### Title

The Ratio of Cross Sections for Double to Single Ionization of Helium by High Energy Photons and Charged Particles




### Journal

Journal of Physics B, 28(6)



### Authors

McGuire, James H.
Berrah, Nora
Bartlett, R.J.
et al.



### Publication Date

1995

LBL-36837
UC-410

# Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

## Accelerator & Fusion Research Division

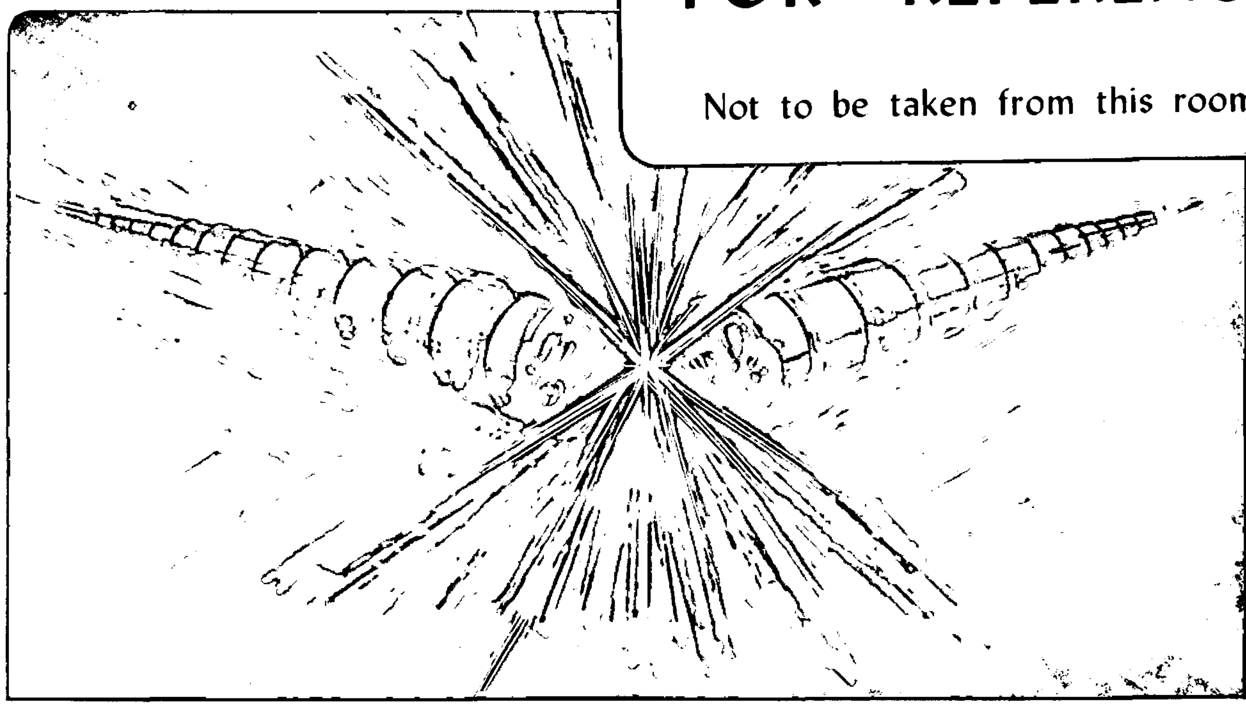
Submitted to Journal of Physics B

# The Ratio of Cross Sections for Double to Single Ionization of Helium by High Energy Photons and Charged Particles

J.H. Mc Guire, N. Berrah, R.J. Bartlett, J.A.R. Samson, J.A. Tanis, C.L. Cocke, and A.S. Schlachter

January 1995







**Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098**

## DISCLAIMER

LBL-36837
LSBL-244
UC-410

# The Ratio of Cross Sections for Double to Single Ionization of Helium by High Energy Photons and Charged Particles

J.H. McGuire
Department of Physics
Tulane University
New Orleans, LA 70118

N. Berrah
Physics Department
Western Michigan University
Kalamazoo, MI 49008-1515

R.J. Bartlett
Los Alamos National Laboratory
Los Alamos, NM 87545

J.A.R. Samson
Behlen Laboratory of Physics
Department of Physics and Astronomy
University of Nebraska
Lincoln, NE 68588-0111

J.A. Tanis
Physics Department
Western Michigan University
Kalamazoo, MI 49008

C.L. Cocke
Caldwell Hall
Kansas State University
Manhattan, KS 66506-2601

A.S. Schlachter
Advanced Light Source
Accelerator and Fusion Research Division
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720

January 1995

This work was supported in part by the Division of Chemical Sciences, Office of Basic Energy Science, Office of Energy Research, of the U.S. Department of Energy under Contract Nos. DE-FG05-92ER14826, DE-FG02-92ER14299, and DE-AC03-76SF00098, and the National Science Foundation under Grant PHY-9017248.

# THE RATIO OF CROSS SECTIONS FOR DOUBLE TO SINGLE IONIZATION OF HELIUM BY HIGH ENERGY PHOTONS AND CHARGED PARTICLES

J.H.McGuire*, N. Berrah**, R.J. Bartlett***, J.A.R.Samson†

J.A.Tanis**, C.L.Cocke†† and A.S.Schlachter†††

(October 11, 1994)

## Abstract

Data and analysis for the ratio of double to single ionization in helium is reviewed for impact by photons and charged particles. In the case of photoionization there are two processes, namely, i) photoionization where the photon is annihilated, and ii) Compton scattering where the photon is inelastically scattered. In the case of charged particle scattering the ratio of total cross sections tends toward an asymptotic high energy value of 0.26% which is well below the value observed for photons of 1.7% at photon energies between 2 and 12 keV. Theoretical relations between various ratios have been predicted and to some extent confirmed by observations.

PACS No. 32.80.Cy,34.50.-s,32.80.Fb

Contents

## I. INTRODUCTION

In the past ten years double ionization of atoms, ions and molecules by high energy photons and charged particles has been studied both theoretically and experimentally. In fast collisions double ionization occurs primarily via an electron-electron interaction following the primary collision with the projectile. Since the collisions are fast, the collision mechanisms are relatively simple. Thus study of such high energy collisions is a sensible way to begin to understand the dynamics of multi-electron interactions. Of interest has been the ratio, $R = \sigma^{++}/\sigma^{+}$, of double to single ionization cross sections in helium which appears to approach a constant value at high projectile energy, E. For impact of charged particles (and anti- particles), the limiting value is $R_Z = 0.26\%$ now in agreement with theory. The corresponding ratio for photon impact was observed to be $R_\gamma = 1.7\%$ at incident photon energies from several hundred eV to 12 keV. This value of 1.7% is in agreement with various predictions for photoionization, where the photon is annihilated during the interaction. However, it has recently been established that Compton scattering dominates over photoionization in helium at photon energies above about 6 keV. In Compton scattering the photon is not annihilated, but is simply inelastically scattered, much like a charged particle.

Photons and charged particles both interact with matter via electromagnetic field. Thus, in principle, cross sections for Compton scattering and photo-excitation and ionization may be related to cross sections for scattering by electrons, protons, and particles of arbitrary charge, Z. On the other hand, Compton scattering, photo-annihilation and scattering from charge particles also differ. For example, Compton scattering differs from photo-excitation and ionization in that: i) the $A^2$ interaction operator for Compton scattering differs from the $\vec{p} \cdot \vec{A}$ term for photo-excitation and ionization, ii) in Compton scattering there is sum over scattering angles of the photon which is absent in photo-annihilation, iii) in high energy Compton scattering large values of the momentum transfer, Q, dominate so that dipole forbidden transitions are important, while total cross sections for photo-excitation and ionization are dominated by the dipole terms. In first order, Compton scattering is simply related to scattering by fast charged particles at all values of Q. At small Q in first order both Compton scattering and charged particle scattering are related to photo-excitation and ionization.

In this paper we review data for the ratio of double to single ionization of helium and provide some theoretical analysis. The data is presented for impact by photons in Sec.II and for charged particle impact in Sec.III. The photon impact data are divided into data for photoionization in Sec II.A, and data for Compton scattering, where the photon is inelastically scattered, are in Sec II.B, In the charged particle case the first data are presented in Sec III.A for total cross section ratios and Sec. III.B contains data for ratios of differential cross sections. The data are followed by a theoretical analysis in Sec.IV, where the mechanisms described by many body perturbation theory (MBPT) are discussed, as well as the relation of the photon data to the charged particle data.

# II. DATA FOR PHOTONS

## A. Energy Dependence of Double Photoionization in Helium

### *1. Overview*

Double photoionization of helium has been used extensively as a testing ground for understanding correlation phenomena since helium is the simplest atom which exhibits electron-electron interactions. Recently, there has been a great deal of progress experimentally and theoretically, both near and far above threshold. From the late seventies until the present the threshold region has been extensively investigated [Schmidt et al. 1976, Wight et al. 1976, Holland et al. 1979, Carter and Kelly 1981, Wuilleumier 1982, Wehlitz 1991] and excellent agreement [Kossman et al. 1988, Lablanquie 1990] was found with the theory of Wannier [1953]. More recently in the high energy limit, measurements of the ratio of double-to-single photoionization at several photon energies, from 2 to 12 keV have been reported by Levin et al. [1993] to be 1.5 (± 0.2)% consistent with a calculated asymptotic value of 1.66% [Byron and Joachain 1967, Aberg 1970, Ishihara et al. 1991, Dalgarno and Sadeghpour 1992]. It was only last year, after much debate over different and in part conflicting theories [Byron and Joachain 1967, Aberg 1970, Carter and Kelly 1981, Ishihara 1991, Dalgarno and Sadeghpour 1992, Amusia et al. 1975], that an understanding of the relative importance of the different processes in double photoionization in the asymptotic limit was achieved. In particular, in this limit only ground state correlation need to be considered when using the acceleration gauge [Dalgarno and Sadeghpour 1992]. From about 2 keV up, new theoretical studies of asymptotic behavior considering also the impact of Compton scattering by Andersson and Burgdoerfer [1993] have been made.

In contrast to the threshold and high energy regime, for intermediate energies few reliable theoretical values exist. Very recent theoretical calculations [Hino et al. 1993, Pan and Kelly 1994] have investigated this range where the available data [Carlson 1966, Bartlett et al. 1994] to test these differing theories are also extremely scarce, in fact previously non-existent from 560 eV to 2 keV. Further progress in understanding requires answering a leading question: what are the dominant correlation effects in the energy gap between threshold and below about 1500 eV, which force the ratio to undergo a significant decrease from about 5% to 2% before settling slowly into the asymptotic limit? Therefore, measurements in this intermediate energy range were of critical importance because they test the capability of the ab initio calculations to describe the transition between the low and high energy regime in an adequate way.

In these section we present measurements between 280 eV and 12 keV that test the most recent theories of Pan and Kelly [1994], of Hino [1993] and Hino et al.[1993] and of Andersson and Burgdoerfer [1993] The aim is to understand how the interplay of electron correlations in both the initial and final states, affects the behavior of the ratio of double-to-single ionization, to understand the relative importance of the basis set, and of higher-order correlation effects.

## 2. Experimental Technique

The intermediate energy measurements were performed at the Berliner Elektronenspeicherring-Gesellschaft für Synchrotronstrahlung m.b.H. (BESSY). Monochromatic light from the high-energy toroidal grating monochromator beamline (HE-TGM-1) operated by the Fritz-Haber-Institute was tuned to several photon energies, $h\nu$, from 280 eV to 1210 eV. The ions were analyzed by a TOF similar to that used by Levin et al. [1992]. Several experimental effects can result in an inaccurate determination of the ratio of double-to-single ionization of helium [Levin et al. 1992, Levin et al. 1993, Berrah et al. 1993]. For these measurements special emphasis was put on suppressing possible higher- order and stray light contributions to the ion signal caused by the monochromator. In order to suppress the stray light effects, measurements were made using a succession of filters to absorb unwanted low energy photons; the measurements were made just below the edge of each filter used. The higher-order light effects were taken into account by taking measurements just above the edge of the appropriate filter.

The high energy measurements by Levin et al. [1993] were obtained at the National Synchrotron Light Source (NSLS) on two beam lines. Monochromatic light from National Institute of Standards and Technology and Argonne National Laboratory (NIST-ANL) beam line X-24A was used in the 2-4 keV range. Focused broadband radiation from Atomic Physics beam line X-26C was employed at higher energies, where the rapidly diminishing He photoionization cross section makes use of monochromatic light less practical (at 10 keV, $\sigma^{++} \leq 3mb$) [Hino et al. 1993]. Data from 8-12 keV were obtained on beam line X26C using broadband light. Because the photoionization cross section at $h\nu = 200$ eV, where the $\sigma^{++}/\sigma^{+}$ ratio peaks (at about 4.8%), is more than five orders of magnitude higher than at 10 keV, steps were needed to assure that no low-energy photons reached the source region to avoid biasing the measurement. The presence of 1.125 mm of Be in several windows along the beam line reduced transmission to below 10-5 at 2.2 keV. Further attenuation of the light was achieved by the use of Cu, Zn, or Al filters, which, in addition, served to help define the photon energy and reduce the energy bandwidth. The He photoionization cross section at 5 keV is less than one order of magnitude higher than at 9 keV, while transmission is more than three orders of magnitude lower.

