The Mean Free Paths of Cesium Atoms in Helium, Nitrogen, and Cesium Vapor

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The mean free paths of cesium atoms in helium, nitrogen, and cesium vapor were measured with a molecular beam apparatus permitting the measurement of scattering deflections of about 5 seconds of arc. Utilizing the free fall of the beam atoms, the variation of the mean free path with the velocity of the beam atoms could be determined directly. This variation was found to be in agreement with the equations calculated on the basis of classical theory treating the atoms as elastic spheres. The measured values of the sums of the effective atomic radii are 12.0×10^{-8} cm for Cs-He, 17.2×10^{-8} for Cs-N_2, and 27.3×10^{-8} cm for Cs-Cs collisions. These values correspond to mean free paths of 2.1 meters in He, 2.2 meters in N_2, and 1.36 meters in Cs-vapor of 10^{-6}-mm Hg pressure for cesium atoms with a velocity corresponding to the maximum intensity of the gravity deflection curve.

AS pointed out in a previous paper,[1] the deficiency of slow atoms in a molecular beam of Cs-atoms can be explained as arising from the scattering of the beam atoms in the immediate vicinity of the oven slit. For a quantitative estimate of the effect of this scattering, it is necessary to know the effective cross section for the Cs-Cs collision. The present investigation was undertaken to measure the mean free path of cesium atoms in cesium vapor, helium, and nitrogen, and to determine the velocity dependence of the mean free path.

1. THEORY

In the quantum-mechanical treatment of scattering, Massey and Mohr[2] consider a beam of atoms with the DeBroglie wave-length $\lambda = h/Mv$ incident upon an atom which is at rest in the coordinate system. The scattering cross section Q is defined as

$$Q = 2\pi \int_0^\pi I(\theta)\, \sin\theta d\theta, \tag{1}$$

where $I(\theta)$ is the scattering intensity per unit solid angle of the beam atoms scattered through an angle θ from the direction of the incident beam. For a hard sphere model, Massey and Mohr[2] have given as a rough approximation the angular dependence of $I(\theta)$ shown in Fig. 1 as curve A. Here r_0 is the sum of the "classical" atomic radii and $k = 2\pi/\lambda$, where $\lambda = h/Mv_R$ is

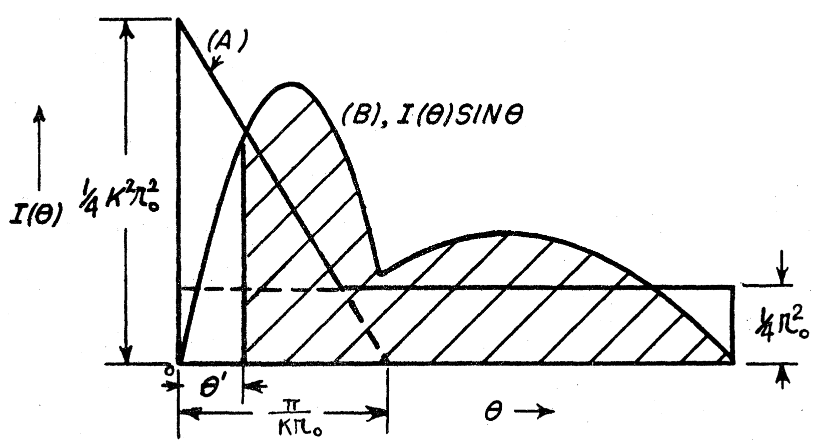

FIG. 1. Scattering intensity as function of angle.

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[1] I. Estermann, O. C. Simpson, and O. Stern, Phys. Rev. **71**, 238 (1947); referred to as I.
[2] H. S. W. Massey and C. B O. Mohr, Proc. Roy. Soc. **141**, 454 (1933).

