

Electron impact ionization cross sections of Cu and Au between 40 and 250 eV, and the velocity of evaporated atoms*

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The total ionization cross sections of Cu and Au were measured with a simple crossed-beam apparatus for bombarding electron energies between 40 and 250 eV. The maximum cross sections were $\sigma(\text{Cu}) = (7.6 \pm 1.1) \times 10^{-16} \text{ cm}^2$ at $95 \pm 10 \text{ eV}$ and $\sigma(\text{Au}) = (15.3 \pm 2.1) \times 10^{-16} \text{ cm}^2$ at $100 \pm 10 \text{ eV}$. The ratio of the cross sections is $\sigma(\text{Au})/\sigma(\text{Cu}) = 2.0 \pm 0.2$ between 50 and 180 eV. The shape of our cross section curve for Cu compares well with a previous measurement by Crawford. An evaporation model which predicts thermal equilibrium between a liquid and the atoms evaporating into a vacuum is discussed, and the average velocity of atoms evaporated from an evaporation boat is calculated.

I. INTRODUCTION

The ionization cross sections of atoms by electron impact have been of long-standing interest. Until fairly recently, the experimental measurement of these cross sections had been restricted, however, to gases, mercury, and a few of the alkali metals. In the last few years numerous experiments have been performed with other elements.

Absolute cross sections have been measured for Zn, Cd, and Te₂ by Pottie,¹ for the alkali metals by McFarland and Kinney,^{2,3} for Ca, Sr, Ba, and Tl by McFarland,⁴ for Ag, Cu, and Pb by Pavlov *et al.*,⁵ for Pb by Beilina *et al.*,⁶ for Ag by Crawford and Wang,⁷ for Cu by Crawford,⁸ for Pb by Wang *et al.*,⁹ and for C by Wang and Crawford.¹⁰

Relative cross sections have been determined for Pb, Ag, Tl, and Ca by Rovner and Norman,¹¹ for Au and Cu by Ackerman *et al.*,¹² and for Ag, Ge, Sn, and Pb by Lin and Stafford.¹³ Numerous other relative cross sections have been measured incidental to other work, and are largely listed in the paper by Pottie.¹ Comprehensive summaries and reviews of the available absolute cross sections have been compiled by Kieffer,¹⁴ and by Kieffer and Dunn.¹⁵ Less comprehensive reviews and summaries are contained in the papers by McFarland,⁴ Pottie,¹ and Mann.¹⁶

Many attempts have been made at calculating the ionization cross sections of atoms theoretically, with various degrees of success.¹⁶⁻²³ Reviews of the theory are given by Rudge,²⁴ and by Massey and Burhop.²⁵ Both Mann¹⁶ and Lotz^{21,22} present rather complete comparisons between their theoretical calculations and published experimental results.

From a study of the literature it becomes apparent that the measurement of ionization cross sections should

be repeated for several elements and extended to many others. For many elements there exist rather large discrepancies between the presently available experimental and theoretical cross sections, and among the experimental values obtained by different investigators. Accurately known cross sections are needed, for example, in mass spectrometry, space physics, and plasma physics.

As a start, it was decided to measure the total cross sections of copper and gold.²² The copper measurements could then be compared with those of Crawford⁸ and Pavlov *et al.*,⁵ while gold had never before been measured directly.

A crossed-beam apparatus, similar to, but simpler than Crawford's,^{7,8,26} was used to determine the cross sections. An electron beam from an electron gun and a beam of metal atoms evaporated from an evaporation boat, containing the molten metal, intersected in the collision chamber to produce the metal ions.

In the past, the source of the atoms in crossed-beam experiments had always been an oven with a small hole. For such a source, the evaporated atoms are in thermal equilibrium with the source. The question arises whether atoms evaporated from a free surface, such as an evaporation boat, are also in thermal equilibrium with the surface. The available experimental evidence,²⁷⁻³⁰ and the model of the evaporation process which is described in Sec. V, indicate that thermal equilibrium most likely exists. We shall assume such equilibrium when we calculate the average velocity of the evaporated atoms in conjunction with computing the ionization cross sections.