## 3. Results and Discussion

Ratios of double-to-single ionization obtained at several hv values in the intermediate energy range are shown in Fig. 1. where they are compared with adjusted data of Bartlett et al. [1992] corrected by them by a factor of 1.3, in order to obtain agreement with earlier near-threshold measurements. As can be seen from Fig. 1 the intermediate energy data agree quite well with the scaled data of Bartlett et al.

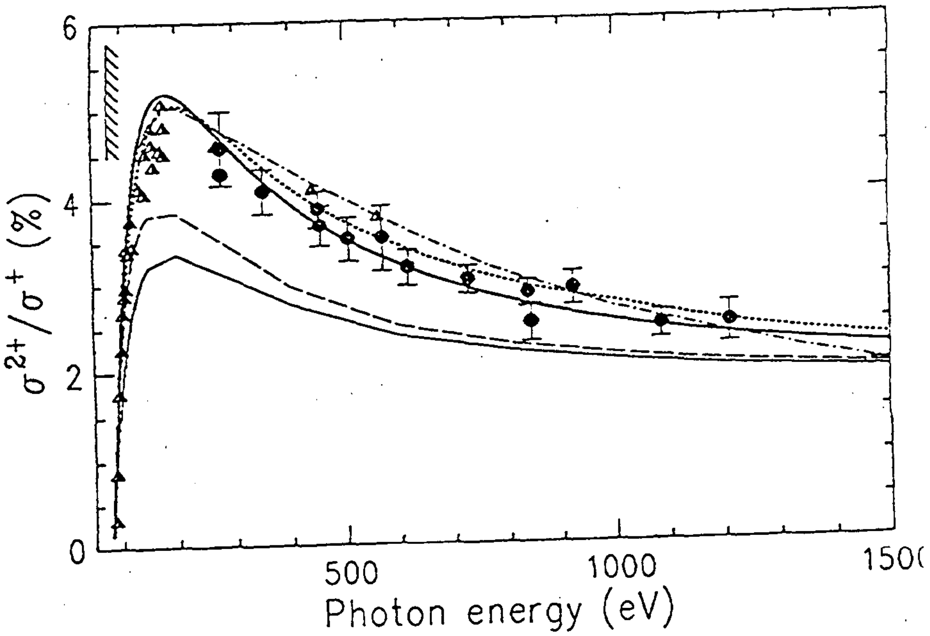



Fig.1 Ratio of double to single ionization as a function of the photon energy comparing the data of Berrah (full circles) with Bartlett et al. (Filled triangles), and theories by Pan and Kelly (heavy solid and short dashed lines) and by Hino and Hino et al. (solid and long dashed lines), and by Samson (dot-dashed line).

The data are also compared in Fig. 1 with recent MBPT calculations (see Sec. IV.A.2). Shown are the results of Pan and Kelly [1994] in the length (heavy short dashed line) and velocity (heavy solid line) along with the MBPT calculations of Hino [1993] and Hino et al.[1993] in the velocity (solid line) and the acceleration form (long dashed line). Pan and Kelly extended Carter and Kelly's threshold energy calculation [Carter and Kelly 1981] up to 14 keV [Pan and Kelly 1994]. Throughout their calculation they included both ground state and final state correlation as in the previous calculations [Carter and Kelly 1981]. They show that while lowest order results show reasonable agreement with experiment, they found that certain higher-order correlation effects are significant. While agreement between the length and velocity forms is very good over this energy range, they find that separate total final state correlation (FSC) and ground state correlation (GSC) diagrams, while individually large and of nearly the same magnitude, are of opposite sign and therefore interfere strongly, with the FSC contribution being the larger of the two. The agreement with the data is therefore very good. The recent MBPT calculations of Hino [1993] and Hino et al. [1993] using the velocity (the length form give the same result as the velocity form) and the acceleration forms of the dipole operator are in fair agreement with experimental data.

If indeed the main difference between Hino et al's MBPT calculation [Hino 1993, Hino et al. 1993] and Pan and Kelly's MBPT calculation [Pan and Kelly 1994] is the inclusion of higher-order effects (both in GSC and FSC) and the use of a different basis set (important since the choice of a pertinent basis set enables the implicit inclusion of higher- order effects), one may conclude that, at intermediate energies (below 1500 eV), unlike the high-energy case, these higher-order effects are very important for the best description of the data. Samson et al. [1992] used a semi-empirical calculation based on a conjecture that there should be a proportionality between production of a doubly charged ion by photon impact on a neutral atom and electron impact on a singly charged ion. Electron impact data of Peart et al. [1969] were scaled to obtain their curve. The chain curve, Fig. 1, shows their calculation, which is in good agreement with the data. Overall good agreement is obtained in the present energy region, however, their model appears to break down above 1300 eV, since their curve decreases rapidly compared to the measured ratios at high energies [Levin et al. 1992, 1993].

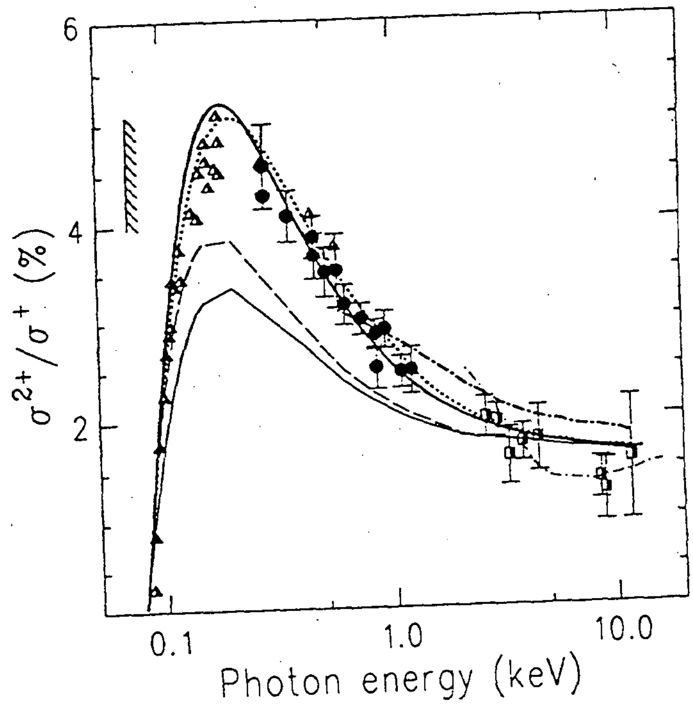



Fig.2 Ratio of double-to-single ionization as a function of the photon energy comparing the data of Berrah et al. (full circles), the data of Levin et al. (half filled squares) and the data of Bartlett et al. (half filled triangles) with calculations by Hino and Hino et al. (solid long dashed and heavy chain lines), by Pan and Kelly (heavy solid and short dashed lines), and by Andersson and Burgdoerfer (chain line). See text for details.

Figure 2 shows the intermediate energy data along with the high energy data [Levin 1992]

They are compared with the recent calculations of Andersson and Burgdoerfer [1993]. of Hino [1993] and Hino et al. [1993] and of Pan and Kelly [1994]. The MBPT calculations of Hino [1993] and Hino et al. [1993] (solid for velocity and long dashed line for acceleration form) are low compared to the data below 2 keV. Hino [1993] also calculates, using the acceleration form using an accurate ground state wave function and a correlated double continuum wavefunction for the final state, the values shown by the heavy chain line. The calculated values by Andersson and Burgdoerfer between 2-18 keV which include contributions due to Compton scattering, agree very well with the high energy measurements. They use the acceleration from of the dipole operator and represent the electron-electron correlation in the final state by a Coulomb distortion factor. They avoid explicit representation of the two-electron continuum by summing ionization-excitation cross sections over all bound He+ states to obtain the single- ionization cross section and by using sum rules.

One may see from Fig. 2 that the length and velocity from calculations of Pan and Kelly (as described above), which converge at about 2 keV, agree quite well with most of the high energy data even through Compton scattering effect is not included. This effect would tend to reduce Pan and Kelly's ratio, moving it in the direction of Andersson and Burgdoerfer value [1993].

### *4. Conclusions*

In summary, by comparing measurements with the most recent calculations [Hino et al. 1993, Pan and Kelly 1994] at intermediate energies (below 1500 eV), both ground state and final state correlation appear to be important, and that inclusion of higher-order effects as well as a judicious choice of basis set used seem to be essential to reproducing the excellent agreement observed [Berrah et al. 1993]. At high energies, the measurements of Levin et al. [1992, 1993] are consistent with an asymptotic value of 1.66% found with older shake calculation [Byron and Joachain 1967 , Aberg 1970] as well as with the most recent calculations [Ishihara et al. 1991, Dalgarno and Sadeghpour 1992, Andersson and Burgdoerger 1993]. In particular, it was found [14] that in this limit, only ground state correlation needs to be considered when using the acceleration gauge and that consideration of the TS1 final state correlation is not essential (see Sec IV.A.3).

## B. Compton Scattering

### *1. Observed Deviations from Photoionization*

The ratio of the He photoionization cross sections for double to single ionization has long been predicted by theory to reach a constant value at high energies. This limiting ratio has variously been calculated at values lying between 1.6% and 2.3% [Byron and Joachain 1967, Aberg 1970, Brown 1970, Amusia et al. 1975, Ishihara et al. 1991]. Recently, there has been a consensus that the ratio should tend to a constant value of 1.66% [Dalgarno and Sadeghpour 1992, Andersson and Burgdoerfer 1993]. Direct measurements of the $\sigma^{++}/\sigma^{+}$ ratio have been made above 2 keV by Levin et al. [1992, 1993] and Bartlett et al. [1994]. Their results suggest a constant $\sigma^{++}/\sigma^{+}$ ratio of about 1.70% has been reached near 4 keV. The expectation that the ratio should reach a constant value is based on the prediction that for high photon energies both the single and double photoionization cross sections vary with energy as $E^{-3.5}$. This energy dependence for total ionization has been shown recently to occur at photon energies greater than about 2.4 keV [Samson et al. 1993,1994b]. These results are shown in Fig. 3.

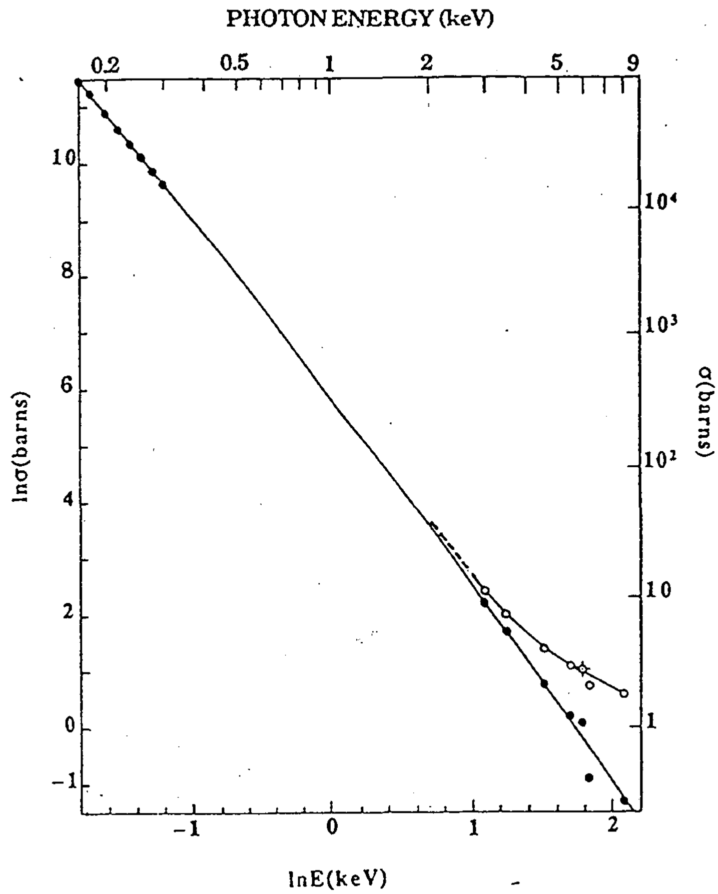



Fig.3. Photoionization cross section (solid line) between 200 eV and 8 keV; small dots at low energy, Samson et al. [1994b], small dots at high energy, Bearden 1966 and McCrary et al. [1970]. Total attenuation cross sections (dashed line); crossed circles, McCrary et al. [1970]; open circles Bearden, [1966].

The open circles represent the corrected attenuation cross sections measured by Bearden [1966] and McCrary [1970]. The solid data points directly below the attenuation data represent the photoionization cross sections obtained after subtracting the total scattering cross sections [Veigele 1973] from the attenuation data. The resulting curve has a slope, on a log- log plot, equal to -3.5 between 3 keV and 8 keV and fits the equation $\sigma = 410E^{-3.5}$, where E is the photon energy in keV units and $\sigma$ is the photoionization cross section in barns ($10^{-24}cm^2$). If this curve is extrapolated smoothly to meet the photoionization data plotted between 180 and 300 eV we find that the slope starts to decrease slowly from -3.5 near 3000 eV to -3.0 at 300 eV. Recent measurements of the attenuation cross sections have been made by Azuma et al. [1994], from 3 keV to 14 keV and are shown in Fig.4. These measurements show that the relation $\sigma = 410E^{-3.5}$ extends to 14 keV.