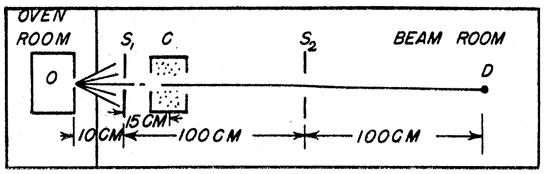

FIG. 2. Schematic diagram of apparatus.

the DeBroglie wave-length associated with the system; $M=m_1m_2/(m_1+m_2)$ being the reduced mass and v_R the relative velocity of the atoms. The large increase in scattering intensity for small angles makes the cross section Q about twice the classical value πr_0^2. Curve B in Fig. 1 represents $I(\theta)\sin\theta$. The experimentally measured cross section depends on the resolving power of the apparatus. If a scattering corresponding to an angle θ' can be detected, the error made in measuring the total scattering cross section is found by comparing the unshaded area under curve B in Fig. 1 with the total area. The resolving power necessary to get meaningful results is reached if an increase in the resolving power will not materially increase the measured cross section. In our experiments, this condition was fulfilled.

If an attractive potential of the form $V=-C/r^S$ exists, the total cross section is given by[3]

$$Q=[\pi(2S-3)/(S-2)]f^{2/(S-1)}(C/k)^{2/(S-1)}, \quad (2)$$

where k is again $2\pi/\lambda=2\pi Mv_R/h$ and f is a known function of s. Assuming dipole-dipole Van der Waals forces with a potential $V=-C/r^6$, Q takes the form

$$Q=Bv_R^{-2/5}, \quad (3)$$

where B is a constant, so that the cross section varies as the inverse $\frac{2}{5}$ power of the relative velocity. The expected dependence of Q on the relative velocity is therefore a slowly varying function, with the result that the variation of Q with the beam atom velocity is still smaller, so that for the range of velocities encountered in the experiments we can consider Q a constant, especially in view of the accuracy of the measurements and the degree of monochromization of the beam.

[3] H. S. W. Massey and C. B. O. Mohr, Proc. Roy. Soc. **144**, 188 (1934).

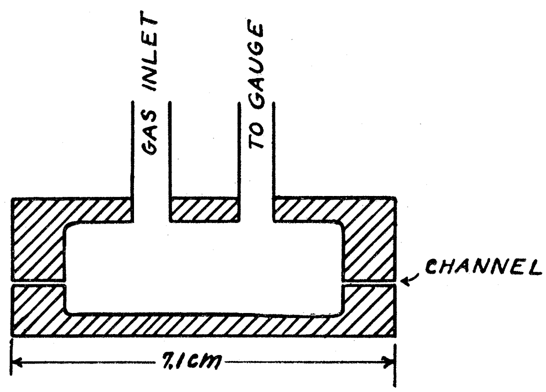

FIG. 3. Scattering chamber for nitrogen and helium.

2. EXPERIMENTAL ARRANGEMENT

The mean free path was determined by measuring the weakening of a molecular beam of cesium atoms passing through a scattering chamber filled with helium, nitrogen, or cesium vapor. The apparatus used for these experiments was the one described in paper I, with a scattering chamber C mounted between the foreslit and the collimating slit of the upper beam, as shown in Fig. 2. For helium and nitrogen as scattering gases, the scattering chamber was made of two brass pieces (Fig. 3). The lower piece had a milled groove 0.4 mm deep and 5 mm wide, which together with the upper piece formed two channels 0.4 mm high, 5 mm wide, and 1 cm long through which the beam passed. The large flow resistance of these channels and the high pumping speed in the rest of the apparatus permitted the maintenance of a pressure ratio of about 1000 to 1 between the chamber and the apparatus. The gas was fed into the chamber through a flexible pipe line from a 5-liter flask outside the apparatus. The pressure in the chamber was controlled by a sensitive needle valve and measured with an ionization gauge connected to the chamber through another tube. A by-pass permitted the outgassing of the ionization gauge.

For the cesium-cesium experiments, the scattering chamber had to be made of a metal not affected by cesium. Monel metal was found to be satisfactory. As shown in Fig. 4, this scattering chamber was machined from a solid metal block. The scattering took place in a tube T 3.5 cm long and 0.476 cm in diameter through which