II. APPARATUS

The crossed-beam apparatus used in this study is shown in Fig. 1 and consists essentially of an electron

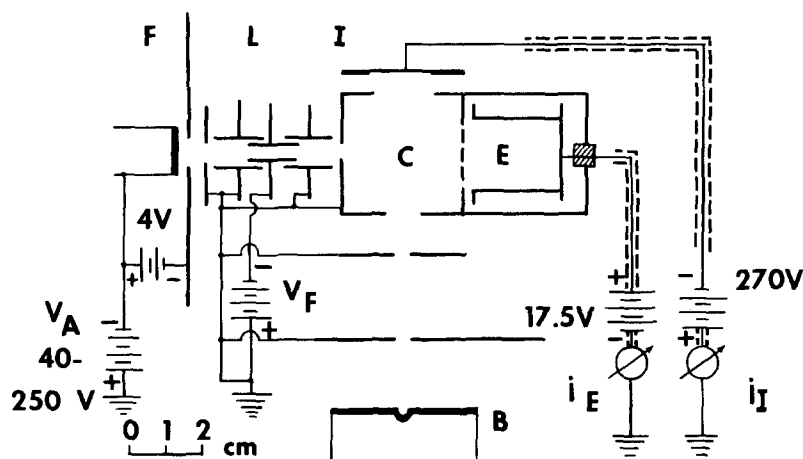


FIG. 1. Schematic diagram of the crossed beam apparatus: F filament, L electrostatic lens, B atomic beam source (evaporation boat), C collision chamber, I ion and atomic beam collector, E electron collector, V_A accelerating voltage, V_F focusing voltage, i_E electron current, and i_I ion current.

gun, an electron collector, an atomic beam source, a collision chamber, and a collector for both the ions and the evaporated atoms. The whole apparatus was constructed from a kit³¹ consisting of ceramic rods, stainless steel cylinders, and prepunched stainless steel plates, and was mounted in a bell-jar vacuum system with a base pressure of 5×10^{-8} torr.

The electron source was a filament made from a 1.02 mm wide, by 0.0013 mm thick, by 13 mm long tungsten ribbon which was heated by a 4 V storage battery. The electron beam was focused by a set of concentric cylinders which formed a modified Einzel lens. The focusing voltage V_F was adjusted for maximum electron current at the electron collector, and was approximately 80% of the electron accelerating potential V_A . A typical current at the electron collector was $i_E = 50 \mu\text{A}$ at 100 eV. The electron collector was biased at -17.5 V with respect to ground to prevent the escape of secondary electrons.

A molybdenum evaporation boat, whose circular depression was filled with molten metal, was used as the atomic beam source. The temperature of the molten metal was determined with an optical pyrometer, and after correcting for light absorption in the walls of the bell jar and for the emissivity of the molten metal, was typically 1730°K. To reduce the emission of charged particles from the boat, it was biased approximately $+12$ V with respect to ground. During evaporation, the pressure in the vacuum system was generally around 5×10^{-7} torr. Between the evaporation boat and the first beam defining aperture was located a beam shutter, not shown in Fig. 1.

The evaporated atoms passing through the collision chamber were collected on an aluminum foil which could be weighed on a microbalance. By experiment it was determined that the sticking factor of the atoms on the foil was 100%.³² The ions were collected on the same foil. A potential of -270 V assured the collection of all ions and also prevented electrons from going from the filament to the ion collector. This high potential had a negligible effect on the current registered at

the electron collector. For copper at an electron energy of 100 eV, typical ion currents were 2.5×10^{-8} A, and typical masses were 0.5 mg collected over 500 sec. For the conditions of this experiment, the secondary electron emission due to the ions incident on the ion collector is zero.³³ Kinetic emission of electrons requires that the ion energy exceeds approximately 500 eV. For Auger electrons to be emitted, the ionization energy of the evaporated atoms must be greater than twice the work function of the collector surface. This condition holds for Au on Al, but not for Au on Au, Cu on Al, or Cu on Cu. The time during which the Au ions strike the clean Al surface of the collector was negligibly short, however, as the surface was quickly covered by evaporated atoms.

However, various other corrections to the apparent ion current had to be made. For Cu they were as follows. A small leakage current of $\sim 3 \times 10^{-12}$ A was observed with both the shutter closed and the electron beam off. Evaporating metal with the electron beam off (by removing the accelerating voltage) enabled us to measure a current of $\sim 5 \times 10^{-11}$ A which was due to ions coming directly from the atomic beam source. Correction was made for the weight of the atoms collected during this time. The ion current due to the ionization of the background gases ranged from 10^{-11} to 10^{-8} A, depending on the system pressure and the electron energy. It was measured by having the boat at full temperature and the electron beam on, but the shutter closed. This background current was measured separately for each experimental point plotted in Fig. 2. The leakage current mentioned above was, of course, included in this measurement. The operating conditions for the gold runs were similar.

