${}^{2}\Sigma_{u}^{+}$ H₂⁻ state have been carried out by Ehrhardt *et al*. [H. Ehrhardt, H. Langhans, F. Linder, and H. S. Taylor, Abstract of the Twentieth Gaseous Electronics Conference San Francisco, 1967 (unpublished), p. 64; see also Ref. 37]. It was found that the cross section for the excitation of the first vibrational level of H₂ becomes appreciable right at the first vibrational excitation threshold (~ 0.5 eV). Examining the relevant portion of the ${}^{2}\Sigma_{u}^{+}$ H₂⁻ curve (i.e., that portion lying below the dissociative attachment threshold) we found that this potential curve predicts moderately well the observed behavior for vibrational excitation. Since the ${}^{2}\Sigma_{u}^{+}$ H₂⁻ state lies about 3.3 eV above the ${}^{1}\Sigma_{g}^{+}$ H₂ state (Fig. 8)

and has a half width $\frac{1}{2}\Gamma$ of about 1.3 eV (Fig. 7), one would expect, based on the Breit-Wigner resonance formula, that resonance effects due to the ${}^{2}\Sigma_{u}^{+}$ H₂⁻ state should become appreciable at an energy below 2.0 eV (i.e., 3.3-1.3 eV).

The small discrepancy can be removed simply by modifying slightly the parameters in Eq. (3.12a) [see J. C. Y. Chen, in Advances in Radiation Chemistry (John Wiley and Sons, Inc., New York, 1968), Vol. I]. Detailed calculation of the resonant vibrational-excitation cross section with explicit consideration of the angular dependence of the scattered electrons is being carried out.

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Lifetime of a Negative Helium Ion

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The lifetime of a negative ion of helium produced from He⁺ by charge exchange in helium gas is measured and found to be $18.2\pm2.7 \mu$ sec. This is consistent with the lower limit of 10 μ sec previously determined. Total loss cross-section measurements in the energy range 20-70 keV are reported and compared with a previous published value at 17.5 keV. An investigation is also made of the destruction of the negative ions by thermal photons; this is found to be negligible at the temperatures involved, enabling an upper limit of 2.0×10^{-15} cm² to be set on the photodetachment cross section of He⁻.

1. INTRODUCTION

C INCE Hiby¹ first proposed the existence of the \mathfrak{I} negative ion of helium, a considerable amount of experimental and theoretical work has been carried out on it. The experimental work has been mainly directed at producing more intense ion beams, which are admirably suited for certain experiments in nuclear structure, and to the investigation of new processes for producing helium negative ions (Donnally and Thoeming²). Theoretical estimates of the binding energy of the ion have been made by Ta-You Wu,3 by Holøien,4 and by Holøien and Midtal,⁵ the latter results being largely borne out by the experimental work of Riviere and Sweetman⁶ and of Smirnov and Chibisov.⁷ Holøien and Midtal find that the only stable electronic configuration is the $(1s2s2p)^4P_{5/2}$ with a binding energy of 0.075 eV with respect to the $(1s2s)^3S$ metastable level. The ${}^{4}P_{5/2}$ state is radiatively metastable and not subject to auto-ionization (Holøien and Midtal⁵). For spon-

taneous transitions from this state, the only available final state, consistent with angular momentum and parity conservation, is then the $(1s^2kf)^2F$ state. In calculating the lifetime of the ${}^4P_{5/2}$ state, the matrix element connecting the stationary state to the ${}^{2}P_{5/2}$ continuum state, the Coulomb, and spin-orbit operators have vanishing matrix elements. However, transitions can still proceed by means of a spin-spin interaction between two of the electrons and calculations on this basis by Pietenpol⁸ yield a lifetime of 1.7 msec. More recent calculations by Laughlin and Stewart⁹ have shown that this value is too large and more refined calculations using more complex wave functions give lifetimes which lie close to 4×10^{-4} sec for uncorrelated wave functions. Introducing correlated wave functions into their calculations increases this value by one order of magnitude. However, it is of interest to determine experimentally the lifetime of the ion.