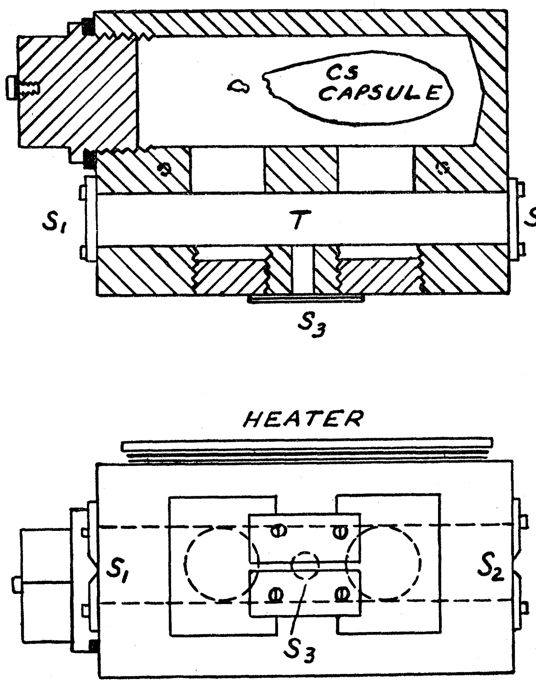

FIG. 4. Scattering chamber for cesium vapor.

the beam passed. The cesium vapor was supplied from a cesium capsule in the second part of the scattering chamber, which was connected to the first by two $\frac{1}{4}$-inch holes. Since the cesium vapor escaping from the scattering chamber was quickly condensed at the cooled copper strip (see Fig. 5 of paper I), it was possible to use slits S_1 and S_2 instead of channels at the ends of the scattering tube without materially increasing the pressure in the apparatus. A Nichrome ribbon heater was fastened to the top of the chamber and a thermocouple to the plug. For better thermal insulation, the chamber was mounted on a glass plate and supported by brass angles attached to the vertical glass plate (see Fig. 4 of paper I), which also carried the slit and detector system.

It was found that the Cs pressure in the scattering chamber could not be calculated reliably from the temperature measured by the thermocouple. In order to measure the pressure of the scattering Cs vapor, a third slit S_3 was attached to the side of the scattering tube and an auxiliary hot tungsten wire detector was mounted 2.3 cm away from the side slit. A collimating slit and a magnetically operated shutter were mounted between this detector and the side slit. From the intensity of the cesium beam striking the side detector and the geometry of the arrangement, the pressure of Cs atoms in the scattering tube could be calculated. It was also calculated from the intensity of the Cs beam which originated from the scattering chamber. This intensity was measured with the beam detector after displacing the oven sufficiently as to cut out all the atoms originating in the oven. Because of the efflux of cesium vapor through the slits at the ends of the scattering tube, the pressure inside the chamber was not quite uniform. An appropriate correction for this effect was applied in the calculations.

3. RESOLVING POWER

The resolving power of the apparatus follows from the geometry of the molecular beam. The source slit (fore slit) and collimating slit were both 0.02 mm wide and 100 cm apart. The detecting wire had a diameter of 0.02 mm and was located 100 cm beyond the collimating slit. The distance between the middle of the scattering chamber and the collimating slit was 85 cm. Hence, if the direction of the velocity vector of a beam atom passing through the scattering chamber is changed by an angle of 2.4×10^{-5} radian or 5 seconds of arc (0.02 mm in 85 cm), the atom cannot pass through the collimating slit and is, therefore, measured as "scattered." The actual resolution will be slightly lower since atoms can be scattered into the beam from the 0.1-mm broad beam wedge inside the scattering chamber, which is formed by the oven slit and the fore slit.

In the theory of scattering, the angle of scattering is the angle through which the relative velocity vector v_R is turned. What is measured experimentally is the deviation of the beam atom. In the case of small angular deviations, this is roughly half the angle through which the relative velocity vector is turned.[4]

By using the hard sphere model and the Massey and Mohr approximation (Fig. 1) for Cs-Cs scattering, the angle $\theta_0=\pi/kr_0$ at which quantum scattering begins to make itself noticed, turns out to be 0.3°, if the effective radius for the Cs atom is taken from our experi-

[4] W. H. Mais, Phys. Rev. **45**, 773 (1934).

ments as equal to approximately $\cdot 10\times 10^{-8}$ cm. This means that beam atom deflections of less than 0.15° will show "quantum scattering" (see Fig. 1). Since the apparatus has a resolving power 100 times larger than this, the interpretation of the measurements seems justified, even allowing for the fact that there is no *a priori* reason for assuming that large atoms like Cs will behave in collisions like hard spheres, and that, therefore, the actual form of the angular function $I(\theta)$ is not known.

