Formation of He($3\,^3D$) by electron capture in collisions of He^+ with various polyatomic molecules

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Electron capture into the $3\,^3D$ state of He was measured by the detection of 5876-Å photons from the $3\,^3D-2\,^3P$ transition following collisions of He^+ with various polyatomic molecules. The photon-emission cross section for this transition was measured for each target over the helium-ion energy range of 100 to 300 keV. The targets used were H_2, N_2, O_2, CO, CO_2, N_2O, CH_4, C_2H_2, C_2H_4, and C_2H_6. For all targets, the cross sections could be fitted very well with a $1/v^2$ function, where v is the helium-ion velocity.

INTRODUCTION

The study of electron capture in ion-molecule collisions in the intermediate energy range has appropriately been centered on the study of the $H^+ + H_2$ system.[1] This was first investigated theoretically by Tuan and Gerjuoy for capture to the ground state using the Born approximation.[2] The initial work of Tuan and Gerjuoy has been refined and extended by Ray and Saha,[3,4] and by Band[5] to capture into various *nlm* states. More recently, Deb, Jain, and McGuire looked at the theory of Tuan and Gerjuoy for cases of fixed orientation of the molecular axis.[6] Sural and Sil also looked at this collision process using an impact-parameter calculation.[7]

An electron-capture collision involves a rearrangement. As such, the use of the Born approximation is not rigorously justified. However, the above-cited works have been notably successful in fitting the experimental data. Due to the complexity of the integrals involved, a closed-form expression for the capture cross section is not possible, but some general statements regarding the theory can be made. First, because of interference effects between the two atomic centers the molecular system cannot be treated as the sum of two atoms; i.e., Bragg additivity with regard to the capture cross section is not valid. This has been shown to be true with more complicated targets also, most recently by Varghese *et al.*[8] Second, the variation of the cross section with projectile velocity is a v^{-12} function in the high-energy limit ($E > 400$ keV). However, there is some disagreement in the lower-energy region which is of interest for the work presented here. Tuan and Gerjuoy's work shows a v^{-12} dependence over the entire range. Johnson shows that using an interaction potential of the form of e^{-R} the cross section in the lower energy region approaches a v^{-2} function.[9] The work of Ray and Saha and Band show a dependence somewhere between v^{-12} and v^{-2}. As is usual in impact parameter calculations, the results of Sural and Sil show a v^{-2} dependence. Third, except in the impact parameter calculation, the theory shows that capture is dominated by capture from gerade states of the target with the ungerade states contributing at most 10% to the cross section, and then only at the higher projectile energies. This effect can be also understood in terms of the interference effects of the wave functions involved. As both Tuan and Gerjuoy, and Ray and Saha, show, the interference is primarily destructive for the ungerade states and constructive for the gerade states for the collision energies of this work. The interference of the ungerade states becomes constructive at collision energies near 900 keV.

As there are no theoretical calculations, and very little data, dealing with electron capture to excited states in more complicated ion-molecule collisions, the work presented here will provide a basis to determine if the general structure of the theory can be extended to more complicated targets and projectiles. This will focus on two aspects; that of the velocity dependence of the cross section and the effect of changing the molecular target used. While direct comparison to theory developed for proton-H_2 collisions is impossible, an attempt is made to use targets that are linear, symmetric molecules, and thus share some similar structural qualities with H_2.

Electron capture into the $3\,^3D$ state of He was measured by the detection of 5876-Å photons from the $3\,^3D-2\,^3P$ transition following collisions of He^+ with various molecular targets. The targets used were symmetrical linear molecules with the exceptions of CO, N_2O, and CH_4. These three molecules were also studied simply to add to the information available on ion-molecule collisions. A He^+ beam was used for several reasons. First, with the hydrocarbons and H_2 as targets there would be significant Balmer emission from the target. The experimental arrangement did not allow for easy separation of target and projectile emission. Second, a helium projectile in the energy range of the accelerator used has velocities that are on the order of an atomic unit where disagreement in the theories is most prominent. Third, very little work has been done with a He^+ projectile on more complicated targets. As far as the author is aware, there has been only one other experimental study of photon emission following electron capture by He^+ impact on molecular targets in this approximate energy range. This is a paper by Head and Hughes which looked at capture from He, N_2, and O_2, in the 20–120-keV range.[10]

