

Then  $(\partial\epsilon_1/\partial E)_z$  is equal to the value of  $d\epsilon_1/dE$  without the general radiofrequency cavity.

Also

$$\begin{aligned}(\partial l/\partial z)_E &= (\partial l/\partial x_1')_E (\partial x_1'/\partial z)_x, \\ (\partial\epsilon_2/\partial E)_z &= (\partial\epsilon_2/\partial x_1)_z (\partial x_1/\partial E)_z.\end{aligned}\quad (54)$$

From Eq. (51), one finds

$$(\partial l/\partial x_1')_E = E_0 (\partial x_1/\partial E)_z. \quad (55)$$

From Eq. (43), one finds

$$(\partial x_1'/\partial z)_x = (1/E_0) (\partial\epsilon_2/\partial x_1)_z. \quad (56)$$

Therefore

$$(\partial l/\partial z)_E = (\partial\epsilon_2/\partial E)_z, \quad (57)$$

$$\alpha_s' = -\frac{1}{2} (\partial\epsilon_1/\partial E)_z = -\frac{1}{2} (d\epsilon_1/dE). \quad (58)$$

Thus, if the fields are not sufficiently strong to change the form of the equilibrium orbit appreciably, the damping rate of the synchronous oscillation is not changed by any form of radio-frequency fields.

Since the total damping rate is invariant, the damping rate of the radial betatron oscillations is also not changed by a generalized radio-frequency field.

## Thermal Diffusion Factors from Column Operation\*†

T. I. MORAN† AND W. W. WATSON

*Physics Department, Yale University, New Haven, Connecticut*

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Extending the measurements of Corbett and Watson, the performance of a carefully-constructed, all-metal, hot-wire thermal diffusion column has been determined for isotope separations in neon, argon, krypton, and xenon. The same quantitative agreement with theory for normal argon gas at the low wall-temperature ratio of 2 is again found. This is probably fortuitous, for in general there is a discrepancy between the calculated and observed separation factors, with a trend in the data indicating that the assumptions of the theory that (1) the molecules are Maxwellian and (2) the thermal diffusion factors are constant, independent of temperature, are at fault.

### INTRODUCTION

THE hot-wire thermal diffusion column has proven to be of great value in the isotopic enrichment of certain gaseous compounds. Many workers have compared the experimental performance of such columns with the theory of Jones and Furry,<sup>1</sup> finding that the agreement is in general only qualitative. Corbett and one of the present authors,<sup>2</sup> however, using a carefully constructed all-metal column, reported very good agreement between experiment and theory, without the inclusion of any parasitic remixing term, when the column was operated with normal argon gas at a temperature ratio of 2 between the hot wire and the cold wall.

This result raises the question as to whether under such conditions one can determine thermal diffusion factors  $\alpha$  from measurements of column performance. To investigate this possibility further, we have studied the performance of this same column for other noble gases. Our results show that, in general, accurate values

of  $\alpha$  may not be so determined, and for the reasons discussed at the end of this report.

Although the Jones and Furry theory considers the gas as a binary isotopic mixture of Maxwellian molecules with a constant thermal diffusion factor to be evaluated at the cold-wall temperature, it is easy to generalize to the case of a multi-isotopic mixture. According to this theory the equilibrium separation factor  $q_{ij}$  for two isotopes  $i$  and  $j$ , defined as the ratio  $c_i/c_j$  of the concentrations at the upper end to that at the lower end, is given by the expression

$$q_{ij} = e^{2A_{ij}l}, \quad (1)$$

where

$$A_{ij} = H_{ij}/(K_c + K_d), \quad (2)$$

and  $l$  is the length of the column. The thermal diffusion factor  $\alpha_{ij}$  is contained in the transport factor  $H_{ij}$ . A reduced thermal diffusion factor  $\alpha_0$  may be defined from

$$\alpha = \alpha_0 (m_i - m_j) / \bar{m}, \quad (3)$$

where the  $m_i$  and  $m_j$  are the masses of any two isotopes in the multicomponent mixture and  $\bar{m}$  is the average of the two. To the sum of the two remixing factors  $K_c$  from convection and  $K_d$  from axial diffusion is usually added of necessity a third,  $K_p$ , from parasitic effects originating in nonuniformities of construction, azimuthal temperature asymmetries, etc. Only in our first

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‡ Now at the General Electric Company, Hanford, Washington.

