Crossed Beam Studies of Low Energy He^+ Ions by He Atoms[†]

TATSUO ARIKAWA*

(Received 6 June 1973)

Differential scattering of He^+ ions by He atoms in the energy range of 10 to 100 eV over the scattering angle from 2° to 20° has been measured using the crossed beam apparatus. Prominent oscillatory structures in the differential cross section resulting from the interference between waves scattered from the lowest gerade $^3\Sigma_u^+$ and ungerade $^3\Sigma_u^+$ states of a pseudo-molecular ion He_2^+ were observed. The rainbow peak due to the elastic scattering of He^+ by He was observed superimposingly upon these oscillatory structures due to the resonant electron capture. The relative differential cross sections were normalized to the absolute data of Lorents and Aberth.

1. Introduction

The measurement of the resonant electron capture on the He^+ -He scattering was first carried out by Ziemba and Everhart¹⁾, who obtained the electron capture probability for a fixed scattering angle of 5° as a function of the incident ion energy. Later Lockwood, Helbig and Everhart²⁾ studied this probability in great detail in the incident ion energy of 0.4 to 250 keV as a function of the scattering angle from 0.4° to 4.4° . These earlier studies^{3), 4)}, however, were confined to a determination of the dependence of the electron capture probability on both velocity and the impact parameter.

Recently, Lorents and Aberth⁵⁾ carried out the measurement of the differential cross section with the same system over a wide angular range from 1° to 36° at relative energies of 20 to 600 eV. In their experiment, however, mass analysis of the incident ion beam was not attempted. Ions produced in the ion source were introduced into the scattering cell where the He target gas was contained. Ions scattered here were collimated by an angle-defining slit grooved on the surface of the cylinder

concentric with the scattering cell, and the determination of the scattering angle was made by rotating this slit around the scattering cell.

In the present experiment measurements of the differential cross sections in rather low energy region were carried out using the crossed beam apparatus⁶⁾, where the mass analyzed primary He^+ ions were crossed at right angles with the secondary beam of the neutral He atoms inside a high vacuum chamber. By this method the collision point was positioned definitely and the reaction products were unlikely to be subjected to further collisions except at the intersecting region, so that the energy and the angular distributions of the collision product were obtained without multiple collisions.

Differential cross sections were measured in the energy range of 10 to 100 eV over the scattering angle of 1° to 20° . These relative cross sections were normalized to the data of Lorents and Aberth⁵⁾. The angular positions of the maxima and minima of the differential cross section were fairly in good agreement with those of Lorents and Aberth. The peak

* Department of Physics, Faculty of Science, University of Tokyo, Bunkyo-ku, Hongo, Tokyo

† Supported in part by the Kurata Research Foundation.

due to the rainbow scattering was also observed in the differential cross section superimposed upon the smooth oscillations. The depth of the intermolecular potential of He_2^+ was estimated from the angular position of the rainbow peak assuming the 12-6-4 potential used by Mason and Schamp⁷⁾ and others⁸⁻¹⁰⁾. Subsidiary oscillations due to the nuclear interchange were not observed under these experimental conditions. This effect is expected at larger scattering angles and at much higher incident energies⁹⁾.

A brief description of the experimental apparatus in association with the experimental procedures will be given in the next section. The experimental results and some discussions will be presented in the last section.

2. Experimental Apparatus

The crossed beam apparatus used in this experiment is shown schematically in Fig. 1. The apparatus mainly consists of an ion beam source, a neutral beam source, a detection system and measuring equipment.

Beam ions were produced in the ion source^{12), 13)} which was composed of an electron bombardment ionizer, an rf quadrupole mass filter¹⁴⁾ and a deceleration lens^{15), 16)}. Transport lenses and beam steering half-plates were placed in front and back of the rf mass filter

to maximize the transmission current into the subsequent deceleration lens. The ion source provides a mass selected ion beam of energies from about 100 to 1 eV or less. The beam intensity ranged from 10^{-9} A at a high energy to 10^{-12} A at a low energy. A more detailed description of the ion source is presented in the previous paper¹⁷⁾.

