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Thermodynamic and Transport Properties of Rubidium Vapor and Cesium Vapor

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Prepared for DEPUTY COMMANDER AEROSPACE SYSTEMS
AIR FORCE SYSTEMS COMMAND
UNITED STATES AIR FORCE
Inglewood, California



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THERMODYNAMIC AND TRANSPORT PROPERTIES
OF RUBIDIUM VAPOR AND CESIUM VAPOR

by

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ABSTRACT

Thermodynamic and transport properties of rubidium and cesium vapors are determined from theory, supplemented by experimental results. The situation for which results are obtained involves negligible ionization of the alkali atoms.

The model used is an imperfect gas obeying the virial equation of state. The second virial coefficient is considered adequate to account for deviations from the ideal behavior in the temperature and pressure range under consideration. It is assumed that the vapors are comprised of alkali atoms whose interactions are the pairwise Lennard-Jones (6 - 12) type.

On the basis of the two parameters which occur in the Lennard-Jones potential for interactions between some alkalis and noble gas atoms, and the known parameters for the noble gas atoms, the parameters are estimated for the alkalis. From these parameters the desired thermodynamic and transport properties of rubidium and cesium vapors are obtained. Numerical values are given for the important properties.

I. INTERMOLECULAR POTENTIALS FROM SPECTRAL SHIFTS

Foreign gases can cause a shift, commonly called the pressure shift, in the central frequency of the spectral distribution from radiating atoms. In the excited state, the radiating atom possesses a different energy of interaction with another atom than when in the ground state. This difference in energy can give rise to a frequency shift and broadening of the spectral line. In the case of some foreign gases the average mutual potential energy can be greater when the radiating atom is in the excited state; for other foreign gases the reverse is true. Hence, the frequency shift may be positive or negative. Measured values of this frequency shift provide information regarding intermolecular potentials.

A simple model of the origin of the pressure shift is shown in Eqs. (1) through (6). The frequency ν_0 emitted by a radiating atom unperturbed by foreign gases is

$$\nu_0 = \frac{E_2 - E_1}{h}, \quad (1)$$

where $(E_2 - E_1)$ is the energy difference between the excited and ground states which gives rise to the frequency ν_0 . In the presence of a foreign atom, both excited and lower levels of the emitting atom will be perturbed, so that the new emitted frequency is

$$\nu = \frac{E_2 + V_2(r)}{h} - \frac{E_1 + V_1(r)}{h}, \quad (2)$$

where $V_2(r)$ is the interatomic interaction for the excited state and $V_1(r)$ refers to the ground state. The preceding equation may be written as

$$\nu - \nu_0 = \Delta\nu = \frac{V_2(r) - V_1(r)}{h} \quad (3)$$

where $\Delta\nu$ is the frequency shift caused by the foreign atoms. If there are n_i foreign atoms at a distance r_i from the radiating atom, the Lennard-Jones potential gives

$$\bar{\nu}_2 - \bar{\nu}_1 = (a_2 - a_1) \sum_i \frac{p_i(r_i)n_i}{r_i^{12}} - (b_1 - b_2) \sum_i \frac{p_i(r_i)n_i}{r_i^6} \quad (4)$$

or

$$\bar{\nu}_2 - \bar{\nu}_1 = 4\pi \left[\Delta a \int \frac{p(r)n(r)r^2 dr}{r^{12}} - \Delta b \int \frac{p(r)n(r)r^2 dr}{r^6} \right] \quad (5)$$

In Eqs. (4) and (5), $p(r)$ is the probability that the radiating atom and the foreign atom are a distance r apart. In general, $p(r)$ will have the following form:

$$p(r) = A e^{-V(r)/kT} \quad (6)$$

where A is a normalization constant.

From the measured values of frequency shift in the hyperfine spectra of alkalis, as well as the temperature coefficients of the shift, this writer¹ has obtained the parameters in the Lennard-Jones potential for the following interactions: rubidium-helium, rubidium-neon, cesium-helium, and cesium-neon. The temperature coefficient of the shift is $\frac{d}{dT}(\bar{\Delta\nu})$. Using a more detailed theory than that suggested by Eq. (4), the results given in Table I were obtained.