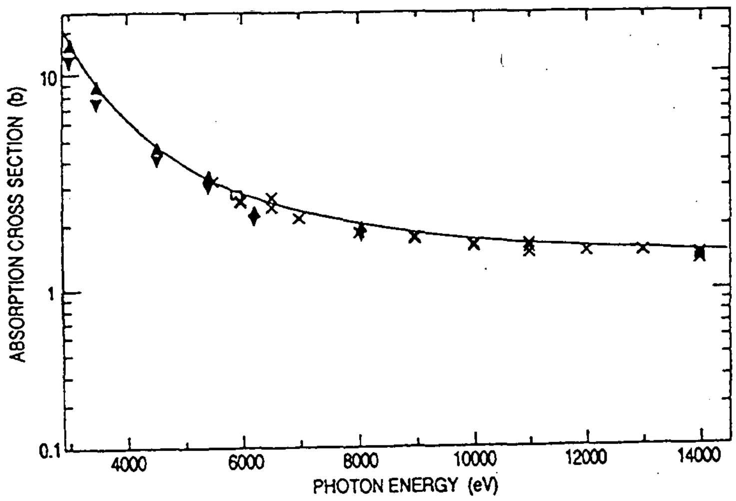



Fig. 4 Total attenuation cross section of photons in helium at energies between 2 and 14 keV as measured by Azuma et al. [1994] (crosses); Bearden [1966] (black erect and inverted triangles); McCrary et al. [1970] (open squares); Veigele [1973] (solid line).

*2. Compton Scattering*

The above studies of photoionization and photo-attenuation cross sections of He have shown that the total coherent (Rayleigh) and incoherent (Compton) scattering become noticeable above 2 keV (see Sec IV, B). This is in agreement with the published compilations of the calculated scattering cross sections by Hubbell et al. [1975], Veigele [1973], and Cullen et al. Also, from the Compton formula for scattering from free electrons, Compton ionization of He is expected to have a threshold at approximately 2.5 keV. Thus, just before the $\sigma^{++}/\sigma^{+}$ ratio is beginning to establish a constant value, some contribution to the $He^{+}$ signal from Compton ionization may be starting. In fact, direct measurements of $He^{+}$ produced by Compton ionization have been reported recently [Samson et al. 1994a]. How will this affect the observed double to single ionization ratio? Preliminary results by Bartlett et al. [1994] and Levin et al. [1993] appear to answer this question. These results are shown in Fig. 5 at energies from 3 keV to 12 keV and are compared to calculated photoionization data of Hino et al. [1994].

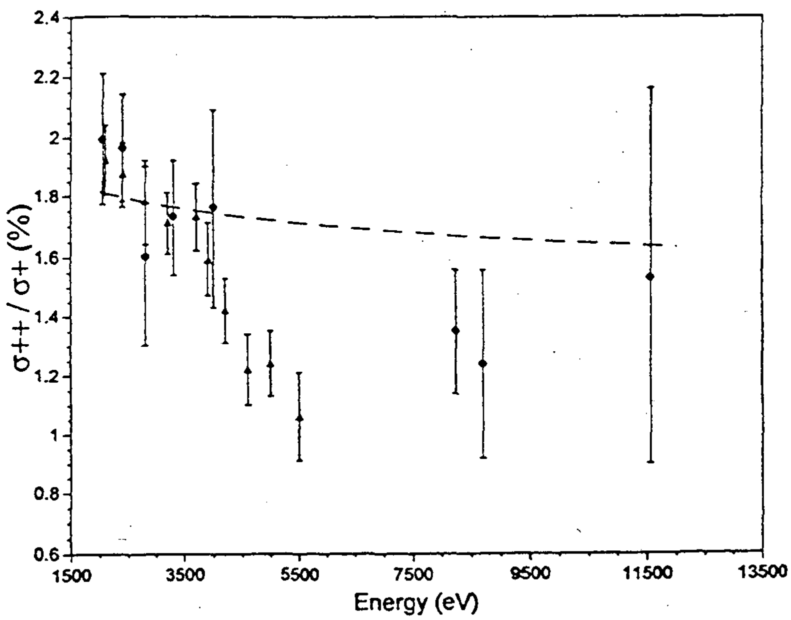



Fig.5 Cross section ratios for double to single ionization of helium by photons. The solid dots are data of Levin et al. [1992,1993] and the solid triangles are from Bartlett et al. [1993]. The dashed curve is the photoionization ratio calculated by Hino et al. [1994]. The initial departure of the data from the photoionization curve is caused by the addition of single ionization from Compton scattering. The measurements of Bartlett et al. give greater weight to the ionization caused by Compton scattering than to photoionization. Thus their data show a stronger departure from the photoionization ratio than actually occurs.

At about 3.8 keV the observed ratio starts to decrease and appears to reach a minimum value. This decrease would be expected on the basis that Compton ionization was producing an increasing amount of $He^+$. Although no direct observation of double ionization production by the Compton effect has ever been made it is reasonable to assume that for photon energies greater than 4.6 keV (the threshold photon energy necessary for the transfer of sufficient energy to produce double ionization by Compton scattering from free electrons) the probability for producing $He^{++}$ would increase. Thus, in Fig. 5 one would expect $R_{obs}$ to increase towards higher energies. The actual magnitude of the decrease in $R_{obs}$ is not known accurately because of possible systematic errors associated with differences in the collection efficiency for $He^+$ ions produced by photoionization and those produced by Compton ionization. The former have recoil energies in the range 0.4 to 1.4 eV, whereas Compton ions have energies close to zero. Nevertheless, it is of interest to estimate the value of $\sigma^+$ at, say 4600 eV, where we might expect the Compton ratio $R_C = \sigma_C^{++}/\sigma_C^{+}$ to be approximately zero. $R_{obs}$ can be written as,

$$R_{obs} = \frac{\sigma_\gamma^{++} + \sigma_C^{++}}{\sigma_\gamma^{+} + \sigma_C^{+}} \tag{1}$$

where the subscripts $\gamma$ and c refer to the photoelectric process and the Compton effect, respectively. Equation (1) can be rewritten in terms of the cross sections $\sigma_\gamma^{+}$ and $\sigma_C^{+}$ and the Compton ratio $R_C = \sigma_C^{++}/\sigma_C^{+}$, thus,

$$\sigma_C = \sigma_\gamma(R_\gamma - R_{obs})/(R_{obs} - R_C) \tag{2}$$

Assuming that the ratio $R\gamma$ follows the dashed curve in Fig. 5, that $R_C \sim 0$, and using the absolute photoionization cross sections $\sigma_\gamma$ reported recently [Samson et al. 1994b], one obtains from Eq.(2) a value for $\sigma_C^{+} = 0.66$ barns at 4.5 keV. Hubbell et al. [1975] quote a value of 0.66 barns for total incoherent scattering and Hino et al. [1994] quote a value of 0.64 barns for Compton ionization. However, no definitive statement can be made about the accuracy of the results at this stage. The results are consistent with the interpretation that the dip in the curve of Fig. 5 is caused by the appearance of Compton ions and in qualitative agreement with the calculations by Hino et al. [1994].

### *3. Summary*

The production of $He^{+}$ via the Compton effect appears to be an important process in the production of the $\sigma^{++}/\sigma^{+}$ ratio in the photon energy region above 3.6 keV. The high energy data of Levin et al [1993] shows the ratio rising from 1.3% above 8 keV to 1.53% at 11.5 keV. Presumably, $He^{++}$ is now being produced by the Compton effect and contributes to the ratio. In fact, we would expect that ratio at 11.5 keV to be produced mainly by Compton ionization because the ratio of incoherent scattering to the photoelectric effect is about a factor of 10 at this energy. Experimental and theoretical work are continuing in this area in an effort to understand the similarity and differences between photoionization and Compton ionization.

## III. DATA FOR CHARGED PARTICLES

### A. Total Cross Section Ratios

#### *1. Direct Target Ionization*

Measurements of the direct ionization of helium by intermediate- to high-velocity projectiles is reviewed in this section. Direct (or pure) ionization is defined as target ionization for which the incident projectile charge state Z remains unchanged. Most of the studies to date have focused on establishing the velocity regimes where different collision mechanisms dominate. Of particular interest are those results obtained for projectiles in high charge states at high velocity, v. In the discussion below, regimes of validity are delineated for each of the mechanisms, and ratios of double-to-single ionization cross sections are presented and compared with theoretical predictions.

Double ionization of helium by charged particles has been the subject of numerous experimental and theoretical investigations for nearly three decades [Mittleman 1966, Byron and Joachain 1966, Horsdal-Pedersen and Larson 1979, McGuire 1982, Knudsen et al. 1984, Andersen et al. 1986, 1987, Reading and Ford 1987a, Reading and Ford 1987b, Giese and Horsdal 1988, Kamber et al. 1988, Reading et al. 1989, Herber et al. 1990]. Much of the interest in double ionization by charged particles stems from its fundamental nature [Mittleman 1966, Byron and Joachain 1966, McGuire 1982] and its relationship to double ionization by photons [Horsdal-Pedersen and Larsen 1979, Samson 1990, Levin et al. 1991] (see Sec. IV.B). While the single ionization of helium by fast, fully-stripped ions has been studied extensively, both theoretically [Inokuti 1971, 1978] and experimentally [Haugen et al. 1982] and is well understood, for double ionization additional physical mechanisms come into play.

As will be discussed in more detail below in Sec. IV, double ionization of He by a fast highly-charged projectile can result from either a single or a double interaction of the projectile with the target electrons. This is in contrast to photoionization (see Sec. II.A) in which the incident photon interacts with only one of the He target electrons. This latter process is generally referred to as a one-step mechanism or shakeoff (SO). For high-velocity charged particle collisions where the collision time is small, there is a small probability for double ionization by separate interactions. In this case the projectile interacts with only one of the target electrons, transferring it to the continuum (see Sec. IV.A.2). Subsequent rearrangement from the two-electron wavefunction of the remaining ion can lead to the ejection of the second electron.

At lower projectile velocities, a two-step process (TS) sometimes referred [Andersen et al. 1987] to as TS2, in which the projectile interacts with each of the target electrons independently, dominates the double-ionization process. In yet another type of process involving a single projectile interaction with a target electron, Knudsen et al. [1987] have suggested that the projectile may interact with one of the target electrons, and this electron may subsequently interact with the second electron in a binary collision, thereby ejecting it. This latter two-step process is labeled TS1 (see Sec. IV.A.2). It is important to note, however, that it may not be possible experimentally [Andersen et al. 1987, Vegh and Burgdoerfer 1990] to distinguish TSl from shakeoff (SO) and, in fact, in the limit of high ejected electron velocities these mechanisms may be identical in magnitude but opposite in sign [Vegh and Burgdoerfer]. Each of these various mechanisms is be discussed more fully in Sec. IV.A.

Ionization (single and double) of neutral targets by charged ions depends on the projectile charge Z and the collision velocity v. It is, in fact, well known that the important parameter governing ionization is Z/v. From various studies [McGuire 1982, Knudsen et al. 1984 Andersen et al. 1987, DuBois and Toburen 1988], it can be inferred that the TS2 mechanism should dominate for projectile charges Z and velocities v (in atomic units) such that $Z/v > 0.2$, while shakeoff should dominate for $Z/v < 0.05$. Both TS and SO are likely to be important in the intermediate range $0.2 < Z/v < 0.05$ where interference effects may be important. The regions of validity for the different double ionization mechanisms are displayed graphically in Fig. 6.

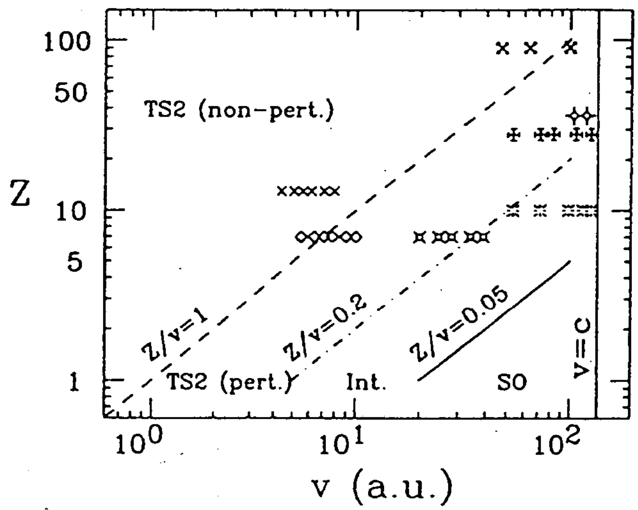



Fig. 6. Plot showing the Z and v (in atomic units) regimes where the two-step and one-step (shakeoff) mechanisms of double ionization are expected to be dominant as well as the intermediate region where both mechanisms are expected to be important. The values Z/v = 1.0, 0.2, and 0.05, represent the approximate maximum Z/v values corresponding to the perturbative TS2, intermediate (interference), and SO regimes, respectively. For $Z/v > 1$ the TS2 mechanism is also dominant, but perturbation methods cannot be used. Z and v values for the following ions are indicated: winged squares - $N^{+7}$; open diamonds - $0^{7+}$; starburst - $Ne^{+10}$; X's - $S^{13+}$; crosses - $Ni^{+23}$ winged diamonds - $Kr^{+36}$; fancy crosses $U^{90+}$.

The dashed line corresponding to Z/v = 1 indicates the approximate upper limit where perturbation methods may be used to treat the TS2 mechanism. The lower and upper limits for the two-step and shakeoff regimes are chosen as Z/v = 0.2 and 0.05, respectively, by noting that at these points the competing process of shakeoff or two-step, respectively, is negligible [McGuire 1982, Knudsen et al. 1984, Anderson et al. 1987, DuBois and Toburen 1988]. These do not represent absolute boundaries; they serve, however, to indicate where one process ceases to dominate and the other becomes non-negligible. Of course, between these two limits, both the two-step and shakeoff terms are important and interference between these two amplitudes can play a significant role.