4. PROCEDURE

(a) Helium and Nitrogen Experiments

With the scattering chamber evacuated, the beam was stabilized in the manner described in paper I, and the gravity deflection curve of the unscattered beam was measured out. Then, a certain gas pressure was introduced into the scattering chamber. The measurement of the gravity curve was repeated for different scattering pressures between 8 and 33×10^{-6} mm Hg.

(b) Cesium Experiments

For the cesium experiments, the scattering chamber was charged with a Cs capsule which was broken in a N_2 atmosphere before assembling the apparatus. After a vacuum of better than 10^{-6} mm was obtained, both the oven and the scattering chamber were heated slowly to a temperature somewhat higher than the operating temperatures and outgassed for several hours. Then the scattering chamber was allowed to cool off to room temperature and the unscattered beam was measured out. The scattering chamber was then heated again until the beam intensity was cut to $\frac{1}{2}$ or $\frac{1}{3}$ of its original value; and after stabilization, the gravity curve was measured again. Since part of the intensity measured by the beam detector came from cesium atoms originating in the scattering chamber, the oven was later displaced sufficiently as to cut out all the atoms originating from the oven, and the residual intensity was measured out. These measurements served as a second determination of the pressure in the scattering chamber. In some experiments, a check of the original beam intensity was made by letting the scattering chamber cool off again to room temperature.

5. RESULTS

(a) Helium and Nitrogen Scattering

Gravity deflection curves obtained in the helium and nitrogen experiments are shown in

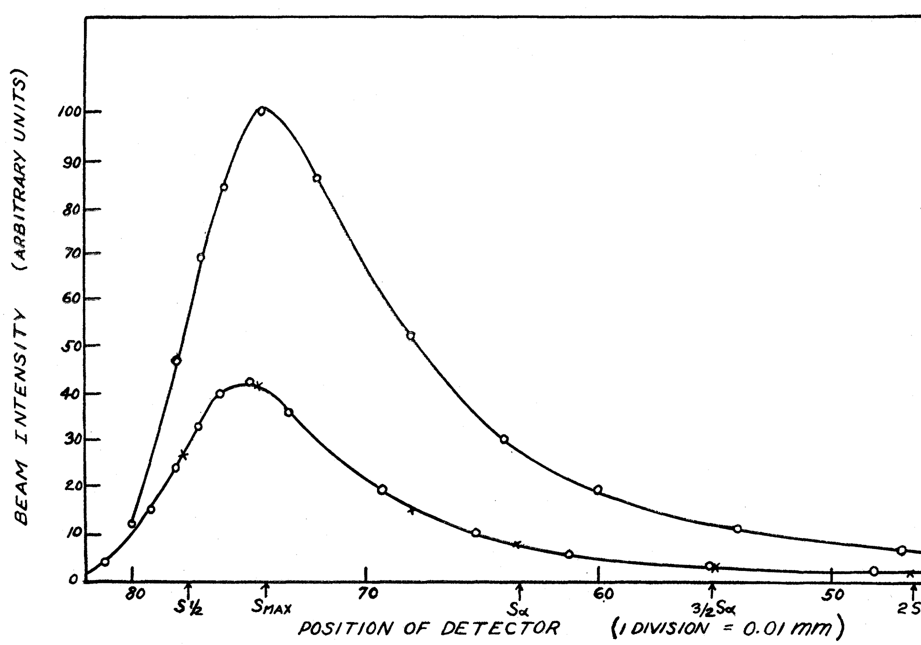

FIG. 5. Intensity distribution in cesium beam scattered by helium (o measured points, x calculated points).