<sup>1</sup> R. Clark Jones and W. H. Furry, *Revs. Modern Phys.* **18**, 151 (1946).

<sup>2</sup> J. W. Corbett and W. W. Watson, *Phys. Rev.* **101**, 519 (1956).

work with this accurate column, using normal argon and a temperature ratio of 2, was it unnecessary to invoke a parasitic remixing factor in order to bring the performance into agreement with the predictions of the theory.

### EXPERIMENTAL RESULTS

To analyze the data on the performance of this column for other noble gases, it was first necessary to measure the isotopic thermal diffusion factors, especially  $\alpha_0$  for krypton and xenon, as a function of temperature. These measurements we have made with a "Trennschaukel" apparatus<sup>3,4</sup> for all of the noble gases in the temperature range 230°K to 600°K.

Repeating first the measurements reported in reference 2, we assured ourselves that the column was mounted properly and that heating the central wire with ac did not change the performance. Then, keeping the ratio of hot-to-cold wall temperatures at 2, we made two sets of measurements of separation factor *versus* gas pressure for each gas, each at a different cold-wall temperature  $T_1$ . The temperature  $T_2$  of the hot wire was determined from its extension as measured with a cathetometer. The experimental values of  $2A$  corresponding to all of these measured separation factors, as well as the values of  $2A$  calculated from the Jones and Furry theory, are summarized in Table I. More illuminating, perhaps, are the plots in Fig. 1.

### DISCUSSION

In general, there is a complete lack of quantitative agreement between the theoretical predictions and the observed column performance, although qualitatively the Furry, Jones, and Onsager theory does describe this rather complex situation rather well. Now this theory requires (1) that the molecules of the gas be Maxwellian insofar as the temperature dependence of the elementary gas coefficients is concerned, and (2) that the thermal diffusion factor is a constant, independent of temperature, for each gas. Since a molecule which is nearly Maxwellian should have a very small thermal diffusion factor, xenon should meet the first requirement the best and neon least well. Inspection of Fig. 1 shows that the column performance with neon does indeed display the greatest divergence from the theoretical predictions, although there seems to be no progressive improvement as we go from neon to xenon. The explanation for this may be that for krypton and xenon requirement (2) is less adequately met, since for these heavier gases the thermal diffusion factors are strongly temperature-dependent.<sup>4</sup>

A noticeable trend in these data is the increasingly better agreement between the observed pressure for maximum separation factor and that predicted by the

theory in going from neon to xenon. This suggests that the assumption that the molecules are Maxwellian is primarily responsible for displacing the theoretical curve towards higher pressure. This displacement would also have the effect of increasing the magnitude of the

TABLE I. Comparison of experimental and theoretical column performance.  $r_1=0.250$  inch,  $r_2=0.010$  inch; length of column = 182 cm;  $T_2/T_1=2.0$ .