II. LENNARD-JONES (6-12) PARAMETERS FOR RUBIDIUM VAPOR AND CESIUM VAPOR

Very little information exists regarding the form of an intermolecular potential which exists between alkali atoms. Because of the increasing importance of alkalis in modern technology, it is desirable to have some knowledge of this intermolecular potential. The Lennard-Jones (6-12) potential is reasonably satisfactory in many cases. However, in the interpretation of some phenomena, including alkalis and large rare gas atoms,¹ the writer has found this potential unsatisfactory in a quantitative way. No conclusive evidence has been found by the writer of the usefulness of the Lennard-Jones potential in alkali-alkali interactions. In this paper, it will be accepted as a satisfactory approximation.

Some published data regarding the effects of foreign gases on the spectral lines of rubidium and cesium can be reinterpreted to obtain the constants in the Lennard-Jones potential for these alkalis. The published results can now be looked upon in a slightly different fashion to obtain the Lennard-Jones parameters for rubidium-rubidium and cesium-cesium, as explained below.

The Lennard-Jones potential for the interaction of two different types of molecular species, i and j , separated a distance r , is given by

$$V_{ij} = 4 \epsilon_{ij} \left[\left(\frac{\sigma_{ij}}{r} \right)^{12} - \left(\frac{\sigma_{ij}}{r} \right)^6 \right] , \quad (7)$$

where all symbols have their usual significance. The attractive (negative) term is the London dispersion term in the van der Waals energy. This dispersion energy term can be written more compactly as

$$V_{ij} \text{ (attraction)} = - \frac{C_{ij}}{r^6} . \quad (8)$$

The various approximate forms which have been used for the C_{ij} give rise to the inequality² that

$$C_{ij}^2 \leq C_{ii} C_{jj} \quad , \quad (9)$$

or equivalently

$$\epsilon_{ij} \sigma_{ij}^6 \leq (\epsilon_{ii} \epsilon_{jj})^{1/2} \sigma_{ii}^3 \sigma_{jj}^3 \quad . \quad (10)$$

The subscripts ii and jj refer to interactions of like pairs of molecules. One would expect the equality in Eqs. (9) and (10) to be approached more closely as the molecules become more and more alike. Alkali and noble gas atoms are as unlike as atoms can be, hence the equality in Eqs. (9) and (10) is not to be expected.

Equation (10) is commonly used as a basis for assigning parameters for unlike interacting partners when the parameters for the individual species are known separately. There seems to be no theoretical basis for the usual assignments that

$$\epsilon_{ij} = (\epsilon_{ii} \epsilon_{jj})^{1/2} \quad (11)$$

and that

$$\sigma_{ij} = \frac{1}{2} (\sigma_{ii} + \sigma_{jj}) \quad (12)$$

Equations (11) and (12) are reasonably satisfactory for similar substances. In this case Eq. (11) will be used, but at the same time the inequality in Eq. (10) will be maintained. Hence only a lower bound on σ_{jj} can be assigned if both σ_{ij} and σ_{ii} are known. The results are far more sensitive to the σ 's than to the ϵ 's. Therefore it seems probable that discrepancies between theory and experiment are the result of uncertainties in the σ 's rather than in the ϵ 's. When the equalities in Eqs. (11) and (12) fail, variations in ϵ 's are usually not large enough to cause sizeable discrepancies. Hence the choice of Eqs (10) and (11), rather than (11) and (12), as the basis of analysis seems reasonable.