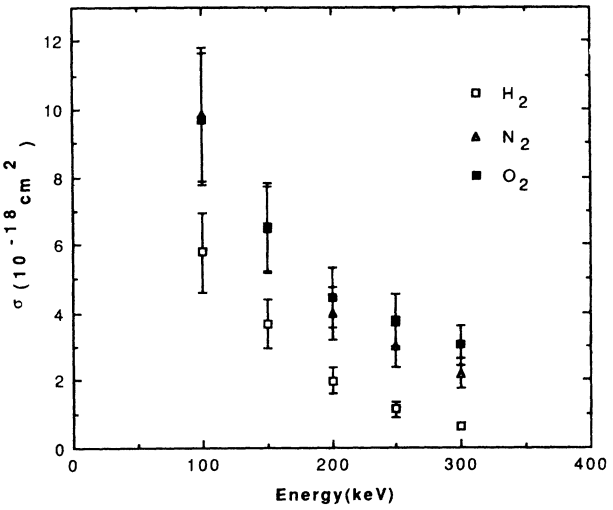

FIG. 1. Photon-emission cross sections as a function of helium beam energy for H_2, N_2, and O_2 targets.

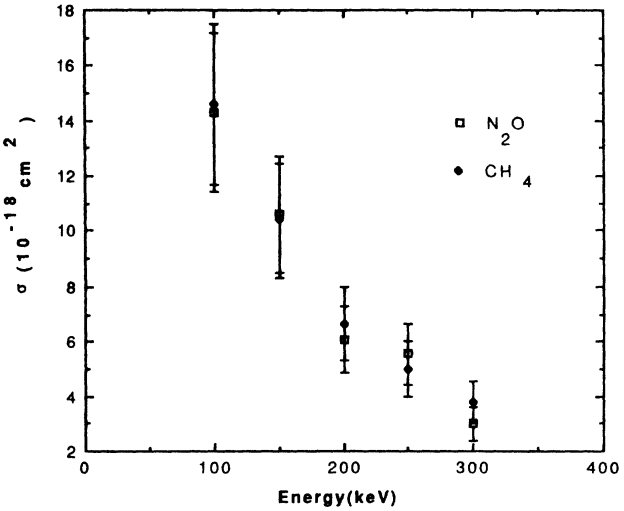

FIG. 3. Photon-emission cross sections as a function of helium beam energy for N_2O and CH_4 targets.

EXPERIMENTAL PROCEDURE

The experimental system has been described in detail in previous publications.[11,12] Briefly, a He^+ beam was obtained from a Van de Graaff accelerator. The beam entered a differentially pumped target chamber after being collimated to a diameter of approximately 1 mm. After passing through a target gas cell the beam was collected by a Faraday cup. Photons emitted at 90° to the beam direction were collected by a lens system and then analyzed by a monochromator. The photons were detected by a cooled photomultiplier tube. The monochromator was set on the 5876-Å line. The resolution of the monochromator was greater than the linewidth so all emitted photons from that line were counted. The photon detection system was calibrated with respect to its absolute sensitivity by standard techniques, and correction made for polarization and anisotropy effects.[11,12]

Target gas pressure was measured by a capacitance manometer. Single-collision conditions were maintained by using a target gas pressure of 3 mTorr. Previous work at this laboratory showed that the cross sections were linear with respect to target gas pressure up to 5 mTorr. The integrated beam current was corrected for neutralization in the target region for each of the targets and at all beam energies. This correction varied from approximately 50% at 100 keV to about 5% at 300 keV; the exact values depended on the target being used.

The estimated error due to experimental systematics in all the absolute photon emission cross sections reported here is 20%. Approximately three quarters of the error is due to measurement of the sensitivity of the optical system, while one quarter is due to statistical uncertainty and error from measurements of the target pressure, temperature, and beam current. No correction was made for cascade contributions to the cross sections. In the previ-

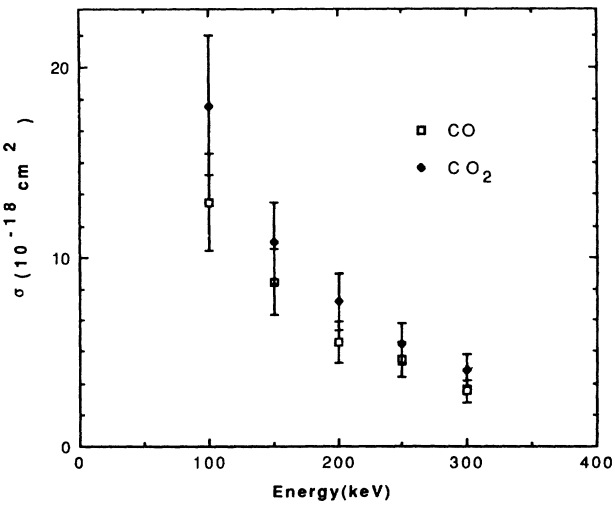

FIG. 2. Photon-emission cross sections as a function of helium beam energy for CO and CO_2 targets.

