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Electron transfer in keV-energy $^4He^{++}$ collisions. III. Experimental tests of the close-coupling calculations for $^4He^{++}$-H(1s) collisions*

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Two experimental tests of the $^4He^{++}$-H(1s) close-coupling collision calculations were performed with a highly mass-resolved $^4He^{++}$ incident ion beam and an atomic hydrogen scattering target. The total single-electron-transfer cross section σ_{21}(H) was measured for $^4He^{++}$ energies between 7 and 144 keV, with the results in better agreement with molecular-state calculations than with theory employing atomic states. Above 25 keV this cross section is 50% above values obtained by Shah and Gilbody for incident $^3He^{++}$ with the same collision velocity. Also measured was the cross section σ^*_{21}(H) for single-electron transfer into the 2s state of He^+ using an ion-photon time-coincidence technique over the $^4He^{++}$ energy range 7 to 70 keV. These data differ with the corresponding $^3He^{++}$-H(1s) data of Shah and Gilbody at the higher energies. For σ^*_{21}(H), the observed energy dependence as well as the magnitude at the lower energies are in marked disagreement with all close-coupling results.

I. INTRODUCTION

The calculation of atomic-collision cross sections is most difficult in the intermediate collision energy region 1–1000 keV, where the collision velocity is not too different from that of atomic electrons. In the case of the H^+-H system, recent close-coupling calculations that include the $n=1$, 2, and 3 atomic states on each center have been quite successful in simultaneously predicting many aspects of the electron transfer cross sections into the resonant 1s state, 2s and 2p states, as well as excitation into 2s and 2p.[1,2] Although some differences remain, generally surprising agreement exists with the recent electron-transfer and excitation data,[3-6] considering that the energies involved are three orders of magnitude larger than that required for excitation of all possible atomic excited states as well as the continuum.

The encouraging theoretical situation for H^+ on H suggests that perhaps reasonably accurate calculations of keV atomic collision cross sections can be made for many other collision systems of practical interest. Among these are He^{++} collisions with atomic hydrogen, where recently several many-state close-coupling calculations have been made. Several particular features distinguish the asymmetric He^{++}-H collision system from that for the symmetric H^+-H case, as can be seen from the molecular energy curves shown in Fig. 1. One needs to first recall that within the molecular collision picture resonance is said to exist when two molecular electronic potential energy curves are asymptotically (internuclear distance $R\rightarrow\infty$) degenerate. It is seen that in the H^+-H(1s) system the ground-state process is resonant and the excited state highly nonresonant, whereas the He^{++}-H(1s) electron transfer collision is asymptotically resonant into the $n=2$ states of He^+ and the He^+(1s) ground state highly nonresonant. Thus total electron transfer in the He^{++}-H system is more complicated in that (i) there are the several final resonant states $He^+(2s, 2p_0, 2p_1)$; (ii) these favored final states are highly coupled by the long range He^+-H^+ Coulomb force; and (iii) trajectory and other effects arising from this Coulomb repulsion need to be considered.

Only two total cross-section experiments have been performed on the $^3He^{++}$-H(1s) system. Fite, Smith, and Stebbings[7] (FSS) have made measurements of σ_{21}(H), the total cross section for single electron transfer in the energy range 0.1 to 36 keV, summed over all final states of excitation. Their cross sections were normalized to their early results for σ_{10}(H), the cross section for single electron-transfer in $H^+ + H(1s)$ collisions, at an energy of 1.9 keV.

Recently Shah and Gilbody (SG) have measured both σ_{21}(H) and σ^*_{21}(H) for incident $^3He^{++}$ over the $^3He^{++}$ energy range 10–55 keV.[8] Their energy dependence for σ_{21} agrees with the earlier result of FSS, but the absolute magnitude of this cross section was lower, supporting the results of close-coupling theory using atomic-basis states instead of the theory using molecular-basis states. The data for σ^*_{21}(H) obtained by SG is a factor of 5 below the predictions of both types of theory.

