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Temperature Dependence of Electron Attachment in I₂ Vapor*

F. K. Truby

Sandia Laboratories, Albuquerque, New Mexico 87115

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The temperature dependence of electron attachment in I₂ vapor has been measured over the temperature range from 253 to 467 °K. The attachment coefficient was found to increase with temperature over the range studied, varying from 0.9×10^{-10} to 4.2×10^{-10} cm³/sec. Studies involving selective heating of the electrons indicate that for a gas temperature of 298 °K the attachment coefficient increases from 1.8×10^{-10} to 2.8×10^{-10} cm³/sec as the electron thermal energy was increased from 0.04 to about 0.27 eV.

I. INTRODUCTION

Previous studies have been made to determine the dependence on electron energy of the dissociative attachment process for iodine molecules at thermal energies.¹⁻³ However, the temperature dependence of the attachment coefficient for the case where the electrons and gas molecules are in thermal equilibrium has not been reported. As an extension of recent studies⁴ concerning dissociative electron attachment in iodine vapor at 295 °K, the current study has involved determination of the thermal electron attachment coefficient for iodine as a function of temperature over the temperature range from 253 to 467 °K. Thus, in the present case the effect on the dissociative attachment coefficient of populating higher vibronic levels in I₂, as well as the effect of shifting the electron energy distribution, can be examined. Such an examination has been carried out by Shipsey,⁵ using the data reported herein.

The dissociative electron attachment processes studied can be described as follows:



This process was found by Fox to be the only significant mechanism by which electrons having en-

ergies up to a few electron volts are attached to iodine.¹ In the present case, the attachment coefficient has been determined under the assumption that electron losses by processes other than dissociative attachment can be essentially ignored.⁴ Under such an assumption, the time dependence of the electron number density can be approximated by

$$n_e \approx n_e(0) e^{-\nu_a t / a}, \quad (2)$$

where n_e is the electron number density at some point in the cavity and $n_e(0)$ is the electron number density at some initial time. In Eq. (2), ν_a is the attachment frequency and t is time. The attachment coefficient k is given as follows⁴:

$$k = \nu_a / n_g, \quad (3)$$

where n_g is the iodine-molecule number density. In the above expressions, Maxwellian energy distributions for the particles are assumed. In order to determine to what extent Eq. (2) was valid, values for the ambipolar diffusion coefficient were obtained at elevated temperatures so that diffusion losses could be separately assessed.

In addition to determining the attachment coeffi-

cients for iodine as a function of gas temperature, values for k were obtained under the conditions that the gas temperature was held at 298 °K and the electron energy was increased by microwave heating. These results will also be discussed.

II. EXPERIMENTAL

A. Gas Temperature Variation Studies

The electron density was obtained by the microwave-cavity method in which the ionization of a small fraction of the iodine vapor is achieved directly by use of a hydrogen flash lamp. Helium is added to the iodine vapor within the microwave cavity to reduce diffusion losses. The time dependence of the electron number density in the cavity is determined after a single photoionization light pulse from the flash lamp. The details of the apparatus used and the methods of determining the electron-attachment frequencies and subsequent attachment coefficients have been previously reported.⁴

For the present study the microwave cavity was heated with an oven. Cavity temperatures were determined by use of thermocouples mounted at various points on the cavity. Cooling of the cavity was accomplished by introducing precooled nitrogen gas into the thermally insulated cavity oven. The flash lamp was shielded from the heat of the oven using a technique similar to that described by Shaw *et al.*⁶

B. Electron-Heating Studies

For the experiments in which electron heating took place, a method similar to that employed by Mahan and Young⁷ was utilized. The microwave-power level used to measure the electron density decay in the cavity was increased so that appreciable electron heating could occur. In the present experiment, the amplitude of the sawtooth voltage applied to the klystron reflector⁴ was reduced, so that at no time in the klystron frequency sweep cycle⁴ was the microwave-power input to the cavity reduced by more than 5% of that value encountered when the microwave frequency matched the resonant frequency of the cavity. Thus, the electron temperature was essentially constant during the afterglow period. The intensity of the H₂ flash lamp (using a 10- μ sec light pulse) was adjusted with appropriate ballast resistors so that the initial electron number density was kept below 3×10^8 electrons/cm³. This was necessary in order to keep the shift in cavity resonant frequency due to the presence of electrons small enough so that the cavity frequency shift could still be followed as previously described.⁴