The primary ion beam emerging from the final slit of the ion source crossed at a right angle to the secondary neutral beam from a multicapillary nozzle¹⁸⁾. The intensity of the neutral beam at the intersecting point was estimated to be 5×10^{13} atoms·cm⁻³ from a separate experiment made with the same geometry as used in the present experiment. Typical widths of both ion and neutral beams at the intersecting point were 0.3 mm (FWHM) for the ion beam and 1 mm (FWHM) for the neutral beam, respectively.

Both ion and neutral beam chambers were connected to the side ports installed at right angles to the main chamber, 500 mm in diameter and 200 mm in height, which was evacuated separately from both beam chambers by using an ion pump and a titanium sublimation pump. Typical operation pressures in the main chamber were of the order of 10^{-9} Torr without gas load and about 10^{-7} Torr in the presence of both beams.

Scattered ions from the scattering point entered into the detection system through angle-defining slits. An energy analyzer of a stopping potential type was located 5 mm behind the slits. After passing through the analyzer ions were focused onto the first dynode of a 16-stage electron multiplier. The entire detection assembly was mounted on an alignment plate inside the main chamber and can be rotated about the scattering point from -10° to $+100^\circ$ with respect to the direction of the incident ion beam. The true zero angle was determined from the maximum of the plot of the incident

Fig. 1. Schematic diagram of the apparatus.

current as a function of the angle scale by scanning the detector in both positive and negative angle regions in each time to change the incident energy of the ion beam. All scattering angles were measured from this true zero angle. The speed of the angular scanning of the detector was designed to be $0.1^\circ \cdot \text{sec}^{-1}$ in normal operation, and the alternation of the speed was made by changing the combination of gear ratios. When the intensity of the scattered ions was not sufficient in the normal scanning mode, the detector was first fixed at a certain angular position and the accumulation of signals was attempted to reduce the statistical errors of the data for a time interval of 10 to 60 sec according to the intensity of the scattered ions. The position of the detector was then advanced by a small amount successively to sweep out the entire angular range.

Pulses from the electron multiplier were fed into a charge sensitive preamplifier which served for the reduction of the stray capacitance of the cable from the electron multiplier to the input of the preamplifier. Output signals delivered from the preamplifier were further amplified and shaped to a convenient form for the subsequent discriminator which was followed by an electronic counter and a digital recorder.

All electrical connections of the detection system were made through vacuum-tight connectors of a manifold chamber which was attached to the bottom of the main chamber. A detailed description of the apparatus will be published shortly.

3. Experimental Results and Discussion

Typical experimental results of the differential cross section measured on the system of He^+ on He are shown in Fig. 2 for three different energies of incident He^+ ions. The curves show prominent oscillatory structures superimposed on monotonically decreasing curves.

In such collisions between two identical particles, the electron may be thought of as oscillating back and forth between two nuclei to form a pseudo-molecular ion He_2^+ during the collision. When the incident energy of He^+ ions is not too high, the wave function of this molecular ion may be described well by two components, one is gerade and the other is ungerade. Oscillatory structures of the differential cross section arise from the interference between the waves scattered from these two potentials of molecular ions¹⁹⁾.

In this experiment the primary ion beam was expected to contain almost the ground $1^2\text{S}_{1/2}$ state alone, since most of the experiments were carried out with the energy of bombarding electrons below the threshold of producing the ion metastable $2^2\text{S}_{1/2}$ state as far as the intensity of scattered ions permitted. However, when the intensity was not sufficient, the bombarding energy was often raised to above the threshold to increase the intensity of the primary ion beam. In these cases the metastable ions can exist to a considerable extent in the production of ions. However, the metastable state $2^2\text{S}_{1/2}$ is situated very close to the excited state $2^2\text{P}_{1/2}$ whose energy differs from the energy of the metastable state only by $5.8 \times 10^{-5} \text{ eV}^{20)}$, and consequently the Stark mixing between two states will easily quench the metastable ions to the ground state by the electric field applied to the electrodes inside the ion source. From the calculation based on sudden approximation, the contribution of the metastable state to the incident beam was estimated to be less than 1 % compared with the ground state. On the other hand, the secondary beam thermally effused from the multicapillary nozzle is thought to be the electronic ground state 1^1S_0 . As these two particles, $\text{He}^+(1s)$ and $\text{He}(1s^2)$, come together, they are combined to form the molecular ion He_2^+ whose electronic configurations are given respectively by $\sigma_g(\sigma_u)^2$ and $(\sigma_g)^2\sigma_u$, i. e. the

