Metastable fractions of fast He atoms produced by electron detachment from He^- ions

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The metastable fraction of He atoms produced by electron detachment from He^- ions in single collisions with H_2, He, N_2, Ne, and Ar has been measured in the energy range 25–400 keV by the beam-attenuation technique, using H_2 as attenuation gas. These data support earlier results, pointing to the existence of a bound, long-lived doublet state of He^-. Further, the electron loss and deexcitation cross sections for metastable He atoms in He, N_2, Ne, and Ar have been measured individually at 50 keV, and it is shown that the deexcitation cross section is comparable to the electron-loss cross section.

INTRODUCTION

Recently, the He^- ion has been the subject of a number of investigations dealing with either formation,[1] destruction[2-5] (electron detachment), or lifetimes, and fine structure[6,7] of this ion which exists in metastable, doubly excited states only.[8]

The electron-detachment studies of He^-, including both collisional[2-4] and electric-field detachment,[5,9,10] have been concentrated on either the total-detachment probability[2,3,9,10] or the relative probabilities of producing metastable and ground-state He atoms.[4,5] In the latter type of experiments, metastable fractions of about 70% or less have been found. The lifetime studies have, on the other hand, shown that two quite distinct lifetime components of 10 and 350 μsec are present. These lifetimes have been associated with the $J=\frac{1}{2}$, $\frac{3}{2}$, and $J=\frac{5}{2}$ fine-structure components of the lowest doubly excited quartet state $1s2s2p\ ^4P^o$, which is known to be bound[8] and metastable against autoionization as well as radiative decay. This identification has, however, been questioned[5] on the basis of the mentioned measurements of metastable fractions, and the suggestion has been made[5] that the short-lived component should be associated with the lowest doubly excited doublet state $1s2p^2\ ^2P^e$, which is metastable against autoionization. The existence of this state has been assumed previously to explain single capture by fast ground-state He atoms in metal vapors.[11]

The present experiment is an extension of our previous study of the production of metastable He atoms by collisional detachment from 50-keV He^- ions in H_2.[4] The energy interval is 25–400 keV and the investigated detachment gases are H_2, He, N_2, Ne, and Ar. The metastable fractions were measured using the beam-attenuation technique[12] with H_2 as attenuation gas since this has been shown to give the true metastable fraction.[5] At the highest energies, where electron-exchange reactions are negligible, the metastable fractions will give information about the initial-spin state of the He^- ion. At the lower energies, on the other hand, electron exchange causes the metastable fraction to decrease at different rates for the different detachment gases, as will be seen by the experimental results.

Since our conclusions on the initial-spin state of the He^- ion reported on previously[5] relied on a correct measure of the metastable fraction, we investigated the attenuation process itself by using attenuation gases other than H_2.

In a series of experiments at 50 keV, He, N_2, Ne, and Ar were used as attenuation gases while H_2 was used as detachment gas. These measurements allowed the separate determination of the electron-loss cross section and the deexcitation cross section for metastable He atoms in He, N_2, Ne, and Ar at 50 keV.

EXPERIMENTAL PROCEDURE

The experimental arrangement was the same as that described in a previous article.[4] A schematic is shown in Fig. 1. He^- ions are produced in the charge-exchange cell and selected electrostatically. The negative ions are partly neutralized in the detachment cell. In the last part of the apparatus, the metastable fraction of the resulting neutrals is measured by the attenuation technique described by Gilbody *et al.*[12]

Briefly, in the approximation used by Gilbody *et al.*,[12] if the initial neutral beam, of intensity I_0, contains a fraction f of metastable atoms, then its intensity after passing through an attenuation gas of thickness μ is

$$I=(1-f)I_0\exp(-\mu\sigma_{01})+fI_0\exp(-\mu\sigma_{0*1}),$$

where σ_{01} and σ_{0*1} are the one-electron-loss cross sections for ground-state and metastable atoms, respectively. If $\sigma_{0*1}\gg\sigma_{01}$, then the two exponen-

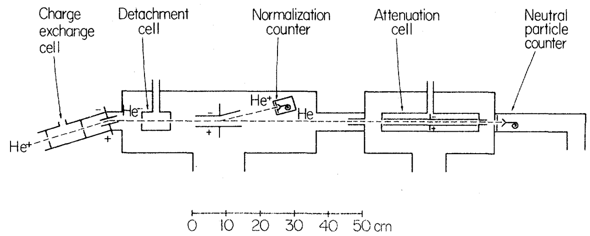

FIG. 1. Experimental arrangement.

tials are easily resolvable in a plot of $\ln(I/I_0)$ against μ, and a value of f is obtained.

