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AN ANALYSIS OF DATA ON DOUBLE TRANSITIONS IN He^+

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This report aims at setting up a net of inter-comparisons that may serve as a framework for further discussion or design of experiments. The numerical values presented here are extremely tentative, especially in Sec. 2, being intended only to exemplify the relationships that may be considered. Since we plan to keep this subject under continuing review, we shall depend heavily on contributions by comment, criticism and additional information.

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In this report we summarize and compare information from various experimental as well as theoretical sources on the probability of double electron jumps in He. These sources are :

a) Photon bombardment

1. Discrete absorption lines observed by Madden and Codling (MC)¹ and related theoretical data by Burke, et al and others.²
2. Ratios of double to single ionization at various photon energies and spectra of secondary electrons for 277 eV photons by Carlson (Ca)³.
3. Theoretical estimates of double processes by Salpeter and Zaidi (SZ)⁴.

b) Particle bombardment

4. Yield of double ionization by electron bombardment in the range 0.3 - 15 keV by Schram et al⁵ as well as by earlier workers.^{6,7}
5. Yield of He⁺ fluorescence by electron bombardment of He in the range ~ 0.1 to 5 keV by St. John et al⁸, deHeer et al⁹ and by Hughes and Weaver¹⁰.

6. Yield of autoionization electrons from H^+ , H_2^+ bombardment of He at 75 keV by Rudd¹¹.
7. Yield of autoionization electrons by electron impact at 4 keV on He by Mehlhorn¹².
8. Yield of double excitations by electron impact at 90 - 500 eV detected by spectrum of energy loss of scattered electrons with $\sim 0^\circ$ deflection (differential in angle) by Silverman and Lassetre (SL)¹³ and Simpson et al¹⁴.

Bombardment by sufficiently fast particles (which often include electrons of ~ 1 keV) generally produces primarily dipole transitions, whose yield can be compared with the yield obtained by photon bombardment. However the double ionizations produced by particles will be shown to arise probably mostly by non-dipole mechanisms, as first suggested by Schram's results⁵. Consequently the evidence from a) and b) may not be directly comparable.

Values of the oscillator strength corresponding to the total photoabsorption throughout the continuum are obtained from recent experiments by Lowry et al (LET)¹⁵ and by Lukirskii et al (LBZ)¹⁶ and have been reduced to a standard curve by Mignerone and Levinger (ML)¹⁷.

1. Oscillator Strengths.

The value of the oscillator strength of the transition $1s^2 \rightarrow 2s2p$ determined by Fano and Cooper (FC)¹⁸ from the experimental data of (MC) is 0.0057. This value is in agreement ($\sim 10\%$) with calculations from first principles². The oscillator strengths of the following lines of the (sp2n+) series appear to fit in a Rydberg progression beginning with $2s2p$ (i.e., $n=2$), namely to vary in proportion to the hydrogenic law $2/(n-\sigma)^3$ where σ is the observed quantum defect. [The recommendation of (FC) that the widths and strengths of successive lines should be in the ratio of line intervals, namely $2(n-\sigma)/[(n-\sigma)^2-1]^2$, is now regarded as incorrect.] Accordingly the oscillator strengths of successive lines of the $2n+$ series are estimated to be

$n =$	2	3	4	5	>5	Total
$10^3 f_n =$	5.7	1.2	0.5	.2	.3	8

The oscillator strengths of the other series in the same range, $2n-$ and " nd ", are lower by two or more orders of magnitude and, accordingly, negligible for our purposes.

The spectral density of df/dE at the continuum threshold of the $sp2+$ channel can be estimated by extrapolation of the f_n given above¹⁸, being equal to $f_n/[2/(n-\sigma)^3]$ if E

is expressed in Rydberg. Thus we have

$$[df_{sp2+}/dE]_{E=65.4\text{eV}} = 0.012 \text{ (Ry)}^{-1} = 0.001(\text{eV})^{-1} \quad (1)$$

Since the total oscillator strength for all transitions at this energy is

$$[df_{tot}/dE]_{E=65.4\text{eV}} = 0.128 \text{ (Ry)}^{-1} \quad (2)$$

the strength of double transitions at this energy amounts to about 10% of the total.

