## Production and Loss of Fast Metastable Helium Atoms in Collisions with Xe, $H_2$ , Ar, and He<sup>†</sup>

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Cross sections for production and destruction of fast  $He^0$  atoms in triplet states have been measured for 10- to 30-keV He<sup>+</sup> ions incident on gas targets of Xe, H<sub>2</sub>, Ar, and He. In a Cs vapor the production of positive and negative ions of He is sensitive to the fraction of metastable triplet atoms in the incident He<sup>0</sup> beam so that it is possible to use a thin Cs vapor target to analyze the fraction of triplet metastable atoms among the fast He<sup>0</sup> atoms emerging from the gas target of Xe, H<sub>2</sub>, Ar, or He. The fraction of neutral He<sup>0</sup> atoms in the triplet metastable state emerging from thin targets of Xe, H<sub>2</sub>, Ar, and He are  $0.19 \pm 0.07$ ,  $0.08 \pm 0.04$ ,  $0.05 \pm 0.02$ , and < 0.02, respectively, for incident He<sup>+</sup> energies of 25 keV.

Charge-changing collisions involving fast neutral He beams and various gas targets have been extensively studied. Most of the significant results through 1958 are summarized in a review article by Allison.<sup>1</sup> The analysis of these collisions to yield charge-changing cross sections is complicated by the fact that fast He<sup>o</sup> beams must be produced by charge-exchange collisions which results in the presence of atoms in long-lived metastable states in the fast He<sup>0</sup> beam. Some effects of these states have been noted by Barnett and Stier<sup>2</sup> and more recently by Gilbody et al.<sup>3</sup> For example a typical experiment to measure  $\sigma_{0+}$  involves neutralizing He<sup>+</sup> in a gas cell and subsequently measuring the cross section,  $\sigma_{0+}$ , by allowing the fast He<sup>o</sup> beam produced in the first cell to enter a second gas target and produce positive ions. It is observed that there is a dependence of the cross section,  $\sigma_{0+}$ , on the particular gas used in the preparation of the fast  $He^{0}$ beam by charge-exchange collisions and on the density of this gas. This variation of  $\sigma_{0+}$  has been attributed to the varying ratio of metastable to ground-state atoms. Gilbody et al.<sup>3</sup> have given a quantitative analysis of their data which yields the ratio of metastable to ground-state atoms in beams formed by neutralization of a 60- to 250-keV beam of He<sup>+</sup> ions in a gas such as  $H_{2}$ .

This paper reports measurements of the cross sections for production and destruction of fast  $He^0$  atoms in triplet states in gas targets of Xe,  $H_2$ , Ar, and He for energies of the incident  $He^+$  ion between 10 and 30 keV.

The measurements were carried out as follows. A He<sup>+</sup> beam is passed through two targets. In the first target the He<sup>+</sup> beam is neutralized by charge exchange. In this experiment the gases used as the first target were Xe, H<sub>2</sub>, Ar, and He. After the beam emerges from the first target any charged particles remaining in the beam are deflected, leaving only a fast He<sup>0</sup> beam. The

He<sup>0</sup> beam subsequently passes into a second target of Cs vapor, and the intensity of the He<sup>0</sup>, He<sup>+</sup>, and He<sup>-</sup> beams emerging from the Cs target is measured. Recent experiments<sup>4</sup> on the production and destruction of He<sup>-</sup>, He<sup>0</sup>, and He<sup>+</sup> from a fast He<sup>+</sup> beam incident on a Cs vapor have shown that the production of He<sup>+</sup> and He<sup>-</sup> from a fast He<sup>0</sup> beam is sensitive to the fraction of triplet metastable atoms in the He<sup>o</sup> beam because the cross sections for the production of He<sup>-</sup> or He<sup>+</sup> from a metastable triplet atom are much larger than the corresponding cross sections from a singlet atom. Therefore the Cs target makes a very useful analyzer for the fraction of triplets in the fast He<sup>0</sup> beam emerging from the first gas target.

The apparatus used in this experiment is essentially the same as that described in Ref. 4 except for the addition of a gas target of known length and density for neutralization of the fast  $He^+$  beam. The density of the target gas is measured with an ion gauge which is calibrated with a McLeod gauge.