Since we are interested mainly in the mechanisms leading to the double ionization of helium, we focus here on the ratio R of double-to-single ionization cross sections which can be written (as shown in Sec.IV) as:

$$R_z = (C_1 + C_{12}Z/v + C_2(Z/v)^2) \quad (3)$$

Extensive measurements of double-to-single ionization ratios in the intermediate- to high-velocity regimes are available only for $H^+$ [Knudsen et al. 1984, Andersen et al. 1987, Shah and Gilbody 1985] and $He^{2+}$ [Knudsen et al. 1984, Andersen et al. 1987, Heber et al. 1990]. Additionally, there are published data for $N^{7+}$ [Heber et al. 1990], $0^{7+}$ [Tanis et al. 1991a], $S^{13+}$ [Tanis et al. 1991b], $U^{90+}$ [Berg et al. 1992], several other highly-charged (not fully-stripped) ions [McGuire et al. 1982]; and recent high velocity data for $Ne^{10+}$,$Ni^{28+}$, and $Kr^{36+}$ ions obtained by Ullnch et al. [1993]. As mentioned above, the ratio of double-to-single ionization is the parameter used most frequently to exhibit and test double ionization mechanisms. From Eq.(3), R is mainly (except for a slowly varying ln v dependence) a function of Z/v (or v/Z) [McGuire 1982], and thus a high degree of universality should be exhibited in a plot of R vs. v/Z. Furthermore, in the perturbative two-step regime, i.e., v/Z = 1-5 (q/Z = 1.0-0.2), R is expected to vary nearly as (Z/v)2 (see Eq.(3) above), and deviations from this dependence may be attributable to the C12 interference term.

Following Tanis et al. [1992], values of R for a diverse set of projectile ions with different velocities and charges are shown in Fig. 7, plotted as a function of v/Z [Schlachter and Tanis 1994]. Most of the available data are seen to fall along a common curve with the "crossover" from the TS2 to the SO regime being clearly evident. Furthermore, in the TS regime, i.e., v/Z = 1-5, the data agree quite well with the predicted (v/Z)-2 dependence (solid line in figure 7); for v/Z > 10, R approaches a constant value in agreement with the prediction of the SO mechanism.

Available data for $0^{7+}$ [Tanis et al. 1991a] and $Si^{13+}$ Tanis et al. 1991b] lie entirely in the TS regime as seen in Fig. 6. In fact, the $Si^{13+}$ data are out of the perturbative two-step regime. In Fig. 7 the $0^{7+}$ data are seen mainly to follow the (v/Z)-2 dependence while the $Si^{13+}$ data deviate strongly from this behavior as might be expected. There is a continuity, however, from the $0^{+7}$ data to the $S^{13+}$ data as the value of v/Z decreases.

It is seen in Fig. 6 that the 10–40 MeV/u $N^{7+}$ data fall well within the two-step perturbative regime, just bordering on the region where interference effects may start to come into play. Although there is scatter in the measured ratios, the trend of these data tend to decrease with increasing v/Z as seen in Fig. 7.

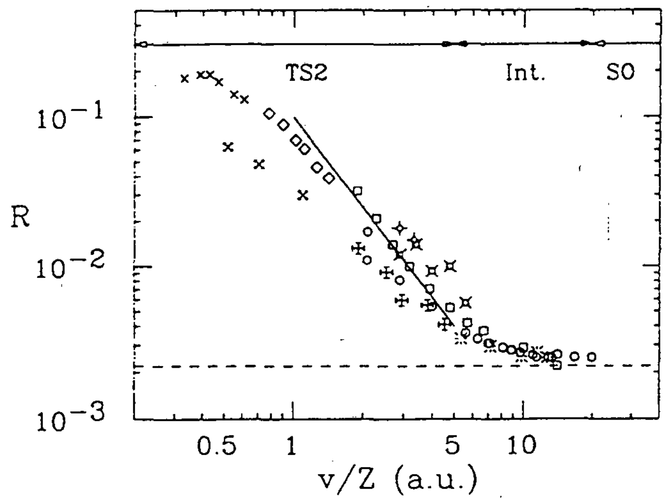



Fig. 7. Double-to-single ionization ratios R of helium by several ions as a function of v/Z (in atomic units). Data are as follows: open circles - $H^+$ (Knudsen et al. [1984], Andersen et al. [1987], Shah and Gilbody [1985]); open squares - $He^{2+}$ (Knudsen et al. [1984], Andersen et al. [1987] and Heber et al. [1990]); winged squares - $N^{+1}$ (Herber et al. [1990]); open *diamonds* - $O^{+7}$ (Tanis et al. [1991a]; starburst - $Ne^{+10}$ (Ullrich et al. [1993]); X - $Si^{+13}$ (Tanis et al. [1991b]); crosses -$Ni^{+28}$ (Ullrich et al. [1993]); winged diamonds - $Kr^{+36}$ (Ullrich et al. [1993]); fancy crosses - $U^{+90}$ (Berg et al. [1992]). The solid line indicates a (v/Z)-2 dependence (see text). The dashed line indicates the shakeoff (SO) limit (Z/v = 0.0022) determined by Knudsen et al. [1984]. The two- step (TS2), shakeoff (SO), and intermediate (Int.) regions are also indicated.

In the work of Berg et al. [1992] for very high charge-state and very high-velocity projectiles, the ratios for double-to-single ionization of helium by 60-, 120-, and 420-MeV/u $U^{90+}$ ions are at Z/v values far removed from the one-step regime in which the asymptotic limit can be tested. However, it is noted that the U9°+ data fall farthest from the common curve and below the (v/Z)-2 dependence in Fig. 7. The deviation of the U9°+ data from universality may be, in part, due to: (l) the fact that these data lie largely outside the perturbative regime (see Fig. 6), (2) the high charge state of these ions, or (3) relativistic effects. In order to differentiate between these possibilities, or, perhaps, to find an alternative explanation, more theoretical and experimental work will be required for ions with very high velocities and charge states.

A summary of existing high energy measurements is given in Table 1. This table shows that, to date, only for $H^+$, $He^{2+}$ and $Ne^{10+}$ projectiles are there double ionization data in the asymptotic regime, i.e., $Z/v < 0.05$. It should be noted, however, that according to Fig. 6, if the theoretical formulations are correct, the asymptotic regime can never be reached for projectiles with charge $Z \gtrsim 10$.

| Z | E (Mev/u) | v (a.u.) | Z/v | R (%) |
|---|---|---|---|---|
| 1 | 15 | 24 | 0.04 | 0.2 |
| 2 | 20 | 28 | 0.07 | 0.2 |
| 7 | 40 | 39 | 0.18 | 1.0 |
| 10 | 1500 | 127 | 0.08 | 0.25 |
| 90 | 420 | 99 | 0.91 | 3.0 |
| γ | ≥ 2 keV | – | – | 1.5 – 3.5 |

Table 1. Measured values R of double-to-single ionization at high energies, where Z is the incident charge of the projectile, E is the maximum kinetic energy for which data are available for the given charge state, v is the (relativistic) velocity in atomic units, and R is the ratio of double-to-single ionization. The ratio for high-energy photons (> 2 keV) is also listed (Hino et al. [1993] and Levin [1991]).

The above results confirm that the charge state of the projectile ion is fully as important as the projectile velocity in determining the regime where double ionization due to the one-step shakeoff mechanism dominates. In fact, by comparison with other data, it is apparent that the asymptotic limit has been reached to date only for incident electrons, protons, alpha particles, and $Ne^{10+}$. By plotting ratios of double-to-single ionization for several ionic species at different velocities and in different charge states it is seen that this ratio exhibits a high degree of universality as a function of v/Z (or Z/v) in agreement with the expectations of the perturbative treatment of double ionization. In summary, the parameter of significance in analyzing double ionization mechanisms is demonstrated to be Z/v as expected.

### *2. Target Ionization Accompanied by Projectile Capture or Loss*

In addition to target double ionization in which the charge state of the projectile remains unchanged, double ionization of helium can also occur in connection with single-electron capture or loss by the projectile. While double ionizatons associated with no projectile charge change is understood quantitatively, both theoretically and experimentally, this same cannot be said for double ionization associated with projectile electron capture or loss.

Compared to "pure" ionization in which the projectile does not change charge, target ionization accompanied by projectile electron capture or loss is expected to occur at smaller average impact parameters since electron capture or loss generally requires a "harder" collision, at least when the projectile electron binding energy is greater than that of the target. Thus, the study of double ionization of helium associated with electron capture, electron loss, or no charge change can be useful in understanding qualitatively the dependence of this process on impact parameter. Additionally, for incident projectiles which carry electrons into the collision, comparison of the resulting double-to-single ionization ratios to those obtained for bare ions gives information on the contribution of the projectile electrons to double ionization [Wang et al., 1990] In the case of double ionization associated with projectile electron capture, the captured electron leaves the collision region with the speed of the projectile, and for high-velocity collisions, this condition satisfies the requirement for shakeoff to occur [Knudsen et al. 1987].

In measurements of Tanis et al. [1991a,b], ionization of helium by 0.125–3.0 MeV/u $He^+$, 0.7.5-2.5 MeV/u $O^{7+}$, and 0.44–1.6 MeV/u $S^{13+}$ was investigated, ~~thus providing information~~ covering charge states and energies over the range Z/v = 0.1–3.0 (v/Z = 0.33–10). In this work, target ionization was measured for projectiles undergoing electron capture, electron loss, or no charge change. The resulting double-to-single ionization ratios were found to depend strongly on the outgoing charge of the projectile as shown in Fig. 8.

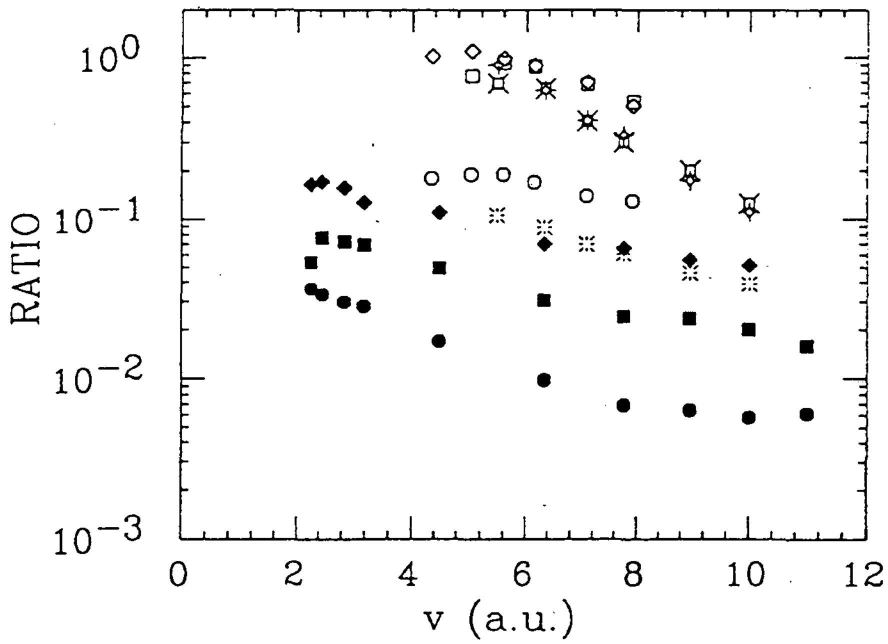



Fig. 8. Measured ratios of double-to-single ionization plotted as a function of v/Z. Symbols represent target ionization associated with outgoing projectile charge states as follows: no charge change, black dots - $He^+$, starburst - $O^{7+}$, open dots - $S^{13+}$; electron capture, solid diamonds - $He+$, winged diamonds - $O^{7+}$, open diamonds - $S^{13+}$; electron loss, black squares - $He^+$, winged squares - $O^{7+}$, open squares - $S^{13+}$.

For target ionization accompanied by projectile electron capture or loss, the ratios are seen to be significantly enhanced and their dependence on v is somewhat different from the case of no charge change. The large enhancement is qualitatively understood from the fact that target ionization accompanied by projectile capture or loss is expected to occur at smaller average impact parameters than ionization without accompanying charge change, thereby giving rise to a higher probability for double ionization in the former cases. It is also noted that the double-to-single ionization ratios associated with projectile electron capture and loss are nearly identical for $O^{7+}$ and for $S^{13+}$. The origin of these various behaviors for the double-to-single ionization ratio is not undertood at present, indicating that double ionization accompanied by projectile capture or loss is not simply described by Eq.(3).

These results show that, while the qualitative features of the double ionization of helium associated with projectile electron capture or loss are understood, the specific behavior of the data are not explained within the framework of existing theories. Thus, these results point to the need for further investigation of the mechanisms leading to the double ionization of helium (see Sec. IV.A.4), especially when target ionization is associated with projectile electron capture or loss.

### B. Differential Cross Sections

For both charged particle impact and photon impact, the ratio of double to single ionization of He appears to reach a limiting value for high energy projectiles. The ratios are quite different, however, near 0.27% for charged particles (see Sec. III.A) and near 1.7% for photons (see Sec. II.A). It is thus immediately clear that a fast charged particle is in no way similar to a high energy photon in its impact upon He. The basic reason for the difference is that high energy photoionization deposits most of the photon energy in a single target electron, whereas the total cross section for charged particle impact at high velocity is dominated by large impact parameter collisions which eject soft electrons from the target with continuum energies of the order of their original binding energies in the He. This point was made in 1984 by McGuire, who used the relationship between the photoionization cross section for producing a photoelectron of a certain energy and that for producing an electron of the same continuum energy by a fast charged particle impact to show that the high velocity charged particle value of R should be much less than that for photon impact (see Sec. IV.B.2). It was suggested that to observe a large value for R for charged particle impact would require a differential measurement which isolated events in which fast electrons were produced.

