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Ionization and charge transfer in He^{2+}—rare-gas collisions

R. D. DuBois
Pacific Northwest Laboratory, Batelle Memorial Institute, Richland, Washington 99352

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Cross sections for all significant channels leading to positive-ion and free-electron production in He^{2+}—rare-gas collisions are presented. The energy range studied is 15—200 keV. It is shown that the electron production is dominated by the single charge transfer plus ionization channels—even for a helium target. It is also shown that charge transfer plus an additional ionization can be more probable than charge transfer by itself. For double capture from argon and krypton, this is observed to occur throughout the entire energy range studied.

INTRODUCTION

It is now well established that several electrons can be simultaneously removed in a single collision between a charged particle and an atom. Even for light-ion impact, multiple ionization can represent as much as 50% of the total ionization cross section.[1] In such cases a comparison between experimental data and theoretical calculations is meaningless unless the ionization state of the target is measured or the theory includes the possibility of multiple ionization events.

Inclusion of multiple ionization into existing theoretical treatments is in itself quite difficult and needs to be accurately tested. Although the first Born approximation can predict single direct ionization cross sections at high energies,[2,3] more complicated theories are required for multiple ionization. McGuire[4,5] has shown that even for the relatively simple case of high-energy proton impact ionization of helium, double ionization needs to be treated in terms of both the "shake-off" and the "two-step" mechanisms. The shake-off mechanism, which corresponds to direct single ionization followed by shaking-off the second electron because of the rapid change in electronic screening of the nucleus, dominates at very high energies. For intermediate energies both mechanisms are important and quite possibly interference between them needs to be taken into account. Although this theoretical approach simulates the observed velocity dependence, calculated cross sections have not been directly compared with experimentally measured values.

Sidorovich and Nikolaev[6] have calculated cross sections for direct double ionization of helium using an independent electron model which is similar to McGuire's two-step model. In this model double ionization is determined from the product of the single ionization probabilities. They used three different models for the single ionization probabilities—one of which produced reasonable agreement with experiment at higher energies.[2] For lower energies, the theory overestimated the cross section.

In both of these theoretical treatments only the direct multiple ionization channel was considered. However, charge transfer collisions also produce considerable multiple ionization. For very low impact energies this "transfer ionization" can be interpreted in terms of a molecular-orbital picture.[7] Within this mechanism, the internal potential energy of the initial and final states differs. This excess energy, obtained by subtracting the ionization energies before and after the collision, is available to cause additional target ionization as opposed to higher impact energies where the projectile possesses sufficient kinetic energy to cause additional ionization.

Until recently no theoretical treatment of charge transfer plus ionization occurring in a single collision was available. However, Sidorovich *et al.*[3] have just published calculations for all channels leading to ionization in He^{2+}-He collisions. The calculations were done using an impact-parameter formulation where the probabilities were calculated using an independent electron approximation. Their calculated direct single-ionization cross sections tended to agree well with experimental data for higher impact energies but overestimated the cross section below $\sim$200 keV/amu. For He^{2+} impact their calculated direct double-ionization cross sections agreed reasonably well with the limited experimental data although theory appeared to underestimate the cross section at low energies. The same was true for the pure single-electron transfer channel and the double-electron transfer channel. However, these two effects—overestimation of lower energy direct single ionization and underestimation of the single-electron transfer—tended to cancel which resulted in their charge transfer plus ionization calculation agreeing reasonably well with experimental data. Thus this calculation of charge transfer plus ionization gave reasonable results, probably because of mutually cancelling errors.

Becker *et al.*[8] have developed an independent Fermi-particle model which they have used to calculate multiple ionization occurring via the direct Coulomb or charge transfer channels. Their results for H$^+$-Ne, although calculated for higher energies, appear to be in good agreement with measurements[9] for single- and double-target ionization but tend to overestimate the cross section for triple ionization. No experimental data were available for higher states of ionization. Similar calculations were also made for He^{2+} impact on neon.

For low-energy collisions where charge transfer and direct ionization cross sections are comparable in magnitude, the total amount of multiple ionization is often quite large.[1] This makes interpretation of the data difficult and sometimes misleading.[9] However, due to experimental complexities in separating the various multiple-ionization channels and the lack of accurate theoretical treatments, very little information about this region is available.