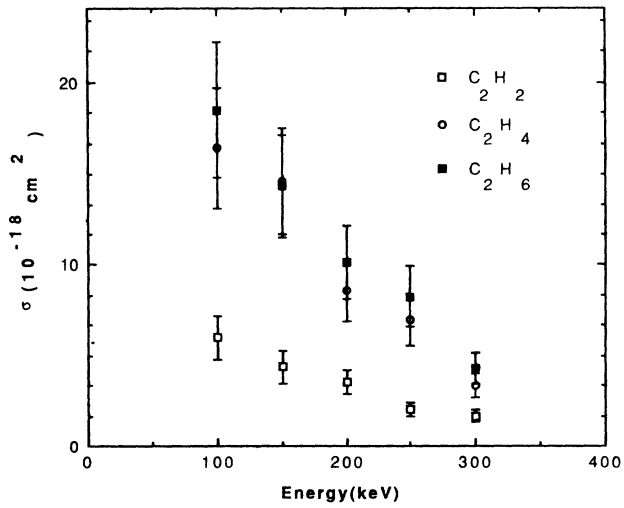

FIG. 4. Photon-emission cross sections as a function of helium beam energy for hydrocarbon targets.

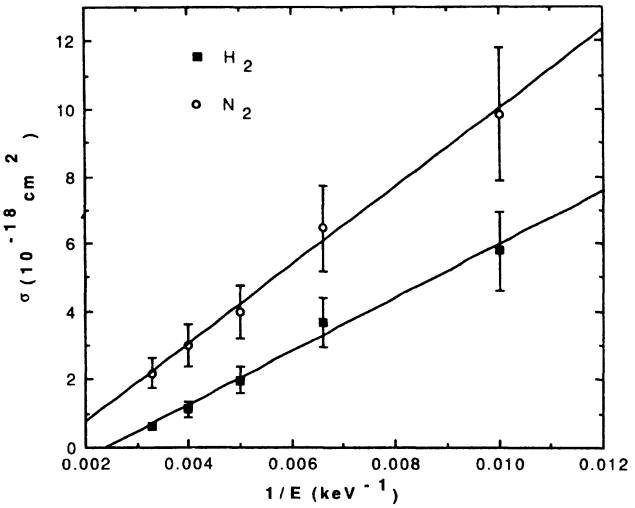

FIG. 5. Photon-emission cross sections as a function of inverse beam energy for H_2 and N_2 targets.

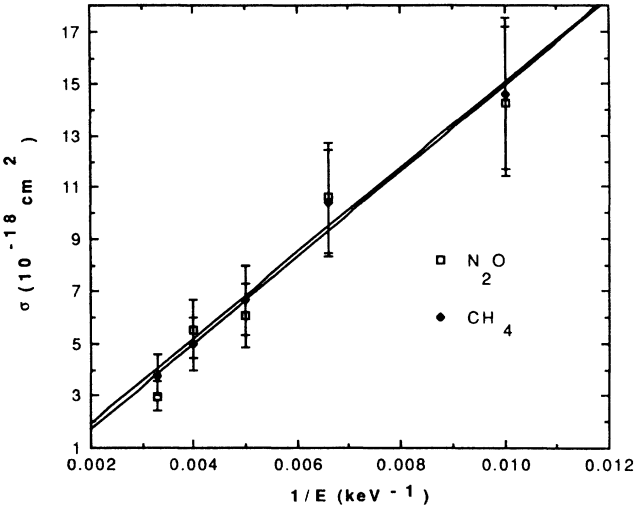

FIG. 7. Photon emission cross sections as a function of inverse beam energy for N_2O and CH_4 targets.

ous work by Head and Hughes it was shown that the major contribution to the population of the $3\,^3D$ state by cascading was from the $5\,^3F$ level. Their work showed that about 30% of the $3\,^3D$-$2\,^3P$ radiation was due to cascading for a beam energy of 50 keV. For the higher beam energies used in this work it is assumed that the contribution would be less than at 50 keV. It was not possible to directly measure the contribution from the $5\,^3F$ cascade as the wavelength of this transition was well outside the range of the instrumentation. Thus, the overall error in the absolute cross sections would be close to 50%, but the error as relative cross sections is 5%.