One mechanism which contributes to the double ionization of He for high energy photoionization is shakeoff (SO) ( see Sec. IV.A.1). If one electron is removed from He suddenly, the remaining electron finds itself moving in a modified potential, with the result that its wave function immediately has non-zero overlap with the new continuum. In this approximation R is the square of this continuum amplitude squared, integrated over all continuum states, and does not depend on how the electron was removed. Thus for this mechanism, one might expect that any process which makes a fast enough primary electron would produce the same value of R. There have been several experiments which have isolated fast primary electrons and which have thus addressed this conjecture. These include experiments identifying kinematically isolated binary ridge electrons, direct measurements in coincidence with of fast electrons, and electron capture, and the results from each of these will be discussed below. However, it is known that shakeoff, in the sense of an overlap of Hartree Fock Wave functions, contributes only about 0.7% [Byron and Joachain 1967, Carlson 1966, Levinger 1953] of the 1.7% limiting value of R for photoionization. The remainder is commonly attributed to ground state correlations (a non-unique designation; see section IV.A.2-4 of this report). Whether it is still to be expected that R should have a universal value for ANY process producing a fast enough primary electron is not at all clear. The subject is discussed by various authors [McGuire 1984, Cocke et al. 1989, Vegh and Burgdoerfer 1990, Aberg 1973]. Arguments have been given that the production of fast electrons via photoionization requires probing of high momentum components of the He wave function, whereas the ejection of a fast electron by a charged particle probes all momentum components. Thus the correlations in the initial wave function might not play identical roles in the two cases.

#### *1. Large-angle charged particle scattering*

The differential cross section for single ionization of He at large scattering angles (large here means outside about 0.2 mrad) and at projectile velocities above about 6 a.u. shows a very clean binary ridge structure, characteristic scattering of the projectiles from a quasi-free electron in the He Kamber et al. 1988]. If one ignores the Compton momentum distribution of the target, this binary encounter scattering is kinematically limited to lie inside an angle of 0.55 mrad (the electron/proton mass ratio) in the laboratory. The data show a kinematic rainbow structure in the laboratory at this angle. Projectiles can also be scattered from the He nucleus into this angular range, but above about 1 MeV the projectile-electron scattering dominates in the single ionization channel in the vicinity of the binary ridge ($0.3 < \theta_{lab} < .55 mrad$). Therefore for this mechanism a particular proton scattering angle corresponds to a unique primary electron energy, and the latter is typically quite fast. Kamber et al. [1988] measured R as a function of proton scattering angle for protons between 3 and 9 MeV and plotted the result versus the energy of the calculated primary electron energy (see Fig. 9), which lay between 1 and 5 keV. The resulting value of R was found to be between 1.8 and 2.4%, much higher than that for total cross sections and rather similar to the corresponding values of R found in photoionization for the same range of photoelectron energies. However, at these projectile energies one has still to contend with double ionization via two separate interactions of the proton with the He electrons, and indeed, Kristensen and Horsdal-Pedersen [1991] found the average probability for ejecting a He electron for small impact parameters to be near 6% at 3 MeV, enough to account for all of the R measured by Kamber et al., and more. On the other hand , if this process were really so important in the data of Kamber et al., the value of R would be expected to vary substantially with proton energy, which was not observed to the the case. No charged particle experiment has really been performed at sufficiently high velocity to avoid criticism on this ground.

The above experiment does not experimentally isolate fast primary electrons, but assumes them. Doerner et al.[1993] have reported measurements of R for proton-electron collisions which experimentally isolate fast electrons produced in collisions of He with 1 and 3 Mev protons. These velocities are not high enough to avoid double ionization by independent interactions of the two electrons with the projectile. Nevertheless, the results show a value of R near 2%, robustly independent of electron velocity above 200 eV and of proton energy.

Many inelastic electron scattering experiments on He have been carried out over the years [Wight et al. 1976, Schmidt 1976, Holland et al. 1979]. These experiments have focused on transverse momentum transfer collisions for which the matrix element for electron scattering in the first Born approximation is known to reduce exactly to that for photoionization for the same primary electron energy. Indeed, before synchrotron sources made available the high quality photoionization data in existence today, these data were generally taken as equivalent to photoionization and filled the gap in the region of photoelectron energies up to 200 eV. The large transverse momentum transfer e,e' experiment corresponding to that of Kamber et al. for proton scattering, does not appear to have been reported. Such an experiment would be of great interest in connection with this problem.

At somewhat lower projectile energies (200 to 500 keV) Giese and Horsdal [1988] pursued R to angles extending beyond 0.55 mrad and found a peak at a scattering angle near 1 mrad. Several theoretical attempts [Olson et al. 1989, Salin 1991, Meng et al. 1993, Fang and Reading 1991, Reading et al. 1989] explain this behavior have been made, including both double collision and shakeoff mechanisms. The most successful descriptions attribute the double ionization in this region to two independent hard scatterings of the projectile from the target electrons. Each individual scattering can carry the projectile only to an angle of 0.55 mrad maximum, but two such scatterings can carry it to a maximum of 1.1 mrad. Projectiles can reach angles beyond this only by hard scattering from the He nucleus, which is much less likely to ionize the He than the hard p-electron scatterings. Thus the differential cross sections for single ionization show a rapid decrease near 0.55 mrad, and for double ionization a similar decrease near 1 mrad, with the result that a rise in the ratio appears near 0.55 mrad, decreasing again near 1.1 mrad and producing a peak slightly inside of 1 mrad. All present calculation based on this mechanism, including CTMC [Olson et al. 1989], dCTMC [Meng et al. 1993], PWBA [Salin 1991] give approximately the correct shape for R versus theta, but the value of R is about a factor of two too large, for which no explanation has been given. The calculation of Fang and Reading include shakeoff in R, but produce the peak at too small theta. The dCTMC calculation of Meng et al. include radial correlation in the He wave function, and thus should have some shakeoff-like mechanism included. It is interesting and perhaps inconsistent that the higher energy results of Kamber et al. seem to be interpretable in terms of a shakeoff-like picture, with no appeal to double hard hits, while the lower energy collisions ignore the shakeoff and still produce too large a value of R. The values of R from both sets of data seem to be too low to accommodate contributions from shakeoff and also from two hard hits.

### *2. Electron Capture*

Kristensen and Horsdal-Pedersen [1991] have measured R as a function of projectile velocity for protons capturing electrons from He. Their results , shown in Fig. 9, when plotted versus primary electron velocity (which is the projectile velocity in this case, since the outgoing electron is captured) , lie remarkably near the photoionization data and a bit above the binary encounter data for higher energy.

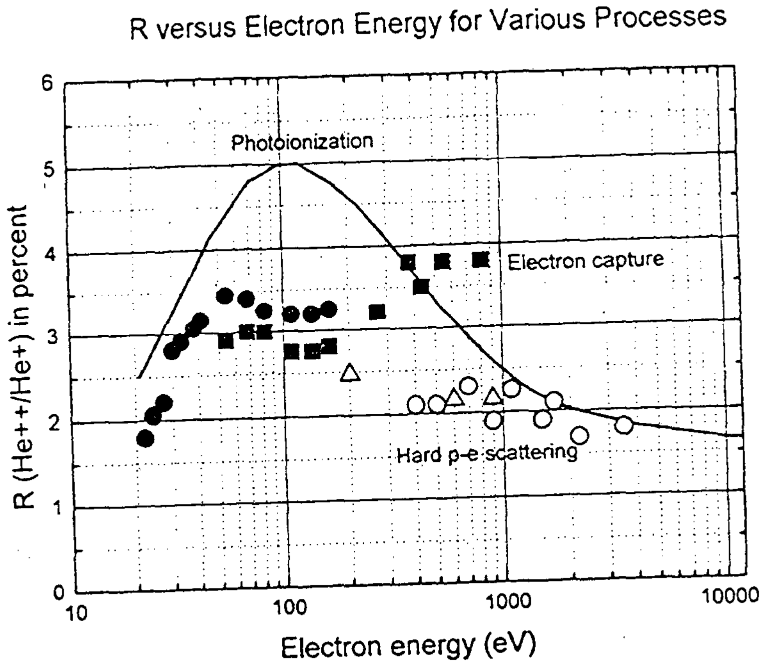



Fig. 9 The ratio R for various processes plotted versus outgoing primary electron energy. The solid line is a curve drawn through the data of Figs. 1 and 2 and is labeled 'photoionization'. For this curve the electron energy is taken to be the photon energy minus the sum of the two ionization potentials of helium. The filled symbols are electron capture: circles, Kristensen and Horsdal-Pedersen [1990]; squares, Horsdal-Pedersen and Larsen [1979]. The open symbols are from hard proton-electron scattering data: circles, Kamber et al. [1988]; triangle, Cocke et al. [1993].

The electron capture mechanism itself is a center of considerable interest in this velocity range . It almost certainly involves both first and second Born processes. The former, commonly described by an OBK amplitude [McDowell and Coleman 1970] is just an overlap in momentum space of the initial and final state wave functions, and the matrix element is somewhat similar to the photoionization matrix element in that no momentum is carried by the transition operator itself. The second Born terms generally involve hard scatterings of the outgoing electron with the projectile , the target nucleus and/or the second electron [McGuire 1995], and the process has similarities to the binary-encounter production of fast electrons. It is interesting that the experimental $R_C$ for capture lies so near the corresponding values $R_\gamma$ and $R_Z$.

It has recently been pointed out that for sufficiently high photon energies, above about 6 keV, the major process whereby He is ionized by photons ceases to be photoionization and becomes Compton scattering. For the former case, the momentum of the outgoing electron is provided by the initial wave function of the He, whereas for the latter case it is provided by the incident photon. Burgdoerfer et al. [1994] have pointed out that, in the Born approximation, the operators for charged particle scattering and Compton scattering are very similar. They show that in this approximation, for a given momentum transfer Q to the outgoing primary electron, the ratio $R_\gamma$ for Compton scattering is expected to be equal to $R_Z$. The comparison shown in Fig. 9 is made under the implicit assumption that the outgoing primary electron has an energy nearly equal to that of the incident photon. For photon energies above about 6 keV, this is definitely not the case, since Compton scattering dominates here and the outgoing primary electrons are low energy. Burgdoerfer et al. point out that the comparison should therefore be made between very high photon energy data and charged particle data for the same Q. For example, the data of Kamber et al., which are differential in Q, extend up to Q=16 a.u., for which an R of 1.7% was obtained. The corresponding photon energy which would produce a Compton electron with Q near 16 a.u. would be around 30 keV. Existing photon data suggest that $R_\gamma$ has saturated near 1.7% well before this limit is reached, in good agreement with the charged particle data. The comparison suggested by Burgdoerfer et al. seems thus to be consistent with experiment. The principle of plotting a universal curve of R versus the energy $\epsilon$ of the primary outgoing electron ($\epsilon \approx Q^2/2m$, where m is the electron mass) remains correct, but it is important to interpret the parameters of the experiment correctly in identifying that energy.

Recent measurements of $R_\gamma$ by Levin et al. [1991,1993] and Bartlett et al. [1994] for high energy photon ionization may show two limits. For photon energies below 4 keV, $R_\gamma$ seems to settle around 1.7%. For higher energy, where Compton scattering turns on, $R_\gamma$ takes a dip, then recovers to a value consistent with 1.7% once more. Recent many body perturbation theory calculations of Hino et al. [1994] predict exactly this kind of behavior. These calculations show that R approaches approximately 1.7% independent of whether photoionization or Compton scattering dominates. This value is approached in either case for outgoing primary electron energies above 1 keV or so. The two processes require quite different photon energies, however. Although no physical explanation of this result seems yet to have appeared in the literature, the observation that this occurs has important implications for the comparison between charged particle and photon data. It appears that the process whereby a fast primary electron is removed is really not important in determine what R will be. This almost certainly is responsible for the result that hard charged particle scattering and electron capture produce values of $R_Z$ and $R_C$ similar to $R_\gamma$ for the same Q. The remaining discrepancies are almost certainly due to the failure of the experiments to exclude multiple interactions with the charged projectiles. One might conclude that a fast moving beagle could gather one electron in its mouth and, provided it moved at $v >> 2$ a.u. and treated the rest of the He gently, would probably elicit a value of $R_B$ around 1.7%. However, the calculations of Burgdoerfer et al. [1994] and Suric et al. [1994] predict a high-energy value of $R_C$ for Compton scattering well below either the photoionization limit or that reached so far in differential charged particle experiments. No universality of $R(Q)$ would be predicted by these calculations, that is, not all processes are expected to give the same value of $R(Q)$.

## IV. ANALYSIS

### A. Collision Mechanisms

In fast collisions the reaction mechanisms are expected to be relatively simple because there is not much time for complicated (e.g. multi-step) processes to occur. For example, single ionization at high v is well described by first-order perturbation theory where the projectile (either a photon or a charged particle) simply knocks a target electron directly into the continuum in a single step. It is widely agreed that in the high-energy limit double ionization is caused by the electron-electron interaction in conjunction with a single interaction with the projectile. In this case the dynamics of the electron correlation interaction which determines the cross section for double ionization also determines ratio of double to single ionization. Thus the high-energy limit of this ratio provides opportunity to understand a relatively simple example of the dynamics of electron correlation.