The experimental data presented here are designed to aid in our understanding of multiple ionization occurring via the direct target ionization and charge transfer channels. Previously such data have been presented for H^+ (Refs. 2, 7 and 9–11) and He^+ (Ref. 9) impact. For He^{2+} impact where the double charge transfer channel can also be significant, only the helium target has been previously studied.[2,12,13] Absolute cross sections for multiple ionization occurring via direct ionization and single and double charge transfer processes are presented—in other words *all* channels leading to ionization of the target are measured. Note that the autoionization contributions are not explicitly separated but rather are observed in the direct or charge transfer channels via the final target ionization state that is produced. $^3He^{2+}$ impact on He, Ne, Ar, and Kr targets is studied for impact energies between 15 and 200 keV.

EXPERIMENTAL PROCEDURE

The experimental apparatus and procedure have been described in detail previously[9,14] so only a brief description will be given here. A collimated beam of $^3He^{2+}$ ions is energy analyzed before passing through a diffuse noble gas and is then electrostatically charge state analyzed and pulse counted by secondary emission detectors. Two postcollision beam components (either the neutral and singly or the neutral and doubly charged components) can be simultaneously measured. Slow target ions having charge state q that result from charge transfer and direct ionization are extracted perpendicular to the projectile beam by an electric field. These ions travel through a field-free drift region before being accelerated to approximately $3kqV$ and detected by a secondary emission detector. As they travel from the interaction region to the detector the target ions separate into their various charge states due to their different flight times.

Coincidences between the postcollision projectile beams and the slow ions are recorded using standard electronics, two time-to-amplitude converters, and a dual multichannel analyzer. The coincidence data consist of target ionization charge state intensities B_q^{2k} resulting from single- and double-charge transfer or direct ionization. The notation used here lists the projectile precollision and postcollision charge states as superscripts and the final target ionization charge state as a subscript. Hence direct ionization and single- or double-charge transferring collisions correspond to $k=2,1,0$, respectively. Note that $(q+k-2)$-free electrons are produced in the collisions.

The target charge state intensities depend on the respective cross sections σ_q^{2k}, the integrated beam intensities $I(He^{2+})$, the target density N, and the detection efficiencies of the beam η and the slow ions η_q. Hence the cross sections can be determined from

$$\sigma_q^{2k}=A\frac{B_q^{2k}}{NI(He^{2+})\eta\eta_q}\ ,$$

where A is a proportionality constant. The total single-electron capture cross section is obtained by summing the appropriate slow ion coincidence signals, i.e.,

$$\sigma^{21}=A\frac{1}{NI(He^{2+})\eta}\sum_q\left[\frac{B_q^{21}}{\eta_q}\right].$$

The detection efficiencies of the beam and the slow ions were investigated in the following manner. The secondary emission beam detectors were calibrated against a surface barrier detector (assumed to have unit efficiency) for H^+, He^{2+}, He^+, and He^0 particles impacting between 100 and 200 keV. The efficiencies were measured to be unity within the measuring errors of 5%. At lower energies the beam detectors were shown to subtend a large enough solid angle that all scattered beam particles were collected. Thus for energies less than 100 keV the detection efficiencies were assumed to be equal, if not unity, for the neutral, singly, and doubly charged beam particles. This assumption appears to be valid since measured charge transfer cross sections[15] where shown to agree with results from other investigators down to a few keV/amu.

The slow ion detection efficiency was shown to be independent of the slow ion extraction and the range of target densities used to within 5%. The measured charge state ratios varied by less than 5% for preacceleration voltages between -2 and -3.8 kV.

