated from Eq. (18)

$$\frac{\Delta H^{\text{vap}}}{RT} = \ln \frac{RT}{PV_1} - \left( \frac{\partial B}{\partial T} \right) \frac{P}{R} + \frac{3y}{(1-y)} + \frac{y_1'}{(1-y)^2}, \quad (18)$$

which follows from Eqs (12)–(16) after the substitution of the hard sphere equation of state of Carnahan and Starling. The function  $y$  depends on the hard sphere diameter  $d$  by the relation

$$y = NV_{\text{HS}}/V = \pi Nd^3/6V. \quad (19)$$

The diameter  $d$  of the representative hard spheres in Eq. (19) was estimated from Lennard-Jones 12–6 potential parameters exploiting the prescription derived by Barker and Henderson<sup>8</sup>

$$d = \int_0^\sigma \{1 - \exp[-\beta w(r)]\} dr; \quad (20)$$

$w(r)$  stands for the pair potential as a function of the intermolecular distance  $r$ .

Results of the vaporization heat calculations are compared in Table I with experimental data<sup>15,16</sup> and with values obtained from Eq. (18) for the diameter estimate  $d = \sigma$ . By the application of Eq. (20) in combination with the equation of state of Carnahan and Starling the standard deviation in  $\Delta H^{\text{vap}}$  decreases from 18.2% to 15.0% for the whole set of 30 compounds, and from 4.8% to 3.1% for systems with spherical nonpolar molecules (with exception of He). The estimation of vaporization heats of spherical molecule systems can be considered as satisfactory; this is, however, not true for nonspherical molecule systems. Eq. (17) was therefore considered which takes into account not only the size but also the shape of molecules. With its application a serious problem arose, namely the determination of the values of the functionals  $R_C$ ,  $S_C$  and  $V_C$  of the representative hard convex bodies. Because of the lack of suitable

TABLE I  
Heats of Vaporization (cal/mol) Calculated from the Hard Sphere Equation of State and Eq. (20)

| Compound                         | $\Delta H_{\text{Obs}}$ | $\Delta H_{\text{CST}}$ | $\Delta H_{\text{CST+Eq(20)}}$ | Dev.,% |
|----------------------------------|-------------------------|-------------------------|--------------------------------|--------|
| He                               | 19.4                    | 26.9                    | 26.5                           | 36.8   |
| Ne                               | 414                     | 424                     | 405                            | - 2.1  |
| Ar                               | 1 558                   | 1 611                   | 1 508                          | - 3.2  |
| Kr                               | 2 158                   | 2 168                   | 2 068                          | - 4.2  |
| Xe                               | 3 020                   | 3 224                   | 3 049                          | 1.0    |
| H <sub>2</sub>                   | 215.8                   | 227.7                   | 222.3                          | 3.0    |
| N <sub>2</sub>                   | 1 333                   | 1 416                   | 1 330                          | - 0.2  |
| O <sub>2</sub>                   | 1 628                   | 1 727                   | 1 629                          | 0.1    |
| F <sub>2</sub>                   | 1 562                   | 1 558                   | 1 475                          | - 5.6  |
| Cl <sub>2</sub>                  | 4 878                   | 5 075                   | 4 798                          | - 1.6  |
| Br <sub>2</sub>                  | 7 170                   | 6 787                   | 6 477                          | - 9.7  |
| I <sub>2</sub>                   | 9 970                   | 12 290                  | 11 105                         | 11.4   |
| NO <sub>2</sub>                  | 3 293                   | 3 054                   | 2 740                          | -16.8  |
| CO                               | 1 444                   | 1 397                   | 1 332                          | - 7.7  |
| HCl                              | 3 860                   | 3 344                   | 3 258                          | -15.6  |
| HI                               | 4 724                   | 5 029                   | 4 738                          | 0.3    |
| CS <sub>2</sub>                  | 6 400                   | 6 255                   | 5 979                          | - 6.6  |
| N <sub>2</sub> O                 | 3 958                   | 3 928                   | 3 687                          | - 6.8  |
| SO <sub>2</sub>                  | 5 960                   | 6 875                   | 6 133                          | 2.9    |
| CH <sub>4</sub>                  | 1 955                   | 2 073                   | 1 962                          | 0.4    |
| C <sub>2</sub> H <sub>4</sub>    | 3 237                   | 3 443                   | 3 219                          | - 0.6  |
| C <sub>2</sub> H <sub>6</sub>    | 3 517                   | 2 973                   | 2 875                          | -18.2  |
| C <sub>4</sub> H <sub>10</sub>   | 5 352                   | 4 640                   | 4 416                          | -17.5  |
| C <sub>6</sub> H <sub>6</sub>    | 7 353                   | 7 201                   | 6 747                          | - 8.2  |
| C <sub>6</sub> H <sub>12</sub>   | 7 190                   | 10 002                  | 8 573                          | 19.2   |
| CH <sub>3</sub> Cl               | 5 150                   | 3 630                   | 3 602                          | -30.0  |
| CHCl <sub>3</sub>                | 7 020                   | 9 193                   | 8 035                          | 14.5   |
| CCl <sub>4</sub>                 | 7 170                   | 10 227                  | 8 741                          | 21.9   |
| CF <sub>4</sub>                  | 2 988                   | 3 752                   | 3 272                          | 9.5    |
| CH <sub>3</sub> OH               | 8 430                   | 5 904                   | 5 755                          | -31.7  |
| C <sub>2</sub> H <sub>5</sub> OH | 9 220                   | 7 066                   | 6 666                          | -27.7  |