The electron-velocity distribution in an applied microwave field has been derived by Margenau.⁸

The average electron energy $\bar{\mathcal{E}}$ may be obtained from the velocity distribution in the present case by the following expression⁹:

$$\bar{\mathcal{E}} \approx \frac{3}{2} k T_g + e^2 M E^2 F(z) / 4 m^2 \omega^2. \quad (4)$$

Here k is Boltzmann's constant, T_g is the ambient gas temperature, e is the electronic charge, M is the atomic mass of helium, E is the peak electric field, m is the electron mass, $F(z)$ is a function having a value close to unity for the current set of experiments, and ω is the radian frequency of the microwaves. In the present case, where the TM₀₁₀ mode has been used, the peak electric field at resonance, E_0 , along the center axis of the cavity is given by the following expression¹⁰:

$$E_0^2 = 7.43 P Q_u / V \epsilon_0 \omega_0, \quad (5)$$

where P is the power absorbed at resonance, Q_u is the unloaded Q of the cavity,¹¹ V is the volume of the cavity, ϵ_0 is the free space permittivity, and ω_0 is the resonant radian frequency of the microwave cavity. In the present case an average electric field has been used to determine the average electron energy in the cavity such that

$$(E^2)_{av} = \int_{vol} E^2(r) n_e(r) dV / \int_{vol} n_e(r) dV, \quad (6)$$

where $n_e(r)$ is the space-dependent electron number density. If the electron number density is assumed to have a fundamental mode distribution, then for the present case, $(E^2)_{av} \approx 0.45 E_0^2$. In using an average electric field to establish an average energy associated with electron heating, an assumption is made that, even though the microwave field varies as a function of position in the cavity, the electron gas exhibits a very high heat conductivity so that most of the electrons present will exhibit the same electron thermal energy.⁹

III. RESULTS

A. Variation of Gas Temperature

The time dependence of the electron number density within the microwave cavity is shown for various temperatures in Fig. 1. For most of the current work, I₂ pressures of approximately 3×10^{-3} Torr were used. For the elevated temperatures utilized in the present study, impurities in the form of metal-iodide vapors (formed by interaction of iodine with the exposed metal surfaces of the cavity components⁴) could be carried through the system if significantly higher I₂ vapor pressures were utilized.¹² Nickel iodide and iron iodide (occurring as a trace impurity in the iodine) appear to be the principal iodides present, as indicated

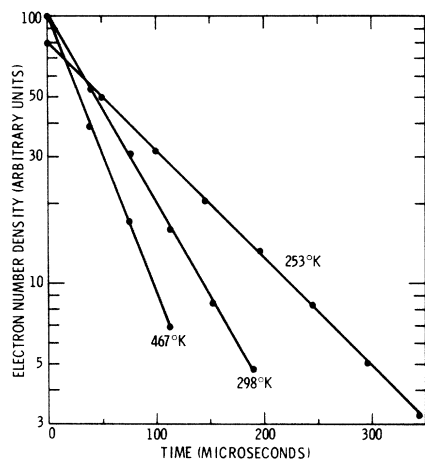


FIG. 1. Electron-number-density decay at different temperatures for an iodine vapor pressure of 2.8×10^{-3} Torr and a helium-buffer gas pressure of 20 Torr.

by chemical analysis of cavity-component surface contamination. For the highest temperatures studied and using I_2 vapor pressures no greater than 3×10^{-3} Torr, quadrupole mass analysis of the gas samples within the cavity⁴ indicated that the amounts of these metal salts in the vapor phase were below the level for detection. This was of the order of 10^{-6} Torr for Fe or Ni under the conditions of the experiment.

