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Laboratory Report No. 66-1

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and
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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
Grant Number: NGR-22-009-121

AFOSR 66-0295

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ABSTRACT

A simple formula in closed form is proposed for the thermal accommodation coefficient from which the accommodation coefficient of a monatomic gas-solid system may be calculated from certain basic parameters of the system. The formula is obtained from consideration of certain aspects of Goodman's lattice theory of accommodation and from conclusions regarding certain properties of available experimental data. Adequate agreement is obtained of the formula with both the lattice theory and the experimental data; in the authors' opinion all the reliable data available are included. Certain corrections are applied to the lattice theory, and the gas-surface potential interaction parameters relevant to the lattice theory are revised.

ACKNOWLEDGEMENT

The calculations for this work were done in part at the Royal Aircraft Establishment, Farnborough, Hants., England; and in part at the Computation Center of the Massachusetts Institute of Technology, Cambridge, Massachusetts, as problem number M4443.

The authors are indebted to Professor L.B. Thomas and to Dr. N. Tho-Nhan for making available some unpublished ac data. The work was jointly supported by the U.S. Air Force Office of Scientific Research under grant number Af 49(638)-1601; National Aeronautics and Space Administration, under grant number NGR-22-009-121; and the Office of Naval Research, under contract number Nonr-3963(03).

INTRODUCTION

The thermal accommodation coefficient (hereafter abbreviated to ac or α) is a measure of the average efficiency of the energy exchange per encounter of a gas molecule with the solid at the gas-solid interface. Experimental methods applied in the measurements of α have been adequately discussed elsewhere (see, for example, the Survey Article by one of us[1]). Here we restrict discussion to a three dimensional (hereafter, "n-dimensional" is abbreviated to "nD") monatomic Maxwellian gas at temperature T which shares an interface with a solid maintained at temperature T_s. If T' is defined by

$$T' = E'/2k \tag{1}$$

where E' is the average energy per reflected gas atom, the thermal accommodation coefficient is defined by

$$\alpha = \frac{T-T'}{T-T_s} \quad . \tag{2}$$

The purpose of this paper is to propose a simple formula in closed form for α from which, it is hoped, the ac of any monatomic gas-solid system may be calculated from certain basic parameters of the system. This formula is partly empirical in origin because, although some of its properties are obtained from existing theory, its final form rests on consideration of certain properties of experimental data.

The first, and perhaps best known, closed-form formula for α was proposed by Baule[2], who considered the interaction in terms of collisions of hard spheres. The accommodation coefficient for the impact of two hard spheres, one representing an incident gas atom of mass M and the other an initially stationary solid atom of mass m, is

$$\alpha = \frac{4\mu}{(1+\mu)^2} \cos^2\eta \qquad (3)$$

where η is the angle between the velocity vector of the incident gas atom and the line joining the centers of the spheres at impact and μ is the mass ratio defined by

$$\mu = M/m \quad . \qquad (4)$$

The Baule formula is obtained by averaging (3) over all η, putting $\overline{\cos^2\eta} = \frac{1}{2}$:

(Baule) $$\alpha = \frac{2\mu}{(1+\mu)^2} \quad . \qquad (5)$$

Landau[3] treated the interaction in terms of classical continuum theory, using an interaction potential V(u) of repulsion only between the gas atom and the surface,

$$V(u) = \varepsilon \exp(-2\kappa u) \qquad (6)$$

where u is the separation of the gas atom from the surface and ε is an energy of arbitrary magnitude. When corrected[4,5], the theory results in the formula

$$\alpha = \frac{3}{mM^{1/2}} \left(\frac{2\pi\hbar^2 \kappa^2 T}{k\,\theta^2} \right)^{3/2} \tag{7}$$

where θ is the Debye temperature of the solid.

Gilbey[5] used a naive continuum model and obtained

$$\alpha = 1 - \exp(-4\xi \cot\xi) \tag{8}$$

where, very approximately

$$\tan^2\xi \sim \mu - 1, \tag{9}$$

(the approximation (9) is not used by Gilbey).

None of the above formulae yields extensive agreement with experimental data, although each may be valid under particular sets of conditions. There are many formulae in the literature which are not in closed form; the best known is, perhaps, the formula of Devonshire[4,5]. However, these also fail to yield extensive agreement with experiment.

A sensible test of any useful formula for acs requires reliable data on the temperature variation of acs of at least two known gases on the same known surface. In our opinion, the most reliable data available are those of Thomas[6] and Silvernail[7] on the temperature dependence of the acs of the inert gases on W, and we have placed considerable reliance on these in this work.

BASIS OF THE FORMULA

Our formula rests heavily on results from the classical lattice theory of acs[8-11] (hereafter referred to as L.T.) and a 3D theory of surface scattering,[12] both developed by one of us, and on conclusions regarding certain properties of available experimental data.

In L.T., the incident gas is monatomic and is represented by a 1D Maxwellian (thermal) gas which impinges normally onto the solid surface, all collisions being head-on with surface atoms; the solid surface is assumed to be clean and is represented by the surface of a simple 3D lattice model, initially at 0°K. It is assumed that α is independent of surface temperature, consistent with data of Thomas and Schofield[13] in the surface temperature range 100 - 300°K and of Watt and Moreton[14] in the range 800 - 1500°K. Interaction potentials of the Morse type are used between the gas atoms and the surface atoms.

The results of L.T. are presented in terms of a parameter $\gamma(t)$ called[10,11] the "effective ac of a single gas atom" and defined by analogy with (2) as follows. Initial and final effective temperatures, t and t_f respectively, of a gas atom interacting with the surface are defined by

$$t = MU_o^2/2k \tag{10a}$$

and

$$t_f = MU_f^2/2k \tag{10b}$$

where U_o and U_f are the initial and final speeds respectively of the gas atom. Accordingly, $\gamma(t)$ for a solid initially at 0°K is defined by

$$\gamma(t) = 1 - t_f/t \tag{11}$$

For a 1D Maxwellian gas at temperature T, the normalized distribution of t is

$$g_{1D}(t) = \frac{1}{T} \exp\left(-\frac{t}{T}\right) \tag{12}$$

and it follows that the ac is given by

$$\text{(1D gas)} \qquad \alpha(T) = \frac{1}{T^2} \int_0^\infty t\, \gamma(t) \exp\left(-\frac{t}{T}\right) dt \quad . \tag{13}$$

This is the formula for $\alpha(T)$ used in L.T.

A typical $\gamma(t)$ curve is shown in Fig. 1; it is determined essentially by four parameters: (i) the critical initial effective temperature, t_c, for trapping, (ii) the effective temperature, t_{min}, at which $\gamma(t)$ has a minimum, (iii) the value of $\gamma(t_{min})$ and (iv) the high temperature limit, $\gamma(\infty)$, of $\gamma(t)$. $\gamma(t) = 1$ for $t \leq t_c$; $\gamma(t) \approx t_c/t$ for values of t just greater than t_c; for larger t, $\gamma(t)$ has a minimum and then approaches the "hard spheres limit", $\gamma(\infty) = \alpha(\infty)$, as $t \to \infty$. An important result of L.T. is that it suggests that the shape of the $\gamma(t)$ curve is independent of the gas-solid system. However, the values of the four parameters of the $\gamma(t)$ curve, t_c, t_{min}, $\gamma(t_{min})$ and $\gamma(\infty)$, depend on the four parameters of the gas-solid system which enter

into the calculation of $\gamma(t)$, viz., M, m, O/a and D, where a and D are parameters of the Morse potential.

L.T. considers only normal, head-on, impact of a gas atom with a surface atom, and if it is generalized to include any type of impact, a more general γ is defined. Accordingly, taking $T_s \approx 0$, we define $\gamma(t,\theta,\phi,x,y)$, where θ is the angle made by the velocity vector of the incident gas atom with the inward normal to the surface, x and y are the coordinates of the initial "aiming point" on the lattice surface (we may take $x = y = 0$ at the center of one of the surface atoms) and ϕ is the incident azimuthal angle (we take $\phi = 0$ as the x-direction). For a 3D Maxwellian gas at temperature T, the normalized distribution of t,θ,ϕ,x and y is

$$g_{3D}(t,\theta,\phi,x,y) = \frac{1}{2\pi AT^2} \sin(2\theta)\ t \exp\left(-\frac{t}{T}\right) \quad (14)$$

where A is the area of the solid surface. It follows that the ac for a 3D Maxwellian gas is

$$\text{(3D gas)} \quad \alpha(T) = \frac{1}{4\pi AT^3} \iiiint t^2 \Gamma(t,\theta,\phi,x,y) \sin(2\theta) \exp\left(-\frac{t}{T}\right) dt\,d\theta\,d\phi\,dx\,dy. \quad (15)$$

We assume that the shape of the $\Gamma(t)$ curve for a 3D gas is also universal and that in addition $\Gamma(t)$ is a sum of two simple and separate parts, a part $\Gamma_1(t)$ significant only at low t and a part $\Gamma_2(t)$ significant only at high t. Accordingly, from (15), $\alpha(T)$ is also a sum of two parts,

$$\alpha(T) = \alpha_1(T) + \alpha_2(T) \ , \tag{16}$$

each defined by analogy with (15).