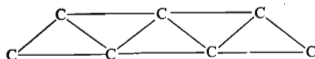


FIG. 1  
Core Models of n-Alkanes and Perfluoro-n-alkanes

data (the determination from virial coefficients of these functionals plus two other parameters is not reliable enough), an attempt was made to exploit structural data of molecules under question and the representative hard sphere diameters of simplest molecules.

Normal alkanes from  $\text{CH}_4$  to  $\text{C}_{12}\text{H}_{26}$  and perfluoro-n-alkanes ( $\text{CF}_4$ — $\text{C}_7\text{F}_{16}$ ) were studied. It was assumed that the core (the hard convex body A) was formed by a carbon chain; thus for  $\text{C}_1$  the core was represented by a point, for  $\text{C}_2$  by a line connecting the C-atoms, for  $\text{C}_3$  by a triangle; the cores of higher alkanes (or perfluoroalkanes) were represented by a planar combination of triangles (Fig. 1). Triangles were replicas of the  $\text{C}_3$ -triangle (with the bond length  $\text{C—C} = 1.533 \text{ \AA}$ ,  $\angle \text{C—C—C} = 109^\circ 30'$ ) in the case of perfluoroalkanes for which some evidence exists<sup>17</sup> that their C-chains are planar. The cores of alkanes with  $m$  carbon atoms ( $m > 3$ ) were represented by a combination of triangles with a side length  $z$  given by Eq. (21)

$$z = 2[2.503 + 1.025(m - 3)]/(m - 1) \text{ (\AA)}, \quad (21)$$

by which the different sterical orientations of the C—C bonds were presumably taken into account<sup>18</sup>. (The increment  $1.025 \text{ \AA}$  is the difference between the average  $\text{C}_1$ — $\text{C}_4$  distance in n-butane and the  $\text{C}_1$ — $\text{C}_3$  distance in propane<sup>17,18</sup>.)

The values of the functionals  $V_A$ ,  $S_A$  and  $R_A$  calculated from the described models are listed in Table II. The functionals of the representative parallel convex bodies (C) were calculated from Eqs (1)–(3) using the tabulated values of  $R_A$ ,  $S_A$ ,  $V_A$  and the radii of the parallel bodies  $\xi_{\text{CH}} = 1.8 \text{ \AA}$  and  $\xi_{\text{CF}} = 2.2 \text{ \AA}$  for n-alkanes and perfluoro-n-alkanes, respectively. The distances  $\xi_{\text{CH}}$  and  $\xi_{\text{CF}}$  were determined as rounded

TABLE II

The Values of the Functionals  $R_A$  and  $S_A$ , and  $V_A$  Used for the Estimation of  $\Delta H^{\text{vap}}$  (in all cases  $V_A = 0.0$ ).