#### *1. Shake*

Perhaps the easiest way to think about the ratio of double to single ionization is to regard double ionization as occurring as a final-state rearrangement of the electron cloud following single ionization [Aberg 1973]. If the electron cloud rearranges due to a change in the screening of the target nucleus, then the final-state eigenfunctions, $|s>$, (without screening) may be expressed as a linear combination of the initial-state eigenfunctions, $|s'>$ (with screening). In particular,

$$|f>=\sum_{s'}|s'><s'|f> \quad .$$

Here $|s'>$ is an eigenstate of the initial Hamiltonian with screening and $|f>$ is an eigenstate of the final Hamiltonian without screening. The probability amplitude for a state $|s'>$, produced before the initial state screening changes, to be in an eigenstate $|f>$ of the final state of the target ion is $<f|s'>$. The so called shake probability [Aberg 1972] is simply $|<f|s'>|^2$ which is the probability that $|s'>$ is in the state $|f>$ long after the projectile has left the target. If $|f>$ is a state in the continuum the process is shakeoff; if $|f>=|nlm>\neq|i>$, then the process is shakeup (or possibly shakedown), and it has been conjectured [McGuire et al. 1988] that if $|f>$ corresponds to transfer of an electron from the target to the projectile then shakeover may also occur. If the projectile leaves quickly, then this probability is independent of the process involved. If shakeover occurs it is expected that the ratio of transfer-ionization to single transfer (see Sec. III.A.2) will increase linearly with the projectile velocity in the high velocity limit non-relativistically.

#### *2. Simple Shake*

In simple shake the states $|i>$ and $|f>$ are products of single electron wavefunctions without exchange. In this simple case the simple shake probability is

$$P_{SimpleShake}=|<f'|i>|^2 \ . \tag{4}$$

All shakeoff process have the same ratio of double to single ionization. In helium, this ratio is easily calculated and found to be about 0.7%. This value lies between the observed photon limit of 1.7 % and the charged particle limit of 0.3 %. The simple shake limit agrees with neither of these observed values. At best it may usefully serve as an order of magnitude estimate of the ratio. Since other amplitudes contribute to the ratio, it is seldom, if ever, the case that the double ionization cross section is simply a factor times the single ionization cross section. At best simple shake usefully serves as an order of magnitude estimate of the ratio.

### *3. Generalized Shake*

Simple shake, using simple product wavefunctions without exchange, depends entirely on the electron-electron screening in the initial and final states. When more accurate wavefunctions are used, the results are often sensitive to electron correlation in these wavefunctions. The accuracy of the results is often significantly improved. Using exact wavefunctions Aberg [1976] introduced the generalized shakeoff probability defined by

$$P_{GeneralizedShake} = |a|^2 = |\int e^{i(\vec{Q}-\vec{k})\cdot\vec{r_1}} \phi_i^*(r_2, r_3, ...r_N)\Phi(r_1, r_2, r_3, ...r_N)|^2 a \ . \tag{5}$$

Here $\Phi(r_1, r_2, ...r_N)$ is the exact wavefunction for the initial state and $\Phi_f^*$ is approximated by $e^{-i\vec{k}\cdot\vec{r_1}}\phi_i^*(r_2, ...r_N)$ for a fast outgoing electron of momentum, $\vec{k}$. All multipoles are included in the $e^{i\vec{Q}\cdot\vec{r_1}}$ operator from the vector potential, $\vec{A}$. The generalized shake probability depends on $\vec{Q} - \vec{k}$, the momentum transferred to the residual target ion. It is the variable $\vec{Q} - \vec{k}$ that may be used to relate the generalized shake probability for different physical processes including, for example, ionization by charged particles, photoionization and ionization by Compton scattering.

### *4. MBPT Mechanisms*

There are various difficulties in discussing the question of mechanisms for double ionization. i) It is not always easy to calculate cross sections for double ionization accurately, and consequently it can be difficult to accurately test the influence of various mechanisms. ii) Atomic collisions are quantum mechanical in nature and the amplitudes for the mechanisms are often added before a square is taken to determine something which may be observed. So it is not always easy to untangle the various amplitudes. iii) Mechanisms are sometimes defined differently by different people so that it can be difficult to understand what is meant. iv) The mechanisms can depend on the gauge used, as discussed in the next subsection.

If one wishes to discuss mechanisms for quantum processes, then there are some reasons for using many body perturbation theory (MBPT) to define mechanisms for collision reactions. First, MBPT is well defined. While there is not always agreement on which diagrams (or mechanisms) are necessary, there is agreement on what the diagrams mean. The rules of computing an amplitude from a given MBPT diagram are well defined. Second, MBPT is widely used. Not only has MBPT been used for over 20 years in atomic physics, but is it used in most other fields of physics as well. On the other hand MBPT provides expansions in both the interaction potential and the correlation interaction for atomic collisions. It is not always the case that a perturbation expansion is applicable, especially in the case of correlation.

Despite the limitations of MBPT, it has been used to describe collision processes for both photon and charged particle impact. If we constrain our attention to first-order terms in the interaction of the projectile, then there are three possible diagrams for double ionization which contribute to first order in the correlation interaction. These are shown in Fig. 10.

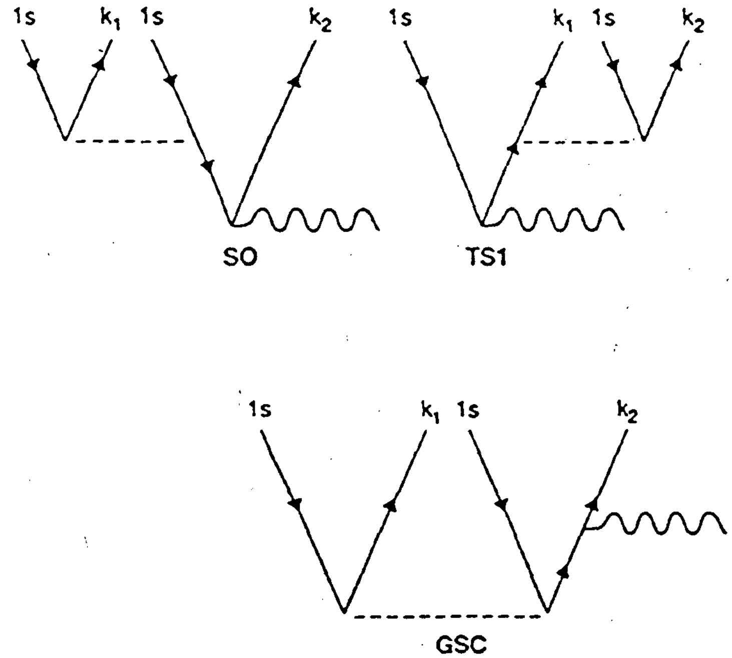



Fig. 10. Lowest order MBPT contributions to double ionization. Time increases in the upward direction and the ground state is not shown. The wavy line represents the interaction with the projectile and the dashed line is the electron-electron interaction. The electrons propagate upward and the holes propagate downward. SO denotes first-order shakeoff, GSC denotes ground-state correlation and TS1 denotes two step 1, meaning the there are two collisions of which 1 is with the projectile (the second is an electron-electron interaction on the way out of the collision).

Since two electrons are involved in double ionization, the simplest description is in terms of a second order (or two step) process. The TS1 corresponds to a collision with the projectile followed by an electron-electron interaction. GSC corresponds to interaction in the ground state followed by a collision with the projectile. SO corresponds to a particle-hole interaction (i.e. the electron is missing in the final state) following an interaction of the target with the projectile. TS1 and SO are sometimes called [Carter and Kelly 1981] final state correlation (FSC).

Many of the calculations for double ionization by photon impact have been done directly using MBPT diagrams for both single and double ionization [Carter and Kelly 1981, Ishihara et al. 1991, Hino et al. 1993, Pan and Kelly 1994]. In the case of charged particle impact no MBPT calculations have yet been completed. In the case of charged particle total cross sections, the outgoing electrons tend to leave slowly for the most part and a MBPT expansion in the electron-electron interaction may not be appropriate (see Sec. IV.A.1.

*5. Gauge Dependence of Mechanisms*

Over the past few years a dispute has developed over the importance of the TS1 mechanism for double photoionization of helium in the high-energy limit. Some theorists [Carter and Kelly 1981, Ishihara et al. 1991] argued that TS1 was the largest contribution, while others [Byron and Joachain 1967, Aberg 1972] argued that TS1 was negligible. Dalgarno and Sadeghpour [1992] resolved this conflict by pointing out that the TS1 amplitude has a different dependence on photon energy in the length, velocity and acceleration forms of the dipole matrix element. Grant [1974] had earlier noted that each form corresponds to a different gauge, and that the velocity form corresponds to the Coulomb gauge, i.e. $\nabla \cdot \vec{A}^V = 0$, where $\vec{A}^V$ is the vector potential in the velocity gauge. It easily confirmed that the quantum gauge transformations from the velocity to the length and acceleration gauges in the dipole limit are $\eta = \vec{r} \cdot \vec{A}^V(t)$ and $\eta = \frac{1}{c}\int^t dt' \vec{A}^V(t') \cdot \vec{p}$ respectively [Wang 1994]. Ishihara [Hino et al. 1993] confirmed this result and was further able to show that the sum of all first-order MBPT amplitudes is gauge invariant. Amusia and McGuire [1994] have conjectured that when all MBPT terms are summed, the summed amplitude is gauge invariant in each order of the interaction potential $V$. The argument is as follows. In an exact calculation gauge invariance requires that the full quantum amplitude is the same (including an overall phase, $e^{i\eta}$, which may be specified) so that one may write $a^V = a^L$, for example. Expanding in the strength,Z, of the interaction, $A^V(Z) = a_0^V + a_1^V Z + a_2^V Z^2 + ... + a_j^V Z^j + ... = A^L(Z) = a_0^L + a_1^L Z + a_2^L Z^2 + ... + a_j^L Z^j + ...$ Since Z may vary arbitrarily, the coefficients are equal, i.e. $a_j^V = a_j^L$. This gauge invariance is expected to hold in each order in the interaction potential. In any case it is clear that the MBPT amplitudes are gauge dependent. And it was noted that [Hino et al. 1993] 'the MBPT terms have no consistent meaning as mechanisms of double ionization unless one defines the form of the dipole interaction'.

We note that there are two types of gauge transformations: electrodynamic and quantum. In an electrodynamic gauge transformation $V \rightarrow V - d\lambda/dt$ together with $\vec{A} \rightarrow \vec{A} + \nabla\lambda$. In a quantum gauge transformation $\psi \rightarrow \psi' = e^{i\eta}\psi$. The form of the Schroedinger equation is unchanged if both transformations are done simultaneously [Schiff 1968]. Physical observables are invariant under both gauge transformations. Gauge dependence in any approximate theory is to be expected. All interaction potentials, V, vary with gauge. If V is changed the corresponding force changes and effects resulting from this force can change. Any first-order perturbation theory in V is likely to be gauge dependent.

This issue has not arisen in the case of the interaction of charged particles because it is conventional to use the Coulomb gauge in atomic physics. In this gauge the interaction between two electrons is $e^2/r_{12}$. In another gauge the correlation interaction will differ from $e^2/r_{ij}$. If one wishes to be consistent in questions relating to gauge, it may be sensible to think in a conventional gauge, i.e. to recognize the Coulomb gauge as the gauge in which to discuss physical mechanisms. The issue of the gauge dependence of interaction mechanisms deserves further discussion.

As the velocity,v, is reduced from its asymptotically large limit, it is possible for a projectile of charge Z to interact more than once with the target before it leaves the collision. It is then appropriate [McGuire and Straton 1994] to expand the probability amplitude for double ionization, $a$, in a Born series in $Z/v \sim \int V dt$, namely,

$$a = c_1 Z/v + c_2 (Z/v)^2$$

with the observable probability,

$$P = |a|^2 = |c_1|^2\, Z^2/v^2 + 2Re(c_1 c_2)\, Z^3/v^3 + |c_2|^2\, Z^4/v^4 \tag{6}$$

and resulting ratio for total cross sections (ignoring a slowly varying ln v term),

$$R_Z = \sigma^{++}/\sigma^{+} = \left(|C_1|^2 + 2Re(C_1 C_2) Z/v + |C_2|^2 Z^2/v^2\right) . \tag{7}$$

Here the C coefficients differ somewhat from the c coefficients due to the integration over impact parameters required to obtain cross sections from transition probabilities. If $Z/v$ is small enough there is a term linear in $Z$ in the ratio, $R_Z$, of double to single ionization since the double ionization probability contains a $(Z/v)^3$ term.

If there is no correlation, then $c_1 = 0$. This is obvious on physical grounds because without correlation the projectile (constrained to interact only once in first-order) could only ionize one electron which could not interact with the second electron. Thus the observable $Z^3$ terms depend on the dynamics of the electron correlation interaction.

