Electron Capture into Excited Helium Levels by Fast He^+ Impact on He, N_2, and O_2†

CHARLES E. HEAD* AND R. H. HUGHES
Department of Physics, University of Arkansas, Fayetteville, Arkansas
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Collision cross sections have been measured for electron capture into the $3\,^3S$, $3\,^1P$, $3\,^3P$, $3\,^1D$, $3\,^3D$, $4\,^1S$, $4\,^3S$, $4\,^1D$, $4\,^3D$, and $5\,^1D$ levels of helium by He^+ impact on He, for electron capture into the $3\,^1D$, $3\,^3D$, $4\,^1D$, and $4\,^3D$ levels of helium by He^+ impact on N_2, and for electron capture into the $3\,^3D$ level of helium by He^+ impact on O_2. These cross sections were obtained in an impact energy range of 20 to 120 keV.

I. INTRODUCTION

ABSOLUTE collision cross sections for electron capture into excited levels of fast helium atoms were obtained by passing a well-defined, magnetically analyzed He^+ beam from a positive-ion accelerator into a differentially pumped collision chamber containing a target gas at low pressures and then by measuring the photon emission from the resulting radiative events.

The primary process of interest here should be governed by the following differential equation for single-collision events: $dn^*/dx=-n^*/v\tau+\sigma\rho F/v$, where n^* is the number density of fast He atoms in the excited state formed by electron capture by He^+ from the target gas in the collision chamber, x is the distance measured from the beam-entrance aperture, v is the velocity of the beam particles, τ is the radiative lifetime of the excited state, σ is the cross section for capture into the state, ρ is the target gas density, and F is the He^+ flux. This equation neglects cascade (radiative transfer) as a population mechanism, and F will be approximately constant if ρx is kept sufficiently small so that beam neutralization is small. Under such conditions the solution is given by $n^*=\rho F\tau[1-\exp(-x/v\tau)]$.

At the point x, $p=n^*A$, where p is the number of emitted photons sec^{-1} cm^{-3} of wavelength λ resulting from the observed transition from the excited state and A is the Einstein coefficient for spontaneous emission of radiation of wavelength λ. Measuring the photon emission involves integrating over the volume of beam under observation. The photon emission P into a 4π solid angle is given by $P=i\rho A\tau L[1-(v\tau/L)e^{-x_0/v\tau}\times(1-e^{-L/v\tau})]$, where i is the number of ions/sec incident and L is the length of the beam segment under observation measured from x_0 to x_0+L. Measurement of P, i, ρ, L, and x_0 allows the determination of σ. Values of A and τ were obtained from the calculated transition probabilities tabulated by Gabriel and Heddle.[1]

If $L\ll v\tau$ then $P\propto(1-e^{-x_0/v\tau})$. This relationship was experimentally verified for the $4\,^1D$, $4\,^3D$, and $3\,^3D$ levels for He^+ on N_2. The collision chamber was fitted with a 25 cm long window running parallel to the beam, which allowed a point-by-point measurement of P versus x_0. The τ necessary to fit the experimental excited atom density buildup agreed with the calculated τ to within 10%. An anomaly, however, occurred in the case of the $3\,^3D$ buildup as measured from the $\lambda5876$ Å ($3\,^3D\rightarrow2\,^3P$) line. At larger x_0, ($x_0>8$ cm at 50 keV) the $3\,^3D$ buildup deviated from a simple buildup indicating that cascade from long-lived higher levels was beginning to show a contribution to the population of the $3\,^3D$ level. Final data were obtained at an x_0 of about 4 cm where the contribution from the cascade of the long-lived level was minimal.

A calibrated JaCo 500-mm Ebert spectrometer using an EMI 6095B photomultiplier provided spectral analysis. The calibration procedure has been previously described.[2]

II. RESULTS

A. He^+ Impact on N_2

Cross sections for the $3\,^3D$ level of He were obtained from measurements on the $\lambda5876$-Å ($3\,^3D\rightarrow2\,^3P$) line, for the $3\,^1D$ level by measurements on the $\lambda6678$-Å ($3\,^1D\rightarrow2\,^1P$) line, for the $4\,^3D$ level by measurements on the $\lambda4471$ Å ($4\,^3D\rightarrow2\,^3P$) line, and for the $4\,^1D$ level by measurements on the $\lambda4922$ Å ($4\,^1D\rightarrow2\,^1P$) line. The level cross sections are numerically equal to the line cross sections for the $3\,^3D$ and the $3\,^1D$, while for the $4\,^3D$ and the $4\,^1D$, the level cross sections were obtained from the line cross sections by using calculated branching ratios. Measurements were taken at pressures which at no time exceeded $3\,\mu$(Hg). In this pressure range the cross sections did not exhibit a dependence on the pressure and current within the experimental error, indicating that no collisional effects were observed other than simple charge transfer. Reproducibility was within 5% with the exception of the 20- and 120-keV data on the $4D$ cross sections where the reproducibility ($\sim$10%) suffered because of weak signals.