Representation of $\alpha_1(T)$.

For convenience we define a Step Function, H(x), of x as follows:

$$x < 0; \qquad H(x) = 0 \tag{17a}$$

and

$$x > 0; \qquad H(x) = 1 \ . \tag{17b}$$

We assume that the mathematical form of $\Gamma_1(t)$ for all t is the same as that of the 1D $\gamma(t)$ at low t; that is,

$$\Gamma_1(t,\theta,\phi,x,y) = t_c/t - (t_c/t-1)\ H(t_c/t-1) \tag{18}$$

where $t_c = t_c(\theta,\phi,x,y)$ in general for a particular gas-solid system. Thus Γ_1 is significant only at low t, as required. We consider three Assumptions as to the form of Γ_1.

Assumption 1: t_c is independent of θ,ϕ,x and y and may be written

$$t_c = T_o \ . \tag{19}$$

This assumption combined with (18) and (15) gives

$$\Gamma_1(t) = t_o/t - (T_o/t - 1)\ H(T_o/t-1) \tag{20}$$

and

$$\alpha_1(T) = 1 - \exp\left(-\frac{T_o}{T}\right) - \frac{T_o}{2T}\exp\left(-\frac{T}{T_o}\right) \ . \tag{21}$$

Assumption 2: t_c depends only on θ, and this dependence is such that Γ_1 depends only on the "normal component", $kt\cos^2\theta$, of the incident energy:

$$t_c = T_o \sec^2\theta \tag{22}$$

$$\Gamma_1(t) = (T_o/t)\sec^2\theta - [(T_o/t)\sec^2\theta - 1]H[(T_o/t)\sec^2\theta - 1] \tag{23}$$

and

$$\alpha_1(T) = 1 - \exp\left(-\frac{T_o}{T}\right) + \frac{T_o}{2T}\, ei\left(\frac{T_o}{T}\right) \tag{24}$$

where ei(x) is an exponential integral[15] of x:

$$ei(x) = \int_x^\infty \exp(-\xi)\, d\xi/\xi \quad . \tag{25}$$

Assumption 3: Each "component" of the incident energy, the "normal component", $kt\cos^2\theta$, and the "tangential component", $kt\sin^2\theta$, is accommodated according to a formula of type (18), and each "component" is associated with the same critical initial effective temperature, t_c, expressed as in (19). This assumption, which is the same as that inherent in the lattice theory, implies that

$$\Gamma_1(t) = 2T_o/t - (T_o/t - \cos^2\theta)H(T_o/t - \cos^2\theta) - (T_o/t - \sin^2\theta)H(T_o/t - \sin^2\theta) \tag{26}$$

and leads simply to

$$\alpha_1(T) = 1 - \exp\left(-\frac{T_o}{T}\right) . \tag{27a}$$

We note that the relation (27) follows also by combining (18) with (13), putting $\alpha = \alpha_1$, $\gamma = \Gamma_1$ and $t_c = T_o$. For low T, L.T. gives a similar result:

(low T) $$\alpha(T) \simeq 1 - \exp - \frac{t_c}{T} \quad . \tag{27b}$$

None of these three Assumptions gives results very different from either of the others in the ranges of interest (T_o appears as a dispensible parameter). We choose (27), that is Assumption 3, to represent $\alpha_1(T)$ on the basis of the simplicity of the result and the fact that it may be considered a first approximation to either of the other results, and in fact, has a value which lies between them.

Representation of $\alpha_2(T)$

We let ω_o be a "characteristic frequency" associated with the solid surface, and τ_i an "interaction time" of the gas-solid interaction[9-11]. We assume that the hard spheres limit is approached asymptotically as $\omega_o\tau_i \to 0$, and a first guess at the form of this approach may be

$$\text{(large t)} \qquad \frac{\Gamma_2(t,\theta,\phi,x,y)}{\Gamma_2(\infty,\theta,\phi,x,y)} = 1 - \exp\left(-\frac{1}{\omega_o\tau_i}\right) \quad . \qquad (28a)$$

However, we find that the experimental data are, in general, approximated better by the form

$$\text{(large t)} \qquad \frac{\Gamma_2(t,\theta,\phi,x,y)}{\Gamma_2(\infty,\theta,\phi,x,y)} = \tanh\left(\frac{1}{\omega_o\tau_i}\right) \qquad (28b)$$

and we use the form (28b) throughout this paper.

We assume that, at least for small values of μ, τ_i is related approximately to the normal component, $U_o\cos\theta$, of the incident velocity of the gas atom normal to the solid surface and to the Morse parameter $\underline{a}$ of the gas atom-solid atom potential by

$$a\tau_i U_o \cos\theta \simeq \text{constant} \tag{29}$$

Also, U_o is related to t via (10a) to obtain

$$\text{(large t)} \qquad \frac{\Gamma_2(t,\theta,\phi,x,y)}{\Gamma_2(\infty,\theta,\phi,x,y)} = \tanh\left[c' \frac{a}{\omega_o} \left(\frac{kt}{M}\right)^{1/2} \cos\theta\right] \tag{30}$$

where c' is a constant of order unity.

L.T. is concerned with evaluation of $\Gamma(t,0,\phi,0,0)$ and for $\mu < \sim 0.84$ the hard spheres limit of this function is[10,11]

$$\Gamma(\infty,0,\phi,0,0) = 4\mu/(1+\mu)^2 \ . \tag{31}$$

The form of $\Gamma_2(\infty,\theta,\phi,x,y)$ is obtained through consideration of results from a 3D theory of surface scattering[12] cited above, an approximate result of which is

$$\frac{1}{A} \int\int \Gamma(\infty,\theta,\phi,x,y)\,dx\,dy \simeq \frac{3.6\mu}{(1+\mu)^2} \cos\theta \ . \tag{32}$$

Oman[16] proposed a similar formula in which the 3.6 on the R.H.S. is replaced by 4. However, we consider (32) more nearly correct. Combining (32) with (30) gives

$$\text{(large t)}\ \frac{1}{A}\int\int\Gamma(t,\theta,\phi,x,y)\,dx\,dy \simeq \frac{3.6\mu}{(1+\mu)^2} \cos\theta \tanh\left[c' \frac{a}{\omega_o} \left(\frac{kt}{M}\right)^{1/2} \cos\theta\right] \tag{33}$$

where we have used the fact that $\Gamma_2(\infty,\theta,\phi,x,y) = \Gamma(\infty,\theta,\phi,x,y)$.

We assume further than an approximate form of $\alpha_2(T)$ for large T may be obtained from (15) and (33) by replacing $\cos\theta$ and t in the integral by their respective mean values via (14).

$$\overline{\cos\theta} = 2/3 \tag{34a}$$

and

$$\bar{t} = 2T$$

to give

$$\alpha_2(T) = \frac{2.4\mu}{(1+\mu)^2} \tanh\left[c \frac{a}{\omega_o} \left(\frac{kT}{M}\right)^{1/2} \right] \tag{35}$$

where c is a constant of order unity. In order that $\alpha_1(T) + \alpha_2(T)$ should not exceed unity at smaller T we multiply α_2 by $\exp(-T_o/T)$ to obtain

$$\alpha_2(T) \approx \alpha(\infty) \tanh\left[\frac{am}{\lambda'} \left(\frac{T}{M}\right)^{1/2}\right] \exp\left(-\frac{T_o}{T}\right) \tag{36}$$

where

$$\alpha(\infty) = 2.4\mu/(1+\mu)^2 \tag{37}$$

and λ' is a parameter whose value depends only on the solid surface.

The arguments above may have to be modified for larger values of μ because of the possibility of several collisions at each encounter between gas atom and solid surface. In principle, the nature of these modifications is not clear. In practice, we find better overall agreement with experimental data, especially those relevant to systems with larger values of μ, when the argument of the tanh function is multiplied by $(1+\mu)^2$, and λ' is replaced by a new λ whose value again depends only on the solid surface.

Our representation of $\alpha_2(T)$ is

$$\alpha_2(T) = \alpha(\infty) \tanh \left[\frac{(MT)^{1/2}}{\alpha(\infty)} \frac{a}{\lambda} \right] \exp\left(-\frac{T_o}{T}\right) \quad (38)$$

where the hard spheres limit $\alpha(\infty)$, given by (37), is assumed valid for all values of μ; this assumption is supported by qualitative evidence from the lattice theory[8-11].