In the limit of weak correlation the coefficient $c_1$ may be calculated in terms of SO, GSC and TS1 MBPT diagrams and $c_2$ will correspond to a MBPT diagram where the projectile interacts with each of the two electrons once and there is no further interaction. This is called TS2 and corresponds to the lowest order term in the independent electron approximation [McGuire 1992]. It has been shown [McGuire and Straton 1994, Stolterfoht 1994] that in this limit there is no $Z^3$ term in Eq.(6) because $c_1$ and $c_2$ are a factor of $i = \sqrt{(-1)}$ out of phase. It is necessary to include time ordering (which corresponds to time correlation, or energy-non-conserving intermediate states in a second-order theory) to produce the $Z^3$ terms. The meaning of the mechanisms here is the same as in the case of photon impact.

### B. Relation of photon impact and impact by charged particles

Now let us consider the relation between interactions of helium with photons and interactions with charged particles. A photon interacts with an atom via its electromagnetic field. A charged particle also interacts via an electromagnetic field. In principle, interactions of photons and charged particles are related. We shall soon make explicit this relationship within the framework of first-order perturbation of the interaction, V, with the projectile.

### 1. Photoionization and Compton Scattering

Photoionization and ionization by Compton scattering are different processes. In photoionization (i.e. the photo effect first described by Einstein), the photon is annihilated, while in Compton scattering the photon is inelastically scattered (and not annihilated). At present there is some disagreement as to what the high-energy limit is for the ratio $R_C$, of double to single ionization for Compton scattering. Some theorists [Hino et al. 1994, Amusia 1994] have suggested that $R_C = R_\gamma$ since the final wavefunctions are similar in the high-energy limit. Others [Burgdoerfer and Andersen 1994, Suric et al. 1994] disagree. As in Aberg's generalized shake description described above, Suric et al. [1994] have used $\Phi_f = e^{i\vec{k}\cdot\vec{r_1}}\phi(r_2, ...r_N)$ and predicted for ionization via Compton scattering that,

$$R_C = 1 - \sum_{bound \quad states} \int d^3r_1 | \int \phi_i^*(r_2)\Phi(r_1, r_2)d^3r_2|^2 \ . \tag{8}$$

This may be compared to the expression from Dalgarno and Sadeghpour [1992] for photoionization that,

$$R_\gamma = 1 - \frac{\sum_{bound \quad states} |\phi_i^*(r_2)\Phi(r_2, r_1 = 0)d^3r_2|^2}{\int |\Phi(r_2, r_1 = 0)|^2 d^3r_2} \ . \tag{9}$$

In the simple shake limit $\Phi = \phi_1\phi_2$ (without exchange), then both ratios reduce to the same simple shake ratio, which for ionization (not including bound states) gives 7.1% if s = 5/16, corresponding to a variational hydrogenic wavefunction with s chosen to minimize the binding energy. For more complete wavefunctions, however, (8) and (9) differ because photoionization depends on the value of $\Phi$ at $r_1 = 0$ while for Compton scattering there are contributions from all values of $r_1$. At present the value of $R_C$ in the high-energy limit is not known experimentally.

It is widely agreed that total ionization cross sections for photoionization are well described by the dipole approximation for the interaction operator with the photon even at high photon energies where $k_i r > 1$. In Compton scattering, on the other hand, the contributions are mostly non-dipole.

### 2. Non-dipole limit

For Compton scattering the cross section for inelastic scattering from an arbitrary initial state $|i>$, to an arbitrary final state, $|f>$, may be expressed [Burgdoerfer et al. 1994] in first order (in both the fine structure constant, $\alpha$, and $1/c^2$) as,

$$\begin{aligned} \frac{d\sigma_C}{d\Omega_f} &= r_{cl}^2 \, \frac{k_f}{k_i} \, |\vec{\lambda}_i \cdot \vec{\lambda}_f^*|^2 \, | < f|A^2|i > |^2 \\ &= r_{cl}^2 \, \frac{(1 + \cos^2\theta_f)}{2} \, | < f|e^{i\vec{Q}\cdot\vec{r}}|i > |^2 \end{aligned} \tag{10}$$

where $r_{cl}$ is the classical radius of the electron, $\vec{k}_{i,f}$ is the initial or final photon momentum, $\vec{\lambda}_{i,f}$ is the polarization vector of the initial or final photon, $\vec{A}$ is the vector potential and

$(\vec{Q})^2 = (\vec{k}_i - \vec{k}_f)^2 = k_i^2 + k_f^2 - 2k_i k_f \cos\theta_f \approx 2k_i^2(1-\cos\theta_f)$. The expression in the second line above is for unpolarized light.

For the impact of a high velocity particle of charge Z the cross section in the plane-wave Born approximation [McDowell and Coleman 1970] is given by,

$$\frac{d\sigma_Z}{d\Omega_f} = a_0^2 \, \frac{K_f}{K_i} \frac{\mu^2}{4\pi^2} \; |<f|\frac{Z}{|\vec{R}-\vec{r}|}|i>|^2$$
$$= a_0^2 \, \frac{4\mu^2 Z^2}{Q^4} \; |<f|e^{i\vec{Q}\cdot\vec{r}}|i>|^2 \tag{11}$$

where $a_0$ is the radius of the ground state of hydrogen, $\vec{K}_{i,f}$ is the initial or final photon momentum, now $\vec{Q} = \vec{K}_i - \vec{K}_f$ is again the momentum transfer of the projectile and $\mu$ is the reduced mass of the system. We note that in this approximation no momentum is transferred to the projectile by the target nucleus, so that the momentum transfer, $\vec{Q}$, here is the momentum transferred to the electron(s).

From Eqs.(10) and (11) it is apparent that both the Compton and the charged particle cross sections above are proportional to the generalized oscillator strength [McDowell and Coleman 1970], $f_{fi}(Q) = \Delta E_{fi}/Q^2| < f|e^{i\vec{Q}\cdot\vec{r}}|i > |^2$. Consequently, the cross section for Compton scattering may be expressed in terms of the first Born cross section for scattering by charged particles. Using $1 + cos^2\theta_f = 1 + (\frac{k_i^2+k_f^2-Q^2}{2k_i k_f})^2$ and $K_i^2/\mu^2 = v^2$ where $v$ is the velocity of the incident charged particle, we obtain from Eq(10) and Eq(11) a useful relation [Burgdoerfer et al. 1994], namely,

$$\frac{d\sigma_C}{dQ} = \frac{r_{cl}^2}{a_0^2} \, \frac{v^2}{8k_i^2 Z^2} \, \left(1 + (\frac{k_i^2+k_f^2-Q^2}{2k_i k_f})^2\right) \, Q^4 \, \frac{d\sigma_Z}{dQ} \; . \tag{12}$$

This relation is valid for arbitrary final states, $|f>$, including both single and double ionization. This relation could be tested experimentally by observing differential cross sections for Compton scattering and comparing them to existing data for differential cross sections by charged particle impact weighted by the factors in the above relation. This relation may also be used to evaluate cross sections for Compton scattering by modifying existing computer codes for single and double ionization by charged-particle impact.

Let us now use the above relation to express the ratio of double to single ionization cross sections by Compton scattering in terms of a corresponding cross section ratio by charged particles. We now assume that the Compton scattering cross section at sufficiently high (but non-relativistic) photon energies is dominated by contributions from large Q, i.e., $Q >> r_{target}^{-1}$, where $r_{target}$ is the radius of the target electron. We also assume that the ratio of double to single ionization, $R_Z(Q) = \frac{d\sigma_Z^{++}}{dQ}/\frac{d\sigma_Z^{+}}{dQ}$, tends to a constant value for charged particles at large values of Q. Then from the above relation we have,

$$R_C = \frac{\sigma_C^{++}}{\sigma_C^{+}} = \frac{\int (d\sigma_C^{++}/dQ)\, dQ}{\int (d\sigma_C^{+}/dQ)\, dQ}$$
$$= \frac{\int \left(1 + (\frac{k_i^2+k_f^2-Q^2}{2k_i k_f})^2\right) \, Q^4 \, (d\sigma_Z^{++}/dQ)\, dQ}{\int \left(1 + (\frac{k_i^2+k_f^2-Q^2}{2k_i k_f})^2\right) \, Q^4 \, (d\sigma_Z^{+}/dQ)\, dQ}$$

$$\approx \frac{\int \left(1+(\frac{k_i^2+k_f^2-Q^2}{2k_ik_f})^2\right) \; Q^4 \; R_Z(Q\approx\infty) \; (d\sigma_Z^+/dQ) \; dQ}{\int \left(1+(\frac{k_i^2+k_f^2-Q^2}{2k_ik_f})^2\right) \; Q^4 \; (d\sigma_Z^+/dQ) \; dQ}$$

$$= R_Z(Q\approx\infty) \equiv \left(\frac{d\sigma_Z^{++}}{dQ} / \frac{d\sigma_Z^+}{dQ}\right)_{Q\approx\infty} . \tag{13}$$

Note that the $R_Z(Q)$ used above differs from the $R_Z = \sigma_Z^{++}/\sigma_Z^+$ for the ratio of total cross sections used in section III.A, and that the $R_Z(Q)$ of Eq.(13) is discussed in section III.B. Here Q is the momentum transferred by the target electron(s) with no internuclear momentum transfer. The status of related experiments is discussed in Sec. III.B above. We encourage further experiments of this type.

*3. Dipole limit*

If the momentum transfer, Q, is small, then $< f|e^{i\vec{Q}\cdot\vec{r}}|i > \approx i < f|\vec{Q}\cdot\vec{r}|i >$, and the generalized oscillator strength reduces to the standard dipole optical oscillator strength. In this limit the matrix elements reduce to the dipole matrix elements used for for photo-excitation and ionization. For charged particle impact, if the Born cross section is expanded in inverse powers of the projectile energy, E, then $\sigma_Z(E) \simeq A\frac{\ln E}{E} + B/E$. The leading $\frac{\ln E}{E}$ (or $\frac{\ln v^2}{v^2}$) contribution to the cross section for charged particles (which is absent both in $d\sigma_Z/dQ$ used above and classically) may be expressed in terms of the cross section for photo-excitation or ionization [Byron and Joachain 1967], namely,

$$\frac{d\sigma_Z}{d\epsilon} = \frac{Z^2}{2\pi\alpha} \frac{\ln v^2}{v^2} \frac{\sigma_\gamma(\epsilon)}{\epsilon} \tag{14}$$

where $v$ is the velocity of the projectile, $\epsilon$ is the energy of the (faster) ejected electron, and $\alpha$ is the fine structure constant. If Q is near $Q_{min}$, then $Q \approx Q_{min} = K_i - K_f = \Delta E_{if}/2v = (I+\epsilon)/2v$, where I is the atomic binding energy. Using this limit, it is straightforward to express the ratio for double to single ionization by charged-particle impact as an integral over the cross section ratio for photoionization [McGuire 1984, Manson and McGuire 1994], namely,

$$R_Z = \int R_\gamma(\epsilon)\rho_Z^+(\epsilon)d\epsilon \tag{15}$$

where $\rho_Z^+(\epsilon) = \frac{1}{\sigma_Z^+}\frac{d\sigma_Z^+(\epsilon)}{d\epsilon}$ is the density of states for single ionization by a charged particle. This relation has been recently used by Manson Manson and McGuire 1994 to obtain the observed ratio of double to single ionization in helium by charged particle impact, $R_Z = 0.26\%$, from the observed ratios for photoionization. Manson has noted two discrepancies. Using the values quoted in the literature for $R_\gamma$, the asymptotic value obtained for $R_Z$ using the above relation is 0.32% somewhat above the observed value of $0.26\% \pm 0.03\%$. Also the high-energy shape of $R_Z(E)$ given by Eq.(15) differs somewhat both from experiment and the Forced Impulse Method calculation of Reading and Ford [1989]. Thus, confirmation of the predicted relation between charged particle impact and impact of photons is not quite complete.

## V. SUMMARY

We have reviewed recent observations of the ratio of cross sections, $R = \sigma^{++}/\sigma^{+}$, for single to double ionization of helium interacting with high-energy photons and charged particles. For photon impact at energies below 6 keV the predominant means of ionization is photoionization (i.e., the Einstein photo-effect) in which the incident photon is annihilated. The high energy limit of this ratio is $1.7\% \pm 0.1\%$ in agreement with theory. At photon energies above 6 keV ionization is dominated by Compton scattering, where the incident photon is inelastically scattered. The high-energy limit of this ratio has not been observed and various calculations give values between the 1.7% limit for photoionization and 0.7%. For impact of charged particles and anti-particles the ratio R of total cross sections has been established by observation as $0.26\% \pm 0.03\%$. If the energy transfer of the collision is fixed, values of R in the neighborhood of 2% have been observed for large energy transfer. Theoretically cross section ratios for photons and charged particles have been related. In the dipole limit photoionization has been related to charged particle impact at a fixed energy transfer. In the limit in which many multipole terms contribute, the ratio in first order theory for charged particles has been related to the ratio for Compton scattering at a fixed momentum transfer. Experimental confirmation of the predicted relations between charged particle impact and impact of photons is not complete even for helium.

## VI. ACKNOWLEDGMENTS

We gratefully acknowledge discussion with R. Pratt, T. Aberg. M. Amusia, S. Manson, L. Nagy, A.F.Starace and J. Wang. This was supported in part by the Division of Chemical Sciences, Office of Basic Energy Science, Office of Energy Research, U.S. Department of Energy, under contracts DE FG05 92ER14286 and DE FG02 92ER14299, and the National Science Foundation under grant PHY-9017248.