The capture cross sections into the $3\,^1D$,$3\,^3D$, $4\,^1D$, and $4\,^3D$ states of He from N_2 bombarded by He^+ are shown in Fig. 1 as a function of the kinetic energy of the He^+. Neither the $3\,^1D$ nor the $3\,^3D$ cross sections appear to peak over the energy range studied although they do go through points of inflection at about 40 keV. On the

† Work supported by the National Science Foundation.
* Present address: Physics Department, Louisiana State University in New Orleans, New Orleans, Louisiana.
[1] A. H. Gabriel and D. W. O. Heddle, Proc. Roy. Soc. (London) **258**, 1241 (1960).
[2] R. H. Hughes, R. C. Waring, and C. Y. Fan, Phys. Rev. **122**, 525 (1961).

other hand, the 4 ^1D function appears to peak at about 60 keV, and the 4 ^3D function peaks at about 70 keV.

It is interesting to compare the relative populations of the ^1D and ^3D levels. The ratio of the 3 ^3D population to the 3 ^1D population is a fairly constant 2.6 over the energy range investigated. The ratio of the 4 ^3D to the 4 ^1D cross sections varies from 2.2 at 20 keV to 2.8 at 100 keV and then drops to 2.7 at 120 keV. Figure 2 includes a plot of the ratio of the population of the 3 ^1D level to the population of the 4 ^1D level as a function of the energy of the He^+, and a corresponding plot for the 3 ^3D and 4 ^3D levels. Both the singlet and triplet ratios seem to asymptotically approach approximately 1.8 at high energies.

B. He^+ Impact on He

As in the case of He^+ impact on N_2, the cross sections for electron capture into the 3 ^1D, 4 ^1D, 3 ^3D, and 4 ^3D levels of helium were obtained by means of measurements on the λ6678 Å, λ4922 Å, λ5876 Å, and λ4471 Å lines, respectively. Capture cross sections into the 3 ^1P, 5 ^1D, and 3 ^3P levels of He were obtained by measurements on the λ5016 Å (3 $^1P \rightarrow 2\,^1S$), λ4388 Å (5 $^1D \rightarrow 2\,^1P$), and λ3889 Å (3 $^3P \rightarrow 2\,^3S$) lines, respectively. Measurements were taken observing the beam at 30° to make use of the Doppler shift in order to separate the radiation of the fast atoms from radiation from the target gas. Pressures less than 3 μ were used. All cross sections were independent of pressure within experimental error. Figure 3 displays the cross sections for these levels.

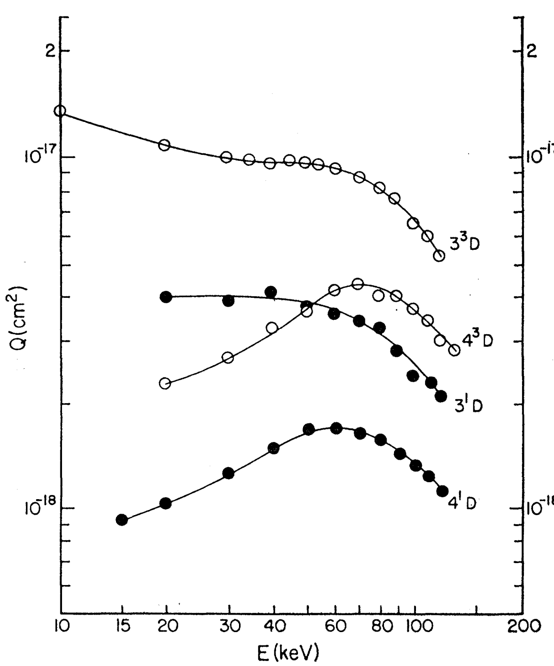

FIG. 1. Cross sections for electron capture into the 3^3D, 4^3D, 3 ^1D, and 4 ^1D levels of helium by He^+ on N_2.