Our experimental work can be divided into two separate sections: (i) the investigation of the fraction of metastable He atoms produced by collisional electron detachment from He^-; (ii) the investigation of the assumptions used in the simple analysis of the attenuation method. These will now be discussed in turn.

(i) In our previous investigation,[5] we were able to show that when H_2 is used as attenuation gas, the above simple analysis gives the true metastable fraction f. Therefore, in the investigation of the metastable fraction of He atoms resulting from collisional detachment, H_2 was always used as attenuation gas. The fractions produced by electron detachment in single collisions of 25–400-keV He^- were measured with the detachment gases H_2, He, N_2, Ne, and Ar.

The single-collision value of f was obtained by measuring $f(\mu)$ as a function of detachment-gas thickness μ near the single-collision region and then extrapolating to $\mu=0$ to obtain $f(0)$, as described in Ref. 4.

The size of the apertures used in this experiment was such that neutral particles scattered through angles larger than 2.5 mrad in the detachment-gas cell would hit the entrance aperture to the attenuation-gas cell. The influence on the measurements of this small acceptance angle was studied at 50 keV for a low and a high Ar detachment-gas pressure by varying the acceptance angle from 2.5 mrad to 0.7 mrad by means of a set of movable slits situated between the two vacuum chambers of the experimental arrangement. No variation of $f(\mu)$ was found, so it was concluded that the measured values of $f(0)$ are representative of the total neutral beam.

(ii) The second part of the experiment was an investigation of the attenuation method itself. The basis of this method has been discussed in detail by Vujović *et al.*[13] They show that if the excitation cross section σ_{00*} for ground-state atoms and the deexcitation cross section σ_{0*0} for metastable atoms in the attenuating gas are not negligible compared to σ_{01} and σ_{0*1}, then the attenuation method will not yield the true metastable fraction f of the neutral beam. Instead, it will give an apparent fraction Φ, related to f by

$$\Phi = f\,\frac{\sigma_{0*1}-\sigma_{01}}{\sigma_{0*1}+\sigma_{0*0}-(\sigma_{01}+\sigma_{00*})} - \frac{\sigma_{00*}}{\sigma_{0*1}+\sigma_{0*0}-(\sigma_{01}+\sigma_{00*})}\ . \tag{1}$$

The rate of decay of the two exponentials seen in the attenuation data is given by $\sigma_{0*1}+\sigma_{0*0}$ and $\sigma_{01}+\sigma_{00*}$ for the metastable and ground-state component, respectively.[13] Only in cases where σ_{0*0} or σ_{00*} are negligible compared to the corresponding electron-loss cross section will σ_{0*1} or σ_{01} be measured in this type of experiment.

Gilbody *et al.*[12] used different attenuation gases in order to check that σ_{0*0} and σ_{00*} were negligible and found no variation in metastable fraction within their experimental accuracy. However, the metastable fractions they measured were 25% or less, whereas in the present work, the fractions are about 70%, so that a more sensitive check is possible.

As we will discuss below, our results show that the metastable fraction does depend on the attenuation gas so that Φ rather than f is being measured.

At a fixed energy of 50 keV, apparent metastable fractions $\Phi(\mu)$ were measured using He, N_2, Ne, and Ar as attenuation gases. In this part of the experiment, the detachment gas was always H_2. Once again, a variation of the metastable content of the He-atom beam could be achieved by varying the detachment-gas thickness μ.