2. PRINCIPLE OF MEASUREMENT

An ion such as He⁻ decaying in flight with lifetime τ will, under idealized conditions of perfect vacuum, undergo loss of intensity such that the current after time t will be proportional to $e^{-t/\tau}$. With the known minimum life of 10⁻⁵ sec, an ion velocity of the order

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⁵ E. Holøien and J. Midtal, Proc. Phys. Soc. (London) A68, 815 (1955).
 ⁶ A. G. Riviere and D. R. Sweetman, Phys. Rev. Letters 5,

^{560 (1960).}

⁷ B. M. Smirnov and M. I. Chibisov, Zh. Eksperim. i Teor. Fiz. 49, 841 (1966) [English transl.: Soviet Phys.—JETP 22, 585 (1966)7.

J. L. Pietenpol, Phys. Rev. Letters 7, 64 (1961).

⁹C. Laughlin and A. L. Stewart, Proc. Phys. Soc. (London) A88, 893 (1966).





of 10^8 cm/sec and a flight path of 1 m, however, the reduction in intensity will be only 10%, so that accurate measurements can only be made if the flux of the daughter products, the fast neutral atoms, is observed directly. In addition, however, we have the production of neutral atoms through electron loss by collision with the molecules of the residual gas, which contributes an attenuation according to $\exp[-(\sigma K p x)]$, where σ is the total electron-loss cross section, p is the mean gas pressure in Torr, and $K=3.2.10^{+16}$ for an atomic absorber. (Additional sources of neutral production, by interaction of He⁻ with the slits defining the beam and by interaction with infrared photons, are discussed later.) Hence if $I^-(x)$ is the current of He⁻ at distance x, then

$$I^{-}(x) = I^{-}(x_0) \exp\left[-\left(\sigma K \rho + 1/v\tau\right)x\right], \qquad (1)$$

where x is in cm, v is the particle velocity, and τ is the lifetime in sec. It is shown (see Sec. 3) that positive ion production is negligible, so that the neutral flux

$$I^{0}(x) = I^{-}(x_{0}) - I^{-}(x).$$
⁽²⁾

The effects of electron loss are small when

$$p \ll 1/(v \tau K \sigma)$$
.

For $\sigma_{tot} = 10^{-15}$ cm² at an ion energy of 50 keV, we have

$$p_{\tau} \ll 3.12 \times 10^{-10}$$
 Torr sec,

so that for $\tau \sim 10^{-5}$, $p \ll 1.5 \times 10^{-5}$ Torr. For the experiments with 1-m flight path, the pressures were between 10^{-7} and 10^{-8} Torr.

The current of He⁻, of the order of 10^{-12} A, was measured directly on one of the detectors marked F (Fig. 1) and was continuously monitored throughout the experiment. The flux of neutrals was measured as a current of secondary electrons produced at the Ridley detector.¹⁰ Similarly, the total current ($I_{tot}=I^0+I^-$) was measured as a current of secondary emitted electrons on the same detector. Determining the ratio $I^0(x)/I^-(x)$ in terms of the neutral current and the total current on the Ridley detector enabled the measurements to be carried out on one detector only, thus introducing only one secondary emitting surface. The results quoted in Sec. 4 for the lifetime depend critically on the assumption that the secondary electron emission coefficients for He⁻ and He⁰ particles of the same velocity are equal. While to our knowledge there is no direct evidence for this, several investigators have determined this ratio as between positive ions and neutrals, using a thermocouple as the basic means of establishing particle flux. With this technique, Stier, Barnett, and Evans¹¹ have shown that between 20 and 200 keV the ratio of emission coefficients for fast helium neutrals to that for positive helium ions was constant at 1.05. It would seem improbable that the ratio of secondary emission coefficients for neutrals and negatives in this energy range was widely different from unity. Assuming, then, a ratio of unity, Eq. (1) may be rewritten as

$$\sigma(E)K \int_{0}^{L} p dx + \frac{L}{q\tau} E^{-1/2} = \ln(1+R), \qquad (3)$$

where

$$R = I^0 / (I_{tot} - I^0).$$

All currents in the above equation represent currents of secondary electrons: L is the interaction length in cm, E is the particle energy in keV, and q is a constant $=2.24\times10^7$. The procedure adopted is therefore (a) to estimate and subtract the pressure effect, and (b) to plot $\ln(1+R)$ against $LE^{-1/2}$.