Table I shows the results of the above calculations. Equation (11) is used to determine the ϵ 's for rubidium-rubidium and cesium-cesium interactions. If both Eqs. (10) and (11) are used, the following relationship is found as a lower bound for σ_{jj} :

$$\sigma_{jj} = \sigma_{ii}^{-1} \sigma_{ij}^2 \quad (13)$$

The parameters obtained in Table I can be checked by calculating the London dispersion parameters and comparing them with the recent calculations of P. R. Fontana³ for both Rb-Rb and Cs-Cs. The results of Table I yield for Rb-Rb

$$4\epsilon\sigma^6 = 673 \times 10^{-60} \text{ erg cm}^6 \quad ,$$

and for Cs-Cs

$$4\epsilon\sigma^6 = 807 \times 10^{-60} \text{ erg cm}^6 \quad .$$

Table I. Parameters in the Lennard-Jones (6-12) Potential

Interaction	$\sigma \times 10^8$ cm	$\epsilon \times 10^2$ ev	Reference
He - He	2.56	0.0882	Footnote 2
Ne - Ne	2.78	0.1406	Footnote 3
Rb - He	3.24	0.416	Footnote 1
Rb - Ne	3.43	0.536	Footnote 1
Rb - Rb	4.10	1.962	From Rb - He
Rb - Rb	4.23	2.031	From Rb - Ne
Rb - Rb	4.17 ± 0.07	2.00 ± 0.03	Average
Cs - He	3.39	0.352	Footnote 1
Cs - Ne	3.58	0.458	Footnote 1
Cs - Cs	4.49	1.406	From Cs - He
Cs - Cs	4.61	1.450	From Cs - Ne
Cs - Cs	4.55 ± 0.06	1.423 ± 0.02	Average

The above calculations are based on the average values of ϵ and σ . Fontana's calculations yield for Rb-Rb and Cs-Cs, respectively

$$4\epsilon\sigma^6 = 1570 \times 10^{-60} \text{ ergs cm}^6$$

and

$$4\epsilon\sigma^6 = 2200 \times 10^{-60} \text{ ergs cm}^6$$

The latter values cannot give both ϵ and σ separately, but only the product as indicated. If one assumes that the values of ϵ are correct as given in Table I, Fontana's result would yield

$$\sigma (\text{Rb-Rb}) = 4.8 \times 10^{-8} \text{ cm}$$

and

$$\sigma (\text{Cs-Cs}) = 5.3 \times 10^{-8} \text{ cm}$$

In both cases the results are about 15 per cent larger than the corresponding values in Table I.

III. THERMODYNAMIC PROPERTIES

Thermodynamic properties of rubidium and cesium vapors are readily obtained by considering vapors as imperfect gases obeying the virial equation of state. The properties below are given per mole: \tilde{S} (entropy), \tilde{E} (internal energy), \tilde{C}_p (heat capacity at constant pressure), and \tilde{H} (enthalpy).

$$(\tilde{S} - \tilde{S}^0)/P + (R/P) \ln (P/P^0) = - \left(\frac{dB}{dT} \right) \quad (14)$$

$$(\tilde{E} - \tilde{E}^0)/P = - T \left(\frac{dB}{dT} \right) \quad (15)$$

$$\left(\tilde{C}_p - \tilde{C}_p^0 \right) / P = - T \left(\frac{d^2B}{dT^2} \right) \quad (16)$$

$$(\tilde{H} - \tilde{H}^0)/P = \left[B - T \left(\frac{dB}{dT} \right) \right] \quad (17)$$

The equations above apply to the regions of temperature and pressure where the second virial coefficient B is sufficient to represent the deviations of the alkali vapors from ideal behavior.

Hirshfelder, Curtiss, and Bird have calculated B^* , $T^* dB^*/dT^*$, and $(T^*)^2 d^2B^*/d(T^*)^2$ where B^* is the second virial coefficient in terms of the universal parameter T^* , assuming the validity of the Lennard-Jones (6-12) potential. The parameter $T^* = kT/\epsilon$, where ϵ is defined in Eqs. (7) and (11). The actual second virial coefficient B is related to B^* as follows: $B(T) = 2/3 \pi \tilde{N} \sigma^3 B^*(T^*)$, where σ is defined in Eqs. (7) and (13) and \tilde{N} is Avogadro's number. In this report, the quantities appearing on the right hand side of Eqs. (14), (15), (16), and (17) are listed in Table II for rubidium and cesium. These results are not appreciably different from those reported by Evans, Jacobson, Munson and Wagman, which were obtained in a different manner