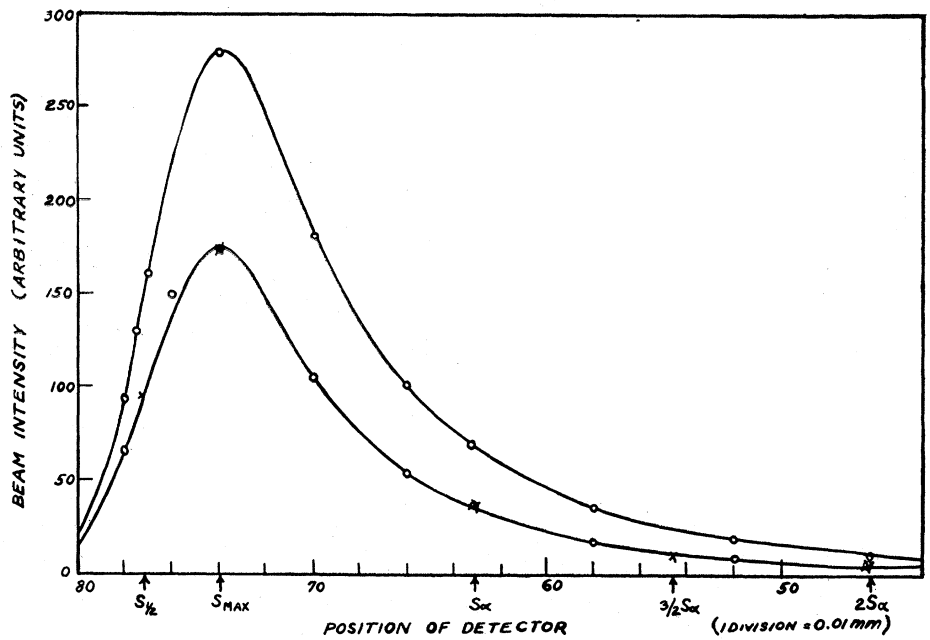

FIG. 6. Intensity distribution in cesium beam scattered by nitrogen (o measured points, x calculated points).

Figs. 5 and 6, respectively. In both figures the higher intensity curve represents the "unscattered" beam. Similar curves were obtained with helium pressures of 8.5×10^{-6} mm and 17.9×10^{-6} mm Hg, and a nitrogen pressure of 18.0×10^{-6} mm Hg.

The points marked "calculated" were found in the following way: The weakening of a homogeneous beam (i.e., a beam in which all atoms have the same velocity) from an intensity I_0 to an intensity I is given by

$$I=I_0\exp(-l/\lambda)=I_0\exp(-lp/\lambda_0), \quad (4)$$

where p is the scattering pressure, l the length of the scattering chamber, λ the mean free path, and $\lambda_0=\lambda/p$ a constant independent of pressure. If we consider the actual beam as a superposition of a number of homogeneous beams of different velocities c, we can write

$$I^c=I_0^c\exp(-lp/\lambda_0^c), \quad (4a)$$

where the superscript c denotes that the symbol pertains to the velocity c. If the weakening of the beam by a certain scattering pressure p is known for two velocities c and c', then

$$\frac{\lambda^{c'}}{\lambda^c}=\frac{\ln(I_0^c/I^c)}{\ln(I_0^{c'}/I^{c'})}. \quad (5)$$

Because of the finite width of the slits, all the atoms striking the detector at a given deflection position S do not have the same velocity. Calculations given elsewhere[5] show, however, that the average of the free paths of all these atoms does not deviate appreciably from the free path of the atoms reaching the position S and originating from the center of the undeflected beam. We may, therefore, define an effective velocity c corresponding to every detector position S, and, since the deflections are inversely proportional to c^2; $c^2/\alpha^2=S_\alpha/S$, where α refers to the most probable velocity of the beam atoms, and S_α to the corresponding deflection. Comparing all the free paths with the free path of the atoms in the maximum of the gravity curve, we get

$$\frac{\lambda_0^m}{\lambda_0^c}=\frac{\ln(I_0^c/I^c)}{\ln(I_0^m/I^m)}, \quad (5a)$$

where the superscript m, refers to the atoms in the maximum of the gravity curve. If the relationship between λ_0^c and λ_0^m is known, the scattered curve can be calculated from the unscattered curve if one point of the scattered curve is known.

[5] S. N. Foner, Thesis, Carnegie Institute of Technology, 1945.

TABLE I. Mean free paths for cesium atoms in helium and nitrogen.