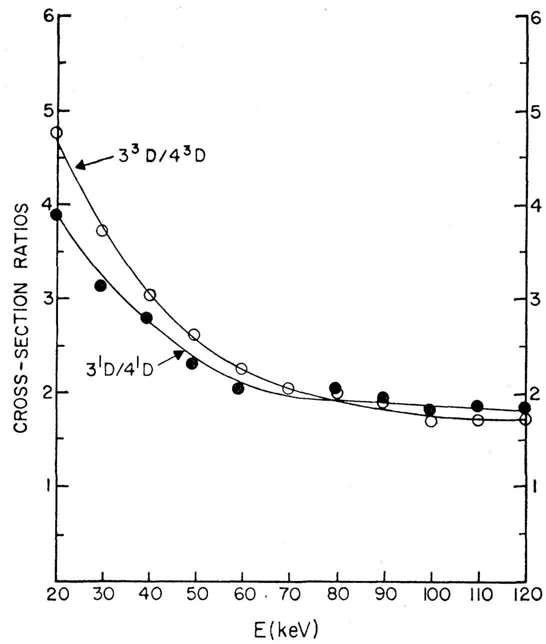

FIG. 2. Ratios of 3*D* to 4*D* capture cross sections as a function of He^+ kinetic energy for He^+ on N_2.

Measurements were also made on the 3 ^3S, 4 ^3S, and 4 ^1S levels via the λ7065 Å (3 $^3S \rightarrow 2\,^3P$), λ4713 Å (4 $^3S \rightarrow 2\,^3P$), and λ5047 Å (4 $^1S \rightarrow 2\,^1P$) lines. Meas-

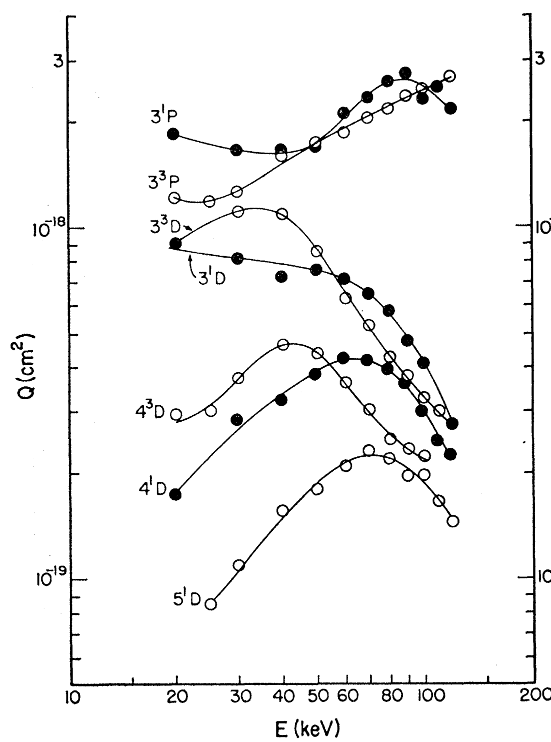

FIG. 3. Cross sections for electron capture into the 3 ^1P, 3 ^3P, 3 ^3D, 3 ^1D, 4 ^3D, 4 ^1D, and 5 ^1D levels by He^+ on He.

urements on the λ4713 and λ5047 Å lines were carried out over a limited energy range because it was possible to Doppler shift the lines into other He lines present in the spectra from the collision chamber. Measurements were taken on the λ4713- and λ5047-Å lines using a different technique. A collision chamber about 5 cm long with $\frac{1}{16}$ in.-diam. apertures at either end was filled with helium. The collision chamber was differentially pumped at both ends. The He^+ beam was allowed to enter the collision chamber and to depart into an evacuated observation region. The excited-atom density outside the collision chamber is given by $n^* = F\sigma\rho\tau(1-e^{-L/v\tau})e^{-x/v\tau}$, where L is the length of the collision chamber and x is the distance from the beam-departure aperture. Making observations in a vacuum has the advantage of getting rid of the spectra produced by the excited target gas. Unfortunately, the emission was too weak to work at sufficiently low pressures where the apparent cross section is independent of the pressure.