The present report describes two experiments done on the $^4He^{++}$-H(1s) collision system. The first is a measurement of the cross-section σ_{21}(H)

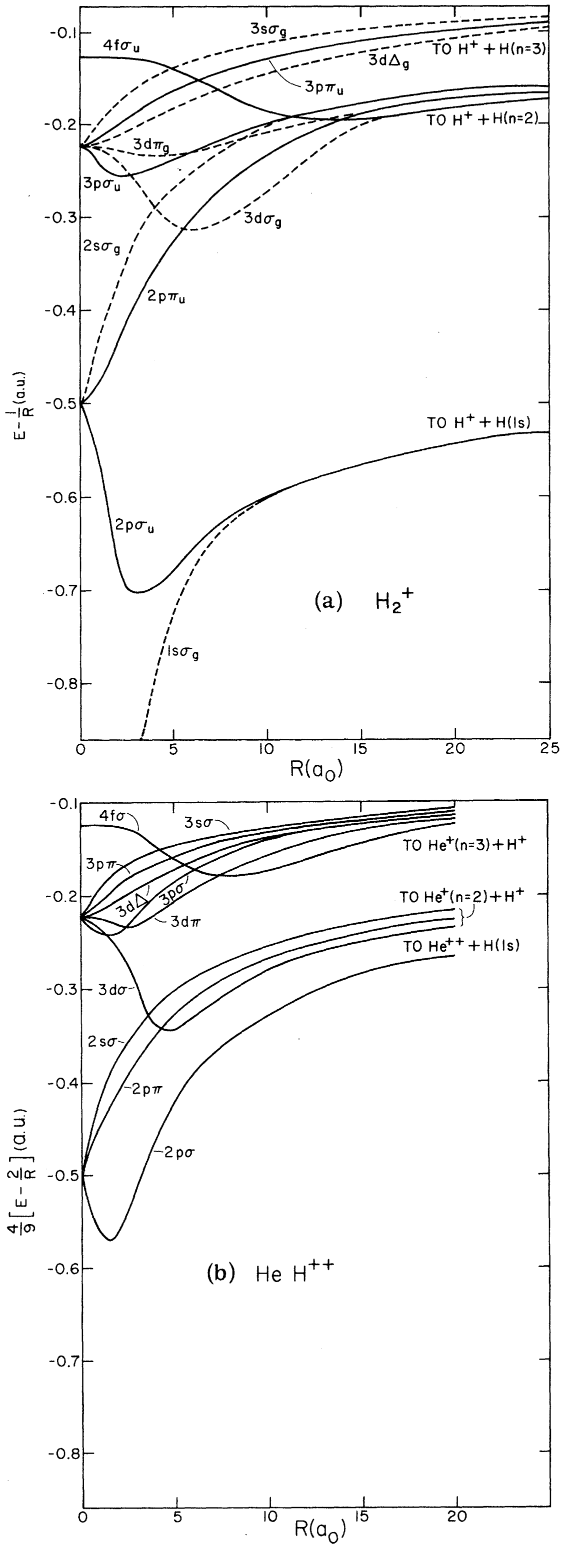

FIG. 1. (a) Born-Oppenheimer (BO) potential energy levels of H_2^+. United atom state labeling of the states is used. Note that the internuclear repulsion R^{-1} (a.u.) has been subtracted. (b) BO levels of HeH^{++}.

for total electron transfer:

$$^4He^{++} + H(1s) \rightarrow {}^4He^+ \text{ (all states)} + H^+.$$

As in paper I of this series,[9] DC detection of the incident $^4He^{++}$ beam and scattered $^4He^+$ beam was possible over the $^4He^{++}$ energy range 32.7–144.6 keV. The second experiment paralleled those of paper II of the series[10] and used single-ion counting techniques to extend the DC data for $\sigma_{21}(H)$ to lower energies; the range 7–70 keV was covered. Also measured was the cross section $\sigma_{21}(H)$ for the collision process

$$^4He^{++} + H(1s) \rightarrow {}^4He^+(2s) + H^+,$$

again over the energy range 7–70 keV.

A preliminary report of the present results was made in 1973.[11]

II. EXPERIMENTAL PROCEDURES

The experimental apparatus was basically the same as in papers I and II with the exception of the scattering target and its associated instruments.