In Fig. 2, the attachment coefficients for I_2 vapor are plotted as a function of temperature. The average random error for the data shown is estimated at less than $\pm 10\%$. The main source of this error arises from variations in the I_2 vapor pressure due to small fluctuations in the temperature of the iodine chamber⁴ which was used to control the I_2 vapor pressure in the cavity. The data shown in the figure were obtained with the assumption that the only electron-loss process in the cavity is by attachment. As previously discussed,⁴ with a helium-buffer gas pressure of 20 Torr and with an I_2 vapor pressure of 2×10^{-3} Torr or greater, losses by diffusion processes can be essentially neglected at 295 °K under the conditions of the experiment.

The positive-ion ambipolar-diffusion coefficient was examined at high temperature in order to establish to what extent diffusion can be ignored in the high-temperature electron-attachment studies. For these studies a low I_2 vapor pressure was used so that electron-number-density loss by diffusion was greater than loss by attachment. The helium pressure was varied for these studies as previously described⁴ in order that the magnitude of the ambipolar diffusion could be separately assessed. A value for the product of the ambipolar diffusion coefficient and the helium pressure thus obtained was $D_{a+}p = 1500 \pm 200 \text{ cm}^2 \text{ Torr/sec}$

at 451 °K compared to the previously obtained value of $730 \text{ cm}^2 \text{ Torr/sec}$ at 295 °K.⁴ The deviation between the measured values for D_{a+} at elevated temperature and those expected for the case of constant mobility where D_{a+} (at constant pressure) increases as the square of the absolute temperature¹³ is not considered significant in the present case.

With the known high-temperature ambipolar-diffusion coefficient, the error in determining the attachment coefficient k at elevated temperature by using Eqs. (2) and (3) (where diffusion is omitted) can be assessed as previously described.⁴ With an iodine vapor pressure of 3×10^{-3} Torr, the error introduced by neglecting diffusion is essentially negligible for the data plotted in Fig. 2.

B. Variation of Electron Temperature

The effect of selective electron heating on the electron attachment coefficient k , where the gas temperature is maintained at room temperature, is shown in Fig. 3. As shown in the figure, k increases from $1.8 \times 10^{-10} \text{ cm}^3/\text{sec}$ at room-temperature electron energies up to $2.8 \times 10^{-10} \text{ cm}^3/\text{sec}$ for electron "thermal" energies in the range of 0.27 eV. Because of the assumptions involved in calculating the electron energy, the accuracy of the energy scale shown in Fig. 3 necessarily reflects the validity of these assumptions.

The increased diffusion encountered in the electron heating experiments can be used as a separate method to determine the electron thermal energy. The equation describing decay of the electron number density when negative ions are present is⁴

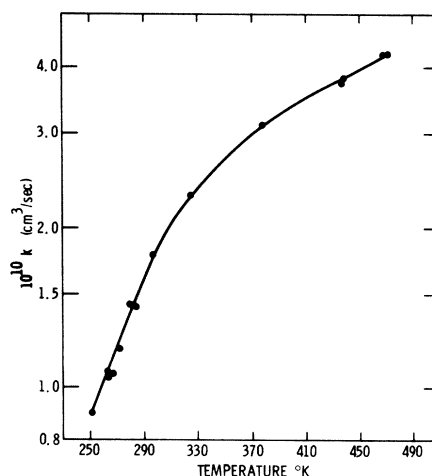


FIG. 2. Observed values for the electron attachment coefficient k for iodine vapor as a function of gas temperature.

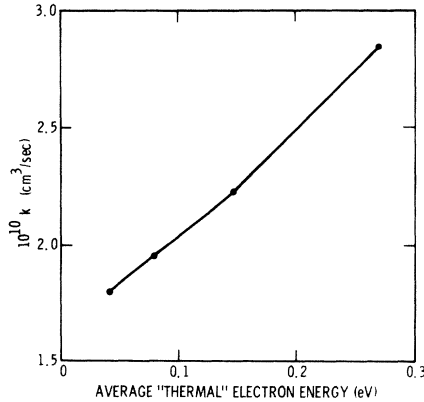


FIG. 3. Effect of selective electron heating on the attachment coefficient k , where the gas temperature was 298 °K, the iodine vapor pressure was 2.8×10^{-3} Torr, and the helium-buffer gas pressure was 20 Torr. The electron energies shown were determined from microwave-power measurements.