T (deg K)	B (cm ³)		$-\frac{dB}{dT} \times 10^2$ (cm ³ deg ⁻¹ K)		$-\frac{d^2B}{dT^2}$ (cm ³ deg ⁻² K)		$-\frac{d^3B}{dT^3}$ (cm ³)		$[B - T \frac{dB}{dT}]$ (cm ³)	
	σ_1	σ_2	σ_1	σ_2	σ_1	σ_2	σ_1	σ_2	σ_1	σ_2
232	-232	-352	-174.6	-265	-405	-615	4.55	6.90	-637	-967
348	-109.8	-166.6	-63.5	-96.3	-221	-335	-1.29	2.32	-331	-502
464	-57.4	-87.2	-32.1	-48.7	-149.1	-226	0.750	1.136	-206	-313
580	-28.6	-43.4	-19.19	-29.1	-111.3	-168.8	0.441	0.669	-139.9	-212
696	-10.54	-15.99	-12.61	-19.14	-87.8	-133.2	0.289	0.438	-98.3	-149.2
812	1.735	2.63	-8.87	-13.45	-72.0	-109.2	0.203	0.308	-70.3	-111.8
928	10.54	15.99	-5.68	-9.89	-52.7	-91.8	0.1502	0.233	-42.2	-75.8
1160	22.3	33.8	-3.89	-5.90	-45.1	-68.4	0.0910	0.1381	-22.8	-34.6
2320	42.2	64.0	-0.693	-1.052	-16.09	-24.4	0.01856	0.0287	+26.1	+39.6
4640	48.1	72.9	-5.65 x 10 ⁻²	-8.58 x 10 ⁻²	-2.62	-3.98	3.36 x 10 ⁻³	5.11 x 10 ⁻³	45.5	68.9
11600	46.2	70.6	+4.08 x 10 ⁻²	+6.18 x 10 ⁻²	+4.73	+7.17	-3.09 x 10 ⁻⁵	-4.70 x 10 ⁻⁵	50.9	77.8
$\sigma_1 = 4.17 \times 10^{-8}$ cm , $\sigma_2 = 4.8 \times 10^{-8}$ cm										
165 0	-302	-477	-319	-504	-526	-832	8.32	13.15	-828	-1309
247 5	-142.7	-226	-116.0	-183.4	-287	-454	2.79	4.42	-430	-640
330	-74.6	-118.1	-58.7	-92.7	-193.8	-306	1.370	2.16	-268	-424
412 5	-37 2	-58.8	-35.1	-55.5	-144.6	-229	0.805	1.275	-181.8	-288
495	-13 70	-21.7	-23.1	-36.5	-114.1	-180.5	0.527	0.832	-127.8	-202
577 5	2.25	3.56	-16.19	-25.6	-93.5	-147.9	0.371	0.587	-91.3	-144.3
660	13 70	21.7	-11.92	-18.85	-78.7	-124.4	0.275	0.435	-65.0	-102.7
825	28.9	45.7	-7.10	-11.22	-58.6	-92.6	0.1663	0.263	-29.7	-46.9
1650	54.5	86.7	-1.267	-2.01	-20.9	-33.1	0.0345	0.0545	+33.9	+53.6
3300	62.5	98.8	-0.1033	-0.1630	-3.41	-5.38	6.15 x 10 ⁻³	9.70 x 10 ⁻³	59.1	93.4
8250	60.4	95.5	+7.44 x 10 ⁻²	+0.1177	+6.14	+9.71	-5.66 x 10 ⁻⁵	-8.95 x 10 ⁻⁵	66.5	105.2
$\sigma_1 = 4.55 \times 10^{-8}$ cm , $\sigma_2 = 5.3 \times 10^{-8}$ cm										

Table II. Thermodynamic Properties of Alkali Vapors

IV. TRANSPORT PROPERTIES

Expressions for the transport properties of vapors can be given in terms of the well-known collision integrals (or omega integrals) of Chapman and Cowling⁴ and of Hirshfelder, Curtiss, and Bird.⁵ The expressions for the diffusion coefficient D , the viscosity coefficient η , and the heat conduction coefficient κ are given below for pure substances. These expressions are based on the assumption of a Lennard-Jones (6-12) interaction between molecules.