The experimental values of $\Phi(\mu)$ are shown in Fig. 2 together with the previously published[4] true

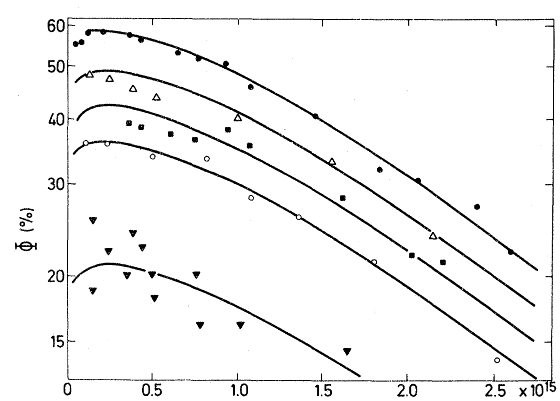

FIG. 2. Apparent metastable fractions Φ of 50-keV He beams prepared by electron detachment from He^- in H_2 at various detachment-gas thicknesses. The apparent metastable fractions were measured as explained in the text, using different attenuation gases: ●, H_2; △, Ar; ■, N_2; ○, He; ▼, Ne.

metastable fractions found using H_2 as attenuation gas. The fully drawn curve through the H_2 data is the best theoretical fit to these.[4] Curves with the same shape as this have been drawn through the experimental points for the other attenuation gases used. The figure shows that $\Phi(\mu)$ is proportional to $f(\mu)$ or, using Eq. (1), that σ_{00*} is negligible compared to $\sigma_{0*1}+\sigma_{0*0}$ in the attenuation gases used. This result is consistent with the fact that the metastable fraction of neutrals is very small at charge-state equilibrium in He, N_2, Ne, and Ar at these energies,[14] implying that the total probability of producing metastables is much less than the probability of destroying them. The ratio between $\Phi(\mu)$ measured in one of the attenuation gases He, N_2, Ne, or Ar and $f(\mu)$ measured in H_2 is then given by

$$R=\frac{\sigma_{0*1}-\sigma_{01}}{\sigma_{0*1}+\sigma_{0*0}-\sigma_{01}}, \tag{2}$$

where the cross sections apply to the particular attenuation gas used.

It is possible to extract the cross sections σ_{0*1} and σ_{0*0} individually from this ratio because $\sigma_{0*1}+\sigma_{0*0}$ and σ_{01} are known from the attenuation data used to determine $\Phi(\mu)$ or from previously published attenuation experiments.[12,15]

It should be noted that our analysis is still an approximation since, as normally accepted, we assume that electron capture and double loss can be ignored at the present energies.

For the lighter gases, attenuation due to scattering was less than 15% at the highest pressures used but was very severe in the cases of Ne and Ar, being a maximum of 50% and 80%, respectively. However, in each case, this attenuation was linear on a semilog plot, showing that the scattering will not influence the measurement of the metastable fraction. As an extra check, $\sigma_{0*1}+\sigma_{0*0}$ and σ_{01} were found for He, Ne, and Ar by correcting the curves for the scattering loss. In Table I, the results are compared with those of previous workers, where it can be seen that the agreement is satisfactory, particularly for Ne and Ar, where the correction was large.

RESULTS AND DISCUSSION

In order to avoid confusion, the results will be discussed in two parts, corresponding to the two sections of the experiment previously described.

(i) The measured true metastable fractions $f(0)$ produced by the various detachment gases are shown in Fig. 3. Since the ratio between the loss cross sections for metastable and ground-state He atoms in H_2 decreases in the present energy range,[12] the attenuation method is more precise at the lower energies than at the higher ones. This is reflected by the error bars shown in Fig. 3, which range from ±5% at the lowest energies to ±10% at the highest. The systematic error is estimated to be less than 5%.

TABLE I. Comparison of the present values of the metastable destruction cross section $\sigma_{0*1}+\sigma_{0*0}$ and the ground-state, one-electron-loss cross section σ_{01} for He atoms at 50 keV with those of other workers. The results of Gilbody *et al*. (Ref. 12) and Tawara (Ref. 15) have been read from figures, and in the former case, for Ne and Ar, have been extrapolated from 60 keV.