Our formula is obtained by combining (16) with (27) and (38):

$$\alpha(T) = 1 - \exp\left(-\frac{T_o}{T}\right) + \alpha(\infty) \tanh\left[\frac{(MT)^{1/2}}{\alpha(\infty)} \frac{a}{\lambda}\right] \exp\left(-\frac{T_o}{T}\right) . \quad (39)$$

The constant c of (35) may be related to the λ of (39) by equating the arguments of the two tanh functions. We assume that the characteristic frequency of the surface, ω_c, is the Debye frequency of the solid; that is

$$\omega_o = k\theta/\hbar \quad (40)$$

and we obtain, for small μ,

$$c \approx \frac{m\theta}{17\lambda} \text{ mole}^{\frac{1}{2}} \text{ gm.}^{-\frac{1}{2}} \text{ deg.}^{-\frac{1}{2}} \text{ A.}^{-1} . \quad (41)$$

PROCEDURES AND COMPARISON WITH EXPERIMENTAL DATA

We use two different types of experimental data, one giving the dependence of α on T for several gas-solid systems, the other consisting of α values for several gas-solid systems, each determined at a single T value. We select only data from experiments in which we think considerable care has been taken to control the state of the surface, and to our knowledge we include all available data of this nature. Discussion of available ac data and the consequences of failing to excercise reproducible control in the course of experiment is given elsewhere[1]. We use one of two procedures, depending on the nature of the data, to check our formula against experiment; these two procedures (A and B) are described below together with a third (C) which is useful in special cases.

Procedures

Procedure A is used when temperature-dependent data are available for at least one gas. If data are available for more than one gas on the same solid, it is convenient to begin with the gas which at any given temperature is suspected to adsorb least on the given surface, that is, the gas associated with the lowest value of T_o (He in all cases we considered here). We then find the "best fit" values of T_o and λ, i.e., the values for which our Formula (39) fits the T-dependent data as well as possible. The resultant value of T_o is appropriate to the gas-solid system and

that of λ to the particular surface under consideration. This value of λ is used with the T-dependent data for other gases on the same surface to obtain the best-fit value of T_o appropriate to each of the other gas-solid systems.

Procedure B is used when ac data on a given surface are available at only one temperature. It may be properly applied only when the ac on a given surface is known for at least one gas for which T_o is small; otherwise the calculated value of λ is liable to considerable error. For any particular surface we use the He ac whenever available to find the value of λ for the surface (setting $T_o = 0$ for He), and then use this value of λ to find the value of T_o for each of the other gases on this surface.

Procedure C provides a quick method for finding a value (approximate in general) of λ. It is applicable when data are available in a temperature range where $\alpha_1(T)$ as defined by (27) is negligible compared to $\alpha(T)$, and where the difference between the tanh function in (39) and its argument is negligible. For an accuracy of 5%, the first conditions implies that

$$T > \frac{20T_o}{\alpha(T)} \tag{42}$$

and the second that

$$\alpha(T) < 0.4\,\alpha(\infty) . \tag{43}$$

When both of these conditions prevail (39) may be approximated by

$$\alpha(T) \approx (MT)^{1/2}\, a/\lambda . \tag{44}$$

Thus one exact value of α in this temperature range is sufficient to obtain a good approximation to the value of λ. In practice, measured ac values may be plotted against $T^{\frac{1}{2}}$ to yield a straight line through the origin whose gradient determines λ. This value of λ may be used with experimental data at lower values of T, where $\alpha_1(T)$ is not negligible compared to $\alpha(T)$, to find T_o; this value of T_o may then be used to correct the previously estimated value of λ, and a new value of T_o determined.

Comparison with experimental data

Most of the data on the temperature dependence of the acs of the inert gases on tungsten are due to Thomas[6] and Silvernail[7]; the others (at higher temperatures for the He-W system) are due to Goldstein and Tho-Nhan[17] and to Wachman[18]. The data of Refs. 6 and 7 were obtained with both simple-filament and potential-lead tubes. The data from simple filament tubes (represented by open circles in Figs. 2-4) have been corrected for end-losses[1,19] while other data (represented by diamonds) are uncorrected from potential lead tubes. The data of Ref. 17 (squares in Fig. 2) are from a potential-lead tube while those of Ref. 18 (Triangles in Fig. 2) are from simple-filament tubes; neither set is corrected for end losses. Failure to apply end-loss corrections to data from simple filament tubes results in ac values which are slightly too large (about 8% for the data of Ref. 18). All the experimental tubes except those of Ref. 18 contained an evaporated mischmetal getter on the tube walls to aid in maintaining clean filament surfaces.

Values of gas pressures used in calculating acs from experimental measurements may require correction for thermal transpiration if pressures are determined when the McLeod gauge and the experimental tube are at different temperatures; such corrections have been applied to the data of Refs. 6, 7 and 18. In our opinion, the only temperature dependent ac data available on clean surfaces are on tungsten and the most reliable are those of references 6 and 7 in the temperature range 77 - 303°K. Procedures used by Raines[20] to obtain the ac of He on Ni (see Fig. 5) suggest to us that the resulting surface is not characteristic of the clean metal, although it may be of ne ly constant composition over the experimental temperature range.

The experimental ac values on Na and K from 77° to 298°K obtained by Petersen[21] were determined on surfaces of the bulk metals (rather than on monolayers of these metals atop a foreign metal) which were presumed to be free of adsorbed gases. Blickensderfer[22] working in the same laboratory after Petersen found that alkali metals interact with glass to form hydrogen and concluded that this gas was very likely present during the ac measurements cited in Ref. 21. Petersen[21] assumed that hydrogen does not chemisorb on bulk potassium at room temperature, and cited the work of Trapnell[24] in partial support of this assumption. However, the reaction vessel used by Trapnell[24] was of Pyrex, and the work of Blickensderfer suggests that hydrogen was very likely present during these measurements also. Indeed, Trapnell[23] noted the presence of a foreign gas, which he

identified as hydrogen, in the process of studying adsorption of C_2H_2 on potassium; however, he attributed its origin to a reaction of K with C_2H_2. Accordingly, the potassium surface used by Trapnell might have been hydrogen saturated before being deliberately exposed to the adsorbate gas. Because of these considerations, it is our opinion that the states of the surfaces in Petersen's experiments are uncertain. Neither the formula nor the lattice theory can be made to agree satisfactorily with the temperature dependence exhibited by these data; for these reasons we have used only room temperature data from Ref. 21 and Procedure B on the assumption that at room temperature each surface is of constant composition in its interaction with He, Ne and A.

We use twenty-one ac values of inert gases, each at one temperature (see Table I), on eight metal surfaces, all determined in Thomas' laboratory except the value for the ac of Ne on Fe due to Eggleton and Tompkins[30]. Another set of ac values we cite below, due to Wachman[31] consists of data on the ac of He and Ne near room temperature on initially clean W deliberately covered with known adsorbates.

The theoretical curves obtained on applying Procedure A to the temperature dependent data for He, Ne, A, Kr and Xe on W [Refs. 6, 7, 17 and 18] and for He - Ni [Ref. 20] are shown as continuous curves in Figs. 2-5. The dash-dot curve in Fig. 3 which agrees better with the Ne-W data than does the continuous curve is obtained when $\alpha_2(T)$ is represented by (36) rather than by the modified form (38). Agreement with the He-W data is

the same regardless of which of the two forms for $\alpha_2(T)$ is chosen because Procedure A begins with the He-W data. Hence, for W, the λ' of (36) is related to that from (38) by

$$\lambda' \approx 2.30\,\lambda \,. \qquad (45)$$

Our reason for selecting, in general, the form in (38) over that in (36) are stated above. For the case of Ne-W, we display $\alpha_1(T)$, $\alpha_2(T)$ and $\alpha(T)$ separately in Fig. 6, using the form (38) for $\alpha_2(T)$.

Dashed curves in Figs. 2-5 represent the best fit curves obtained from L.T. as it stands in Refs. 10 and 11, where the fitting procedure is described. Forms of the response function of the lattice model used in L.T. calculations for A, Kr and Xe on W are approximate[10,11] while those used for He on W and Ni and Ne on W are more exact[32], in all cases the lattice model is that described in Refs. 10 and 11 (all the spring constants associated with a given metal have the same value).

Tables I and II contain lists of the systems treated by Procedures A and B respectively. Appropriate values of α and T are included in Table II. The sources of data are given in the Tables, together with the computed values of λ and c for each surface and the values of μ, $\alpha(\infty)$ and T_o for each gas-surface system. The values of λ' and T_o' for He-W and Ne-W in Table I are those values appropriate when $\alpha_2(T)$ is represented by (36). Estimates of T_c for a given gas by Procedure B are obtained where α values on the same surface are known for that gas and for another gas having $T_o \approx 0$ (i.e., He in all cases

we have treated). In estimating T_o we arbitrarily set $\alpha_1 \approx 0$, implying $T_o \approx 0$, if α_2 is found to be within 10% of the experimental value of α. We have no data for the system He-Fe, and we assume that $T_o = 0$ for Ne-Fe.

L.T. is applied only to these systems for which ac data are available at several temperatures. Accordingly the values of the dispensible parameters of the theory, θ/a and D, and the resulting value of t_c derived from comparison of these data with L.T. are included in Table I. The values of θ/a and D in Table I for He-W, He-Ni and Ne-W are not the same as those cited in Refs. 8-11. The differences in the values associated with He-Ni arise because this system was previously treated by a perturbation theory[9] which is not used here; the differences in the values associated with He-W and Ne-W arise because certain errors in values of experimental data used in Refs. 9-11 have been eliminated in the present case. Also included in the Tables are relevant values of θ and $\underline{a}$. The values of $\underline{a}$ in Table II and those in Column 4 of Table I are calculated from literature values of the $\underline{a}$'s appropriate to the gas atom-gas atom and solid atom-solid atom interaction by means of a combination rule[9]. Values of θ from the sources cited are used to calculate c from (41) and to derive the values of $\underline{a}$ appropriate to L.T. (column II, Table I) from L.T. values of θ/a.