S (10^{-2} mm)	S_α/S	c/α	Scattering gas: Helium I^c/I_0^m		Nitrogen I^c/I_0^m	
			I^c/I_0^m	λ_0^c/λ_0^m	I^c/I_0^m	λ_0^c/λ_0^m
3.0	5.70	2.38	0.268	1.460	0.346	1.212
6.4	2.67	1.63	0.415	1.000	0.620	1.000
12.8	1.34	1.16	0.148	0.711		
14.1	1.00	1.00	0.069	0.613	0.125	0.700
22.7	0.67	0.82	0.019	0.503	0.040	0.586
28.2	0.50	0.71	0.007	0.435	0.017	0.519

For helium scattering, the cesium atoms traversing the scattering chamber are essentially stationary targets for the much faster He atoms, hence the probability of a cesium atom being scattered out of the beam is simply proportional to the time spent in the chamber $t=l/c$. Therefore, $\lambda_0^m/\lambda_0^c=c^m/c$, and the intensity distribution in the scattered beam is given by

$$\ln(I_0^c/I^c)=(c^m/c)\ln(I_0^m/I^m)$$
$$=(S_\alpha/S^m)^{\frac{1}{2}}/(c/\alpha)\ln(I_0^m/I^m). \quad (6)$$

For the collisions between cesium atoms and nitrogen molecules, the relationship between free path and velocity is more complicated. Calculations for this case are given elsewhere.[5–7]

The absolute value of the mean free path of the cesium atoms is given for those atoms which make up the maximum of the gravity curve, since there the measurements are most accurate. For these atoms, and a scattering pressure of 10^{-6} mm Hg, the mean free path is

$$\lambda_0^m=209\text{ cm}/10^{-6}\text{ mm Hg for cesium in helium,}$$

and

$$\lambda_0^m=216\text{ cm}/10^{-6}\text{ mm Hg for cesium in nitrogen.}$$

The calculated values for the intensity I^c in the scattered beam as function of the deflection S are indicated on Figs. 5 and 6. They are also given in Table I together with the relative values for the mean free path for cesium atoms of different velocities in helium and nitrogen.

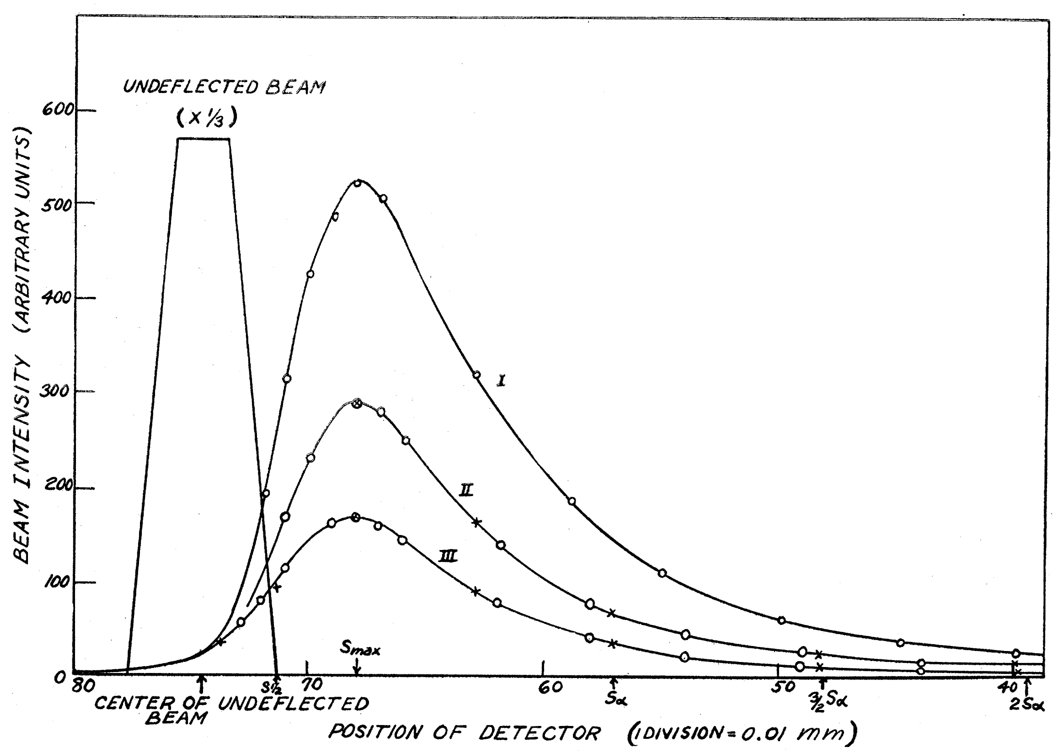

FIG. 7. Intensity distribution in cesium beam scattered by cesium vapor (0 measured points, x calculated points).