$$D = \frac{3}{16} \frac{(2\pi k^3 T^3 / \mu)^{1/2}}{P \pi \sigma^2 \Omega^{(1,1)*}} \quad (18)$$

In Eq. (18), μ is the reduced mass, P is the pressure, and σ is defined in Eqs. (7) and (13). All of the omega integrals, $\Omega^{(s,t)*}$, will be given later.

$$\eta = \frac{5}{16} \frac{(\pi m k T)^{1/2}}{\pi \sigma^2 \Omega^{(2,2)*}} \quad (19)$$

In Eq. (19), m is the mass.

$$\kappa = \frac{25}{32} \frac{(\pi m k T)^{1/2}}{\pi \sigma^2 \Omega^{(2,2)*}} \frac{s_v}{\eta} \quad (20)$$

In Eq. (20), s_v is the specific heat of the vapor at constant volume. The collision integrals $\Omega^{(s,t)*}$ are given by

$$\Omega^{(s,t)*} = \left(\frac{kT}{2\pi\mu} \right)^{1/2} \int_0^{\infty} e^{-\gamma^2} \gamma^{2t+3} Q^{(s)} d\gamma \quad (21)$$

where $\gamma = v_r (\mu/2kT)^{1/2}$ is the reduced (dimensionless) initial relative speed of the colliding partners, with actual initial relative speed v_r .

$$\Omega^{(s)} = 2\pi \int_0^\infty I(v_r, \theta) (1 - \cos^s \theta) \sin \theta d\theta \quad (22)$$

In Eq. (22), $I(v_r, \theta)$ is the differential scattering cross section. The collision integrals $\Omega^{(s, t)*}$ can be evaluated in terms of a single universal parameter $T^* = kT/\epsilon$, where ϵ is defined as in Eqs. (7) and (11). Hirshfelder, Curtiss, and Bird have also tabulated the pertinent $\Omega^{(s, t)*}$. On the basis of these tabulations, the transport coefficients given in Eqs. (18), (19), and (20) can be calculated for both rubidium and cesium.

In this paper the viscosity coefficients of both rubidium vapor and cesium vapor have been calculated and compared with one other known calculation for cesium. In a paper called Some Selected Properties of Cesium,⁶ Irving Granet calculated transport properties of cesium vapor in a straightforward fashion. He used the mean free path theory from the simple kinetic theory of gases. The diameter of cesium atoms was estimated from the density of solid cesium. Granet assumed that cesium vapor is essentially monatomic. This is certainly suggested by every estimate obtained for the value of σ . Granet found for cesium $\eta = 0.98 \times 10^{-5} \sqrt{T}$ poise. The present calculations give $\eta = 1.45 \times 10^{-5} (\sqrt{T}/\Omega^{(2, 2)*})$ poise for $\sigma = 4.55 \times 10^{-8}$ cm and $\eta = 1.12 \times 10^{-5} (\sqrt{T}/\Omega^{(2, 2)*})$ poise for $\sigma = 5.3 \times 10^{-8}$ cm. The collision integral $\Omega^{(2, 2)*}$ is of the order of unity. The present calculation agrees quite well with the results of Granet's simple calculation.

Table III lists the transport properties given in Eqs. (18), (19), and (20) for various values of the reduced temperature, T^* . To convert T^* to degrees Kelvin, one must multiply by 232 for rubidium and by 165.0 for cesium. A simplifying assumption, $s_v = 3R/(2M)$, has been made in Eq. (20); R is the molar gas constant and M is the atomic mass number (atomic weight) in grams.