Gas	Pressure in. of Hg-absolute	$2A$ ( $10^{-3}$ cm $^{-1}$ ) theoretical	$2A$ ( $10^{-3}$ cm $^{-1}$ ) experimental
Neon	10	0.63	0.87
$T_1=298^\circ\text{K}$	15	1.42	1.86
$\eta=314$ $\mu\text{poise}$	20	2.48	3.15
$D=0.531$ cm $^2$ /sec	23.5	3.34	4.00
$\alpha=2.15\times 10^{-2}$	30	5.07	4.48
	35	6.28	4.72
	39	7.05	4.66
	45	7.88	3.63
	50	8.00	2.42
Neon	10	0.36	0.78
$T_1=358^\circ\text{K}$	15	0.82	1.50
$\eta=353$ $\mu\text{poise}$	20	1.45	3.29
$D=0.718$ cm $^2$ /sec	30	3.16	3.81
$\alpha=2.25\times 10^{-2}$	40	5.21	5.85
	52	7.52	5.00
Argon (normal)	10	3.42	3.7
$T_1=288^\circ\text{K}$	15.5	4.85	4.8
$\eta=220$ $\mu\text{poise}$	20.0	5.05	4.6
$D=0.171$ cm $^2$ /sec	26	3.50	2.8
$\alpha=(1.32)\times 10^{-2}$	30	3.00	2.2
Argon (9.70% A <sup>36</sup> )	8	2.20	3.3
$T_1=288^\circ\text{K}$	11	4.00	4.04
$\eta=218$ $\mu\text{poise}$	13.25	4.58	3.98
$D=0.172$ cm $^2$ /sec	15.5	4.70	3.83
$\alpha=1.32\times 10^{-2}$	17.5	4.58	3.61
	20.25	4.10	2.95
Argon (9.70% A <sup>36</sup> )	5	0.50	1.0
$T_1=358^\circ\text{K}$	10	1.93	1.83
$\eta=261$ $\mu\text{poise}$	12	2.70	2.28
$D=0.255$ cm $^2$ /sec	13.5	3.31	3.61
$\alpha=(1.62)\times 10^{-2}$	14	3.48	3.61
	16	4.30	4.11
	17	4.66	4.22
	18	5.00	4.56
	20	5.51	4.56
	22	5.85	4.39
	25	5.99	4.28
Krypton	2	2.38	4.1
$T_1=298^\circ\text{K}$	3	5.28	7.8
$\eta=255$ $\mu\text{poise}$	4	9.13	11.1
$D=0.093$ cm $^2$ /sec	5	13.2	15.4
$\alpha_0=7\times 10^{-2}$	8	24.6	20.8
	12	18.4	15.4
	15	15.5	12.0
Krypton	6	12.4	16.0
$T_1=358^\circ\text{K}$	10	26.3	29.1
$\eta=308$ $\mu\text{poise}$	15	32.6	23.1
$D=0.136$ cm $^2$ /sec	20	25.2	17.3
$\alpha_0=9\times 10^{-2}$	25	17.9	16.2
Xenon	2	3.75	7.55
$T_1=358^\circ\text{K}$	5.5	22.7	13.5
$\eta=272$ $\mu\text{poise}$	8	28.2	19.2
$D=0.080$ cm $^2$ /sec	10	24.9	16.3
$\alpha_0=7.7\times 10^{-2}$	15	14.1	10.1
	20	8.3	6.68

<sup>3</sup> K. Clusius and M. Huber, Z. Naturforsch. **10a**, 230 (1955).

<sup>4</sup> T. I. Moran and W. W. Watson, Phys. Rev. **109**, 1184 (1958).