| Homologue       | n-Alkanes          |                      | Perfluoro-n-alkanes |                      |
|-----------------|--------------------|----------------------|---------------------|----------------------|
|                 | $R_A, \text{ \AA}$ | $S_A, \text{ \AA}^2$ | $R_A, \text{ \AA}$  | $S_A, \text{ \AA}^2$ |
| $\text{C}_1$    | 0.0000             | 0.0000               | 0.0000              | 0.0000               |
| $\text{C}_2$    | 0.3832             | 0.0000               | 0.3832              | 0.0000               |
| $\text{C}_3$    | 0.6961             | 2.2152               | 0.6961              | 2.2152               |
| $\text{C}_4$    | 0.9713             | 4.1630               | 1.0090              | 4.4304               |
| $\text{C}_5$    | 1.2370             | 6.0444               | 1.3219              | 6.6456               |
| $\text{C}_6$    | 1.4983             | 7.8944               | 1.6348              | 8.8608               |
| $\text{C}_7$    | 1.7589             | 9.7395               | 1.9476              | 11.0760              |
| $\text{C}_8$    | 2.0178             | 11.5728              |                     |                      |
| $\text{C}_9$    | 2.2761             | 13.4015              |                     |                      |
| $\text{C}_{10}$ | 2.5340             | 15.2272              |                     |                      |
| $\text{C}_{11}$ | 2.7914             | 17.0496              |                     |                      |
| $\text{C}_{12}$ | 3.0484             | 18.8720              |                     |                      |

values of the representative hard sphere diameters of  $\text{CH}_4$  and  $\text{CF}_4$ , respectively, obtained from the Lennard-Jones 12-6 parameters with help of Eq. (20). From the parallel convex body functionals  $R_C$ ,  $S_C$ ,  $V_A$ , the  $\gamma$  parameter was determined; its values together with the hard body volumes are listed in Tables III and IV. The standard deviation for the set of perfluoro-n-alkanes is 5.9%, for n-alkanes 1.3%. In the case of n-alkanes, the determination of representative hard convex bodies can be compared with that of Connolly and Kandalic<sup>19</sup> who correlated second virial

TABLE III

Heats of Vaporization (cal/mol) of n-Alkanes Calculated from the Hard Convex Body Equation of State

| Compound                     | $\gamma$ | $V_C, \text{\AA}^3$ | $\Delta H_{\text{obs}}$ | $\Delta H_{\text{Eq}(17)}$ | Dev., % |
|------------------------------|----------|---------------------|-------------------------|----------------------------|---------|
| $\text{CH}_4$                | 3.000    | 24.43               | 1 955                   | 1 887                      | -3.5    |
| $\text{C}_2\text{H}_4$       | 3.166    | 40.03               | 3 517                   | 3 529                      | 0.4     |
| $\text{C}_3\text{H}_8$       | 3.273    | 56.76               | 4 487                   | 4 424                      | -1.4    |
| $\text{C}_4\text{H}_{10}$    | 3.444    | 71.47               | 5 352                   | 5 412                      | 1.1     |
| $\text{C}_5\text{H}_{12}$    | 3.641    | 85.67               | 6 160                   | 6 206                      | 0.7     |
| $\text{C}_6\text{H}_{14}$    | 3.853    | 99.64               | 6 896                   | 6 908                      | 0.2     |
| $\text{C}_7\text{H}_{16}$    | 4.074    | 113.57              | 7 575                   | 7 571                      | -0.1    |
| $\text{C}_8\text{H}_{18}$    | 4.302    | 127.41              | 8 214                   | 8 199                      | -0.2    |
| $\text{C}_9\text{H}_{20}$    | 4.534    | 141.22              | 8 777                   | 8 812                      | 0.4     |
| $\text{C}_{10}\text{H}_{22}$ | 4.769    | 155.01              | 9 390                   | 9 406                      | 0.2     |
| $\text{C}_{11}\text{H}_{24}$ | 5.007    | 168.77              | 9 924                   | 9 994                      | 0.7     |
| $\text{C}_{12}\text{H}_{26}$ | 5.247    | 182.53              | 10 441                  | 10 566                     | 1.2     |