		He	Ne	Ar
$\sigma_{0*1}+\sigma_{0*0}$ (10^{-17} cm^2)	Present work	60 ± 6	32 ± 3	100 ± 10
	Gilbody *et al*.[a]	34	39	95
	Tawara[b]	50	20	148
σ_{01} (10^{-17} cm^2)	Present work	8.3 ± 0.3	9.2 ± 0.9	10 ± 1
	Gilbody *et al*.[a]	5.8	10	12
	Tawara[b]	5.0	12	7

[a]Reference 12.
[b]Reference 15.

The energy variation of $f(0)$ is very different in the various detachment gases used, as is seen in Fig. 3. While there is a weak minimum between 50 and 100 keV in H_2 and a plateau in the range 25–100 keV in N_2, $f(0)$ rises monotonically in the other gases in the present energy range. For low energies, $f(0)$ depends strongly on the detachment gas used. At 25 keV, for example, values of $(63.0\pm2.5)\%$ and $(35.5\pm2.5)\%$ are found in H_2 and Ne, respectively. At high energies, however, $f(0)$ approaches the same value, 70–80%, in all detachment gases. In a previous study, in which electric-field detachment was used,[5] the same metastable fraction $(71\pm2)\%$ was found. It was concluded[5] that this fraction is characteristic for the population of the spin states of the He^- ions entering the detachment cell, and it was further suggested that the short-lived component of He^- should be associated with the $[(1s2p,\ ^3P), 2p]^2P^e$ state and the long-lived component of He^- should be associated with the $1s2s2p\ ^4P^o$ state.

A simple model, in which it is assumed that this identification of the two lifetime components of He^- is correct, was developed to explain the energy variation shown in Fig. 3. Within this model, electron detachment from He^- ions in the $^4P^o$ state leads to metastable He atoms only, while electron detachment from He^- ions in the $[(1s2p,\ ^3P), 2p]^2P^e$ state leads to ground-state atoms in half of the events and to metastable He atoms in the rest, according to whether the two remaining electrons have antiparallel or parallel spins. The He^- lifetime studies have shown that the two components of He^- are initially populated to approximately the