In this paper the symbol  $\sigma_{if}$  will be reserved for cross sections of fast He incident on the first gas target which contains one of the gases Xe, H<sub>2</sub>, Ar, or He, and  $\Sigma_{if}$  will refer to cross sections for He incident on Cs. Throughout this paper the subscripts refer to the initial and final states of the He ion or atom. The subscripts +, -, t, or s refer, respectively, to He<sup>+</sup> ions, He<sup>-</sup> ions, He<sup>0</sup> atoms in the triplet states, and He<sup>0</sup> atoms in the singlet states as discussed in Ref. 4.

It is known that the production of He<sup>-</sup> or He<sup>++</sup> ions in Xe, H<sub>2</sub>, Ar, and He targets is negligible at these energies.<sup>1</sup> Therefore the production and loss of He<sup>+</sup>, He<sub>t</sub><sup>0</sup>, and He<sub>S</sub><sup>0</sup> within the gas target can be described by the following differential equations;

$$dF_t/d\pi = -(\sigma_{t+} + \sigma_{ts})F_t + \sigma_{st}F_s + \sigma_{+t}F_+, \quad (1)$$

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$$dF_{s}/d\pi = \sigma_{ts}F_{t} - (\sigma_{s+} + \sigma_{st})F_{s} + \sigma_{+s}F_{+}, \qquad (2)$$

$$dF_{+}/d\pi = \sigma_{t}F_{t} + \sigma_{s}F_{s} - (\sigma_{+t} + \sigma_{+s})F_{+}, \qquad (3)$$

where  $F_t$ ,  $F_s$ , and  $F_+$  denote, respectively, the fractions of the He beam in the metastable triplet  $He^{0}$  state, the singlet  $He^{0}$  state, and the  $He^{+}$ state.<sup>4</sup> The quantity  $\pi = \overline{\rho} \ l$  is the target thickness in atoms per cm<sup>2</sup>,  $\overline{\rho}$  being the average particle density of the gas and l being the path length in the gas target. Using the normalizing condition

$$F_t + F_s + F_+ = 1$$
 (4)

these equations can be reduced to two simultaneous differential equations and solved using appropriate boundary conditions.<sup>1</sup>

The Cs analyzer is very thin for the fast neutral atoms incident on it. However, the Cs analyzer is not thin for  $He^+$  or  $He^-$  ions produced in it. Under these conditions it can be shown that

$$\frac{G_{+}\Sigma_{+0}}{1 - \exp(-\pi_{Cs}\Sigma_{+0})} = \frac{F_{t}}{F_{t} + F_{s}} \Sigma_{t+} + \frac{F_{s}}{F_{t} + F_{s}} \Sigma_{s+}$$
(5)

and

$$\frac{G_{2}\Sigma_{-0}}{1 - \exp(-\pi_{Cs}\Sigma_{-0})} = \frac{F_{t}}{F_{t} + F_{s}}\Sigma_{t-} + \frac{F_{s}}{F_{t} + F_{s}}\Sigma_{s-},$$
(6)

where  $G_+$  and  $G_-$  denote the respective ratios of fast He<sup>+</sup> and He<sup>-</sup> to fast He<sup>0</sup> in the beam emerging from the Cs analyzer. By measuring  $G_+$  and  $G_$ as functions of the density,  $\pi$ , of the gas target and fitting the data to Eqs. (5) and (6), the ratios  $F_t/(F_t+F_s)$  and  $F_s/(F_t+F_s)$  can be determined. When He was used as the gas target  $G_+$  and  $G_$ did not vary with the gas density. Figure 1 shows typical data illustrating this result at 30 keV. This means that  $F_t/F_s$  is independent of the gas density in the first target. If we were to assume that the entire values of  $G_+$  or  $G_-$  resulted from metastable triplets produced in the He gas target (i.e., if we assume  $\Sigma_{S+} = \Sigma_{S-} = 0$  and if we use the values of  $\Sigma_{t+}$ ,  $\Sigma_{t-}$ ,  $\Sigma_{+0}$ , and  $\Sigma_{-0}$  from Ref. 4) then we could conclude that  $F_t/F_s \sim 0.02$  at all energies. However it would require a remarkable situation for  $F_t/F_s$  to be about 0.02 independent of the gas target density. We believe that it is more logical to assume that  $F_t < 0.02$ and that most of the production of positive or negative ions in the Cs analyzer is due to groundstate singlet He<sup>0</sup> atoms. If we assume that  $F_t/F_s$ =0 for the beam emerging from the He gas target then we can find  $\Sigma_{S+}$  and  $\Sigma_{S-}$  directly from the values of  $G_+$  and  $G_-$ . These values of  $\Sigma_{S+}$ and  $\Sigma_{S-}$  are given in Table I. A small value of  $F_t/F_s$  for He<sup>0</sup> beam formed in a He gas target

would be expected since the collision producing a ground-state singlet is resonant. This result is also consistent with other experimental measurements.<sup>3</sup>