In order to estimate the integrated strength for double photoionizing transitions which leave He^+ excited at $n=2$, we require at least a second value corresponding to (1) at an energy substantially higher than 65.4 eV. The only experimental information available for this purpose is provided by Carlson's³ finding that (6 ± 1) percent of the photoelectrons ejected by the C_K line (277 eV) have an energy corresponding to excitation of He^+ to $n=2$. [To this accuracy it does not matter that 6% represents the $(n=2)/(n=1)$ ratio rather than $(n=2)/(\text{total})$.] The SZ calculations yield for the ratio $[df_{2s, \epsilon p}/dE]/[df_{1s, \epsilon p}/dE]$ just the experimental values of the $(n=2)/(n=1)$ ratio at 65.4 and at 277 eV. Therefore, the values of this SZ ratio at all other energies were adopted as representative of the $(n=2)/(n=1)$ ratio. The standardized ML spectrum of df/dE was multiplied by this ratio and integrated from 65.4 eV to infinity to yield

$$f_{n=2, E>65.4\text{eV}} = 0.03_3 \quad (3)$$

Addition of the discrete spectrum contribution given above yields

$$f_{n=2} = 0.04_1 \quad (4)$$

Consider now possible methods of estimating the ratio $f_{n>2}/f_{n=2}$. For 277 eV photons we have the experimental ratio³ 0.5. If the distribution of n values of the He^+ were to follow the $1/n^3$ law corresponding to the values of H-like wave functions at the nucleus [$R_{n0}^2(0) = 4/n^3$], the ratio would be $0.077/0.125 \sim 0.6$. On the other hand the "sudden approximation" estimates obtained by projecting $\text{He}(1s)$ wave function $\exp(-27r/16a)$ onto R_{30} and R_{20} of He^+ are in a ratio of 0.167, which may be compared to $0.037/0.125 = 0.30$ from the $1/n^3$ law. On the whole we regard the experimental value for 277 eV as fairly representative and take

$$f_{n>2} \sim 0.5 \times 0.04_1 = 0.02 \quad (5)$$

With regard to double ionization, two kinds of experimental data are provided by (Ca). On the one hand, the ion production ratio $\text{He}^{++}/\text{He}^+$ measured by charge spectrometer increases from 0.023 near the 79 eV threshold to 0.035 at ~ 225 eV and thereafter remains constant up to 600 eV. On the other hand, at 277 eV the integrated number of photo-

electrons from double ionization amounts to $(4.5 \pm 2)\%$ of the total. We have taken the df_{tot}/dE values of ML for various energies, multiplied them by the $\text{He}^{++}/\text{He}^+$ ratio at those energies and integrated from 79 eV to infinity. The result

$$f_{++} \sim 0.007 \quad (6)$$

appears a little low as compared to (4) and (5). To some extent double photoionizations are made less likely by the higher threshold (79 eV against 60 eV) for double excitation; yet the ratio of total strengths $f_{>79\text{eV}}/f_{>60\text{eV}}$ amounts to $\sim 2/3$. Pending additional data we set

$$f_{++} = 0.01 \quad , \quad (6')$$

and, therefore,

$$f_{\text{all doubles}} = 0.07 \quad . \quad (7)$$

2. Particle Collisions.

a) Double ionization. The experimental data on the production of double ionization in He by electron collisions^{5,6,7} can be summarized very roughly by the following statement: In the energy range from ~ 300 to ~ 1000 eV the production ratio $\text{He}^{++}/\text{He}^+$ is 0.0055 ± 0.001 ; from 1 to 15 keV this ratio decreases gradually by a factor of ~ 2 . We wish to compare this result with predictions based on the assumption that most of this ion yield derives from dipole processes ($\Delta l=1$) in accordance with Born's approximation theory. According to this theory the cross section for single or double ionization should (at sufficiently high energy) obey the law

$$\sigma T / 4\pi a^2 I_H \sim [f I_H / \langle E \rangle] \log(T/E_{\text{thr}}) + C \quad (8)$$

where T is the collision energy, $\langle E \rangle$ a mean energy transfer in the collisions under consideration, E_{thr} the threshold for these collisions, and C is a constant that includes the contribution of other processes. The assumption we wish to test is that C contributes little to the experimental value of (8) in the range we are considering.

The ratio of the coefficients $[f I_H / \langle E \rangle]$ for the production of He^{++} and He^+ can be estimated as follows. The value of f for production of He^{++} is $\sim .01$ according to (6'),

the corresponding value of $\langle E \rangle_{++}/I_H$ is

$$f_{++} / \int_{79\text{eV}}^{\infty} (df_{++}/dE) I_H dE/E = 8 \quad (9)$$

The corresponding values for production of He^+ are $f_+ = 1.6$ and $\langle E \rangle_+/I_H = 3.3$. Therefore

$$[f/\langle E \rangle]_{++}/[f/\langle E \rangle]_+ = .01 \times 3.3/1.6 \times 8 = 0.0026. \quad (10)$$

Since the ratio of $\log (T/E_{\text{thr}})$ for the two processes is 0.7 at $T = 1$ keV, the ratio σ_{++}/σ_+ should be $0.0026 \times 0.7 = 0.0018$ at this energy if C were negligible, whereas its experimental value is ~ 0.005 .