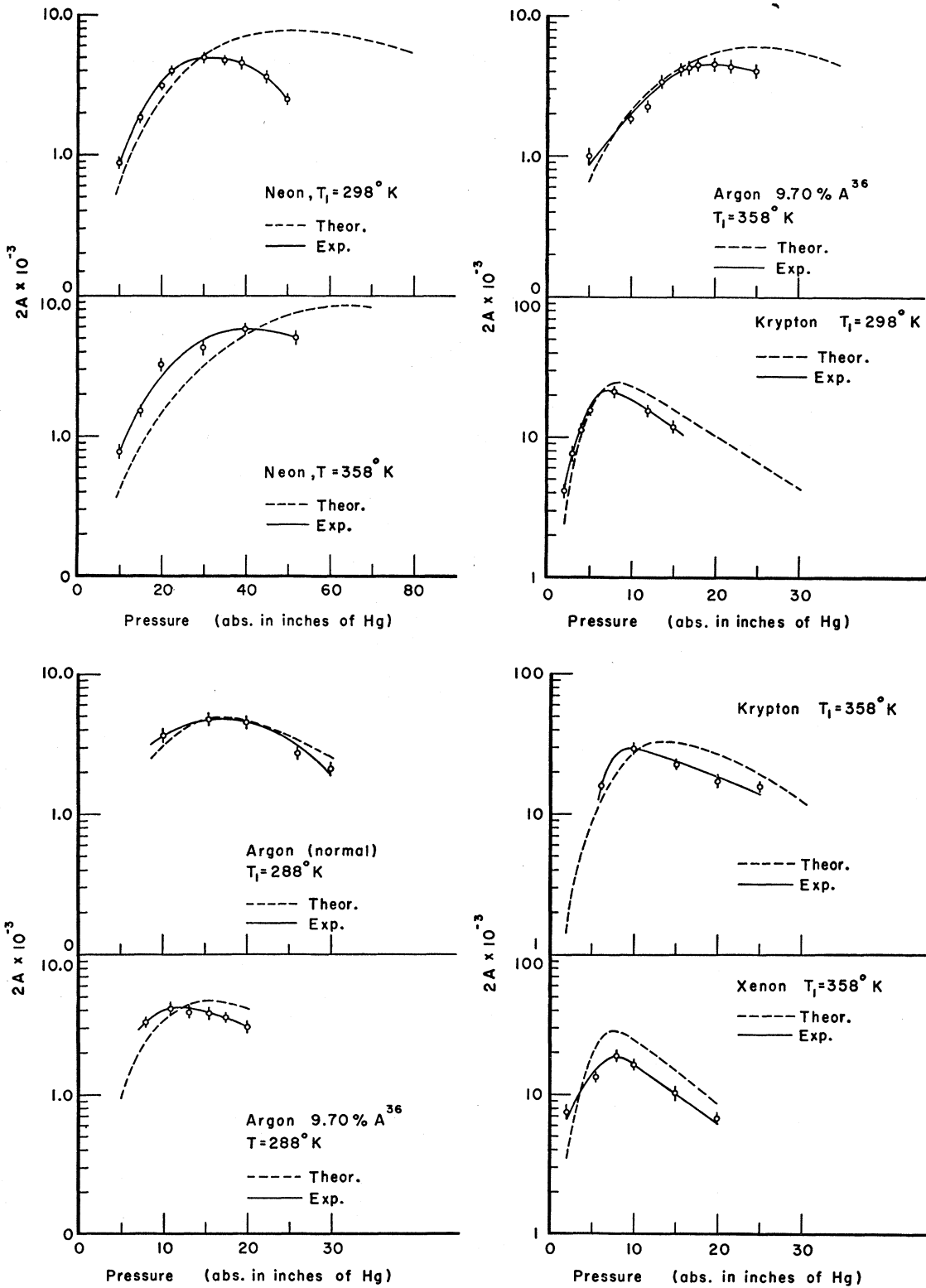


FIG. 1. Plots of the equilibrium separation coefficient  $2A$  versus gas pressure in the column for neon, argon, krypton, and xenon. The ratio  $T_2/T_1$  of the wall temperatures is 2 in every case.

calculated maximum separation factor. For xenon there is actually good agreement between the observed and calculated optimum gas pressures, although the observed maximum factor is smaller than calculated. Most likely this discrepancy arises from the failure of the theory to include any temperature variation of  $\alpha_0$ , which for xenon is quite large ( $\alpha_0=0.08$  at 350°K and approximately 0.16 at 700°K). Requirements (1) and (2) then are mutually contradictory; the gases which are more nearly Maxwellian also display the greatest temperature variation of the thermal diffusion factor, while the gases for which  $\alpha$  is more nearly constant are the least Maxwellian.