When Xe,  $H_2$ , or Ar is used as the gas target the measured values of  $G_+$  and  $G_-$  are relatively high at low target densities and decrease as the target density increases until at high densities  $G_+$  and  $G_-$  have nearly the same values as for a beam neutralized in He. Typical data illustrating the results are shown in Fig. 1. We interpret this as indicating that at low gas target densities fast metastable atoms are formed and escape from the target, and as the target thickness increases these are destroyed during subsequent collisions. For very thick targets the ratio of metastable atoms to ground-state atoms in the beam is very small.

The analysis of our data utilizing the values of  $\Sigma_{+0}$ ,  $\Sigma_{-0}$ ,  $\Sigma_{t+}$ , and  $\Sigma_{t-}$  from Ref. 4 and the values of  $\Sigma_{S+}$  and  $\Sigma_{S-}$  obtained using a He gas target yields values of  $F_t/(F_t+F_S)$  and  $F_S/(F_t+F_S)$  for Xe, H<sub>2</sub>, and Ar targets. At low gas target densities the ratios  $F_t/(F_t+F_s)$  and  $F_s/(F_t+F_s)$  approach  $\sigma_{+t}/\sigma_{+t}$  $(\sigma_{+t}+\sigma_{+s})$  and  $\sigma_{+s}/(\sigma_{+t}+\sigma_{+s})$ , respectively. Therefore we can obtain directly only ratios of cross sections. We obtain the individual cross sections by utilizing values of cross sections measured by other workers.

Barnett and Stier<sup>2</sup> have measured  $\sigma_{+0}$  and  $\sigma_{0+}$ in Ar and H<sub>2</sub>. Since they used equilibrium ratios of  $F_+$  and  $F_0$  produced in thick gas targets to determine cross sections, we assume that their



FIG. 1. The values of

and

$$G_{+}\Sigma_{+0}/[1-\exp(-\pi_{Cs}\Sigma_{+0})]$$

 $G_{\Sigma_{0}} [1 - \exp(-\pi_{CS} \Sigma_{0})]$ in units of  $10^{-17}$  cm<sup>2</sup> as a function of the number of collisions in a thin gas target  $(\pi \sigma_{\pm 0})$ , for incident 30-keV  $\mathrm{He}^+$  ions. The symbols are  $\Box$  -He gas target,  $\triangle$  -Ar gas target,  $\times -H_2$  gas target, and  $\bigcirc$ -Xe gas target. The values of  $\sigma_{+0}$  used in plotting the ordinate for He, Ar, and H<sub>2</sub> are taken from Barnett and Stier.<sup>2</sup> The value of  $\sigma_{\pm 0}$  for Xe is taken from Stedeford.<sup>5</sup>