III. IONIZATION CROSS SECTION MEASUREMENTS

The total ionization cross section σ can be calculated from

$$\sigma = i_I / (i_E n l), \quad (1)$$

where i_I is the net ion current measured at the ion collector, i_E is the electron current measured at the electron collector, n is the number of gas molecules per unit volume in the collision region, and l is the path length of the electrons through the atomic beam. The total ionization cross section is the charge-weighted sum of the cross sections σ_s for s fold ionization,¹⁵

$$\sigma = \sum_s s\sigma_s. \quad (2)$$

Over the energy range under consideration here, only single and double ionization are of significance.

Equation (1) needs to be rewritten in terms of the quantities actually determined in this experiment. Let N be the total number of atoms evaporated onto the atom collector in time t . $N' = fN$ atoms out of the N atoms passed through the volume of intersection of the atom and electron beams. The rate at which atoms enter and leave the volume of intersection is $r' = N'/t$.

The rate is also given by

$$r' = n\bar{v}ld, \quad (3)$$

where \bar{v} is the average velocity of the atoms in the beam, and d is the diameter of the electron beam. Substituting $r' = N'/t$ into (3) we obtain $n = N'/(t\bar{v}ld)$, which when substituted together with $N' = fN$ into (1) gives

$$\sigma = i_I \bar{v} d / (i_E f N). \quad (4)$$

d and f were determined from the geometry of the apparatus; N was easily obtained from the weighings of the aluminum foil; t was the time over which the metal atoms were collected on the foil; and i_I and i_E were measured with electrometers.

We assume that the atoms evaporated from the molten metal in the evaporation boat are in thermal equilibrium with the liquid. In that case, the evaporation is equivalent to molecular effusion through a hole in an oven, and the average velocity, as shown in Sec. V, is

$$\bar{v} = \frac{3}{4}(\pi)^{1/2}(2kT/m)^{1/2}, \quad (5)$$

with T the absolute temperature of the molten metal surface, k Boltzmann's constant, and m the mass of an evaporated metal atom.

Our experimentally determined total ionization cross sections for Cu and Au, using Eqs. (4) and (5) are listed in Table I and plotted in Fig. 2 against the kinetic energy of the bombarding electrons from 40 to 250 eV. The error bars in Fig. 2 were calculated from the random uncertainties in i_I , i_E , t , N , and T for each data point. In addition, there are possible systematic errors in d , f , i_E , i_I , and T common to all our data points, and which add up to an estimated $\pm 10\%$.

IV. DISCUSSION OF CROSS SECTIONS

The solid line through our Cu data represents Crawford's⁸ cross section for Cu multiplied by a factor of 1.85. Since he actually obtained the cross sections

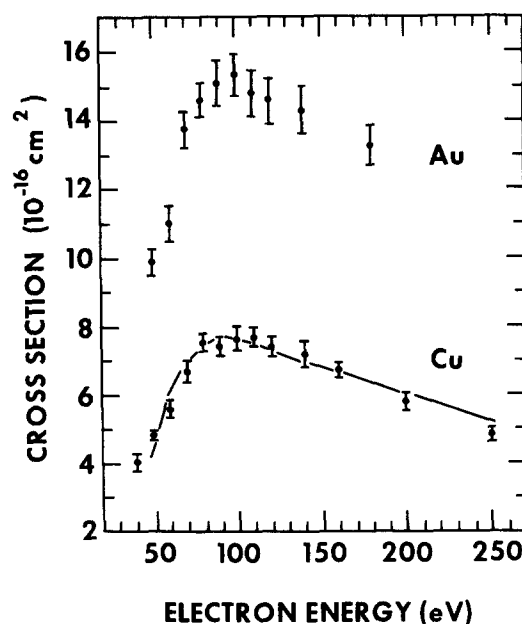


FIG. 2. The total ionization cross sections of Cu and Au as a function of the energy of the bombarding electrons. The error bars indicate random uncertainties. In addition, there may be systematic errors of up to $\pm 10\%$, common to all points. The solid line represents Crawford's total cross section for Cu (see Ref. 8) multiplied by 1.85.

for single and double ionization separately, they were combined for our purposes according to Eq. (2). We see that the variation of our cross section measurements with electron energy agrees with his. Our measured cross section reaches a maximum of $\sigma(\text{Cu}) = (7.6 \pm 1.1) \times 10^{-16} \text{ cm}^2$ at $95 \pm 10 \text{ eV}$, taking into account both random and systematic uncertainties. Crawford's equivalent values are $(4.1 \pm 1.0) \times 10^{-16} \text{ cm}^2$ at $95 \pm 5 \text{ eV}$.