As the target assembly has been discussed in detail,[12] the description here will be brief except where major alterations were made. Basically the system consisted of a radiatively heated 20-cm-long 2.5-cm-diameter double-walled scattering cell assembly placed inside a horizontal open-ended vacuum furnace. The vacuum furnace consisted of a 0.013-cm-thick tantalum resistive tube that was heated by a 700-A pulsed current. The heater was radiatively shielded from the rest of the vacuum chamber by seven concentric layers of 0.025-cm thick tantalum. These were in turn surrounded by a double-walled copper shell through which cooling water flowed. For atomic hydrogen in the target the scattering cell was heated to 2700°K by the pulsed current, with the cell mass being large enough to make the cell temperature not responsive to the furnace heater-current time dependence at the 10-Hz frequency used. The pulsed heater current feature was used to avoid magnetic-field effects on the beam, the latter being pulsed out of phase with the current and delayed by about 13 msec to avoid the remanent Eddy currents in the scattering cell walls. The newer all-tungsten scattering cells[13] were used in the present experiments.

The acquisition of data and its analysis parallels that of papers I and II, except that the target-gas ratio technique used earlier[4] to obtain H^+-H cross sections from H^+-Ar and H^+-H_2 cross sections is now employed for incident He^{++}. In essence, for a hot-argon target the thickness was determined using the $^4He^{++}$-Ar single-electron-transfer cross sections measured in paper I for the cold-argon target. The earlier corresponding $^4He^{++}$-H_2 data are needed for a small (typically ≤30%) correction due to incomplete H_2 thermal dissociation within the target. The dissociation fraction was measured using a variation of the usual double-electron-transfer technique.[14,4] We now utilized the fact that neutral helium atoms could not be formed in a single He^{++}-H collision, whereas they can be in a He^{++}-H_2 single collision. The dissociation fraction was measured to be 0.86 ±0.03, hence 93% of the particles in the hot target cell were hydrogen atoms and 7% were H_2 molecules. This value agrees with our earlier results, given the same H_2 flow rate and vacuum furnace current. A higher target pressure of about 10^{-4} Torr was used in these He^{++} experiments than in the earlier H^+ work, accounting for a lower present value for f. As expected from theoretical work and recent experiments on other target gases,[28] collisional destruction of $He^+(2S)$ having cross sections of about 10 Å^2/atom was found to be less than a 5% effect at our operating pressures.

The cross section $\sigma_{21}(H)$ was measured using the Faraday cup beam-detection techniques of paper I over the energy range 36–144 keV. Between 7 and 70 keV the coincidence techniques of paper II yielded values of $\sigma_{21}(H)$ and $\sigma_{21}^*(H)$. In the energy range of overlap, the values of $\sigma_{21}(H)$ using the two techniques agreed within ±8%. One of the important checks made was that values for $\sigma_{21}(Ar)$ and $\sigma_{21}^*(Ar)$ obtained with the target cell cold and hot agreed to within the reproducibility of the data, ±15%. Sources of error not explicitly mentioned in this paper are those already discussed in papers I and II.

III. RESULTS AND DISCUSSION FOR TOTAL ELECTRON TRANSFER

A. Comparison with $^3He^{++}$ experimental results

The present data for $\sigma_{21}(H)$ are shown in Fig. 2 along with various theoretical curves. The error bars indicate a 90% confidence level, and amount to at most ±30% above 10 keV. For comparison the low-energy $^3He^{++}$ data of Fite *et al.* (FSS) and the $^3He^{++}$ data of Shah and Gilbody (SG) are shown scaled for the same incident $^4He^{++}$ velocity. Reasonable agreement between the present data and those of FSS is found over the 7–48 keV range of overlap in equivalent incident $^4He^{++}$ energy. Our values are some 50% larger than those of SG. The latter authors suggest a renormalization down of the FSS data by 20 or 30%. The FSS data was normalized to early values of the total $H^+ + H$ electron transfer cross section at 2 keV, now believed to be smaller in value on the basis of the more recent work of McClure.[3] If this renormalization is done, then the present $^4He^{++}$ cross section is significantly larger than the $^3He^{++}$ one, and an apparent isotope effect exists. We point out that the data of SG also depend upon the same data obtained by McClure. However, the present data ultimately depends instead upon the accuracy of the better established total single-electron-transfer cross section for $^4He^+ + ^4He$, as discussed in paper I. We presently prefer a skeptical position on any possible isotope effect in σ_{21},[15] and await a direct $^3He^{++}$-H to $^4He^{++}$-H cross-section ratio measurement under otherwise identical conditions.