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He energy	ັ   ບ້	esium		Xenc	ų			Hydrog	ue			Argoi	d		11
(keV)	$\Sigma_{S^+}$	2 <sub>8-</sub>	$F_t/(F_t+F_s)$	$\sigma_{+t}$	$\sigma_{+S}$	$\sigma_{t+}^{}+\sigma_{ts}$	$F_{t}^{\prime}^{\prime}(F_{t}^{+}F_{s}^{})$	$\sigma_{+t}$	$\sigma_{+s}$	$\sigma_{t+}^{+\sigma_{ts}}$	$F_{t}^{\prime }/\left( F_{t}^{+}F_{s}^{-}\right)$	$\sigma_{+t}$	$\sigma_{+S}$	$\sigma_{t^+}^{}+\sigma_{ts}^{}$	
30	3.6	0.32	0.21	25	85	140	60.0	2.0	21	60	0.06	4	66	100	
	$\pm 0.5$	± 0.10	$\pm 0.08$	<b>4</b> 9	± 16	<b>±</b> 70	$\pm 0.04$	± 0.8	± 2	±30	$\pm 0.02$	$\pm 2$	± 7	$\pm 50$	
25	2.6	0.34	0.15	16	88	150	0.08	1.6	18	70	0.05	ŝ	70	130	
	± 0.5	± 0.10	± 0.07	<del>+</del> 9	± 17	<b>±</b> 70	$\pm 0.04$	± 0.7	± 2	$\pm 30$	$\pm 0.02$	± 1	± 7	<b>±</b> 60	
20	2.0	0.3	0.14	14	84	150	0.13	2.3	16	70	0.04	57	75	100	
	± 1.0	± 0.1	± 0.06	<b>∓</b> 6	± 14	± 70	$\pm 0.04$	± 0.7	± 2	$\pm 30$	$\pm 0.02$	± 1	# 8	± 50	
15	1.0	0.2	0.09	10	95	200	0.07	1.0	13	70	0.03	1.8	62	150	
	± 0.3	± 0.1	$\pm 0.03$	± 4	± 16	± 100	± 0.03	± 0.5	$\pm 2$	$\pm 30$	$\pm 0.01$	± 0.8	± 6	± 100	
10	0.4	0.2	0.04	4	104	250	0.07	0.9	12	80	0.012	0.7	55	150	
	± 0.1	± 0.1	$\pm 0.02$	± 2	± 16	$\pm 100$	$\pm 0.03$	$\pm 0.4$	$\pm 1$	$\pm 30$	$\pm 0.008$	$\pm 0.5$	<b>±</b> 6	$\pm 100$	

values are  $\sigma_{+S}$  and  $\sigma_{S+}$ .

The values for  $\sigma_{+t}$  for He<sup>+</sup> incident on Ar or H<sub>2</sub> are given in Table I. Gilbody et al. have cast some doubts on the correctness of Stier and Barnett's values for  $\sigma_{+s}$ . Our values of  $\sigma_{+t}$  will be in error by an amount equal to any error in  $\sigma_{+s}$ .

The value of  $\sigma_{\downarrow 0}$  in Xe was measured by Stedeford<sup>5</sup> using a thin gas target. It is assumed in this case that  $\sigma_{+0} = \sigma_{+t} + \sigma_{+s}$ . From our low density data and  $\sigma_{+0}$  we can obtain  $\sigma_{+t}$  and  $\sigma_{+s}$  in Xe. These are given in Table I.

By varying  $\sigma_{t+}$  and  $\sigma_{ts}$ , the best fit to the dependence of our data on  $\pi$  was computed. It was found that  $\sigma_{t+}$  could be varied over several orders of magnitude without noticeably affecting the fit, and only the sum  $\sigma_{t+} + \sigma_{ts}$  can be determined with any precision. Figure 1 shows computed fits to a typical set of data at 30 keV. Values of  $\sigma_{t+} + \sigma_{ts}$ for Xe, H<sub>2</sub>, and Ar are given in Table I. In calculating  $\sigma_{t+} + \sigma_{tS}$  for  $H_2$  and Ar we have used Barnett and Stier's value of  $\sigma_{0+}$  as  $\sigma_{S+}$ . However, because  $\sigma_{S+}$  is very small the resulting values of  $\sigma_{t+} + \sigma_{ts}$  are not sensitive to  $\sigma_{s+}$ . For Xe no measured value of  $\sigma_{S+}$  was available so we computed  $\sigma_{t+} + \sigma_{ts}$  setting  $\sigma_{s+}$  equal to zero.

We should like to comment on the major difference between our analysis and that of Gilbody et al. We measured the He<sup>-</sup> ions produced in the Cs vapor. Since the state of He<sup>-</sup> is (1s)(2s)(2p) ${}^{4}P_{5/2}$ , it is formed predominantly in collisions of He atoms in the (1s)(2s)  ${}^{3}S_{1}$  state with Cs<sup>0</sup>.<sup>4</sup> The values of  $F_t$  (both magnitude and variation with  $\pi$ ) resulting from the He<sup>-</sup> yield are such as to indicate that almost all He<sup>+</sup> ions produced in the Cs<sup>o</sup> are the result of ionization of He<sup>o</sup> atoms in the triplet metastable state. For some reason (unknown to us) the singlet metastable state (1s)(2s)  ${}^1S_0$  of He<sup>0</sup> seems to play no role in production of He<sup>+</sup> in Cs. This is similar to the results of our previous experiments involving He<sup>+</sup> ions incident of Cs where the data could be analyzed ignoring effects due to metastable singlet atoms.<sup>4</sup> This may be because the singlet metastable state is not produced with a large cross section or perhaps because it is converted into the ground state with a much larger cross section than into He<sup>+</sup> ions. Gilbody *et al.* did not measure the He<sup>-</sup> yield and they assumed that metastable singlet atoms may be produced and may play a role in the production of He<sup>+</sup> ions in the second target. At the energy of our experiment (much lower than the energy for the experiments of Gilbody *et al.*) we do not think this is the case.