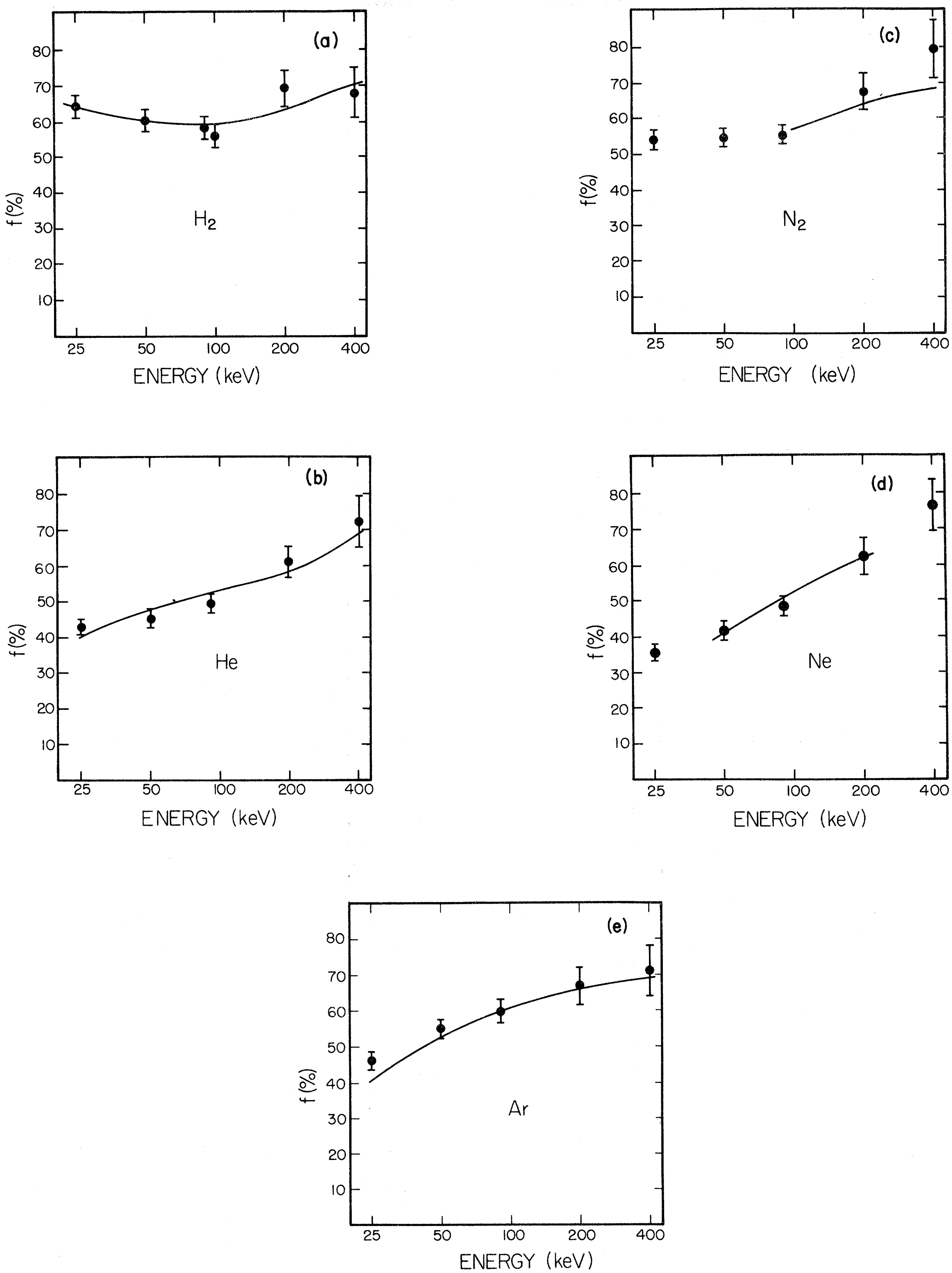

FIG. 3. Energy variation of the metastable fraction $f(0)$ of He beams prepared by electron detachment from He^- ions under single-collision conditions in H_2, He, N_2, Ne, and Ar. Fully drawn curves are semiempirical theoretical results calculated as explained in text. Φ, experimental results.

same degree.[6,7] The present model will thus predict a metastable fraction of approximately 75% at high energies where exchange reactions can be neglected. However, for lower energies, where such reactions attain their maximum value, $f(0)$ deviates appreciably from 75% in some gases. This is explained by introducing into the model the quenching mechanism first discussed by Hughes and Kisner[16] in connection with destruction cross sections for metastable hydrogen atoms. According to this mechanism, ground-state He atoms are formed by electron capture to the ground state of the He^+ core which, during a collision, will be screened to a small extent only by the two excited electrons. The cross section for this indirect detachment process is assumed to equal the electron-capture cross section for He^+ ions. The last assumption, which has to be made in order to make it possible to calculate the energy variation of $f(0)$, is that the cross sections for the direct detachment processes discussed above all vary in the same way with energy in the present energy range. Since the binding energies of the electrons involved are approximately the same, this assumption is probably satisfied.

The probabilities P^* and P of producing metastable and ground-state He atoms by electron detachment from He^- under single-collision conditions are thus given by

$$P^* = \mu(\sigma^q_{-10*} + \sigma^d_{-10*}) ,$$
$$P = \mu(\sigma^d_{-10} + \sigma_{10}) ,$$

where μ is the target-gas thickness measured in units of cm^{-2}, σ^q_{-10*} is the electron-detachment cross section for He^- ions in the $^4P^o$ state, σ^d_{-10*} and σ^d_{-10} are the electron-detachment cross sections leading to metastable and ground-state He atoms for He^- ions in the $^2P^e$ state, and σ_{10} is the total electron-capture cross section for He^+ ions.