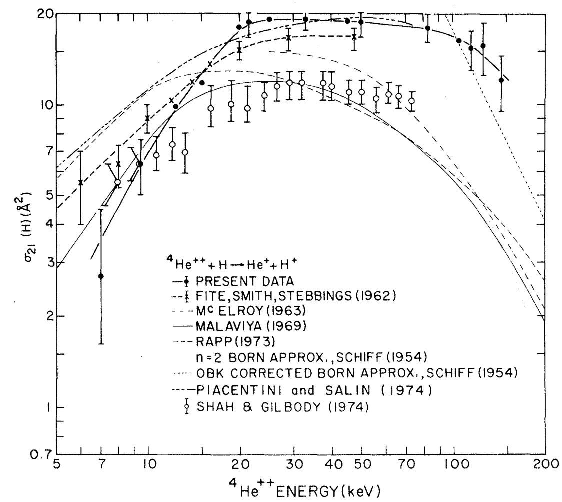

FIG. 2. Total electron transfer in He^{++}-H(1s) collisions. The data of Fite, Smith, and Stebbings (Ref. 7) and of Shah and Gilbody (Ref. 8) are for incident $^3He^{++}$, plotted here for equivalent relative collision velocity. The theoretical references are McElroy, Ref. 26; Malaviya, Ref. 20; Rapp, Ref. 21; Schiff, Ref. 19; and Piacentini and Salin, Ref. 24.

The discrepancy in $\sigma_{21}(H)$ between the present results and those of SG partially can be ascribed to a similar discrepancy in $\sigma_{21}(Ar)$ as shown in Fig. 8 of paper I. Below 20 keV, there is good agreement in the cross section ratio $\sigma_{21}(H)/\sigma_{21}(Ar)$ of the results of the two laboratories. Above 20 keV the present values for this ratio are higher by 20–25%. Because of the future possibility of resolving the $\sigma_{21}(Ar)$ discrepancy more easily than for $\sigma_{21}(H)$, we include in Table I our values for the ratio. These are believed to be accurate to within ±15% determined primarily by long-term reproducibility.

B. Comparison with $^4He^{++}$ theoretical predictions

The large difference between atomic-state $^4He^{++}$-H close-coupling theory and the present results

TABLE I. The measured cross sections $\sigma_{21}(H)$ and $\sigma_{21}^*(H)$ for $^4He^{++}+H(1s)$ collisions. [Total uncertainty in $\sigma_{21}(H)$ is ±30%, and in $\sigma_{21}^*(H)$, ±40%.] Also listed are values for $\sigma_{21}(H)/\sigma_{21}(Ar)$ with an uncertainty ±15%.

Energy (keV)	$\sigma_{21}(H)$ (Å^2/atom)	$\sigma_{21}^*(H)$ (Å^2/atom)	$\sigma_{21}(H)/\sigma_{21}(Ar)$
7	2.7	0.50	1.8
9	6.0	0.70	2.2
10	7.3	0.95	2.2
12	9.6	0.86	2.1
12.8	10.3	1.05	2.2
14.5	11.8	1.06	2.1
15	11.7	1.12	1.8
18.9	16.3	1.14	1.8
20	17.2	1.34	1.9
22	18.4	1.16	1.7
25	19.0	1.37	1.7
35	18.8	1.95	1.45
46	18.8	1.96	1.37
55	18.5	2.31	1.32
70	18.3	2.35	1.31
82	17.9	···	1.32
104	16.2	···	1.26
115	15.4	···	1.28
126	15.6	···	1.30
145	12.1	···	1.1