The cross sections for the $3\,^3S$, $4\,^1S$, and $4\,^3S$ levels appeared to be linear with pressure, whether measured in the conventional way or by the method just described, and the values reported are the extrapolated values for zero pressure. This procedure adds considerably more uncertainty to the reported cross sections for these levels, a fact which should be kept in mind. The fact that the cross sections agreed whenever both methods could be used gives support to the unconventional method. The $3\,^3S$, $4\,^3S$, and $4\,^1S$ level cross sections are displayed in Fig. 4.

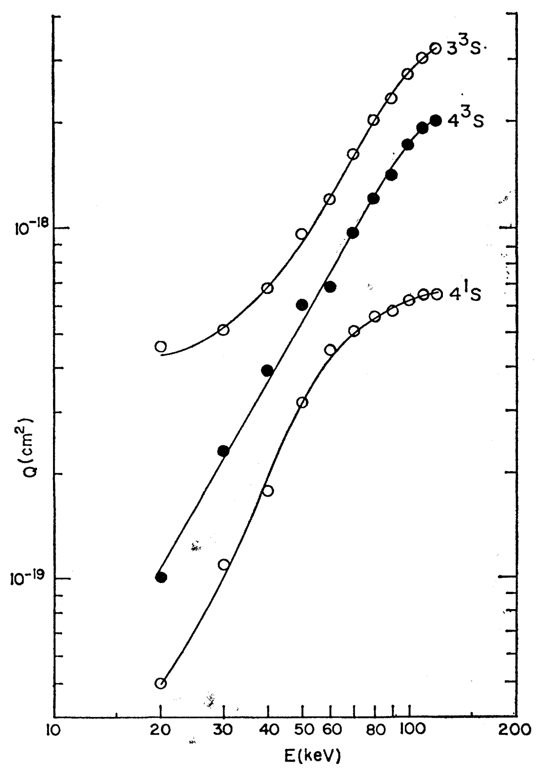

FIG. 4. Cross sections for electron capture into the $3\,^3S$, $4\,^3S$, and $4\,^1S$, of helium by He^+ on He.

C. He^+ Impact on O_2

Cross sections for capture into only one level ($3\,^3D$) of helium were obtained for He^+ impact on O_2. These are displayed in Fig. 5.

III. DISCUSSION

If the capture cross section primarily depends on the orbital angular momentum of the state, then one might expect the ratio of the triplet-to-singlet capture cross section to be roughly 3 to 1 for the same principal quantum number and the same orbital quantum number. Indeed this seems to be nearly borne out in the D states for He^+ on N_2. The ratio is about 2.7 to 1 at the higher energies. In the case of He^+ on He, the triplet P and D state cross sections are strikingly similar to those of the corresponding singlet P and D states. The ratio of the $4\,^3S$ to the $4\,^1S$ cross sections appears to approach 3 to 1 at the higher energies, but the accuracy of the S-state data is limited. In order to determine whether a preferential population of certain magnetic substates was contributing to the results because of momentum conservation laws in the collisions, the polarization of the radiation was checked. No polarization of any of the radiation was observed that could not be attributed to polarization effects caused by window reflections.

An attempt was made to find a simple relationship between the cross section and the effective principal quantum number: $n_{\text{eff}} = (R/T)^{1/2}$, where R is the Rydberg constant and T is the term value. In the case of

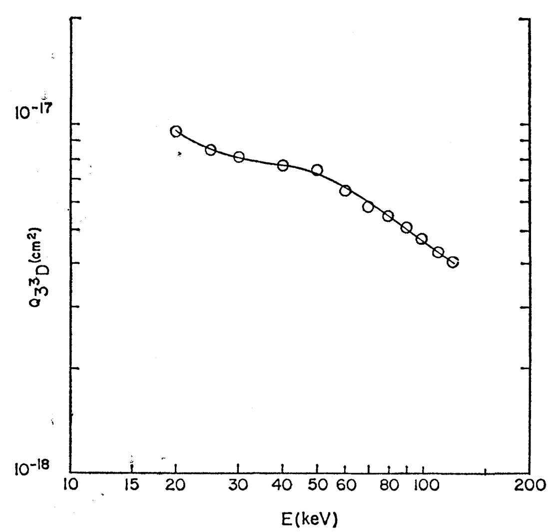

FIG. 5. Cross sections for electron capture into the $3\,^3D$ level of helium by He^+ on O_2.