[6] O. E. Meyer, *Kinetic Theory of Gases* (London, 1899).
[7] J. H. Jeans, *Dynamic Theory of Gases* (Cambridge, 1916) second edition.

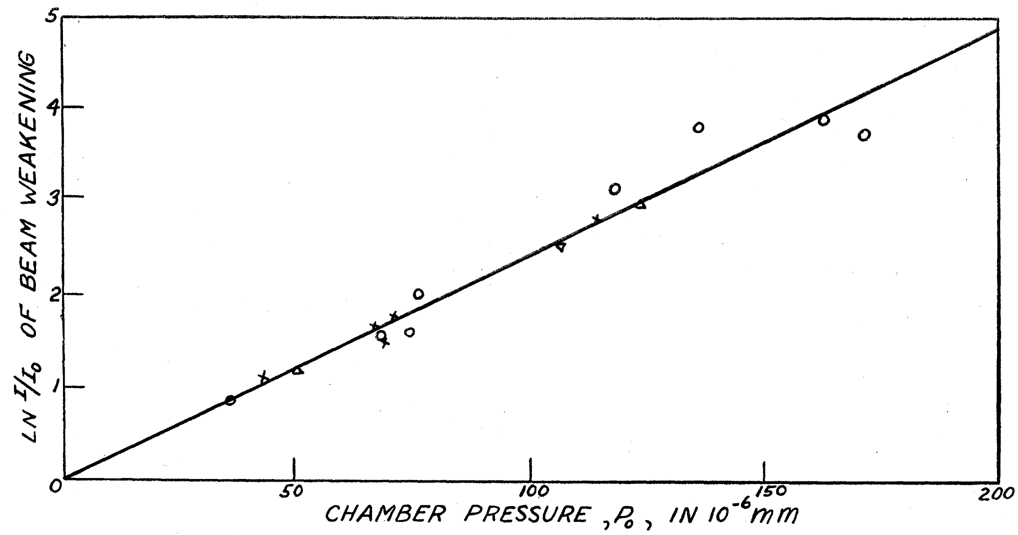

FIG. 8. Weakening of cesium beam scattered by cesium vapor as function of scattering pressure.

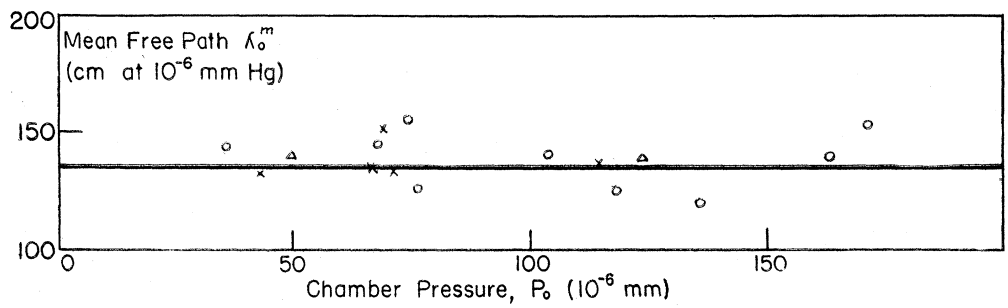

FIG. 9. Values of λ_0^m of cesium atoms in cesium vapor as a function of the scattering pressure.

(b) Cesium-Cesium Scattering

Gravity deflection curves obtained in Cs-Cs scattering experiments are given in Fig. 7. Curve I shows the unweakened beam, curves II and III the beam passing through the scattering chamber filled with cesium vapor of a pressure of 4.9×10^{-6} and 12.4×10^{-6} mm Hg, respectively. The "undeflected" beam curve, as calculated from the slit geometry and the maximum of the "unweakened" beam curve and indicated by the trapezoid, has been reduced by a factor of three to allow a better representation of the gravity curves. The calculated points of the weakened beam curves were obtained by the same method as in the nitrogen scattering experiments. The intensity of the unweakened curve as calculated on the basis of a Maxwellian velocity distribution in the undeflected beam indicates a deficiency of slow molecules (9 percent at $S=S_\alpha$, 30 percent at $S=2S_\alpha$). This effect is discussed in detail in paper I. Otherwise, the agreement between measured and calculated values is very good.