Table III. Transport Properties of Alkali Vapors

	T*	$\eta \times 10^4$ (poise)		$\mu \times 10^5$		PD		b	
		σ_1	σ_2	σ_1	σ_2	σ_1	σ_2	σ_1	σ_2
Rubidium	1	1.362	1.028	1.187	0.896	4.01×10^{-2}		3.03×10^{-2}	
	1.5	2.01	1.520	1.756	1.325	8.85×10^{-2}		4.39×10^{-2}	
	2	2.60	1.963	2.27	1.711	1.518×10^{-1}		8.42×10^{-2}	
	2.5	3.13	2.36	2.73	2.06	2.28×10^{-1}		1.360×10^{-1}	
	3	3.60	2.72	3.14	2.37	3.28×10^{-1}		1.981×10^{-1}	
	3.5	4.04	3.05	3.52	2.66	4.14×10^{-1}		2.70×10^{-1}	
	4	4.46	3.36	3.88	2.93	5.22×10^{-1}		3.51×10^{-1}	
	5	5.21	3.93	4.54	3.43	7.66×10^{-1}		5.38×10^{-1}	
	10	8.29	6.26	7.23	5.45	2.46		1.943	
	20	13.00	9.81	11.34	8.56	7.77		6.81	
50	23.5	17.73	20.48	15.46	35.43		35.50		

$$\sigma_1 = 4.17 \times 10^{-8} \text{ cm}, \sigma_2 = 4.8 \times 10^{-8} \text{ cm}$$

Cesium	1	1.203	0.886	0.675	0.497	1.621×10^{-2}		1.195×10^{-2}	
	1.5	1.779	1.311	0.998	0.735	3.58×10^{-2}		2.64×10^{-2}	
	2	2.30	1.693	1.288	0.950	6.14×10^{-2}		4.52×10^{-2}	
	2.5	2.76	2.03	1.549	1.141	9.22×10^{-2}		6.80×10^{-2}	
	3	3.18	2.35	1.785	1.315	1.277×10^{-1}		9.41×10^{-2}	
	3.5	3.57	2.63	2.00	1.476	1.675×10^{-1}		1.234×10^{-1}	
	4	3.94	2.90	2.21	1.627	2.11×10^{-1}		1.557×10^{-1}	
	5	4.60	3.39	2.58	1.903	3.10×10^{-1}		2.28×10^{-1}	
	10	7.32	5.40	4.11	3.03	9.94×10^{-1}		7.32×10^{-1}	
	20	11.49	8.46	6.44	4.75	2.93		2.16	
50	20.8	15.29	11.64	8.58	14.33		10.56		

$$\sigma_1 = 4.55 \times 10^{-8} \text{ cm}, \sigma_2 = 5.3 \times 10^{-8} \text{ cm}$$

a Units are $\text{cal g}^{-1} \text{sec}^{-1} \text{deg}^{-1} \text{K}$

b Units are $\text{cm}^2 \text{sec}^{-1} \text{atm}$

Figure 1 shows the results of the viscosity calculation for both rubidium and cesium. In each case the values of σ obtained from Eq. (13), as well as those obtained from Fontana's calculations, were used as is shown on the graph. T^* is converted to degrees Kelvin as explained above. Although no final conclusions can be drawn, the evidence seems to favor the larger values of σ . The following procedure is suggested for obtaining σ_{jj} and ϵ_{jj} when C_{ij} of Eq. (8) is known, as well as σ_{ii} and ϵ_{ii} . Calculate ϵ_{jj} from Eq. (11), calculate C_{jj} from the second order quantum mechanical perturbation theory, and then solve for σ_{jj} .

It may appear somewhat strange that ϵ is larger for rubidium than for cesium. However, there is apparently no theoretical reason why this should not be so. These results suggest that if Rb_2 and Cs_2 are formed by "Lennard-Jones binding," Rb_2 has a stronger binding energy than Cs_2 .