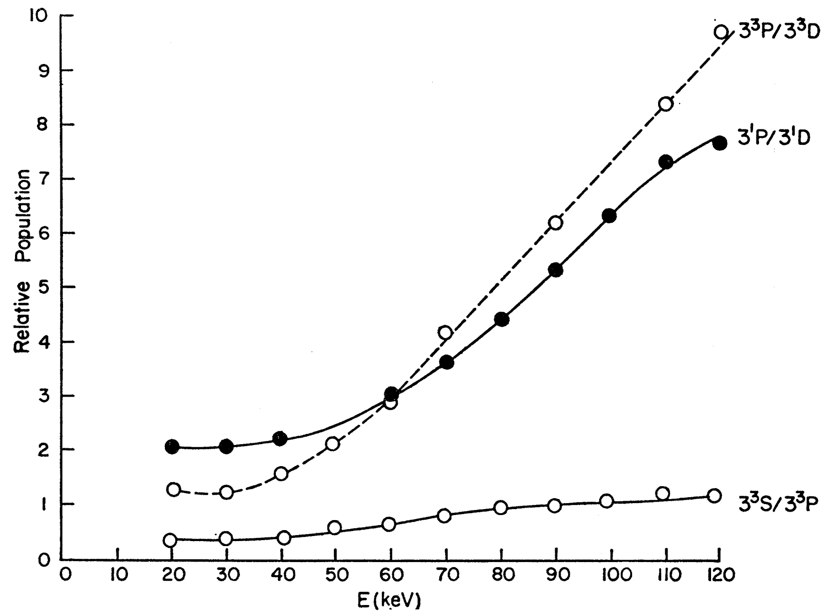

FIG. 6. Relative populations for certain $n=3$ levels of helium by He^+ impact on He.

He^+ on N_2, the cross-section ratio for $3D$ to $4D$ capture approaches 1.8 at the higher energy in both singlet and triplet systems. This is very near the value 1.78, which is obtained by using an n_{eff}^{-2} dependence. It is not evident *a priori* that such a simple relationship should exist.

No simple dependence on n_{eff} was found for He^+ on He. The well-known n^{-3} dependence in S-state capture found in capture by H^+ impact[3] does not seem to apply in the case of 4 ^3S to 3 ^3S capture in this energy range. The ratio of 3 ^3S to 4 ^3S capture seems to approach 1.6 rather than the 2.6 required for an n_{eff}^{-3} dependence. The accuracy of the S measurements is limited but it should be also pointed out that the n^{-3} dependence was derived by using the Born approximation which is not valid in this velocity range.

It appears that at high energies the levels with low angular-momentum quantum numbers become more heavily populated. This may not be surprising since the Born approximation for H^+ impact generally predicts this.[4] Figure 6 compares the ^3S, ^3P, ^3D, ^1D, ^1P cross sections for $n=3$ as a function of energy for He^+ on He.

He^+ impact on N_2 and O_2 has some interest in the investigation of the aurora. In particular if He^+ is an appreciable constituent of the auroral stream, its presence could be best detected by the presence of the λ5876 Å line.[5] It appears that the λ5876 Å line cross sections are of the order of 10^{-17} cm^2 over a large energy range. These are quite large cross sections, relatively speaking, and are comparable to estimated cross sections for the production of H_α emission from H^+ on N_2 in this same energy region.[6] There is almost no background around the λ5876 Å region for impact on N_2 and very little for impact on O_2 when the spectral window does not exceed 10 Å. With our apparatus the λ5876 Å emission is quite strong. It would appear, therefore, that if the only mechanisms operative in the production of the aurora were charge transfer and direct excitation, and if apparatus with sufficient resolution and response were available, then the λ5876 Å line should be detectable in the aurora if an appreciable number of He^+ particles are present. These assumptions are quite naive, however. For a fairly complete discussion of the aurora and the airglow, see Chamberlain.[7]

Although the cross sections presented here are generally good relative to one another (within 5%), the absolute error is considerably larger. It is almost an impossible task to ascertain some of the systematic errors which may be introduced in the course of data measurements. In addition to the usual errors encountered in measuring excitation cross sections, in this investigation added uncertainty is introduced into the results because of the small uncertainty in the value of the distance x the ion beam has traversed through the target gas before it enters the observation region. The usual errors arise through such things as calibration, current fluctuations, pressure fluctuations, optical alignment, and signal noise. Signals were weak for all except the λ5876- and λ6678-Å lines, and at the energy extremes the signal-to-noise ratio sometimes dropped as low as three to one. The measurements on the signals were

[3] J. D. Jackson and H. Schiff, Phys. Rev. **89**, 359 (1953).
[4] R. A. Mapleton, Phys. Rev. **122**, 528 (1961).
[5] C. Y. Fan, Phys. Rev. **103**, 1740 (1956).
[6] J. L. Philpot, Ph.D. thesis, University of Arkansas, 1965 (unpublished).
[7] Joseph W. Chamberlain, *Physics of the Aurora and Airglow* (Academic Press Inc., New York, 1961).