## REFERENCES


* Department of Physics, Tulane University, New Orleans, LA 70118-5698

** Department of Physics, Western Michigan University, Kalamazoo, MI 49008

*** Los Alamos National Laboratory, Los Alamos, NM 87545

† Department of Physics, University of Nebraska, Lincoln, NE 68588-0111

†† Department of Physics, Kansas State University, Manhattan, KS 66506

††† Advanced Light Source, MS 2-400 Lawrence Berkeley Laboratory, University of California, Berkeley, CA 94720



Aberg T 1970 *Phys. Rev. A* **2** 1726

Aberg T 1972 in *Proceedings of the International Conference on Inner Shell Ionization Phenomena and Future Applications* Atlanta, Georgia; Fink R, Manson S T, Palms I M, and Rao R V, eds. CONF-720 404 (U S Atomic Energy Commission, Oak Ridge, TN 1973) 1509

Aberg T 1973 *USAEC Rept. CONG-720404 1509* Atlanta Mtg.,Inner Shell Processes

Aberg T 1976 in *Photoionization and Other Probes of Many Electron Interactions*(Plenum Press) Wuilleumier F,ed. 49

Amusia M, Drukarev E G, Gorshkov V G, and Kazachkov M P 1975 *J. Phys. B* **8** 1248

Amusia M 1994 private communication

Amusia M and McGuire J H 1994 unpublished

Andersen L H, Hvelplund P, Knudsen H, Miller S P, Elsener K, Resfelt K-G, and Uggerhoj E 1986 *Phys. Rev. Lett.* **57** 2147

Andersen L H, Hvelplund P, Knudsen H, Miller S P, Sorensen A H, Elsener K, Rensfelt K-G, and Uggerhof E 1987 *Phys. Rev. A* **36** 3612

Andersson L R and Burgdoerfer J 1993 *Phys. Rev. Lett.* **71** 50

Azuma Y, Berry H G, Gemmel D S, Suleiman J, Westerlind M, Sellin I A, Woicik J C, and Kirkland J 1994, submitted to Phys. Rev. A.

Bartlett R J, Sagurton M, Samson J A R and He Z X 1994 *J. Opt. Soc. Am.* (to be published)

Bartlett R J, Walsh P J, He Z X, Chung Y, Lee E M, and Samson J A R 1992 *Phys. Rev. A* **46** 5574

Baym G. 1974 *Lectures on Quantum Mechanics* (Addison Wesley, Inc. NY), Eq (13-132)

Bearden A J 1966 *J. Appl. Phys.* **37** 1681

Berg H et al 1992 *J. Phys.* **B25**

Berg H, Jagutzki O, Doerner R, DuBois R D, Kelbch C, Schmidt-Boecking H, Ullrich J, Tanis J A, Schlachter A S, Blumenfeld L, d'Etat B, Hagmann S, Gonzales A, and Quinteros T 199~~1~~2 *Phys. Rev. A* **46** ~~in press~~ 5539

Berrah N, Heiser F, Wehlitz R, Levin J, Whitfield S B, Viefhaus J, Sellin I A, and Bekcker U 1993 *Phys. Rev. A* **48** R1733

Burgdoerfer J, Andersson L R, McGuire J H, and Ishihara T 1994 *Phys. Rev. A* **50** 349

Brown R L 1979 *Phys. Rev. A* **1** 586

Byron F W Jr and Joachain C J 1966 *Phys. Rev. Lett.* **16** 1139

Byron F W and Joachain C J 1967 *Phys. Rev.* **164** 1

Carlson T A 1966 *Phys. Rev.* **156** 142

Carter S L and Kelly HP 1981 *Phys. Rev. A* **24** 170

Charlton M, Andersen L H, Brun-Nielsen L, Deutsch B I, Hvelplund P, Jacobsen F M, Knudsen H, Lariccha G, Poulsen M R, and Pedersen J O 1988 *J. Phys. B* **21** L545

Cocke C L, Kamber E Y, Cheng, S and Varghese S L 1989 *Nucl. Instru. Meth. B* **42** 545

Cullen D E, Chen M H, Hubbell J H, Perkins S T, Plechaty E R, Rathkopf J A, and Scofield J A *Lawrence Livermore National Laboratory Report UCRL-50400* **6** Part A Rev 4 (Nat. Tech. Info. Service, US Dept. Commerce, Springfield, VA)

Dalgarno A and Sadeghpour H R 1992 *Phys. Rev. A* **46** R3591

Doerner R, Horsdal-Pedersen E, and Cocke C L 1993 *ICPEACXVII contr. abstracts*

DuBois R D and Toburen L H 1988 *Phys. Rev. A* **38** 3960

Fang X and Reading J F 1991 *Nucl. Inst. Meth. B* **53** 453

Ford A L and Reading J F 1990 *J. Phys.* **B23** 2569

Forest J L, Tanis J A, Ferguson S M, Haar R R, Lifrieri K, and Plano V L *Phys. Rev. A* to be published

Giese J P and Horsdal E 1988 *Phys. Rev. Lett.* **60** 2018

Grant I P 1974 *J. Phys. B* **7** 1458

Haugen H K, Andersen L H, Hvelplund P, Knudsen H 1982 *Phys. Rev. A* **26** 1950

Heitler W *The Quantum Theory of Radiation* (Oxford University Press, London 195) **Sec 5** Eq(4) and **Sec 22** Eq(37)

Heber O, Bandong B B, Sampoll G, and Watson R L 1990 *Phys. Rev. Lett.* **64** 851; 1991 *Nucl. Instrum. Meth. Phys. Rev. B* **56/57** 15

Hino K, Bergstrom P, and Macek J H 1994 *Phys. Rev. Lett.* **72** 1620

Hino K 1993 *Phys. Rev. A* **47** 4845

Hino K,Ishihara T, Shimizu F, Toshima N, and McGuire J H 1993 *Phys. Rev. A* **48** 1271

Holland D M P, Codling K, West J B, and Marr G V 1979 *J. Phys. B* **12** 2465

Horsdal-Pedersen E H and Larsen L 1979 *J. Phys. B* **12** 4099

Hubbell J H, Veigele W J, Briggs E A, Brown R T, Cromer D T, and Howerton R J 1975 *J. Phys. Chem. Ref. Data* **4** 471

Inokuti M 1971 *Rev. Mod. Phys.* **43** 297

Inokuti M 1978 *Rev. Mod. Phys.* **50** 23

Ishihara T, Hino K, and McGuire J H 1991 *Phys. Rev. A* **44** R6980

Kamber E Y, Cocke C L, Cheng S, and Varghese S L 1988 *Phys. Rev. Lett.* **60** 2026

Kossmann H, Schmidt V, and Andersen T 1988 *Phys. Rev. Lett.* **60** 1266

Knudsen H, Andersen L H, Hvelplund P, Sorensen J, and Ciric D 1987 *J. Phys. B* **20** L253

Knudsen H, Andersen L H, Hvelplund P, Astner G, Cederquist H, Danared H, Liljeby L, and Rensfelt K G 1984 *J. Phys. B* **17** 3545

Kristensen F G and Horsdal-Pedersen E 1991 *J. Phys. B* **23** 4129

Lablanquie P, Ito K, Morin P, Nenner I, and Eland J H D 1990 *Z. Phys. D* **16** 77

Levin J C, Lindle D W, Keller N, Miller R D, Azuma Y, Berrah N, Berry H G, and Sellin I A 1992 *Phys. Rev. Lett.* **67** 968

Levin J C, Sellin I A, Johnson B M, Lindle D W, Miller R D, Berrah N, Azuma Y, Berry H G, and Lee D H 1993 *Phys. Rev. A* **47** R16

Levinger J S 1953 *Phys. Rev.* **90** 11

Manson S T and McGuire J H 1994 *Phys. Rev. A* to be published.

McCrary J H, Looney L D, and Atwater H F 1970 *J. Appl. Phys.* **41** 3570

McDowell M R C and Coleman J P 1970 *Introduction to the Theory of Ion-Atom Collisions* (North-Holland, NY)

McGuire J H 1982 *Phys. Rev. Lett.* **49** 1153; and Proceedings of the Second US-Mexico Symposium on Atomic and Molecular Physics: Two-Electron Phenomena, Cocoyoc, Mexico, 1986, published in 1987 *Notas de Fisica* Alvarez I, Cisneros C, de Urquijo J, and Morgan TJ eds. (Instituteo de Fisica, UNAM, Laboratorio Cuernavaca, Mor., Mexico,), **Vol. 10, No. 2** 129-142

McGuire J H 1984 *J. Phys.B* **17** L779

McGuire J H 1992 *Adv. Atomic, Molecular, and Optical Physics* **29** 217

McGuire J H 1995 *Handbook of Atomic, Molecular and Optical Physics* (American Institute of Physics) Drake G, ed. Chapter 40

McGuire J H, Deb N C, Atkas Y, and Sil N C 1988 *Phys. Rev. A* **38** 3333

McGuire J H, ~~Miller~~ Müller A, Schuch B, Groh W, and Salzborn E 1987 *Phys. Rev. A* **35** 2479

McGuire J H and Straton J C 1994 *Nucl. Instru. Meth. B* **87** 17

Meng L ,Olson R E, Doerner R, Ullrich J, and Schmidt-Boecking H 1993 *J. Phys. B* **26** 3387

Mittleman M H 1966 *Phys. Rev. Lett.* **16** 49"

Olson R E, Ullrich J, Doerner J, and Schmidt-Boecking H 1989 *Phys. Rev. A* **40**

Peart B, Walton D S, and Dolder K T 1969 *J. Phys. B* **2** 1247

Pan C and Kelly H P 1994 *Phys. Rev. A* submitted

Reading J F and Ford A L 1987 *Phys. Rev. Lett.* **58** 543

Reading J F and Ford A L 1987 *J. Phys. B* **20** 3747

Reading J F and Ford A L 1988 *J. Phys. B* **21** L685

Reading J F, Ford A L, and Fang X 1989 *Phys. Rev. Lett.* **62** 245

Salin A 1991 *J. Phys. B* **24** 3211

Samson J A R 1990 *Phys. Rev. Lett.* **65** 2861

Samson J A R, Bartlett R J, and He Z X 1992 *Phys. Rev. A* **46** 7277

Samson J A R, Greene C H, and Bartlett R J 1993 *Phys. Rev. Lett.* **71** 201

Samson J A R, He Z X, Bartlett R J, and Sagurton M 1994a *Phys. Rev. Lett.* **72** 3329

Samson J A R, He Z X, Yin l, and Haddad J 1994b *J. Phys. B* **27** 887.

Schiff L I 1968 *Quantum Mechanics* (McGraw-Hill, NY) Chapter 11

Schlachter A S and Tanis J A 1994 Phys. Rev. Lett. 73 3596.

Schmidt V, Sandner N, Kuntzem Hüller Dhez P, Wuilleumier F, and Källne E 1976 *Phys. Rev. A* **13** 1748

Schmidt-Boecking H 1994 private communication

Stolterfoht N, Mattis A, Schneider D, Schiweitz G, Skogvall B, Sulik B, and Rizc S 1993 *Phys. Rev. A* **48** 2986

Stolterfoht N, Mattis A, Schneider D, Schiweitz G, Skogvall B, Sulik B, and Rizc S 1994 submitted to *Phys. Rev.*

Stolterfoht N 1994, private communication.

Straton J C, McGuire J H, and Chen Z, Zheng 1992 *Phys. Rev. A* **46** 5514

Suric T, Pisk K, Logan B A, and Pratt R H 1994 *Phys. Rev. Lett.* **73** 790

Shah M B and Gilbody H B 1985 *J. Phys.B* **18** 899

Tang Z and Shakeshaft R 1993 *Phys. Rev. A* **47** R3487

Tanis J A, DuBois R D, and Schlachter A S 1992 *Phys. Rev. Lett.* **68** 897

Tanis J A, Bernstein E M, Clark M W, Ferguson S M, and Price R N 1991a *Phys. Rev. A* **43** 4723

Tanis J A, Ferguson S M, Forest J L, Haar R R, Lifrieri K, and Plano V L 1991b *Atomic and Molecular Physics: Third US/Mexico Symposium* Cisneros C, Alvarez I, and Morgan T J, eds. (World Scientific), Singapore 83-95

Ullrich J, Doerner R, Berg H, Cocke C L, Euler J, Froschauer K, Hagmann S, Jagutzki O, Lencinas S, Mann R, Meergel V, Moshammer R, Schmidt-Boecking H, Tawara H, and Unverzagt M 1994 *Phys. Rev. Lett.* in press

Ullrich J, Moshammer R, Berg H, Mann R, Tawara H, Doerner R, Euler J, Schmidt-Boecking H, Hagmann S, Cocke C L, Unverzagt M, Lencinas S, and Mergel V 1993 *Phys. Rev. Lett.* **71** 1697

Vegh L and Burgdoerfer J 1990 *Phys. Rev. A* **42** 655

Veigele W J 1973 *Atomic Data* **5** 51

Wang J 1994 private communication

Wannier G H 1953 *Phys. Rev.* **90** 817

Wehlitz R, Heiser F, Hemmers O, Langer B, Menzel A, and Becker U 1991 *Phys. Rev. Lett.* **67** 3864

Wight G R and Van der Wiel M J 1976 *J. Phys. B* **9** 1319

Wuilleumier F J 1982 *Ann. Phys. Fr.* **4** 231

Wang Y D, Straton J C, McGuire J H and DuBois R D 1990 J. Phys. B 23 L133

LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
TECHNICAL AND ELECTRONIC
INFORMATION DEPARTMENT
BERKELEY, CALIFORNIA 94720