at high energies is much larger than initially expected via a comparison with the overall situation for the H^++H system. In the latter, the inclusion of higher excited states did not greatly alter theoretical results for the favored ground-state process, and cascade effects were less than 30%. Also, inclusion of $n=3$ state does not greatly alter the $n=2$ cross sections. The interesting question now arises as to what aspect of the He^{++}-H system makes the comparison of theory with experiment less satisfactory than for H-H^+. Certainly to be investigated is the possible importance of the $n=3$ and higher states, both as providing important additional channels contributing to the experimental curve, and also as possibly introducing significant corrections to the $n=1$ and 2 results computed theoretically. This has been done within close-coupling theory, to be discussed below. In addition, several very-high energy collision calculations suggest that $n=3$ production may be significant, although the approximations used are not expected to be good in the present energy range. Relevant here are the results of the OBK approximation[16] known to give results an order of magnitude too large at high principal quantum number[13] n; the impulse approximation results[17] and the continuum distorted wave (CDW) results recently reported.[18] Summing the CDW results for $n=1$, 2, and 3 gives values $\sigma_{21}(H)$ $=23.5$ Å^2 at 100 keV and 457 Å^2 at 25 keV, much larger than the present results. In addition, the CDW value for 3s state production at 100 keV is forty times larger than that predicted by the impulse approximation. At present, it seems that the Born-approximation results of Schiff[19] are as helpful as these more recent high-energy approaches. Shown in Fig. 2 are Schiff's results first for only the $n=2$ state, a curve close to the close-coupling calculations curves, and then a higher total cross-section curve obtained by Schiff using the OBK n^{-3} rule. The latter curve crosses the present experimental results at 105 keV.

Four $^4He^{++}$-H(1s) atomic-state close-coupling calculations have been performed for the total electron-transfer cross section $\sigma_{21}(H)$. Malaviya[20] used four hydrogenic states $He^+(1s, 2s, 2p_0, 2p_1)$ centered around the He^{++} nucleus. Rapp[21a] used four hydrogenic states centered on each nucleus. More recently Msezane and Gallaher[22] (MG) have used the same states as Rapp, and in addition have performed another calculation using the pseudostate expansion method. The results of MG are not shown since they differ insignificantly from the earlier results of Rapp, the former being always slightly smaller by about 10%. Note that both Rapp and MG compared their $^4He^{++}$ results with the $^3He^{++}$ experimental results at the same incident energy rather than at the same incident velocity. Then the agreement with the results of FSS is excellent at energies below 15 keV. Yet in their papers they both indicated that they were going to compare with the results of FSS at the same incident velocity.[23] Rapp[21b] later corrected the situation and included the effect of the $He^+(3s, 3p)$ states in his 11-state calculation. Figure 2 indicates that the earlier conclusions of Rapp and MG were incorrect and fortuitous and that the disagreement of these two theoretical results with the present data is as poor as their disagreement with the data of FSS. At low energies the results of Malaviya seem to agree with, or at least lie in between, the present results and those of FSS. It should be emphasized that the close-coupling results being quoted involve just the sum of cross sections for electron transfer into the $n=1$ and $n=2$ states of He^+.

Thus high-energy approximations to the theory and our data both join in suggesting that the $n=3$ and possibly higher states might need to be included in the atomic state close-coupling calculations for $^4He^{++}$-H electron-transfer collision processes. On the other hand, the 11-state results of Rapp yield a total $n=3$ contribution to σ_{21} of only 1%.

The results of the impact parameter molecular-state $^4He^{++}$ close-coupling calculation of Piacentini and Salin[24] are also plotted in Fig. 2. Their curve lies higher than the atomic-state curve and seems to agree with the present data above 15 keV. This theoretical work included the $2p\sigma$, $3d\sigma$, and $2p\pi$ states of $^4HeH^{++}$ which were found to be the most strongly coupled states, see Fig. 1. At an internuclear distance $R \sim 6.5a_0$, there is a long-range $2p\sigma$-$3d\sigma$ radial coupling; at $R \sim 4.5a_0$ rotational coupling near the $2p\pi$-$3d\sigma$ level crossing is important, and all three states are simultaneously coupled at smaller R. The results for σ_{21}(H) were believed by Piacentini and Salin to be reliable within this three-state calculation in spite of uncertainty in the nature of the electron-momentum-transfer terms that were included, for they could define unambiguously the total electron-transfer probability to be one minus the total excitation probability. They did state some misgivings about their individual probability amplitudes for transfer into definite final states, such as the 2s state of the He^+ ion.