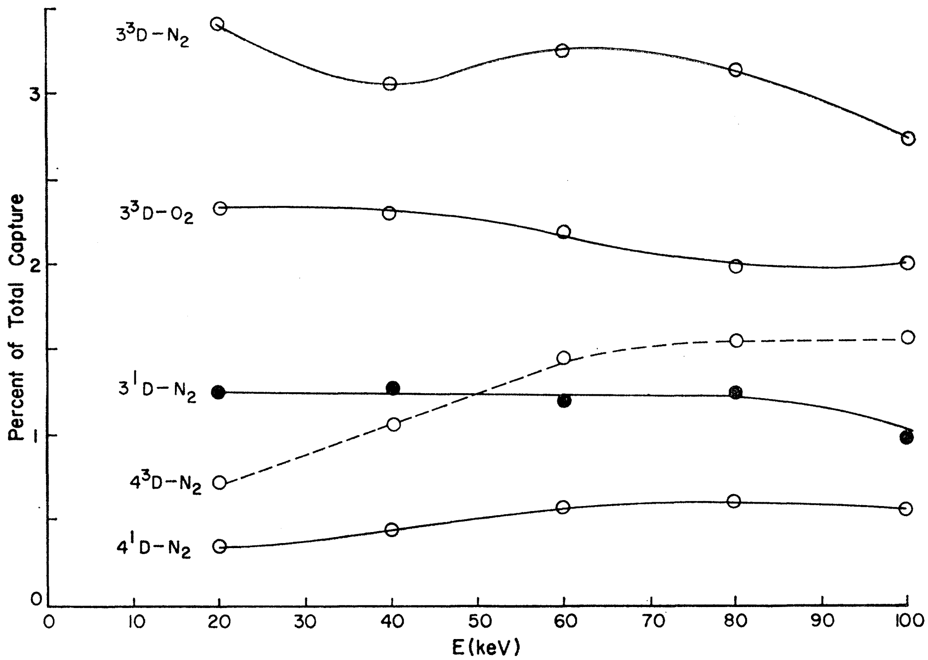

FIG. 7. Percent of total capture into certain D levels of helium by He^+ impact on N_2 and O_2.

better than this signal-to-noise ratio might indicate, for the measurements represent average signals. It is, however, probably wise to estimate the absolute uncertainty in the measurements to be no better than 50%.

There apparently are no published cross sections for capture into excited helium levels from He, N_2, or O_2 gases bombarded by He^+ beams with which to compare these measurements. Experimental values for the total-charge-transfer cross sections, however, have been measured by several investigators and are tabulated by Allison and Garcia-Munoz.[8] Figures 7 and 8 display the percentages of the total capture for various levels and target gases. The relative amount of capture into the excited states increases as the energy increases as shown in Fig. 9.

Perhaps a word regarding cascading to the $3\ {}^3D$ level is in order. An attempt to identify the level(s) involved was made by measuring the decay of the $3\ {}^3D$ state atoms leaving a 12 cm long collision chamber into an evacuated observation chamber. The excited atom density will decay according to $\exp(-x/v\tau)$, where x is now the distance measured from the beam departure aperture. The λ5876 Å decay could be fitted with a two-mode decay: one having a time decay constant of 15 nsec and one having a decay constant of 140 nsec. The shorter decay constant corresponds to the natural life-