3. EXPERIMENTAL TECHNIQUE

The apparatus is illustrated in Fig. 1: positive helium ions were produced in a cold-cathode mercury pool discharge and negative helium ions formed by charge exchange at energies between 6 and 45 keV. This produced a beam of He⁻ ions in the energy range 13–90 keV which was analyzed by a 20° magnet and passed through an interaction chamber that could be either 1 or 2 m in length and whose pressure could be varied over the range 10^{-3} – 10^{-8} Torr. After further analysis by a second bending magnet, the charged and neutral components of the beam were measured, the charged beams by means of a dc amplifier and the neutral components by means of a modified Ridley-type detector. This latter detector was a wide-aperture neutral ion detector with a copper-beryllium secondary emitting

¹⁰ B. W. Ridley, Nucl. Instr. Methods 14, 231 (1962).

¹¹ P. M. Stier, C. F. Barnett, and G. E. Evans, Phys. Rev. 96, 973 (1954).



FIG. 2. Neutral beam versus pressure for various beam energies.

surface and capable of count rates down to a few electrons per second. During the major part of the experiment it was found that the secondary electron beam was large enough for a conventional dc amplifier to be used. It was also found necessary to increase the angular acceptance of the neutral detector due to the large solid angles encountered in the second part of the experiment.

The entire system was constructed of stainless steel and pumped by six diffusion pumps. The first section of the interaction chamber could also be pumped by two cryogenic pumping units operated at liquid-helium temperatures. This enabled an ultimate pressure of approximately 10^{-8} Torr to be obtained in the 18-liter volume of the interaction chamber.

Pressure was measured by means of five Bayard-Alpert-type gauges effectively immersed in the evacuated volume and calibrated, with the usual precautions, against a McLeod gauge. The extreme variation in pressure as measured on the Bayard-Alpert gauges varied by no more than 4%. For the 2-m flight path, the pumping was poor compared to the 1-m path, and the minimum mean pressure was 1.0×10^{-5} Torr. However, such parameters as the electron-loss cross section (see Sec. 4A) were all measured using the shorter flight path.

To apply Eq. (3), we need to know the effectiveinteraction length L. This cannot be taken to be the distance between the defining apertures since neutrals formed between the first bending magnet and the first aperture, and between the second aperture and the second magnet, will contribute to the total. These effects were eliminated by deflecting plates at each end of the chamber, application of voltage to one or other of these plates gave the neutral contribution from one or other region, and correction to the experimental results could therefore be made.

Another experimental uncertainty concerned neutral production at the edges of the beam-defining apertures.

This effect was shown to be quite small (a) by steering a fine beam across the apertures, and observing a plateau over which the current did not change; since the beam is known to be narrower than the aperture, it is improbable that there is any interaction between beam and aperture, and (b) by careful collimation of the beam before the first bending magnet and removal of the defining apertures. The results, within experimental error, were the same.

Under normal conditions of observation (i.e., minimum pressure), the positive current was 1000 times less than the neutral current, so that $\sigma_{\bar{1},1} \ll \sigma_{\bar{1},0}$, $\sigma_{tot} \simeq \sigma_{\bar{1},0}$ and the beam could be assumed to be a two-component system consisting effectively of negative ions and neutrals.

4. RESULTS

A. Variation of $\sigma_{I,0}$ with Energy

The pressure correction for the 1-m flight path was small (a few per cent), but for the 2-m flight path the correction amounted to half the total. Therefore, the electron-loss cross section was determined directly as a function of energy. These measurements were carried out as follows: When the pressure in the interaction chamber is increased the pressure term on the left side of Eq. (3) dominates. Provided that the target thickness is accurately known, the electron-loss cross section may be determined from the slope of the $\ln(1+R)$ pressure graph. These results are shown in Fig. 2. Since values for the electron-loss cross section over this energy range have not been reported elsewhere, they are shown in Fig. 3, together with a single value reported by Windham et al.12 at 17.5 keV. The errors in measured loss cross section (on a least-squares fitting to the results of Fig. 2) vary from about $\pm 3\%$ at the higher energies

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¹² P. M. Windham et al., Phys. Rev. 109, 1193 (1954).



to approximately $\pm 7\%$ at the lower energies. These are indicated by error bars in Fig. 3.