Thus the above formulas reduce to

$$\sigma_q^{2k}=A\frac{B_q^{2k}}{NI(He^{2+})}$$

and

$$\sigma^{21}=A\frac{\sum_q B_q^{21}}{NI(He^{2+})}\ .$$

Experimentally target gas density and beam current fluctuations were removed by simultaneously measuring σq^{21} and σq^{20} and then σq^{22} and σq^{20}. Both sets of data were normalized to the same neutral beam intensity $I(He^0)$ which is proportional to $I(He^{2+})$. These relative cross sections were placed on an absolute scale by normalizing to the single-electron transfer cross sections of Itoh and Rudd:[16]

$$\sigma_q^{22}=\frac{\sigma^{21}}{\sum_q B_q^{21}}B_q^{22}\quad\text{(direct ionization)}\ ,$$

$$\sigma_q^{21}=\frac{\sigma^{21}}{\sum_q B_q^{21}}B_q^{21}\quad\text{(single transfer)}\ ,$$

$$\sigma_q^{20}=\frac{\sigma^{21}}{\sum_q B_q^{21}}B_q^{20}\quad\text{(double transfer)}\ .$$

Even though the beam intensity varied by approximately two orders of magnitude between the σ_q^{22}, σ_q^{20} and the σ_q^{21}, σ_q^{20} measurements, the measured total double charge transfer cross sections $\sigma^{20}=\sum_{q>1}\sigma_q^{20}$ differed by less than 5%. Experimental uncertainties consisting of beam and slow ion detection efficiencies, normalization errors, σ^{21}

uncertainties, statistical, and background subtraction uncertainties indicate that the present absolute cross sections have total uncertainties of approximately 25% except for the smallest cross sections presented where larger uncertainties are possible.

RESULTS

Helium

The results for He^{2+}-He collisions are shown in Fig. 1. Cross sections for direct ionization as well as for single- and double-electron transfer are graphed as a function of energy/mass of the projectile. The total positive ion and electron production cross sections measured by Itoh and Rudd[16] using the condenser plate method are also included. As can be seen, double ionization of helium is dominated by the double-electron capture channel σ_q^{20} throughout the entire energy range shown although the single capture plus ionization channel σ_q^{21} is increasing in importance at higher energies. Direct double ionization σ_q^{22} is negligible throughout the entire energy range.

Included in Fig. 1 are the measurements of Shah and Gilbody[2] and Afrosimov *et al.*[12] The present direct single ionization cross sections σ_1^{22} are slightly smaller than those previously measured. Agreement with the single-electron transfer data of Shah and Gilbody is good but poorer agreement is found for the data of Afrosimov *et al.* The present double capture cross sections are approximately 50% larger than the measurements of Itoh and Rudd[16] and Afrosimov *et al.*[12] but have the same energy dependence. This was also found to be true for measurements involving a lithium target[14] and will be shown to occur for the other targets presented in this paper. This strongly suggests a detection or efficiency problem with one of the beam detectors in the present experiment, although none could be identified when searched for. So the reason for our overestimation of the double transfer cross sections is unknown.

An additional consistency test of the present data can be made by comparing the total positive ion and electron production cross sections with those measured independently.[16] As can be seen from the data, the positive ion production

$$\sigma_+ = \sum_q q\sigma_q^{22} + \sum_q q\sigma_q^{21} + \sum_{q>1} q\sigma_q^{20}$$

is dominated by the charge transfer channels. Good agreement with the data of Itoh and Rudd[16] is found. The total electron production