Finally, why does this column performance for normal argon at  $T_1=288^\circ\text{K}$  agree fairly well with that calculated from the theory, while there is no comparable agreement for the argon analyzing 9.70%  $\text{A}^{36}$  under similar conditions? Since the shape of the calculated  $2A$ -versus- $p$  curve depends in a sensitive manner on the values of  $D$  (self-diffusion) and  $\eta$  (viscosity), these coefficients must be known to say one percent for accurate comparison. In calculating the performance curve for the argon enriched in  $\text{A}^{36}$ , we used the coeffi-

cients given in the literature for normal argon, but corrected for the change in the average molecular mass. Small but appreciable errors may be present in any or all of these coefficients. It is quite possible that the good agreement for the case of normal argon is the fortuitous consequence of slightly inaccurate gas coefficients. Or, of course, argon may represent a compromise in satisfying requirements (1) and (2) of the theory.

The introduction of a remixing factor  $K_p$  will often improve somewhat the fit between the theoretical and experimental separation-factor curves, of course, and one must admit that there may be small parasitic convection currents in any column operation. Examination of Fig. 1, however, shows that this cannot be a general solution of the problem.

We conclude that measurements of the performance of a thermal diffusion column are not at present a good way to determine thermal diffusion factors with any accuracy. The next step to be taken, if column performance data are to be so used, is to extend the theory to include other than Maxwellian molecules as well as a temperature dependence for the thermal diffusion factor.

## Theory of Sputtering by High-Speed Ions

DAVID T. GOLDMAN\* AND ALBERT SIMON  
Oak Ridge National Laboratory, Oak Ridge, Tennessee  
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The theoretical treatment of ion sputtering at energies above about 50 kev is simplified by the fact that the emergent particles originate at depths in the material which are small compared to the range of the incident particles. The displacement rate is nearly constant over this region and this enables one to obtain relatively simple solutions of the diffusion problem for emission.

The emission problem is reduced to an effective one-velocity diffusion calculation by an artifice. The volume displacement rate is increased by the factor  $\bar{\nu}$ , where  $\bar{\nu}$  is the average number of secondaries, and the macroscopic absorption and scattering cross sections are adjusted to make the average number of collisions of each particle equal to the actual average over the secondaries. The resultant sputtering ratio,  $\mathfrak{R}$ , varies with incident energy  $E$ , incident angle  $\psi$ , and mass ratio  $\mu=M_1/M_2$  (where  $M_1$ =mass of incident particle,  $M_2$ =mass of target particle), as  $\mathfrak{R} \propto \mu(\ln E/E) \sec \psi$ .

### I. INTRODUCTION

**M**OST of the theoretical treatments of ion sputtering appear to be limited to the region below a few kilovolts.<sup>1</sup> In this case, the ions have a mean free path of only a few atomic layers and the phenomenon involves a complicated analysis of surface interactions. As the energy of the bombarding ion increases above about 50 kev, however, a simplifying feature emerges. The range of the incident particle increases greatly ( $\cong 10^{-4}$  cm at 500 kev) while the knock-on particles

maintain an approximately constant mean free path ( $\cong 10^{-7}$  cm). Hence, only a small portion of the initial track length of the incident particle produces knock-on particles which can re-emerge from the surface. The displacement rate is nearly constant over this region and this enables one to obtain relatively simple solutions of the diffusion problem for emission.

In Sec. II of this report an expression is obtained for the volume density of primary particles produced by the incident beam. The mean free path for primary production is calculated in Sec. III, as well as the average energy of the primaries. The average number of secondaries is calculated in Sec. IV, the diffusion problem for re-emission is solved in Sec. V and the results summarized in Sec. VI.

\* Now at Department of Physics, University of Maryland, College Park, Maryland.

<sup>1</sup> D. E. Harrison, Phys. Rev. **102**, 1473 (1956); E. B. Henschke, Phys. Rev. **106**, 737 (1957).