Fig. 2 (a). Typical differential cross sections of He^+ ions by He atoms versus laboratory scattering angles for the incident ion energy $E=50$ eV. The relative cross sections ($\bullet$) are fitted to the absolute data of Lorents and Aberth (----), and the rainbow maximum lies very close to the valley of the oscillation ($n=7$) and no prominent rainbow maximum is observed (see also Fig. 3 (b)).

lowest gerade ($^2\Sigma_g^+$) and ungerade ($^2\Sigma_u^+$) states of He_2^+ . The oscillations which appear in the cross section result from the interference between the waves scattered from these two potentials.

The relative differential cross sections were normalized to the absolute ones obtained by Lorents and Aberth⁹⁾. As can be seen from Fig. 2, the positions of the peaks and valleys of the oscillation are slightly shifted in comparison with those of Lorents and Aberth. This discrepancy would be primarily due to the slight difference of the incident ion energies which

were measured in this experiment by the stopping potential energy analyzer. The determination of energy by such an analyzer is not a reliable procedure in a precise measurement.

The peaks in the electron capture probability obtained by Ziemba and Everhart¹⁰⁾ correspond to the valleys of the present ion scattering cross section. However, as pointed out by Lorents and Aberth, an exact agreement between these two sets of results would not be expected, since the envelope of the scattering cross section is a steeply decreasing function of the scattering angle. Consequently

Fig. 2 (b). Typical differential cross sections of He^+ ions by He atoms versus laboratory scattering angles for the incident ion energy $E=30$ eV. The relative cross sections ($\bullet$) are fitted to the absolute data of Lorents and Aberth (----), and the rainbow maximum coincides with that of the oscillation ($n=9 \frac{1}{2}$) and the peak is emphasized (see also Fig. 3 (a)).

the angles at which the maxima occur are shifted toward angles smaller than the actual peaks of the electron capture probability and in the same way the minima toward larger angles. These phenomena of oscillations of the cross section were well interpreted by Marchi and Smith²¹.

The rainbow maximum²² was observed superimposingly on the oscillatory structure. It should be noted here, however, that the real rainbow maximum is shifted slightly owing to these oscillations. An attempt to estimate the depth of the potential well for the molecular

ion He_2^+ was made by separating the real rainbow peak from the oscillatory structure. The shifted amount of the angular position at which the rainbow maximum occurs was estimated to be at most 3 % or less. The 12-6-4 potential was employed to derive the well depth. At an intermediate distance the term of the charge-induced dipole interaction is expected to dominate over the other two terms, which means the force parameter γ of the potential to be zero. The resulting well depth is 1.9 eV, the value of which is 18 % smaller than the well depth, 2.3 eV, of Reagen, Browne and Matsen²³

Fig. 2 (c). Typical differential cross sections of He^+ ions by He atoms versus laboratory scattering angles for the incident ion energy $E=20$ eV. The relative cross sections ($\bullet$) are fitted to the absolute data of Lorents and Aberth (----), and the rainbow maximum is located between the peak ($n=11 \frac{1}{2}$) and the valley ($n=11$) and the height of the rainbow peak is lowered.

This fact suggests that an attempt to estimate the well depth from the rainbow angle alone may not be adequate and a more exact treatment such as an inversion of the differential cross section would be needed especially for such symmetric systems between two identical particles²⁴⁾.

The differential cross section increases rapidly as the scattering angle approaches to the rainbow scattering region, at which the positions of the maxima and minima of the cross section are fairly shifted. Out of this region the extrema of the oscillation show good agreement with those of Lorents and Aberth. The

extrema in the rainbow region are plotted against the incident energies of He^+ ions in Fig. 3. However, it is difficult to distinguish the rainbow peak from peaks of oscillation due to charge exchange especially when they lie close to each other. An experiment with much higher resolution would be required to obtain more accurate information about this.