IV. RESULTS AND DISCUSSION FOR THE 2s STATE CROSS SECTION

A. Comparison with $^3He^{++}$ experimental results

The present results for σ^*_{21}(H) are tabulated in Table I and shown in Fig. 3 along with the $^3He^{++}$ velocity scaled data of SG and with the relevant $^4He^{++}$ theoretical predictions. The rms experimental uncertainty in our cross section is about ±40%, with a possible additional +15% error due to the polarization effect of the stimulated Lyman-alpha radiation.[10]

We observe that our data disagrees with SG not so much in magnitude as in energy dependence. In paper II a discrepancy barely within experimental error of a factor of 3 was found for σ^*_{21} for incident He^{++} on Ar, H_2, N_2 and He targets; Lyman alpha detector calibration uncertainties could thus explain the difference in magnitudes between our data and SG. However, the energy dependences of the two sets of data in paper II were similar, whereas now for the atomic hydrogen target there is qualitative disagreement: the SG curve is fairly flat whereas our data rises with energy.

The contribution to the present discrepancy in σ^*_{21}(H) both associated with that for σ_{21}(H) and originating in the different values for σ_{21}(Ar) is only about $\frac{1}{3}$ of the discrepancy seen in Fig. 3 at 70 keV. Thus the difference in values for σ_{21}(H) is not the principal source of differences in σ^*_{21}(H). On the other hand, the agreement in energy dependences for σ^*_{21}(Ar) seen in paper II seems to rule out major error in the energy dependence of the Lyman-alpha detector efficiency. The presently observed rising values of σ^*_{21}(H) with energy is qualitatively but not quantitatively similar to that observed for

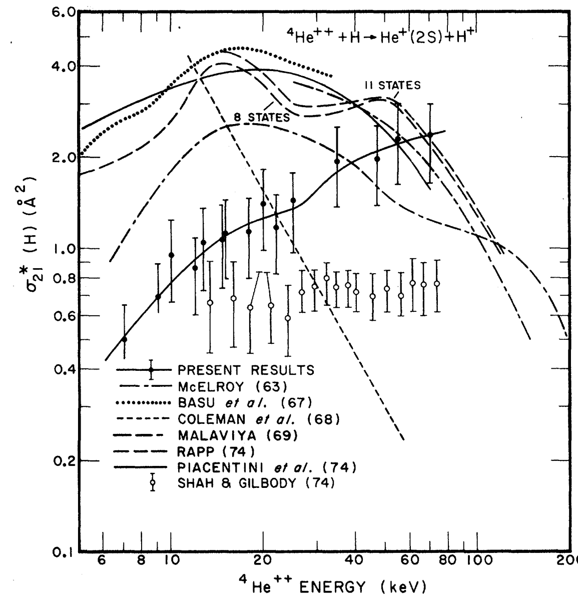

FIG. 3. Theory vs experiment for $He^+(2s)$ production in He^{++}-H(1s) collisions. The data are "present results," uncertain by ±40%; and "Shah and Gilbody," Ref. 8, uncertain by a factor of 2. Those theoretical references different than in Fig. 2 are Basu *et al.*, Ref. 27, and Coleman *et al.*, Ref. 17.

$He^{++}+H_2$.[10] Our values for H are three or more times larger than for H_2 below 20 keV, while only 30% larger at 60 keV, so the energy dependences for H and H_2 are quite different. We have no reason to distrust our direct procedures used to determine the dissociation fraction f, which attained values higher than in the experiments of SG. Thus the differences in high-energy behavior between the $^4He^{++}$ and $^3He^{++}$ data are unexplained.

An additional complication of the present experimental situation can be seen by studying the cross-section ratio K defined by

$$K = \sigma^*_{21}(H)/\sigma_{21}(H)$$

as a function of energy. The experimental and theoretical ratios are plotted in Fig. 4 as a function of $^4He^{++}$ energy. One observes agreement within experimental errors between the two sets of data. This is, however, not the case for the target gases used in the work of papers I and II, where the factor-of-3 difference in σ^*_{21} values reflects itself in the corresponding K values. This would suggest that the Lyman-alpha detector efficiency changed in at least one of the two laboratories between taking data on Ar, H_2, N_2 and He and taking data on H. However, the present data for H was taken point-by-point relative to hot Ar using the ratio technique (and the same target