The value θ = 380°K is chosen for W rather than the value θ = 330°K used in Refs. 9-11 because the former gives better overall agreement between $\underline{a}$ values derived from L.T. and the calculated values. The value θ = 220°K is chosen for Ni from Ref. 28 rather than the value θ = 375°K from Ref. 29 because the former gives the same value of $\underline{a}$ for He-Ni as that for He-W.

DISCUSSION

The formula (38), represented by the continuous curves in Figs. 2-5, agrees well with the ac data used, which cover wide ranges in α and T. The best agreement is with the He data on W and on Ni; we include the Ni data notwithstanding our reservations regarding its quality, on the assumption that surface conditions are constant throughout the temperature range.

As stated above the representation of $\alpha_2(T)$ for Ne-W by (36) (dash-dot curve in Fig. 3) instead of (38) results in better agreement with the experimental data. It is evident from Figs. 2-5 that agreement between the Formula and L.T. (dashed curves in Figs. 2-5) is very good, the best agreement being for Ne-W when $\alpha_2(T)$ is represented by (36). In all cases except those of He the curves from L.T. fit the data slightly better than those from the Formula. An important difference between the Formula and L.T. is in $\alpha(\infty)$ which has the value $2.4\mu/(1+\mu)^2$ in the formula and $4\mu/(1+\mu)^2$ (for small μ) in L.T. As a consequence of this difference, L.T. in its present form must, at large values of T, result in larger values of α than does the Formula. The divergence between them begins to become evident not far above room temperature for He on W and Ni (Figs. 2 and 4). Differences between values of t_c from L.T. and the corresponding values of T_c from the Formula arise in part from the difference in the $\alpha(\infty)$ values. Although in no case is t_c very different from T_o (Table I) it is found that $t_c < T_o$ for

data to the left of the minimum in $\alpha(T)$ (A, Kr and Xe on W) and $t_c > T_o$ for other data (He on W and Ni, and Ne on W). When for the system Ne-W $\alpha_2(T)$ is represented by (36) the resulting value of T_o' is very close to the corresponding t_c (Table I).

From the values of T_o of Table II, we note that in no case does the value of α_2 calculated from the Formula for Ne differ from the experimental value of α by more than 10%. This result lends considerable support to the Formula especially in view of the large ranges of the relevant parameters (e.g., mass ratio μ) through which such agreement is obtained. From Tables I and II we note that the general trends in T_o values for A, Kr on Xe are reasonable; i.e., T_o increases with increasing gas molecular weight on a particular surface. However, the values of T_o for Li seem very low and T_o = 900°K seems unreasonably large for Xe-Mo. This last T_o value, however, may be too large because of the very large effect which a relatively small experimental error in the α value may have on it; e.g., a 15% reduction in the cited experimental value of the ac of Xe-Mo would reduce T_o to 460°K.

Values of $\underline{a}$ for He in Table I calculated from L.T. are too low in comparison with those of the other gases, and we consider this entire set of $\underline{a}$ values unsatisfactory (see the discussion in Ref. 9). The values of $\underline{a}$ are likely to be in error because L.T. gives too high a value of $\alpha(\infty)$, and as a consequence too large a slope of the $\alpha(T)$ curve just to the right of α_{min}. The effect is most pronounced for those systems

for which data are available in this range, viz., He-W and He-Ni. Since, in general, large $\underline{a}$ values lead to large slopes, agreement of L.T. with the He data is forced by selecting an unrealistically small value of $\underline{a}$. Realization of these facts suggests a method to be presented in a later publication for improving L.T.

Values of the parameter λ and the parameter c calculated for each surface from λ using (41) appear in the Tables. We have no idea about what are realistic values for λ. However, it is remarkable that in nearly every case the value of c is very close to unity, the notable exceptions being c = 0.2 for Li and c = 2.4 for Pt. If the proper value of c for each surface is indeed unity, then we must suppose that the value α = 0.038 for He on Pt[34] is too high. However, if we were to postulate on the same bases that the value of c for Li should be closer to unitywe could have to make the assumption that the experimental value of the ac of He-Li is too low, which is difficult to justify.

Several authors (cited in Ref. 35) have noted that the ratio of the ac of Ne to that of He on the same surface is approximately equal to the square root of the mass ratio of Ne/He, i.e.,

$$\frac{\alpha_{Ne}}{\alpha_{He}} \simeq 2.2 \qquad (46)$$

We believe that this result is fortuitous in the sense that

room temperature, where most of the relevant data were taken, is in the range where (44) applies approximately to both He and Ne on many surfaces. Accordingly, using (44) and the approximation $\underline{a}_{He} \approx \underline{a}_{Ne}$ gives

$$\frac{\alpha_{Ne}(T)}{\alpha_{He}(T)} \approx \left(\frac{M_{Ne}}{M_{He}} \right)^{1/2} \tag{47}$$

Fig. (7) contains a plot of $\alpha_{Ne}(T)/\alpha_{He}(T)$ for a tungsten surface, using the Formula, for $0 < T < 2500°K$. It is clear from the plot that (46) may be said to hold over the entire range of experimentally accessible temperatures above about 250°K. It is interesting that (46) holds not only on clean surfaces but also on gas covered surfaces. For example, according to Ref. 31 the experimentally determined ratio α_{Ne}/α_{He} at room temperature is 2.73 on hydrogen covered tungsten, 2.53 on deuteruim covered tungsten and 2.47 on oxygen on tungsten. We are not yet able to deal with systems in which the surface is gas covered because we do not know how to estimate $\alpha(\infty)$ for such cases. However, on the basis of available data it is reasonable to expact that a plot similar to that in Fig. 7 would apply to these systems also.

Perhaps the most reliable prediction that can be made with the formula at this time is about the behavior of $\alpha(T)$ for He^3 on tungsten. The value of c for a tungsten surface is known and the value T_o and hence $\alpha_1(T)$ for He^3-W is likely to be small, therefore $\alpha(T)$ can be estimated with some confidence.

Since Thomas[6] has expressed his intention to study this system experimentally, it appeared worthwhile to include a comparison (Fig. 8) of the $\alpha(T)$ curve for He^3-W with that for He^4-W, using the same value of T_o = 0.2°K for both. The $\alpha(T)$ for He^3-W is not evaluated below 50°K where a large error in T_o would affect the results significantly.

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TABLE I

Results of Analysis of T-Dependent Data

System	Source Ref.	μ[a]	$\alpha(\infty)$[b]	a A^{-1}	θ deg.	λ[e] $\frac{gm^{\frac{1}{2}}\,cgs^{\frac{1}{2}}}{mole^{\frac{1}{2}}\,A}$	T_o[e] deg.	c[f]	θ/a[g] deg.A	D[g] $\frac{kcal}{mole}$	a[h] A^{-1}	t_c[g] deg.	λ'[i] $\frac{gm^{\frac{1}{2}}\,deg^{\frac{1}{2}}}{mole^{\frac{1}{2}}\,A}$	T_o'[i] deg.
He-Ni	20	0.0682	0.143	1.64[c]	320[28]	740	0.7	1.5	240	0.18	1.3	1.1	--	--
He-W	6	0.0218	0.0500	1.64[c]	380[29]	3200	0.2	1.3	290	0.21	1.3	0.30	7400	0.2
Ne-W	6	0.110	0.214	1.60[d]	380[29]	3200	1.8	1.3	220	0.50	1.7	2.5	7400	2.6
A-W	7	0.217	0.352	1.44[d]	380[29]	3200	70	1.3	250	2.4	1.5	62	--	--
Kr-W	7	0.456	0.516	1.34[d]	380[29]	3200	155	1.3	250	4.0	1.5	140	--	--
Xe-W	7	0.714	0.583	1.33[d]	380[29]	3200	380	1.3	250	6.4	1.5	350	--	--

(a) From values of M and m in Ref. 25.

(b) From equation (37) of this paper

(c) Evaluated from the combination rule and the value of a for He-He in Ref. 9 and the values of a for Ni-Ni and W-W in Ref. 26.

(d) From the combination rule in Ref. 9 and the data on a for the gas-gas and solid-solid systems in Refs. 26 and 27.

(e) From application of Procedure A.

(f) From (41) of this paper

(g) From the lattice theory calculation

(h) Calculated by combining Columns 5 and 9 of this table.

(i) From application of procedure A where $\alpha_2(T)$ is represented by (36).