Acknowledgment

The author wishes to thank Dr. T. Mochizuki for his helpful discussions and experimental support throughout this work.

Crossed Beam Studies of Low Energy He⁺ Ions by He Atoms

Fig. 3. Plots of angular positions of the maxima (a) and the minima (b) of the differential cross section as a function of the incident ion energy in the rainbow scattering region. In the figure either (a) or (b), the present results are denoted by closed circles and compared with those of Lorents and Aberth shown by open circles. The maximum corresponds to the minimum of the electron capture probability of Lockwood, Helbig and Everhart (Ref. 2). Integers and half-integers denoted in the figures are consistent with those of Ref. 2.

References

- 1) F. P. Ziemba and E. Everhart, *Phys. Rev. Letters*, **2**, 299 (1959).
- 2) G. J. Lockwood, H. F. Helbig, and E. Everhart, *Phys. Rev.*, **132**, 2078 (1963).
- 3) F. P. Ziemba, G. J. Lockwood, G. H. Morgan, and E. Everhart, *Phys. Rev.*, **118**, 1552 (1960).
- 4) G. J. Lockwood and E. Everhart, *Phys. Rev.*, **125**, 567 (1962).
- 5) D. C. Lorents and W. Aberth, *Phys. Rev.*, **139**, 1017 (1965).
- 6) T. Arikawa, T. Mochizuki and G. Miyamoto, *Japan. J. Appl. Phys.*, **11**, 1212 (1972).
- 7) E. A. Mason and H. W. Schamp, *Ann. Phys.*, **4**, 233 (1958).
- 8) E. A. Mason and J. T. Vanderslice, *J. Chem. Phys.*, **31**, 594 (1959).
- 9) G. E. Ioup and B. S. Thomas, *J. Chem. Phys.*, **50**, 5009 (1969).
- 10) F. A. Herrero, E. M. Nemeth, and T. L. Bailey, *J. Chem. Phys.*, **50**, 4591 (1969).
- 11) R. L. Champion, L. D. Doverspike, W. G. Rich, and S. M. Bobbio, *Phys. Rev.*, **6**, A2327 (1970).
- 12) T. Arikawa, T. Mochizuki and G. Miyamoto, *Mass Spectroscopy*, **20**, 11 (1972).
- 13) T. Arikawa, T. Mochizuki, H. Hirakawa and G. Miyamoto, "Proc. 2nd Int. Conf. on Ion Sources", Vienna (1972), p. 675.
- 14) W. Paul and H. Steinwedel, *Z. Naturforsch.*, **8a**, 448 (1953).
- 15) E. Gustafsson and E. Lindholm, *Ark. Fys.*, **18**, 219 (1960).
- 16) Z. Herman, J. D. Kerstetter, T. L. Rose, and R. Wolfgang, *Rev. Sci. Instrum.*, **40**, 538 (1969).
- 17) T. Mochizuki and T. Arikawa, *Japan. J. Appl. Phys.*, **12**, 1 (1973).
- 18) J. B. Anderson, R. P. Andres and J. B. Fenn, "Advances in Atomic and Molecular Physics", Vol. 1, Academic Press (1965), p. 345.
- 19) E. Everhart, *Phys. Rev.*, **132**, 2083 (1963).
- 20) W. E. Lamb, Jr. and M. Skinner, *Phys. Rev.*, **78**, 539 (1950).
- 21) R. P. Marchi and F. T. Smith, *Phys. Rev.*, **139**, A1025 (1965).
- 22) K. W. Ford and J. A. Wheeler, *Ann. Phys.*, **7**, 259 (1959); **7**, 287 (1959).
- 23) P. N. Reagen, J. C. Browne, and F. A. Matsen, *Phys. Rev.*, **132**, 304 (1963).
- 24) R. E. Olson and C. R. Mueller, *J. Chem. Phys.*, **46**, 3810 (1967).