$$\frac{\partial n_e}{\partial t} = -\left(\frac{D_{ae}}{\Lambda^2} + \nu_a\right)n_e, \quad (7)$$

where D_{ae} is the electron ambipolar-diffusion coefficient and Λ is the characteristic diffusion length of the cavity. D_{ae} is defined as follows¹⁴:

$$D_{ae} \approx D_+ [1 + (T_e/T_g)(1 + 2\alpha)]. \quad (8)$$

In Eq. (8), the free positive-ion diffusion coefficient D_+ is assumed to be equal to the free negative-ion diffusion coefficient. Also in Eq. (8), T_e and T_g are the electron and gas temperatures, respectively, and α is the ratio of the negative-ion number density to the electron number density. Equation (8) is valid only for small values of α . For the measurements involved, the time dependence of the electron number density was measured for the various microwave-power levels utilized. A very low iodine vapor pressure was used so that the diffusion term in Eq. (7) dominates. Also, under such conditions, α in Eq. (8) is kept small during the measured decay of the electron number density. Comparing experimentally determined values for the time dependence of the electron number density with those obtained by numerically integrating Eq. (7) in conjunction with Eq. (8), the ratios of T_e/T_g were obtained at different microwave-power levels. The attachment frequencies ν_a used for Eq. (7) were obtained by first determining a low-power value for

this quantity under the conditions of the experiment. The variation of ν_a with microwave power was then obtained from the attachment-frequency data previously determined for Fig. 3. The values for T_e thus obtained are compared in Table I with those from the microwave-power measurements by use of Eq. (4). From the data shown in the table, good agreement appears to exist between the two methods of electron-thermal-energy measurement.

IV. DISCUSSION

Examination of the data presented in Fig. 2 shows that the attachment coefficient k for I₂ increases from 0.9×10^{-10} to 4.2×10^{-10} cm³/sec as the temperature is increased from 253 to 467 °K. If assumptions are made that at any given gas temperature the electrons have an average velocity \bar{v} and that a cross section for attachment, σ_a , is essentially constant over the velocity distribution, then $\sigma_a = k/\bar{v}$. If σ_a was plotted as a function of gas temperature, this quantity would increase by a factor of about 3.5 over the temperature range studied. A similar determination of σ_a for the selective electron-heating experiments shows that this quantity decreases slightly for increasing electron energies out to about 0.15 eV. The value for σ_a obtained at 0.15 eV was essentially identical to that obtained at 0.27 eV. It appears that since σ_a does not change substantially with increasing electron energy much of the increase in σ_a with increasing gas temperature is due to the change in the population distribution in the I₂ vibronic levels with increasing gas temperature.

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TABLE I. Comparison of calculated and inferred electron temperature (in eV).

T_e Microwave-power measurements	T_e Inferred from diffusion measurements
0.04	0.04
0.08	0.08
0.15	0.16
0.27	0.25

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Thermomagnetic Torque in He³†

D. A. Avery and T. W. Adair, III

Texas A&M University, College Station, Texas 77843

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The thermomagnetic torque or Scott effect has been observed in a number of polyatomic molecules.¹⁻³ Each molecule reported to produce the effect possesses a net magnetic moment and is in a rotational level above the ground state. An investigation of He³ may be significant because the He³ atom has a net magnetic moment but lacks rotational degrees of freedom.

Measurements have been made of He³ with a sensitive torsion-pendulum apparatus described elsewhere.⁴ This apparatus is capable of mea-

asuring a torque as small as 6.4×10^{-7} dyn cm. Measurements at pressures of 0.061 and 0.072 Torr, with magnetic fields from 0 to 150 Oe and temperatures of 310 to 350 K, indicate that no torque is present.

In view of this negative result for He³, we conclude that although the possession of a magnetic moment may be a necessary condition which a molecule or atom must have in order to exhibit the Scott effect, it is not a sufficient condition.

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