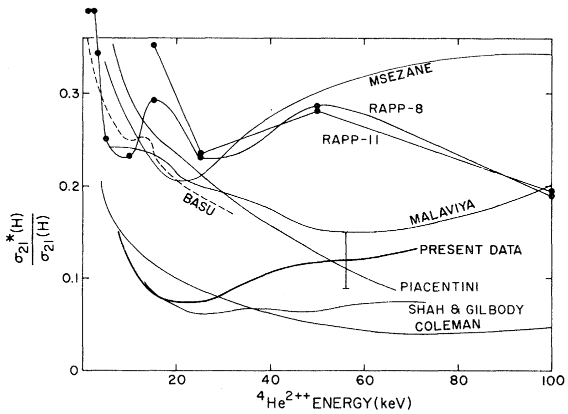

FIG. 4. Fractional electron transfer into $He^+(2s)$ in $He^{++} + H(1s)$ collisions, theory and experiment. The references are as in Fig. 2 and 3, plus "Msezane," Ref. 22.

assembly as for cold H_2 and N_2), with results for H independent of a long-term detector efficiency change.

B. Comparison with $^4He^{++}$ theoretical predictions

When comparing the present results for $\sigma^*_{21}(H)$ with the theoretical curves of Fig. 3, we note better agreement with the results of the seven atomic-state calculation of Rapp than with his eight and eleven atomic-state calculations.[21b] The close-coupling results of Malaviya performed in the four atomic state approximation[20] seem to come closest to the present experimental curve in magnitude but not in shape.

The observed discrepancy between atomic-state close-coupling theory and experiment at both low and high energies suggests that perhaps higher excited states of both the H target and the final-state He^+ ion are playing a more important role than henceforth expected. Since both σ_{21} and σ^*_{21} are smaller than theoretical values at low energies, one suspects that target excitation to n larger than 2 plays the important role there. On the other hand the excited states of He^+ must play a larger role at the high energies (≥ 40 keV) where both measured cross sections are larger than calculated atomic-state close-coupling values.

Figure 4 also reveals that all close-coupling results fail as well to predict the ratio K correctly. This is due either to overestimating σ^*_{21} or to underestimating σ_{21} (or both). The ion-photon coincidence technique used both here and in paper II determines K rather directly. Equation (13) of that paper was

$$K = \frac{1}{\epsilon_L}\left[\frac{C}{M^+}\right],$$

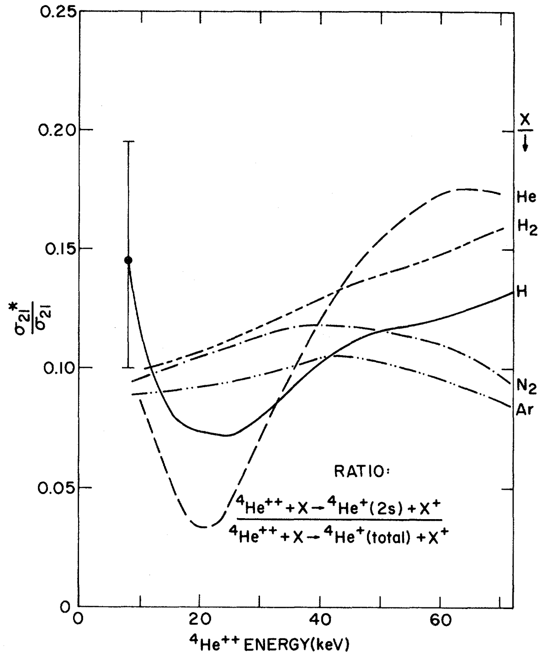

FIG. 5. Results for the fractional electron transfer into $He^+(2s)$ for He^{++} incident on various gases.

where ϵ_L is the overall Lyman alpha detection efficiency, M^+ is the He^+ ion detector count rate and C is the Lyman alpha-ion coincidence count rate. The ratio C/M^+ is independent of the efficiency of the He^+ detector as well as target pressure and other factors. The quantity K depends most critically upon ϵ_L, which would have to be in error roughly a factor of 2. To us this seems possible but unlikely.

It is interesting to note that the results of Coleman and Trelease[25] using the high-energy impulse approximation are completely incorrect in the case of $\sigma^*_{21}(H)$, but give good values for K. At present this should be considered fortuitous.

Figure 5 shows our present values of the cross-section ratio K for all target gases studied to date. These values are tantalizingly large and provide encouragement for the x-ray laser project of Louisell, Scully, and McKnight.[25] Nevertheless, a direct measurement of the partial $He^+(2p)$ production cross section is needed to verify population inversion.

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