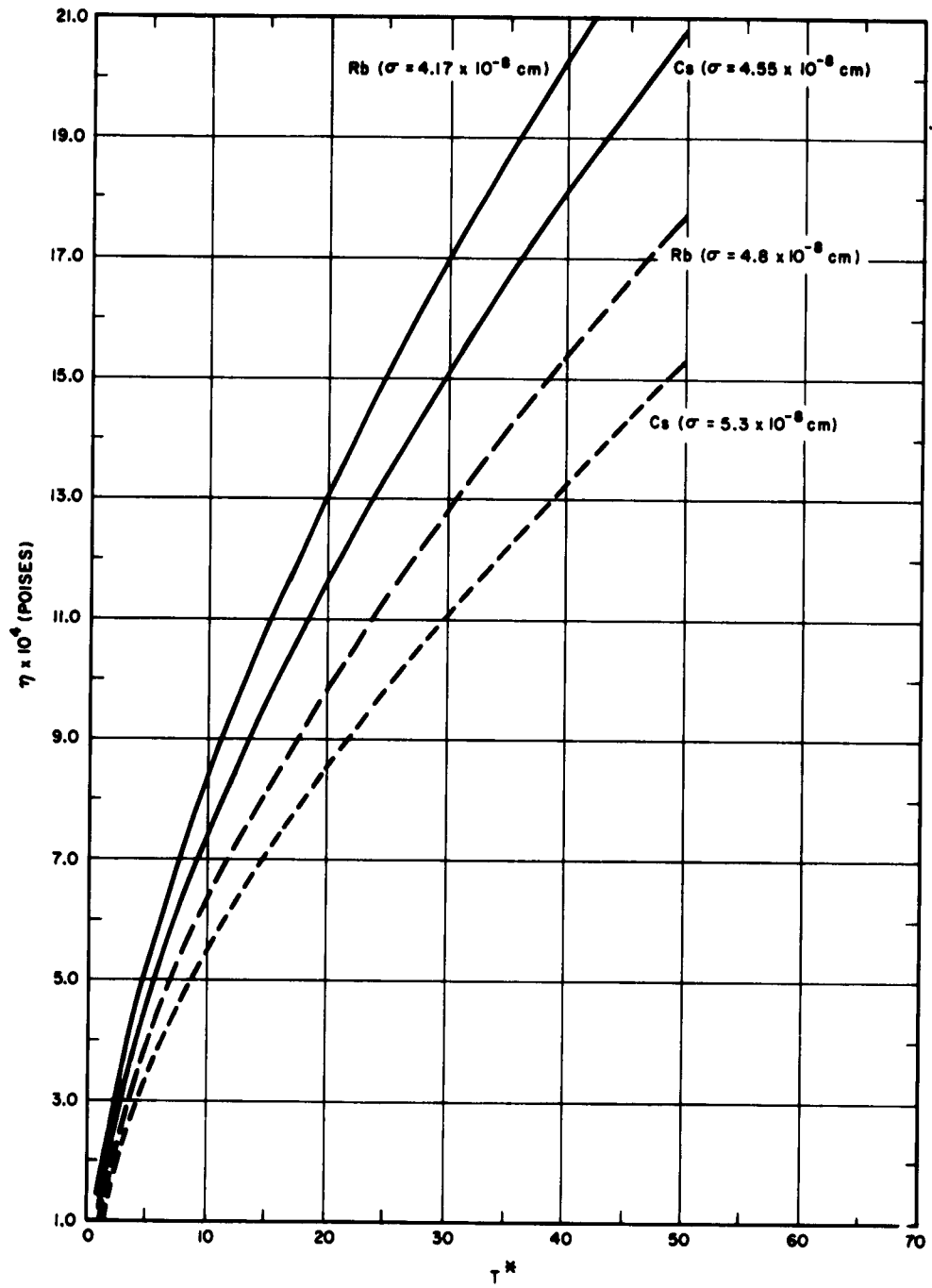


Figure 1. Results of Viscosity Calculation for Rubidium and Cesium

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UNCLASSIFIED	<p>Aerospace Corporation, El Segundo, California. THERMODYNAMIC AND TRANSPORT PROPERTIES OF RUBIDIUM VAPOR AND CESIUM VAPOR, by L. B. Robinson. 19 December 1961. [15]p. incl. illus. 33, 48. (Report TDR-930(2220-50)TR-1; DCAS-TR-61-28) (Contract AF 04(647)-930). Unclassified report</p> <p>Thermodynamic and transport properties of rubidium and cesium vapors are determined from theory, supplemented by experimental results. The situation for which results are obtained involves negligible ionization of the alkali atoms. The model used is an imperfect gas obeying the virial equation of state. The second virial coefficient is considered adequate to account for deviations from the ideal behavior in the temperature and pressure range under consideration. It is assumed that the vapors are comprised of alkali atoms whose</p> <p style="text-align: right;">(over)</p>	UNCLASSIFIED
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