Table II Results of Analysis of Data Independent of T

System	Source Ref.	α_{exp}	T deg.	μ[b]	$\alpha(\infty)$[c]	a A^{-1}	θ[29] deg.	λ[h] $\frac{gm^{\frac{1}{2}}\,deg^{\frac{1}{2}}}{mole^{\frac{1}{2}}\,A}$	T_o deg.	c[j]
He-Al	33	0.073	305	0.148	0.270	1.45[d]	390	670	0[e]	0.92
He-Be	34	0.145	305	0.444	0.511	1.64[e]	1000	380	0[e]	1.4
He-K	21	0.083	298	0.102	0.202	0.79[d]	100	310	0[e]	0.74
He-Li	6	0.024	80	0.577	0.557	1 [e]	360	750	0[e]	0.20
He-Mo	6	0.0261	298	0.0417	0.0927	1.70[d]	380	2200	0[e]	1.0
He-Na	21	0.090	298	0.174	0.303	0.91[d]	150	340	0[e]	0.60
He-Pt	34	0.038	305	0.0205	0.0473	1.64[f]	230	1100	0[e]	2.4
Ne-Al	33	0.159	305	0.748	0.587	1.42[g]	390	670	$\sim$ 0[i]	0.92
Ne-Be	34	0.315	305	2.24	0.512	1.60[e]	1000	380	$\sim$ 0[i]	1.4
Ne-Fe	30	0.053	300[a]	0.361	0.468	1.58[g]	420	2300	$\sim$ 0[i]	0.60
Ne-K	21	0.199	298	0.516	0.539	0.78[g]	100	310	0[e]	0.74
Ne-Li	6	0.049	80	0.577	0.557	1 [e]	360	750	$\sim$ 0[i]	0.20
Ne-Mo	6	0.055	298	0.210	0.345	1.66[g]	380	2200	$\sim$ 0[i]	1.0
Ne-Na	21	0.198	298	0.878	0.598	0.89[g]	150	340	$\sim$ 0[i]	0.60
A-K	21	0.444	298	1.02	0.600	0.75[g]	100	310	90[i]	0.74
A-Li	6	0.290	80	5.76	0.303	1 [e]	360	750	20[i]	0.20
A-Mo	6	0.3155	298	0.416	0.498	1.50[g]	380	2200	90[i]	1.0
A-Na	21	0.459	298	1.74	0.557	0.84[g]	150	340	100[i]	0.60
Kr-Li	6	0.400	80	12.1	0.170	1 [e]	360	750	30[i]	0.20
Kr-Mo	6	0.510	298	0.873	0.597	1.39[g]	380	2200	180[i]	1.0
Xe-Mo	6	0.956	298	1.37	0.585	1.37[g]	380	2200	900[i]	1.0

TABLE II

(a) Not stated in Ref. 30.

(b) From values of M and m in Ref. 25.

(c) From equation (37) of this paper

(d) Evaluated from the combination rule and the value of $\underline{a}$ for He-He in Ref. 9 and the values of $\underline{a}$ for Ni-Ni and W-W in Ref. 26.

(e) Guessed.

(f) Guessed as in Ref. 9.

(g) From the combination rule in Ref. 9 and the data for the gas-gas and the solid-solid systems in Refs. 26 and 27.

(h) From application of Procedure B.

(i) Estimated by Procedure B.

(j) From (41) of this paper.

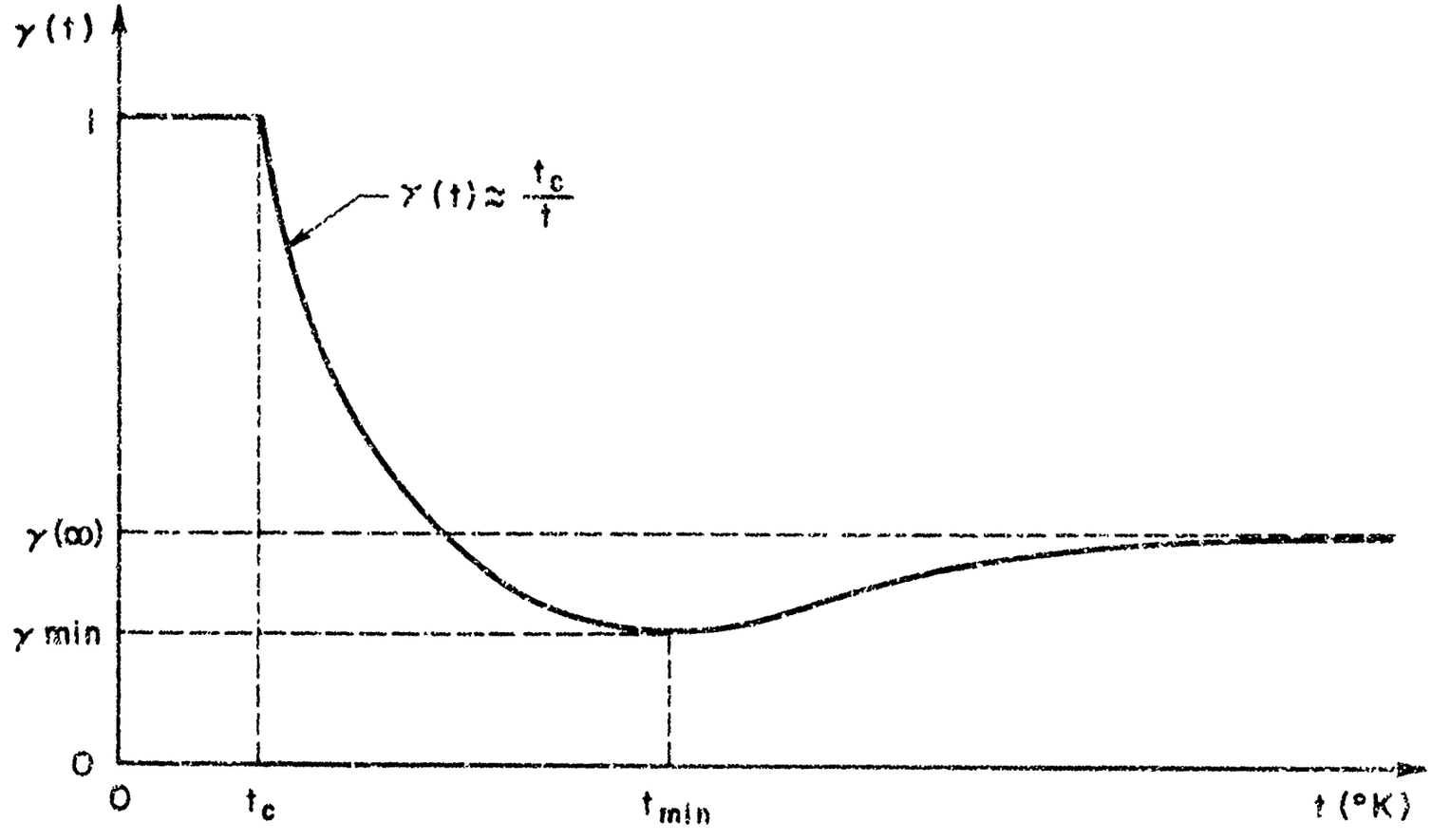

Fig. 1 Typical γ(t) curve from the lattice theory.