RESULTS AND DISCUSSION

The absolute photon-emission cross sections for the various target molecules as a function of He^+ energy are shown in Figs. 1–4. A comparison to the work of Head and Hughes can be made at 100 keV with the N_2 and O_2 targets. The cross section in the present study is about 9.7×10^{-18} cm^2 for both molecules. The values obtained by Head and Hughes are 5.1×10^{-18} cm^2 N_2 and 4.6×10^{-18} cm^2 for O_2. The differences are reasonable given different techniques of calibration of the optical systems. Also, the previous workers did not correct for any anisotropy of the radiation, or for beam neutralization.

To determine the velocity dependence of the cross sections the data was plotted as a function of the inverse of the beam energy. This is shown in Figs. 5–8. As can be seen, the data could be fitted extremely well with a linear function. The best fit lines were determined by the least-squares method. The correlation coefficient (r^2) for the fits varied from a minimum of 0.916 for the ethylene tar-

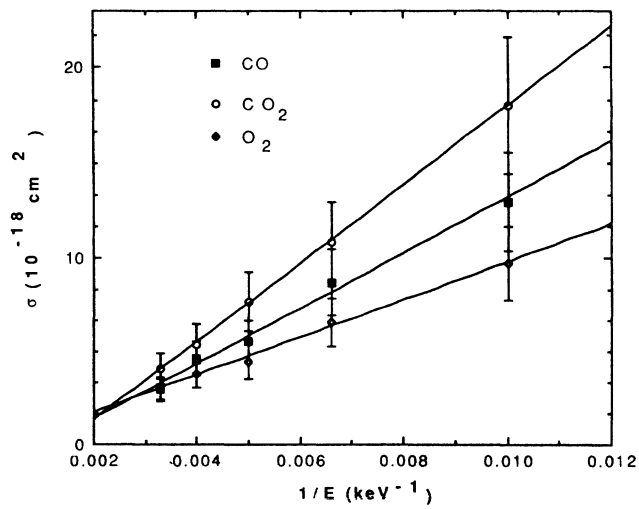

FIG. 6. Photon-emission cross sections as a function of inverse beam energy for CO, CO_2, and O_2 targets.

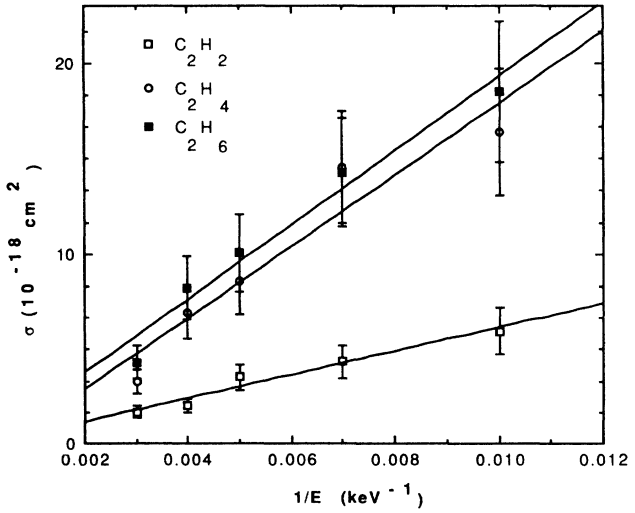

FIG. 8. Photon emission cross sections as a function of inverse beam energy for hydrocarbon targets.

get to a maximum of 1.00 for the carbon dioxide target. Thus, it appears that the cross section varies as v^{-2} in this projectile energy range.

A search was made to determine if there was any general trend in the cross-section data with regard to the target species used. The first correlation tested was that of the dependence of the cross section on the total number of electrons in the target molecule. This showed a general trend of increasing cross section with the number of electrons, but there was a large amount of scatter in the data. A best-fit line had a correlation coefficient of only 0.79. A plot of the cross sections versus the number of valence electrons also showed little correlation ($r^2=0.56$).

CONCLUSION

The absolute photon emission cross section for the 5876-Å line of He was measured following collisions of He^+ with various molecular targets. The cross section showed a v^{-2} dependence with regard to the incoming ion velocity. This is consistent with impact parameter calculations, and also with Born theory depending on the interaction potential that is used.

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