Our cross section curve for Au has its maximum of $\sigma(\text{Au}) = (15.3 \pm 2.1) \times 10^{-16} \text{ cm}^2$ at $100 \pm 10 \text{ eV}$. The only absolute cross section determination available for comparison is a derived value of $11.6 \times 10^{-16} \text{ cm}^2$ at 75 eV ,¹ which compares with our cross section at 75 eV of $(14.5 \pm 2.0) \times 10^{-16} \text{ cm}^2$.

The ratio of our cross sections is

$$\sigma(\text{Au})/\sigma(\text{Cu}) = 2.0 \pm 0.2 \quad (6)$$

between 50 and 180 eV, considering only random uncertainties; any systematic errors cancel out. This ratio compares favorably with Ackerman *et al.*'s ratio of 1.9 to 2.1 at 70 eV.¹²

Pavlov's⁵ experimental total cross section measurements on copper are at complete variance with our and Crawford's results, reaching a maximum of $3.1 \times 10^{-16} \text{ cm}^2$ at 30 eV.

Lotz's²² semiempirical cross section calculations for Cu and Au do not agree with our curves in either magnitude or shape. His maximum cross sections for

TABLE I. Total electron-impact ionization cross sections of Cu and Au.

Electron energy (eV)	$\sigma(\text{Cu})$ (10^{-16} cm^2)	$\sigma(\text{Au})$ (10^{-16} cm^2)
40	4.06 ± 0.24^a	
50	4.86 ± 0.15	9.90 ± 0.37^a
60	5.58 ± 0.24	11.02 ± 0.48
70	6.69 ± 0.29	13.76 ± 0.52
80	7.55 ± 0.23	14.58 ± 0.52
90	7.45 ± 0.23	15.09 ± 0.67
100	7.67 ± 0.30	15.37 ± 0.61
110	7.69 ± 0.27	14.82 ± 0.67
120	7.38 ± 0.23	14.58 ± 0.67
140	7.18 ± 0.33	14.30 ± 0.71
160	6.75 ± 0.21	
180		13.23 ± 0.61
200	5.81 ± 0.25	
250	4.86 ± 0.21	

^a Only the random uncertainties are listed. There may be additional systematic errors of up to $\pm 10\%$, common to all experimental points.

single ionization are $\sigma_1(\text{Cu}) = 3.7 \times 10^{-16} \text{ cm}^2$ at 45 eV, and $\sigma_1(\text{Au}) = 5.0 \times 10^{-16} \text{ cm}^2$ at 70 eV. Above 60 eV, his cross section ratio is $\sigma_1(\text{Au})/\sigma_1(\text{Cu}) = 1.3$ to 1.4, in disagreement both with Mann's¹⁶ theoretical ratio of 1.6 and our experimental ratio of 2.0 ± 0.2 . Mann's¹⁶ maximum cross sections for single ionization are $\sigma_1(\text{Au}) = 6.46 \times 10^{-16} \text{ cm}^2$ and $\sigma_1(\text{Cu}) = 4.06 \times 10^{-16} \text{ cm}^2$. Lin and Stafford²³ made some classical calculations of the maximum cross sections for single ionization and obtained $\sigma_1(\text{Au}) = 13.3 \times 10^{-16} \text{ cm}^2$ and $\sigma_1(\text{Cu}) = 14.7 \times 10^{-16} \text{ cm}^2$. Both their absolute cross section for Cu and the cross section ratio of Au to Cu are in serious disagreement with our measurements. In considering these calculated cross sections, it is necessary to keep in mind that they are all for single ionization, while the experimental cross sections are total cross sections as defined by Eq. (2). In the electron energy range 70 to 100 eV the total cross sections are 20% to 40% larger than the cross sections for single ionization, but the ratios are essentially the same for single and total cross sections.^{7,8}

Because of the consistency between the shape of our and Crawford's⁸ cross section curves for Cu, we believe that our observed variation of cross section with electron energy is correct within a few percent. For the same reason, and because of the agreement with Ackerman *et al.*'s¹² ratio, we believe our measured ratio of the cross sections of Cu and Au to be correct within our indicated error limits. However, the absolute cross sections for Au and Cu are still rather uncertain. Since the cross section ranges for Cu as measured by us and by Crawford⁸ do not overlap, there must still be some systematic errors in our and/or Crawford's data that have not been taken into account.