In fact Eq. (8) is fulfilled by σ_+ rather well for $T > 1$ keV but σ_{++} is found by both Ref. 5 and Ref. 7 to remain constant in this range instead of increasing linearly with $\log T$. These results disagree with the assumption we wished to test but would be consistent with theory under the opposite assumption that the main contribution to σ_{++} at $T \sim 1$ keV arises from non-dipole or non-Born processes.

b) Fluorescence from $n=4$ levels of He^+ . Observation of this fluorescence (emitted in transitions to $n'=3$ or possibly $n'=1,2$), following electron collisions with neutral He, implies the initial occurrence of a double transition in

which one electron has been ejected and the other raised to $n=4$. (The role of cascades from $n>4$ is ignored here.) Cross sections for the production of this fluorescence (refs. 8,9,10) are of the order of 10^{-21} - 10^{-20} cm^2 and vary by factors ≤ 2 from one experiment to another.

The interpretation of these cross sections is complicated by the fact that emissions from states of equal n and different ℓ are superposed but the branching ratio $\eta_{4 \rightarrow 3}$ [i.e., the probability of $(n=4) \rightarrow (n=3)$ transitions divided by that of all decays of $n=4$] depends on ℓ . The values of this ratio, well established by hydrogenic theory¹⁹, are

$\ell =$	0	1	2	3
$\eta_{4 \rightarrow 3}$	0.4	0.04	0.3	1

Since the production of $\ell=3$ seems unlikely, and since the $\ell=1$ value is far smaller than the others and the values for $\ell=0$ and 2 are comparable, the main uncertainty concerns the relative probability of excitation of $\ell=1$. In fact one might regard the effective $\langle \eta_{4 \rightarrow 3} \rangle$ as equal to 0.4 multiplied by this probability. Utilizing cross sections obtained by a Dalgarno-McDowell semi-realistic theory²⁰, one finds $\langle \eta_{4 \rightarrow 3} \rangle \sim 0.3$.

Firstly we compare the fluorescence cross section $\sigma_{4 \rightarrow 3}$ with the double ionization cross section σ_{++} for incident electrons in the range $T \sim 0.3$ - 0.5 Kev where both

cross sections are fairly constant. We take $\sigma_{4 \rightarrow 3} \sim 4 \times 10^{-21}$ cm² from refs. 8a and 9 and $\sigma_{++} \sim 8 \times 10^{-20}$ cm² from ref. 5. The cross section $\sigma_{4+} = \sigma_{4 \rightarrow 3} / \langle \eta_{4 \rightarrow 3} \rangle$ is then $\sim 1.3 \times 10^{-20}$ cm² so that

$$\sigma_{++} / \sigma_{4+} \sim 6 \quad (11)$$

This value decreases to less than 3 if one adopts a more recent value $\sigma_{4 \rightarrow 3} = 9.8 \times 10^{-21}$ cm² 8b or the earlier Hughes-Weaver value 8.4×10^{-21} cm².

The ratio (11) may be compared with the corresponding ratio for photon bombardment. Carlson³ gives $\sigma_{n>2,+} / \sigma_{++} \sim 1$; of this ratio, perhaps 1/3 might be attributed to $n=4$, which would yield $(\sigma_{++} / \sigma_{4+})_{\text{photon}} \sim 3$. That is, the ratios for electron and photon bombardment are quite comparable if one adopts the larger values for the fluorescence cross section $\sigma_{4 \rightarrow 3}$.

Some preliminary measurements by de Heer⁹ give values of $\sigma_{4 \rightarrow 2}$ between 1.5 and 2.2×10^{-20} cm² for electron collisions in the 300-500 eV range. Here we have $\eta_{4 \rightarrow 2} \sim 0.6$ ($\ell=0$), 0.12 ($\ell=1$), 0.7 ($\ell=2$), 0 ($\ell=3$). Taking an average value $\langle \eta_{4 \rightarrow 2} \rangle \sim 0.5$ yields then $\sigma_{4+} \sim 4 \times 10^{-20}$ cm², a value that exceeds those obtained from $\sigma_{4 \rightarrow 3}$ data.