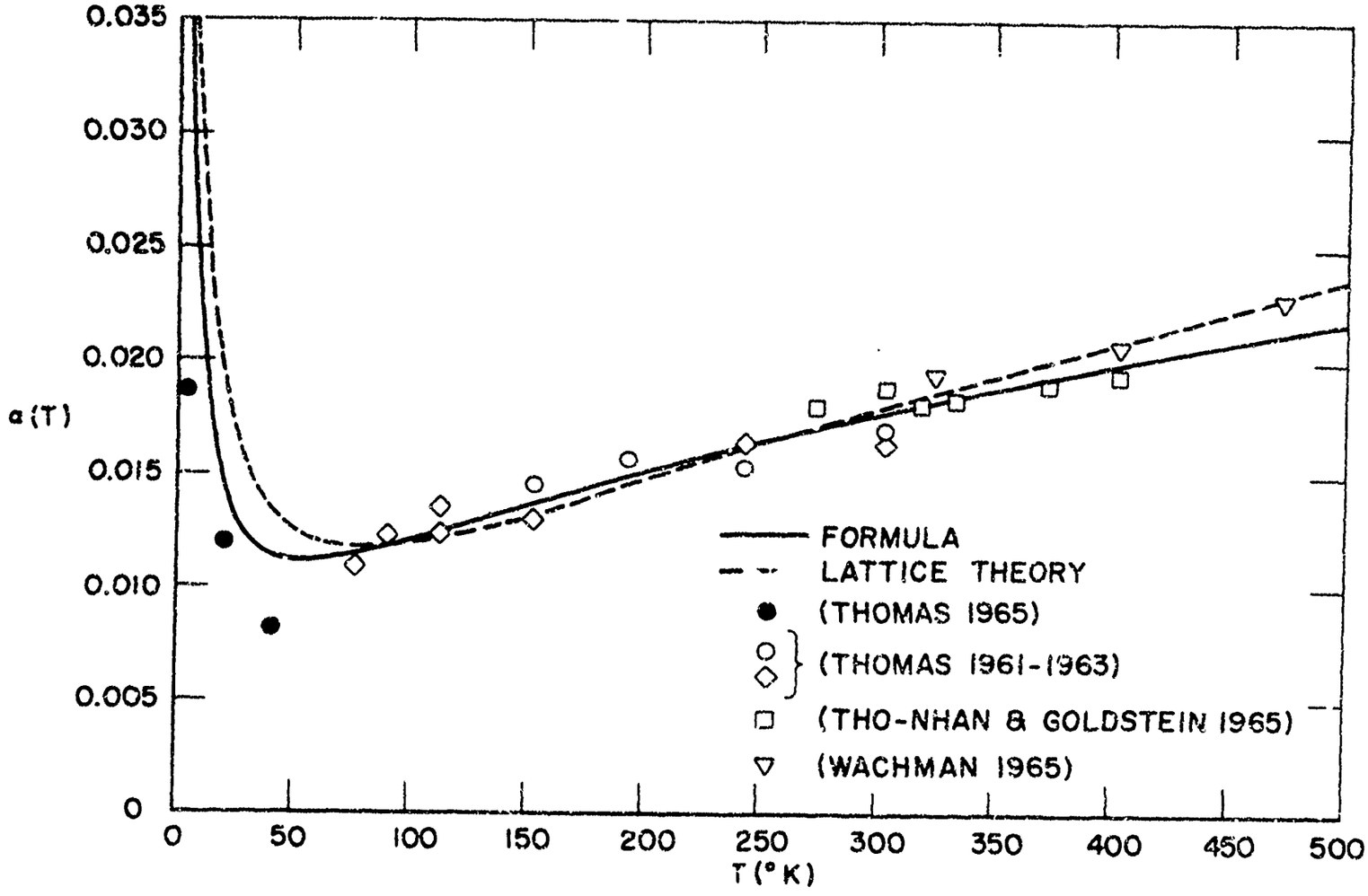

Fig. 2 Comparison of the theories with experimental data on $\alpha(T)$ for He-W.

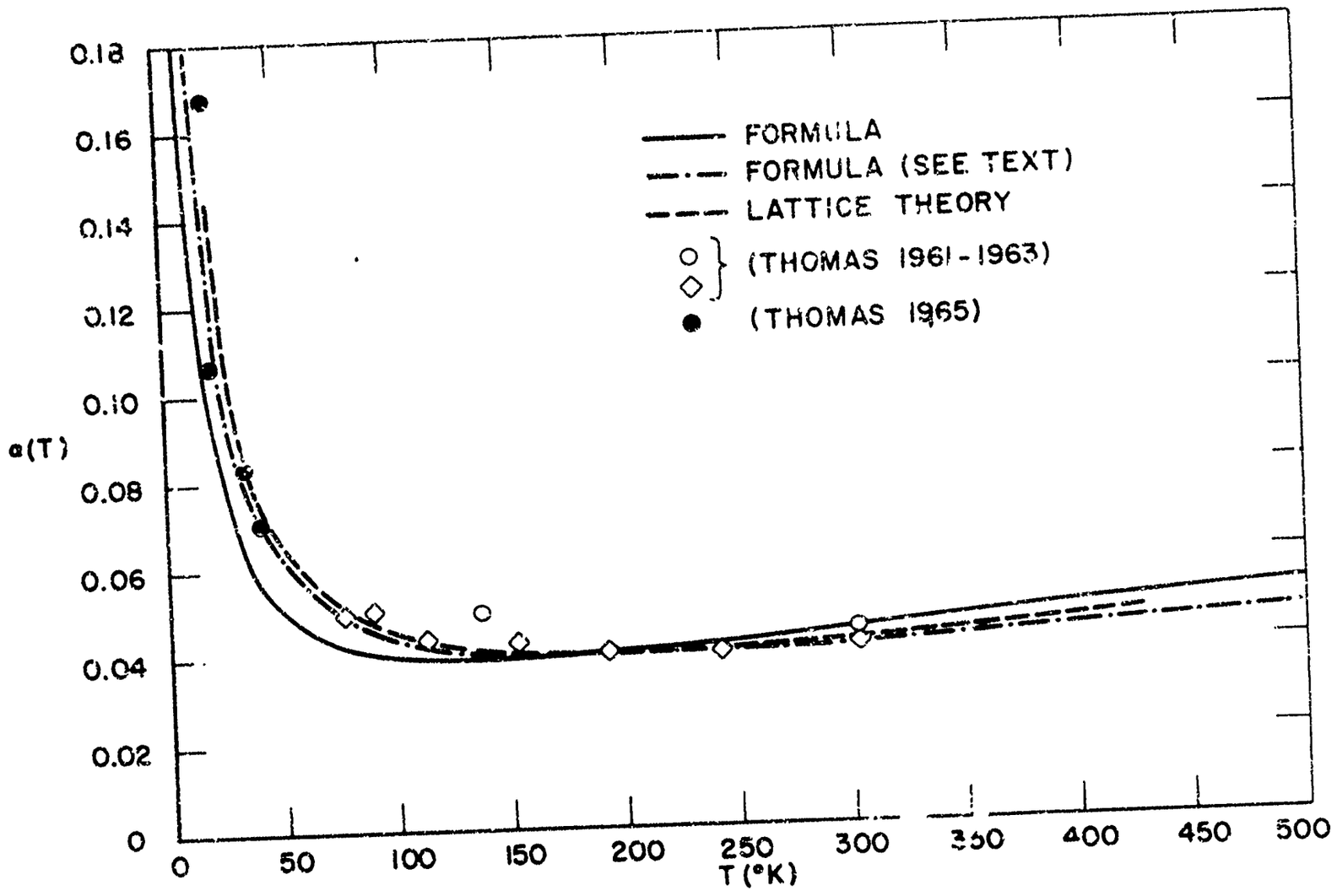

Fig. 3 Comparison of the theories with experimental data on $\alpha(T)$ for Ne-W.

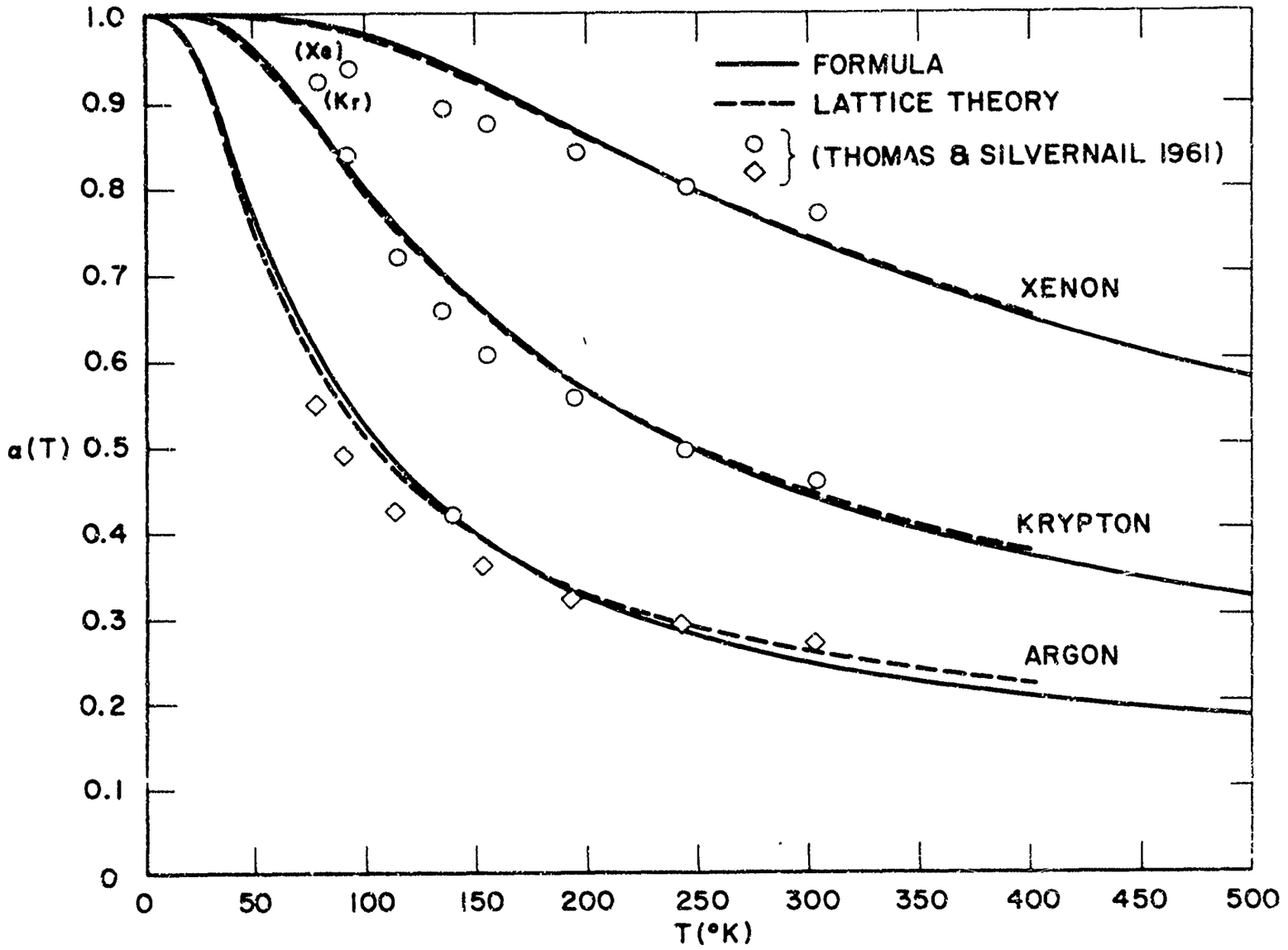

Fig. 4 Comparison of the theories with experimental data on $\alpha(T)$ for A, Kr and Xe-W.