The possible systematic error in σ of $\pm 10\%$ mentioned up to now do not include any error due to any deviation from thermal equilibrium between the molten metal in the evaporation boat and the evaporated atoms. As discussed in Sec. V, such deviations are unlikely. We cannot, at the moment, find any possible systematic errors in our or Crawford's data which might bring the absolute values of our cross section measurements for Cu closer together.

V. VELOCITY OF EVAPORATED ATOMS

To the best of our knowledge there has been only one measurement of the velocity of atoms evaporated from an open liquid. Willis³⁰ determined the velocity distribution of atoms evaporated from molten Pb and In in an open crucible. He found that the evaporated atoms had a Maxwellian velocity distribution with a characteristic temperature equal to that of the molten metal, except that the part of the velocity spectrum below a certain minimum velocity was missing.

On the other hand, Mar and Searcy²⁷ and Rothberg *et al.*²⁸ found for the sublimation of solid gallium nitride and solid alkali halides, respectively, that the vapors were in thermal equilibrium with the surface, i.e., that they had a complete Maxwellian velocity distribution. Thermal equilibrium was also observed by McFee *et al.*²⁹ for molecular beams scattered from a surface.

If the evaporated atoms are in thermal equilibrium with the liquid metal surface, then the evaporation from an evaporation boat is equivalent to the diffusion of a Maxwellian gas into a vacuum from a reservoir of temperature T through an aperture of size equal to the pool of liquid. In this case, the average velocity can be calculated easily as follows.

Let us choose a coordinate system with the $+x$ axis pointing away perpendicularly from the liquid surface, and with the y - z plane parallel to the surface. For a Maxwellian gas of temperature T , the number dN of atoms of mass m crossing unit area each second in the $+x$ direction with velocity components between v_x and $v_x + dv_x$, v_y and $v_y + dv_y$, and v_z and $v_z + dv_z$ is³⁴

$$dN(v_x, v_y, v_z) = n(m/2\pi kT)^{1/2} v_x \times \exp[-m(v_x^2 + v_y^2 + v_z^2)/2kT] dv_x dv_y dv_z, \quad (7)$$

where n is the number of atoms per unit volume of vapor in the reservoir. The average velocity in the $+x$ direction is therefore

$$\bar{v}_x = \int_0^\infty \int_{-\infty}^\infty \int_{-\infty}^\infty v_x dN(v_x, v_y, v_z) \times \left(\int_0^\infty \int_{-\infty}^\infty \int_{-\infty}^\infty dN(v_x, v_y, v_z) \right)^{-1}. \quad (8)$$

The atoms evaporating from the liquid and striking the collector define a cone of half angle $\theta \sim 7^\circ$. In order that the evaporating atoms should not leave the cone

during their travel from the boat to the collector, their velocity component parallel to the surface must not exceed $\pm v_x \tan \theta = \pm \gamma v_x = \pm \delta$. Since $v_y \ll v_x$ and $v_z \ll v_x$, v_y and v_z can be dropped in the exponent of expression (7). Equation (8) now becomes

$$\bar{v}_x = \int_0^\infty \int_{-\delta}^\delta \int_{-\delta}^\delta v_x^2 \exp(-mv_x^2/2kT) dv_x dv_y dv_z \times \left(\int_0^\infty \int_{-\delta}^\delta \int_{-\delta}^\delta v_x \exp(-mv_x^2/2kT) dv_x dv_y dv_z \right)^{-1} \quad (9)$$

$$= \int_0^\infty v_x^4 \exp(-mv_x^2/2kT) dv_x \times \left(\int_0^\infty v_x^3 \exp(-mv_x^2/2kT) dv_x \right)^{-1} \quad (10)$$

$$= \frac{3}{4}(\pi)^{1/2} (2kT/m)^{1/2} = 1.33(2kT/m)^{1/2}. \quad (11)$$

To see how such thermal equilibrium between the evaporated atoms and the surface might come about, let us consider a model of the evaporation process in which we assume the atoms in the liquid to be a Maxwellian gas. We will then calculate the average velocity of the atoms evaporating in a direction perpendicular to the surface, after they have overcome a potential energy barrier of height ϕ .