TABLE IV

Heats of Vaporization (cal/mol) of Perfluoro-n-alkanes Calculated from the Hard Convex Body Equation of State

| Compound                  | $\gamma$ | $V_C, \text{\AA}^3$ | $\Delta H_{\text{obs}}$ | $\Delta H_{\text{Eq}(17)}$ | Dev., % |
|---------------------------|----------|---------------------|-------------------------|----------------------------|---------|
| $\text{CF}_4$             | 3.000    | 44.60               | 2 988                   | 2 992                      | 0.1     |
| $\text{C}_2\text{F}_6$    | 3.120    | 67.91               | 3 857                   | 3 886                      | 0.7     |
| $\text{C}_3\text{F}_8$    | 3.202    | 91.81               | 4 690                   | 4 672                      | -0.4    |
| $\text{C}_4\text{F}_{10}$ | 3.357    | 115.72              | 5 872                   | 5 410                      | -7.9    |
| $\text{C}_5\text{F}_{12}$ | 3.545    | 139.62              | 6 680                   | 6 118                      | -8.4    |
| $\text{C}_6\text{F}_{14}$ | 3.754    | 163.53              | 7 374                   | 6 909                      | -6.3    |
| $\text{C}_7\text{F}_{16}$ | 3.974    | 187.42              | 7 988                   | 7 506                      | -6.0    |

coefficients for  $C_3-C_8$  alkanes by the Kihara expression with a 12-6 potential. They interpreted the cores by combinations of rectangles, each for one C—C bond. The value  $\xi \leq 1.35 \text{ \AA}$  resulting from their treatment is improbably small when comparing with the bond length C—H = 1.12–1.20  $\text{\AA}$ . We believe this is a consequence of the compensation of the hard body core overestimation.

The model used in this work represents the structure of the alkanes more closely than the previous one and the  $\xi_{CH}$  value is quite realistic. A good agreement of the calculated and experimental data within the experimental error supports our belief that the functionals,  $R_A$ ,  $S_A$  and  $V_A$  listed in Table II together with the  $\xi$  values can be considered as a reliable representation of the shapes of the given chain molecules. It has been shown that the vaporization heat estimation for systems with spherical molecules depends considerably on the choice of the reference hard sphere radius and that in the case of nonspherical molecules the reference system must reflect the shape of molecules under consideration.

#### REFERENCES

1. Reiss H., Frisch H. L., Lebowitz J. L.: *J. Chem. Phys.* 31, 369 (1959).
2. Frisch H. L., Lebowitz J. L.: *The Equilibrium Theory of Classical Fluids*. Benjamin, New York 1964.
3. Wertheim M. S.: *Phys. Rev. Letters* 10, 321 (1963).
4. Thiele E. J.: *J. Chem. Phys.* 38, 1959 (1963).
5. Yosim S. J., Owens B. B.: *J. Chem. Phys.* 39, 2222 (1963).
6. Carnahan N. F., Starling K. E.: *J. Chem. Phys.* 51, 635 (1969).
7. Boublík T.: *Chem. Phys.* 53, 471 (1970).
8. Barker J. A., Henderson D.: *J. Chem. Phys.* 47, 2856, 4714 (1967).
9. Ritchie A. B.: *J. Chem. Phys.* 46, 618 (1967).
10. Gibbons R. M.: *Mol. Phys.* 17, 81 (1969); 18, 809 (1970).
11. Hadwiger H.: *Altes und Neues über Konvexe Körper*. Birkhäuser, Basel 1955.
12. Kihara T.: *Advan. Chem. Phys.* 5, 147 (1963).
13. Hála E., Boublík T.: *Einführung in die Statistische Thermodynamik*. Academia, Prague 1970.
14. Rigby M.: *J. Chem. Phys.* 53, 1021 (1970).
15. Dreisbach R. R.: *Physical Properties of Chemical Compounds — II* *Advan. Chem. Ser.* 22. Am. Chem. Soc., Washington 1959.
16. Timmermans J.: *Physico-Chemical Constants of Pure Organic Compounds*. Elsevier, Amsterdam, Vol. I, 1950, Vol. II 1963.
17. *Interatomic Distances* (L. E. Sutton, Ed.) London, Vol. I 1958, Vol. III, 1965.
18. Bonham R. A., Bartell L. S.: *J. Am. Chem. Soc.* 81, 3491 (1959).
19. Connolly J. F., Kandalic G. A.: *Phys. Fluids* 3, 463 (1960).

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