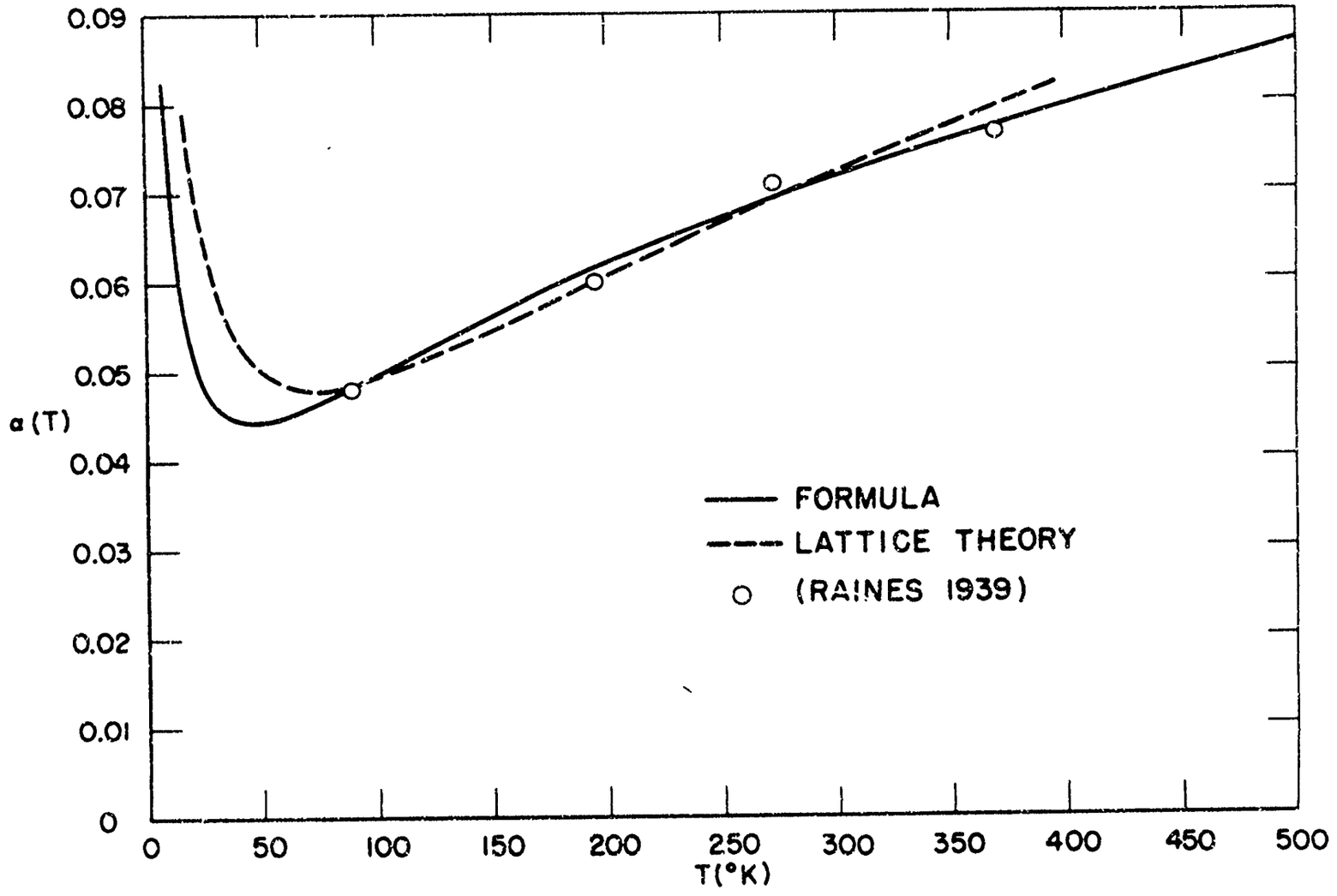

Fig. 5 Comparison of the theories with experimental data on $\alpha(T)$ for He-Ni.

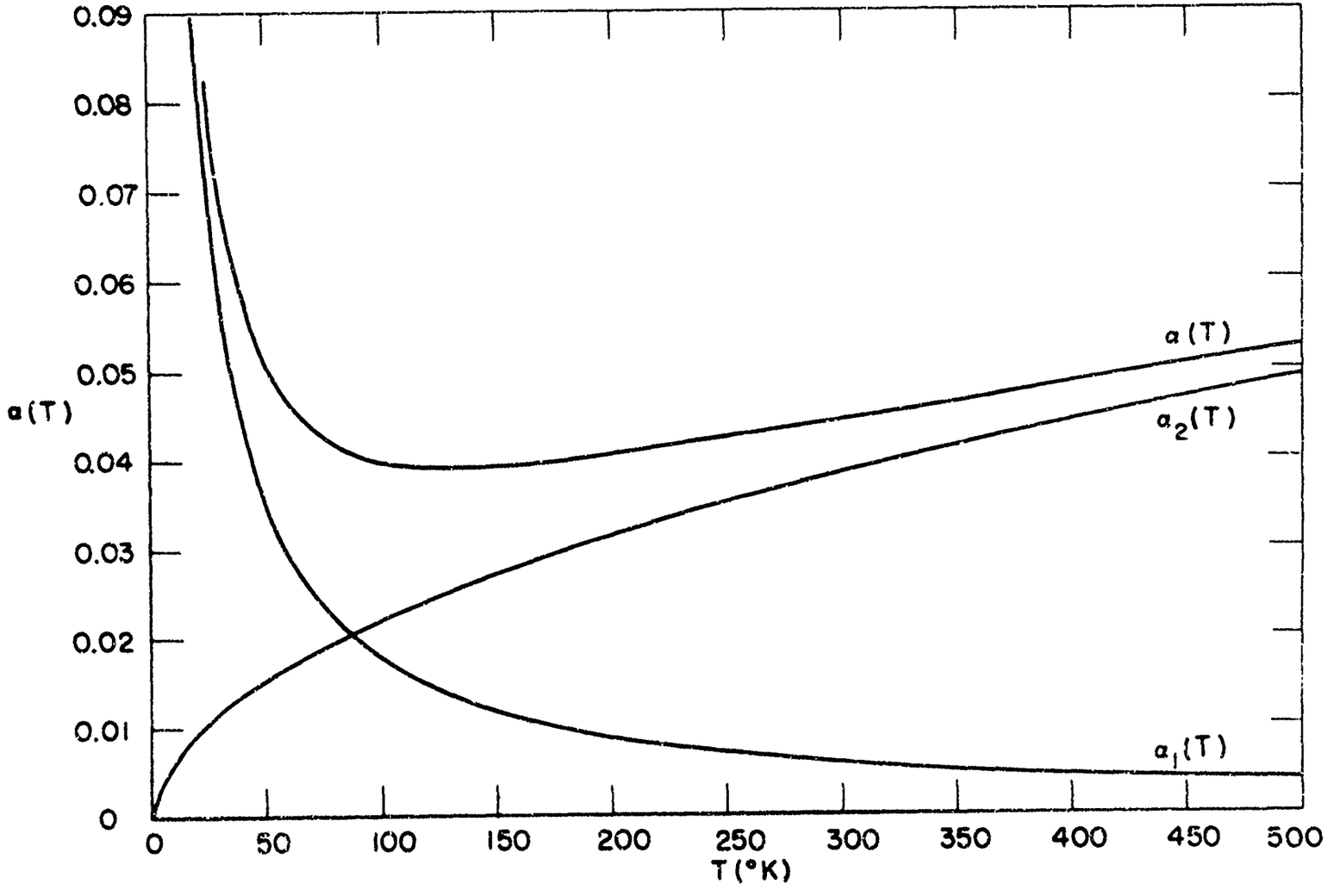

Fig. 6 $\alpha_1(T)$, $\alpha_2(T)$ and $\alpha(T)$ for Ne-W from the Formula.