We have observed above that our measurements depend critically on the assumption of equality of secondary emission coefficients for negative ions and neutrals. In principle, making this assumption can be avoided by the following procedure. The pressure is increased to the region of 10⁻⁴-10⁻³ Torr, when equilibrium is reached: Most of the beam then consists of neutrals, and, subtracting the directly measured positive and negative fractions, and knowing the original current (at zero pressure) one can deduce the secondary emission coefficient for neutrals. In practice, however, this procedure proved difficult: Apart from problems of reproducibility, questions of scattering effects were uncertain, so that as pressure increased, the current to the detector, with the second magnet off, was not flat, but reached a peak and then declined, and extrapolations to the "plateau" were uncertain. The errors, therefore, are appreciably greater than in Sec. 4B. Reference is made to the experiments, however, since they are not inconsistent with the other observations, and they therefore confirm that no major error is involved in the assumption of equal emission coefficients for negative ions and neutrals.

B. Results of Lifetime Measurements

The principal results of the experiments are shown in Fig. 4. In accordance with Eq. (3), $\ln(1+R)$ is plotted against $LE^{-1/2}$:

- (i) for hydrogen, 1-m flight path only, crosses;
- (ii) for helium, 1-m flight path, circles;
- (ii) for helium, 2-m flight path, triangles.

All pressure corrections have been subtracted. Ideally we should find for hydrogen zero net neutral current, while for helium we should obtain a set of points falling on a straight line including the origin. Comment on Fig. 4 is as follows.

1. Hydrogen

The hydrogen minus ion can reasonably be assumed to be stable, and the calibration of the apparatus with this ion is a useful check. The very small residual current of H^0 can be attributed to uncertainties in the gas species remaining at the best vacuum conditions; the variation in H^0 over a factor of 3 in flight time was negligible. These results are in accordance with the postulate of a stable hydrogen ion.

2. Helium, 1-m Flight Path

These results, often and accurately repeatable in conditions of a good vacuum, are shown in circles in Fig. 4: It is clear that they are fairly accurately colinear (correlation of experimental points is 0.98) and as expected include, within experimental error, the origin. From these results we deduce a lifetime of 18.2 μ sec for He⁻.

3. Helium, 2-m Flight Path

The results of a very limited set of experiments (corrected for pressure contribution) are shown as triangles in Fig. 3. Pumping in the long tube was relatively poor, and the pressure correction was half the total. The errors are clearly much greater than for the 1-m flight path, and all that can be said is that the results are not inconsistent with those for the 1-m flight path.

C. Photodetachment

In a blackbody at room temperature there are approximately 10^8 photons per cc. The binding energy



FIG. 4. Neutral beam versus time of flight (at two flightpath lengths).

of He⁻ has been estimated at 0.075 eV,⁷ while at 300°K the energy of the most intense photons is 0.1 eV approximately. Therefore it is necessary to demonstrate that no significant fraction of the neutral helium atoms were produced by photodetachment. The chamber was cooled in dry ice (195°K); no change in neutral current was detected to within 2%. From an analysis similar to that described by Branscomb and Smith¹³ we can place an upper limit of 2×10^{-15} cm² on the photodetachment cross section.

5. DISCUSSION

A. Errors

Basing our results on the 1-m flight path, we deduce a random error of 5%. Systematic errors may well be appreciably greater. These must include, for example, the ignorance of the secondary emission coefficients, together with any systematic errors for which there is no direct evidence. While it is difficult to make allowance for these factors, we consider it unlikely that the total error exceeds 15%.

B. Type of He- Ion

We have referred consistently to the "He⁻ ion" as if there were only one type, with one decay time. It could be that there were other excited states of the ion, with a much shorter life, which would decay before reaching the first bending magnet (contrast, however, the observations of Sweetman¹⁴), or other states, of much longer life, whose decay in a few microseconds was small. All we can conclude from the accurate colinearity of the 1-m flight-path results is that we were dealing with one type of ion only. The negative helium ions we are concerned with have been produced by charge exchange. Other methods of observation (e.g., inelastic electron scattering^{15,16}) yield negative states of the ion but the lifetimes of these states have not been directly determined.

C. Comparison with Theory

In conclusion, the final result of $18.2\pm2.7 \ \mu sec$ is considerably less than 400 μ sec, the least of the values resulting from theoretical studies.

¹⁵ G. J. Schulz, Phys. Rev. Letters 10, 104 (1963).
 ¹⁶ C. E. Kuyatt, J. A. Simpson, and S. R. Mielczarek, Phys. Rev. 138, A385 (1965).

¹³ L. M. Branscomb and S. J. Smith, Phys. Rev. 98, 1028 (1955).

¹⁴ D. R. Sweetman, Proc. Phys. Soc. (London) 76, 998 (1960).