An atom with velocity components u_x, u_y, u_z inside the liquid will have velocity components v_x, v_y, v_z outside the metal. The probability that an atom of velocity u_x will escape through the surface is $P(u_x)$, with $P(u_x) = 0$ for $u_x < u_{xm}$, and $P(u_x) = 1$ for $u_x \geq u_{xm}$ where u_{xm} is the minimum velocity required for an atom to overcome the potential energy barrier and is given by $u_{xm} = (2\phi/m)^{1/2}$. v_x is calculated from u_x by considering conservation of energy,

$$\frac{1}{2}mv_x^2 = \frac{1}{2}mu_x^2 - \phi. \quad (12)$$

The velocity components parallel to the surface are unaffected by the barrier, so that $v_y = u_y$, and $v_z = u_z$.

The average velocity now becomes

$$\bar{v}_x = \int_0^\infty \int_{-\delta}^\delta \int_{-\delta}^\delta v_x(u_x) P(u_x) dN(u_x, u_y, u_z) \times \left(\int_0^\infty \int_{-\delta}^\delta \int_{-\delta}^\delta P(u_x) dN(u_x, u_y, u_z) \right)^{-1}, \quad (13)$$

where $dN(u_x, u_y, u_z)$ is given by (7) on substituting u_x for v_x , etc. Equation (13) is easily integrated and gives again (11).

An upper limit to the error introduced by dropping v_y and v_z in the exponents of Eq. (8) can be estimated by setting $v_y = v_z = \gamma v_x$ in the exponents of Eq. (8) and integrating as before. The result is

$$\bar{v}_x' = \frac{3}{4}(\pi)^{1/2} [2kT/m(1+2\gamma^2)]^{1/2}. \quad (14)$$

For $\theta \sim 7^\circ$, \bar{v}_x' is 1.4% less than \bar{v}_x .

At the vapor pressure employed by us and by Crawford⁸ ($\sim 5 \times 10^{-4}$ torr) and the corresponding molecular mean-free paths of about 5 cm, deviations from a Maxwellian velocity distribution due to collisions between beam atoms near the source are negligible, amounting to an increase in the average velocity of less than 1%.³⁵⁻³⁸

Willis³⁰ discusses an evaporation model very similar to ours. Experimentally he finds a deviation from a Maxwellian velocity distribution for evaporated Pb and In. At the temperatures employed by him, the vapor pressures are high enough (0.1 to 1 torr) to produce a significant depletion of beam atoms of low velocities (at least 50% at one-half of the most probable velocity, and more at lower velocities),³⁵⁻³⁸ which is what he observed, at least qualitatively.

On the basis of the above considerations of Willis' work, our own model, and the evidence from the sublimation of solids and the scattering of atomic beams, as mentioned at the beginning of this section, we believe that it is appropriate to assume a complete Maxwellian velocity distribution for our evaporated Cu and Au atoms.

VI. CONCLUSIONS

On the basis of the results presented here and in the literature we feel that the variation of the ionization cross sections of Cu and Au with bombarding electron energy is known to within $\pm 5\%$, the ratio of the two cross sections to within $\pm 10\%$, and the absolute cross sections to within $\pm 50\%$. Agreement with theory is, in general, poor.

It would be very desirable to improve the accuracy of the absolute cross section determinations. The collision chamber could be redesigned to reduce the uncertainty in the collision volume. A more intense electron beam, a chopped atomic beam with phase sensitive detection, a mass spectrometer, and a lower background pressure in the collision region through use of differential pumping would reduce the uncertainty in the ion current.

Further careful experiments are necessary to definitely determine the proper model which describes the sublimation and evaporation of atoms from a free surface under all conditions. While the exact velocity distribution of these atoms is, so far, still somewhat uncertain, the average velocity is not very sensitive to the exact distribution and can be calculated to within a few percent of the experimentally measured values on the basis of either our or Willis' model.

Experimentally the average velocity of evaporated atoms could possibly be determined by measuring the ionization cross sections of several metals, once using an open liquid as the atom source, and once using a Knudsen cell. Since the velocity of atoms coming from a Knudsen cell is well known, the velocity of the atoms

from the open liquid could then be obtained by comparing the ionization efficiencies using the two sources.

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