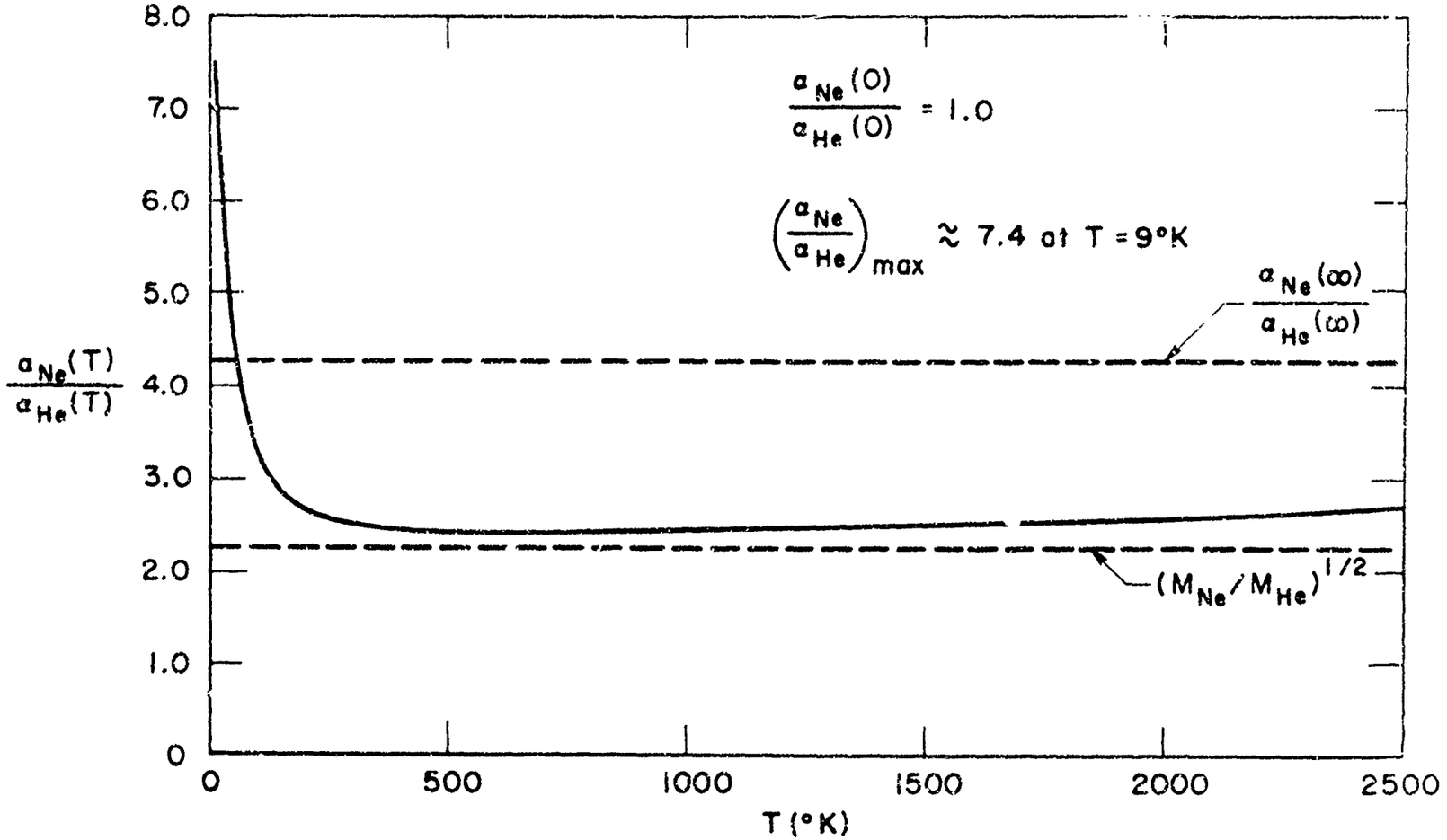

Fig. 7 The ratio $\alpha_{Ne}(T)/\alpha_{He}(T)$ on W from the Formula.

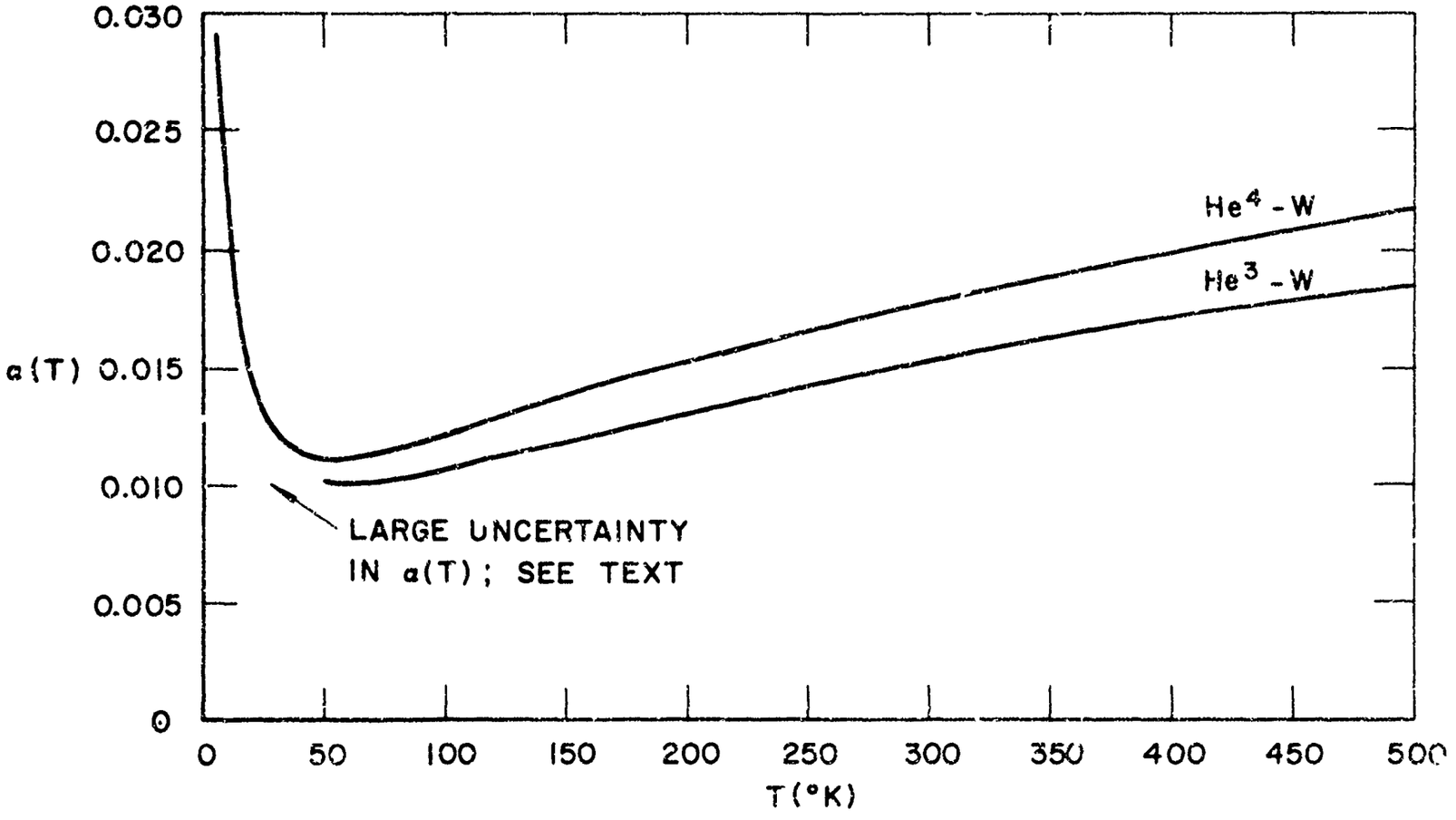

Fig. 8 Prediction of $\alpha(T)$ for He^3-W compared to $\alpha(T)$ for He^4-W from the Formula.

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(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1 ORIGINATING ACTIVITY (Corporate author)

Massachusetts Institute of Technology
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Cambridge, Massachusetts

2A REPORT SECURITY CLASSIFICATION
☒ Unclassified
☐ Other — Specify

2B GROUP

3 REPORT TITLE

FORMULA FOR THERMAL ACCOMMODATION COEFFICIENT

4 DESCRIPTIVE NOTES (Type of report and inclusive dates)

☒ Scientific Report ☐ Final Report ☐ Journal Article ☐ Proceedings ☐ Book

5 AUTHOR(S) (Last name, first name, initial)

Goodman Frank O
Wachman Harold Y
(Trilling Leon Prof (PI))

6 REPORT DATE AS PRINTED	7A. TOTAL NO. OF PAGES	7B. NO OF REFS
February 1966	41	35

8A CONTRACT OR GRANT NO	9A. ORIGINATOR'S REPORT NUMBER(S) (If given)
AF 49(638)1501	
B PROJECT NO. 9781-01	
C 61445014	9B OTHER REPORT NO.(S) (Any other numbers that may be assigned this report) AFOSR 66-0295
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13 ABSTRACT

A simple formula in closed form is proposed for the thermal accommodation coefficient from which the accommodation coefficient of a monatomic gas-solid system may be calculated from certain basic parameters of the system. The formula is obtained from consideration of certain aspects of Goodman's lattice theory of accommodation and from conclusions regarding certain properties of available experimental data. Adequate agreement is obtained of the formula with both the lattice theory and the experimental data; in the authors' opinion all the reliable data available are included. Certain corrections are applied to the lattice theory, and the gas-surface potential interaction parameters relevant to the lattice theory are revised.

DD FORM 1473
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14 KEY WORDS	LINK A		LINK B		LINK C	
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