For the determination of the absolute value of the mean free path of cesium atoms in cesium vapor, a series of measurements of $\ln(I_0^m/I^m)$ in the maximum of the gravity curve was carried out, with the pressure in the scattering chamber varying between 35 and 200×10^{-6} mm Hg. These results are given in Fig. 8, while Fig. 9 shows the resulting values of λ_0^m as a function of the scattering pressure. The mean free path λ_0^m at a pressure of 10^{-6} mm Hg is 136 cm. Values for other velocities are given in reference (5) and can be calculated from Eq. (7).

(c) Collision Cross Sections for Cesium Atoms

From the mean free path at a given pressure, the classical collision cross section $\pi\sigma_{AG}^2$ can be calculated[7] by means of Eq. (7),

$$\lambda_x = x^2/(\pi^{\frac{1}{2}}\nu_G\sigma_{AG}^2\psi(x)), \qquad (7)$$

where

$$x = c_A(m_G/2kT_G)^{\frac{1}{2}},$$

and ν_G is the number of scattering atoms per cm^3, c_A the velocity of the beam atoms, T_G the absolute temperature of the scattering gas, k Boltzmann's constant, m_A and m_G the mass of beam and scattering gas atoms, respectively, and

$$\psi(x) = xe^{-x^2} + (2x^2+1)\int_0^x e^{-y}dy.$$

Since

$$\tfrac{1}{2}m_G\alpha_G^2 = kT_G \quad \text{and} \quad \tfrac{1}{2}m_A\alpha_A^2 = kT_A,$$

where the subscripts G and A refer to the scattering gas and the beam atoms, respectively, we have $x = (c_A/\alpha_A)(T_A m_G/T_G m_A)^{\frac{1}{2}}$. For c_A, we choose the velocity of the atoms in the maximum of the intensity curve. From this curve, we take $c_A = 1.64\alpha_A$. Using the data for $\lambda_x = \lambda_0^m$ corresponding to this velocity, we obtain the values for the cross sections given in Table II, where σ_{AG} is the sum of the effective radii, $\pi\sigma_{AG}^2$ the quantum-mechanical collision cross section and r_0 the classical sum of the atomic radii calculated under the assumption[2] that the classical cross section is one-half of the quantum-mechanical cross section.

The values of the mean free path are estimated to be accurate within 10 percent, hence the accuracy of the effective radii should be 5 percent. It is interesting to compare the classical radii for the Cs-Cs and Cs-He collisions. The radius of the Cs atom in the latter is 9.6×10^{-8} cm, which is larger than the sum of the radii of the Cs-He collision, which is 8.5×10^{-8} cm. This means, of course, that attractive forces are present in the Cs-Cs encounters.

TABLE II. Effective collision radii and cross sections.

Encounter	$\sigma_{AG}^2\times10^{16}$ cm	$\sigma_{AG}\times10^8$ cm	$\pi\sigma_{AG}^2\times10^{16}$ cm	$r_0\times10^8$ cm
Cs-He	142	12.0	446	8.5
Cs-N_2	298	17.2	936	12.1
Cs-Cs	743	27.3	2350	19.3

The effective collision radius for Cs-He encounters was measured by Rosin and Rabi,[8] who found $\sigma_{AG} = 7.18\times10^{-8}$ cm, which is about 40 percent lower than our value. Their apparatus was much smaller (beam length about 10 cm), and the resolving power of the order of 1 minute of arc as compared with 5 seconds of arc in our apparatus. A possible explanation for this discrepancy is that the scattering intensity for small angles may be much higher than is expected on the basis of the simplified quantum theoretical treatment of the atoms as hard spheres.

[8] S. Rosin and I. I. Rabi, Phys. Rev. **48**, 373 (1935).