Preliminary measurements by de Heer et al⁹ of $\sigma_{4 \rightarrow 3}$ extend to electron collisions up to 5 keV. These results show a clear increase roughly linear with $\log T$ above 1 keV, at variance with the corresponding results for σ_{++} ^{5,7}, which is somewhat surprising. The coefficient of the log factor in (8), which is now $f_{4+} I_H \langle \eta_{4 \rightarrow 3} \rangle / \langle E \rangle$ amounts to $\sim 5 \times 10^{-5}$. Taking $\langle \eta_{4 \rightarrow 3} \rangle \sim 0.3$ and $\langle E \rangle \sim 100$ eV yields $f_{4+} \sim 1 \times 10^{-3}$, which appears quite low as compared to (5), $f_{n>2} \sim 0.02$.

c) Double excitations by particle impact. Silverman and Lassettre¹³ observed the excitation of 2s2p and sp23+ through the spectrum of forward scattered electrons at an impact energy of 500 eV. Under these conditions the angular momentum transfer is certainly 1 unit (optically allowed transition) except as noted below, but the magnitude of the momentum transfer ΔP is not very small (0.55 a.u.) [No optically forbidden transition was detected in this experiment.] Because of the rather large value of ΔP , the intensity ratio of double excitation to simple ionization with equal energy loss and the interference of the two processes need not be the same as for photon bombardment. In fact, the rough estimates of resonance width and profile obtained from SL data²¹ are not inconsistent with those of MC for photon absorption. The cross section for 2s2p excitation in this experiment is $\sim 2.0 \times 10^{-19}$ cm²/ster.

Mehlhorn¹² observed autoionization electrons from excitation of $2s2p$ by 4 keV electron impact, i.e., under conditions where Born approximation is well applicable and the process should be similar to photon absorption. The profile of the spectrum of these electrons appears consistent with this expectation.

Simpson et al (SCM)¹⁴ observed several optically allowed and forbidden double excitations near 60 eV under conditions similar to SL's but at impact energies ranging from 400 down to 90 eV. The intensity ratios of forbidden and allowed transitions were observed as functions of energy, tending to zero at high energy, as expected. However the forbidden excitation of $2p^2 \ ^1D$ at 60.0 eV, which is not easily resolved from that of $2s2p$ at 60.1 eV, should still be appreciable at 500 eV and should have contributed to the " $2s2p$ peak" observed by SL. At the lower energies the intensity ratio of optically forbidden and allowed transitions is of the order of 50%. Theoretical calculations of these intensities in Born approximation are contemplated.

The production of double excitations near 60 eV by proton impact has been observed only by detecting autoionization electrons emitted at 160° from the incident beam¹¹. The impact energy of ~ 75 keV corresponds to the same velocity as the impact of ~ 40 eV electrons. The intensity ratio of various transitions (optically forbidden by $\Delta L = 1$ and allowed) is comparable with expectation for 40 eV electrons based on extrapolation of SCM data. The

intensity ratio of double and single transitions with equal energy is also comparable for protons and slower electrons. However, no asymmetry is detected in Rudd's spectral profiles of autoionization electrons, at variance with electron impact results. This suggests that single and double transitions caused by proton impact differ in angular momentum or other parameter to an extent that hinders interference of the corresponding probability amplitudes (i.e., lowers the value of $\rho^{18,22}$ greatly).

An absolute cross section can be estimated for the double transitions produced by proton impact on the basis of that for simple ionization²³. The angular distribution of ejected electrons is peaked forward but flat (for ~ 60 eV transitions) in the backward hemisphere. Presumably the forward peak derives from singly ionizing collisions with large transfer of angular momentum, whereas the double transitions absorb 0-2 units only of angular momentum and should yield an angular distribution without major peaks. If, then, the intensity of autoionization electrons per steradian at 160° is multiplied by $\sim 4\pi$ one obtains a total cross section for double transition to $2s2p \sim 1.5 \times 10^{-19} \text{ cm}^2$. (We thank A.R.P. Rau for this value.)

A very rough comparison of this value with the cross section for double excitation by electrons can be attempted as follows. The cross section for 500 eV electrons at 0° ; differential in angle, was given above as $2 \times 10^{-19} \text{ cm}^2/\text{ster}$;

this cross section is lower by a factor of ~ 2 at $\sim 8^\circ$.
On this basis an integral value of $\sim (1/5) \times 10^{-19} \text{ cm}^2$ might
be estimated. The resulting ratio of ~ 8 between protons
and electrons may be compared to the v^2 ratio which is
 $40/500 \sim 1/12$. These ratios are approximately reciprocal
in the Born approximation.

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