It is interesting to note that the cross section  $\sigma_{\pm t}$  in the gases measured seems to be large if the polarizability of the gas is large and if the energy defect for the collision is small. In addition  $\sigma_{+t}$  increases with energy, whereas in Cs where the pick up into the metastable state is

TABLE I. Cross sections for charge exchange of He ions in Cs vapor  $(\Sigma_{ij})$  and in gas targets  $(\sigma_{ij})$ , in units of 10<sup>-17</sup> cm<sup>2</sup>. Also listed is  $F_t/(F_t+F_s)$  the fraction

He<sup>0</sup> beam in triplet metastable state for low gas densities. The value of  $\sigma'_{+s}$  is due to Barnett and Stier for H<sub>2</sub> and Ar targets.<sup>2</sup> In Xe  $\sigma_{+s}$  is Stedeford's  $\sigma_{+0}$ 

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nearly resonant,  $\sigma_{+t}$  decreases with energy. The total destruction cross section for metastable atoms in the triplet state  $\sigma_{t+} + \sigma_{tS}$  is almost independent of the energy.

In conclusion we would like to stress the fact that the large cross sections  $\Sigma_{f+}$  and  $\Sigma_{f-}$  in Cs

vapor make Cs a very useful analyzer of the rate of production and loss of metastable triplet He<sup>0</sup> atoms in various gas targets. Thus one can measure the cross section  $\sigma_{+t}$  and can obtain  $\sigma_{t+} + \sigma_{ts}$  in various gas targets.

<sup>†</sup>Supported by a grant from the Research Committee of the University of Wisconsin with funds provided by The Wisconsin Alumni Research Foundation.

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PHYSICAL REVIEW

## VOLUME 183, NUMBER 1

5 JULY 1969

## Charge Equilibrium Fractions of a Helium Beam in Hydrogen, Nitrogen, Neon, and Argon from 60 to 850 keV\*

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Equilibrium fractions of charge 0, +1, and +2 of helium beams traversing the gas targets  $H_2$ ,  $N_2$ , Ne, and Ar have been measured. Primary beams of  $(He^4)^+$  and  $(He^3)^{++}$  were used. The measurements cover ion velocities corresponding to energies of  $(He^4)^+$  between 60 to 850 keV. The charge-equilibrated beam was magnetically separated and charge spectra were recorded by means of a movable detector of equal sensitivity for all three charge components. In the energy interval of overlap, that is between 60 and 200 keV, agreement with Barnett *et al.* is excellent in the fractions  $F_{0\infty}$  and  $F_{1\infty}$ , which make up almost the entire charge-equilibrated beam. The results show a strong dependence of the equilibrium charge distributions on the chemical nature or electronic configuration of the gas target.

## I. INTRODUCTION

Charge equilibrium fractions of a He beam in a He gas target were presented in an earlier paper,<sup>1</sup> henceforth referred to as I. The present measurements have been performed with the same basic equipment. Some improvements, to be described below, have been introduced.

The information on charge equilibrium fractions of He beams traversing different gaseous targets, available up to 1962, has been reviewed by Allison and García Muñoz.<sup>2</sup> From 8 to 200 keV this information is based essentially on measurements of Stier, Barnett, and Evans,  $^{\rm 3}$  and Barnett and Stier.  $^{\rm 4}$ 

For the target gases  $H_2$  and He, from 200 to 480 keV the tabulated values are calculated from charge changing cross-section measurements of Allison and others.<sup>5</sup>

For the case of a 200-keV-He beam on a He gas target the discrepancies between the computed charge equilibrium fractions and the corresponding values measured by Barnett *et al.*<sup>3,4</sup> amount up to 15%, as already mentioned in I. Due to systematic discrepancies the measurements of