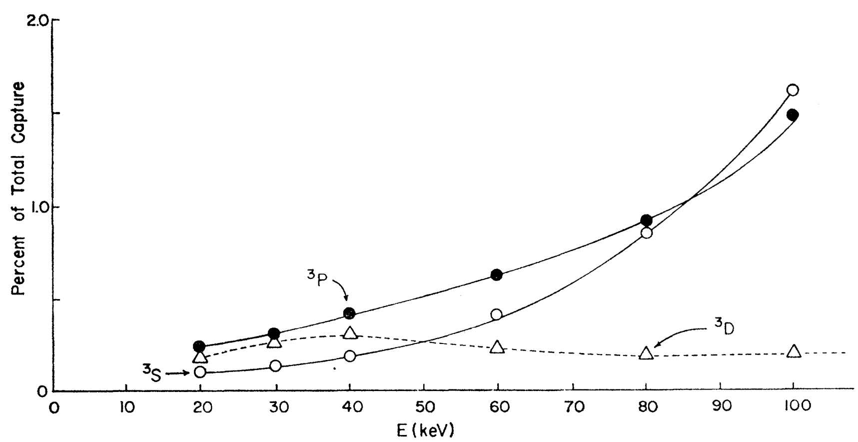

FIG. 8. Percent of total capture into the $n=3$ triplet levels of helium by He^+ impact on He.

[8] Samuel K. Allison and M. Garcia-Munoz, in *Atomic and Molecular Processes*, edited by D. R. Bates (Academic Press Inc., New York, 1962), pp. 756–766. (We used the data tabulated from the work of Stier and Barnett.)

time of the $3\,^3D$ and represents the decay of the $3\,^3D$ level populated by direct capture. The other time constant may be ascribed to a higher level cascading to the $3\,^3D$ level.

Our analysis of the decay curves indicates that if equilibrium were reached the cascading portion would contribute roughly, about $\frac{1}{3}$ to the total $3\,^3D \rightarrow 2\,^3P$ cross section for 50-keV impact on either He or N_2. If a single-level cascade is assumed, then two nearby levels are brought to mind: the $5\,^3F$ level and the $4\,^3P$ level, both of which have lifetimes of around 140 nsec. No 72 nsec $4\,^3F$ decay was observed in $3\,^3D$ decay and only the decay from the directly excited $4\,^3D$ atoms was observed in the $4\,^3D \rightarrow 2\,^3P$ transition, which would seem to rule out F-state decay. On the other hand, the $4\,^3P$ branching ratio does not particularly favor cascade to the $3\,^3D$ level with only 9% of the radiative transfer going to the $3\,^3D$. The amount of cascade is much too large to be consistent with the $4\,^3P$ capture cross section.[9] Thus, given the two choices we are forced to conclude that there may be an appreciable $5\,^3F$ excitation in the capture process. This time constant (140 nsec) also appears in the $3\,^3D$ decay when He is excited by electrom impact.[10] It could be that the $5\,F$ decay in the $4\,^3D$ decay was simply missed in this experiment. The technique yielded 33 and 36 nsec for the lifetimes of the $4\,^3D$ and $4\,^1D$ levels, respectively, which is in agreement with theory and with other experiments.

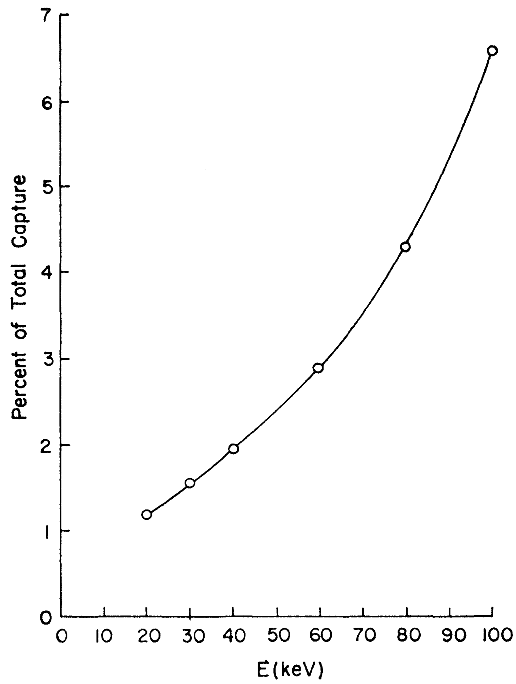

FIG. 9. Percent of total capture into the $3\,^3S$, $3\,^1P$, $3\,^3P$, $3\,^1D$, $3\,^3D$, $4\,^1S$, $4\,^3S$, $4\,^1D$, $4\,^3D$, and $5\,^1D$, levels of He for He^+ impact on He.

[9] The $4\,^3P$ cross section has been measured by F. J. deHeer and his colleagues (Amsterdam). We appreciate Dr. deHeer's giving us their preliminary measurements.

[10] W. R. Pendleton, Jr., and R. H. Hughes, Phys. Rev. 138, A683 (1965).