The metastable fraction $f(0)$ is now given by

$$f(0) = \frac{P^*}{P^* + P} = \frac{\sigma^q_{-10*} + \sigma^d_{-10*}}{\sigma^q_{-10*} + \sigma^d_{-10*} + \sigma^d_{-10} + \sigma_{10}} .$$

Introducing the total electron-detachment cross section,

$$\sigma_{-10} = \sigma^q_{-10*} + \sigma^d_{-10*} + \sigma^d_{-10} + \sigma_{10}$$

and the high-energy value of $f(0)$,

$$f_\infty(0) = (\sigma^q_{-10*} + \sigma^d_{-10*})/(\sigma^q_{-10*} + \sigma^d_{-10*} + \sigma^d_{-10}) ,$$

leads to

$$f(0) = f_\infty(0)(1 - \sigma_{10}/\sigma_{-10}) .$$

The fully drawn curves in Fig. 3 were calculated on the basis of this formula by using available experimental values of the total collisional electron-detachment cross section[2,3,23] and the total electron-capture cross section[17,18] as well as the value 0.71 for $f_\infty(0)$ found in our previous work.[5]

The excellent agreement between the experimental results and the curves in Fig. 3 strongly suggests that the model used is essentially correct.

(ii) The results of the measurements at 50 keV, in which apparent metastable fractions were obtained in the attenuation gases He, N_2, Ne, and Ar, with H_2 as detachment gas, are shown in Table II. The values of R [Eq. (2)] and the corresponding uncertainties were obtained directly from Fig. 2, while the values of σ_{01} and $\sigma_{0*1} + \sigma_{0*0}$ are those of Gilbody *et al.*[12] The cross sections listed in Table II show that deexcitation is a very important quenching mechanism for 50-keV metastable He atoms in the particular gases studied. The ratio $\sigma_{0*0}/\sigma_{0*1}$ varies from about 0.9 in Ne to 0.18 in Ar.

This is in disagreement with the finding of Gilbody *et al.*[12] However, more recent data of Blair *et al.*[19] and McCullough *et al.*[20] on the excitation of He atoms using fast He($1\,^1S$) and He($2\,^3S$) atoms show a remarkably large value of the cross section for the process,

$$He(2\,^3S) + He(1\,^1S) \rightarrow He + He(4\,^3S \text{ or } 4\,^3D) ,$$

of the order of 10^{-17} cm^2 at 50 keV. Using the Wigner-spin conservation rule and their experimental findings, Blair *et al.*[19] show that this process leaves the fast He atoms in the ground state and so is part of the cross section σ_{0*0}. The results of Blair *et al.* and McCullough *et al.*[20] apply for excitation of the target to the $n=4$ level alone. An estimate of the total deexcitation cross section using the n^{-3} scaling rule yields a value of the order of 10^{-16} cm^2, in agreement with the present findings.

Thus our results show that, at low energies (probably less than 100 keV) for He, N_2, Ne, and Ar targets, attenuation data do not yield σ_{0*1} but the sum $\sigma_{0*1} + \sigma_{0*0}$, which, for He, N_2, Ne, and Ar is about 43%, 34%, 90%, and 18% larger than

TABLE II. Electron-loss and deexcitation cross sections σ_{0*1} and σ_{0*0} in units of 10^{-17} cm^2 for 50-keV metastable He atoms in He, N_2, Ne, and Ar as found in the present experiment. Values of the ratios R [Eq. (2)] are also listed. The theoretical cross sections are calculated in the classical-impulse approximation of Bates and Walker (see Ref. 22).

	He	N_2	Ne	Ar
R	0.63 ± 0.02	0.70 ± 0.02	0.38 ± 0.04	0.83 ± 0.02
σ_{0*0}	10 ± 2	28 ± 6	19 ± 4	15 ± 3
σ_{0*1}	23 ± 5	82 ± 16	21 ± 4	85 ± 17
σ_{0*1}(Theor)	29	75	22	120

σ_{0*1}, respectively. The agreement between the present experimental values of σ_{0*1} for metastable He atoms and the corresponding theoretical values[21] calculated in the classical impulse approximation of Bates and Walker[22] and listed in Table II is generally better than that found earlier.[12] In particular, the discrepancy of about a factor of 2 at low energies in Ne is removed.

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