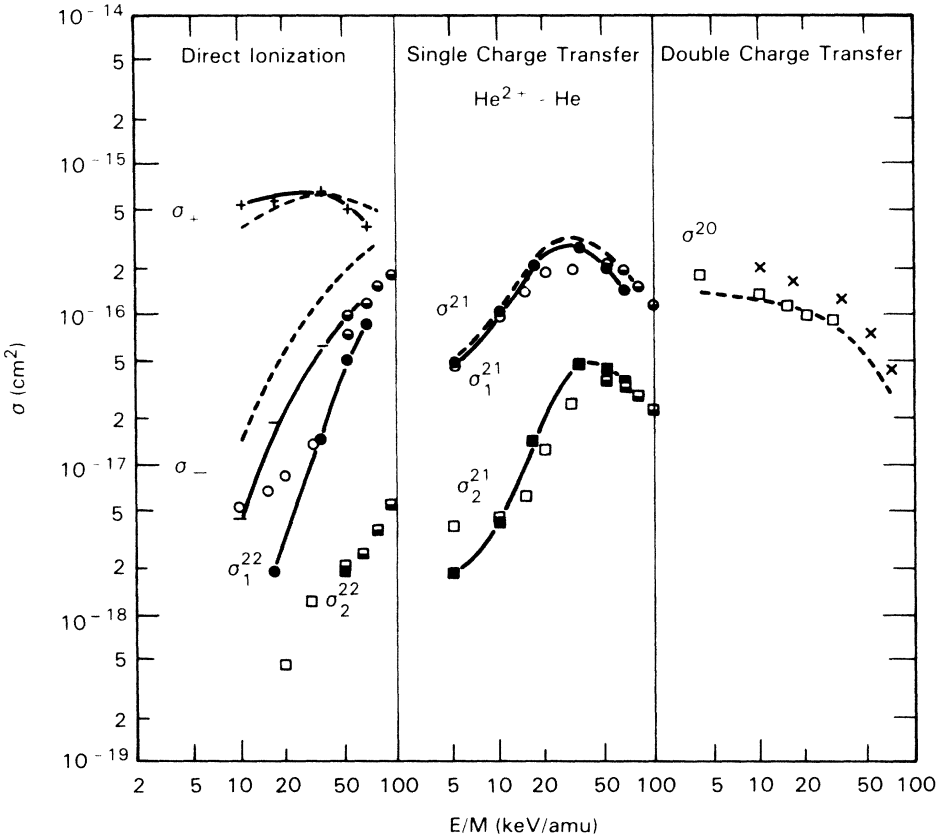

FIG. 1. Cross sections for ionization of helium by $^3He^{2+}$ impact. Total positive ion yields σ_+: +, present data; — — —, Ref. 15. Total free electron production σ_-: —, present data; — — —, Ref. 15. σ^{21}: — — —, Ref. 15, used for normalization. σ^{20}: ×, present data; — — —, Ref. 15; □, Ref. 12. σ_q^{22} and σ_q^{21}: $q=1$: ●, present data; ○, Ref. 12; ◒, Ref. 2. $q=2$; ■, present data; □, Ref. 12; ⬓, Ref. 2. Solid curves through present data are only to guide the eye.

$$\sigma_- = \sum_q q\,\sigma_q^{22} + \sum_{q>1} (q-1)\sigma_q^{21} + \sum_{q>2} (q-2)\sigma_q^{20}$$

is dominated by the single charge transfer plus ionization channel σ_2^{21} for most of the energy range shown. This was also found to be true for H^+ and He^+ impact[9] but only for heavier targets.

The total electron production in the present work is approximately a factor of 2 smaller than that measured previously by a more accurate method.[16] This again suggests a detection efficiency problem in one of the beam detectors. However, a 0.5 to 0.7 efficiency would be required for the He^+/He^{2+} detector (assuming unit efficiency for He^0 detection) to bring the present σ^{20} cross sections in compliance with previous data.[16] This is well outside the uncertainty of the 100% efficiency that was measured. Even if this measurement were in error, the σ_q^{22} and σ_q^{21} cross sections would not change because of the normalization procedure used. Only σ_q^{20} would be effected which would not alter σ_- for the helium target.

Neon

Results for neon are shown in Fig. 2. Below 40 keV/amu the double charge transfer channel σ_2^{20} again dominates the total double ionization. At higher energies the single charge transfer plus ionization channel σ_2^{21} is the most important. Note that double ionization of neon via the single charge transfer channel σ_2^{21} is approaching the pure single charge transfer cross section σ_1^{21} at higher energies but the direct double ionization channel σ_2^{22} is still unimportant. Triple ionization of neon is now becoming important. It occurs via the double charge transfer channel σ_3^{20} below 40 keV/amu and via the single charge transfer channel σ_3^{21} for higher energies.

Again we find that the total double charge transfer cross sections σ^{20} measured by the present method are larger than previous measurements by 30—50%. Also, as before, both the positive ion and electron production are dominated by the charge transfer channels with σ_2^{21} being the most important channel in producing free electrons. Again, good agreement is found between the present and previous data for total positive ion production although the total electron production is underestimated by approximately 50%.

Note in addition that there is good agreement between the present data for the single-electron transfer channel with the measurements of Groh *et al.*[7] Their measurements were placed on an absolute scale by normalizing to the same total single-electron transfer cross sections that

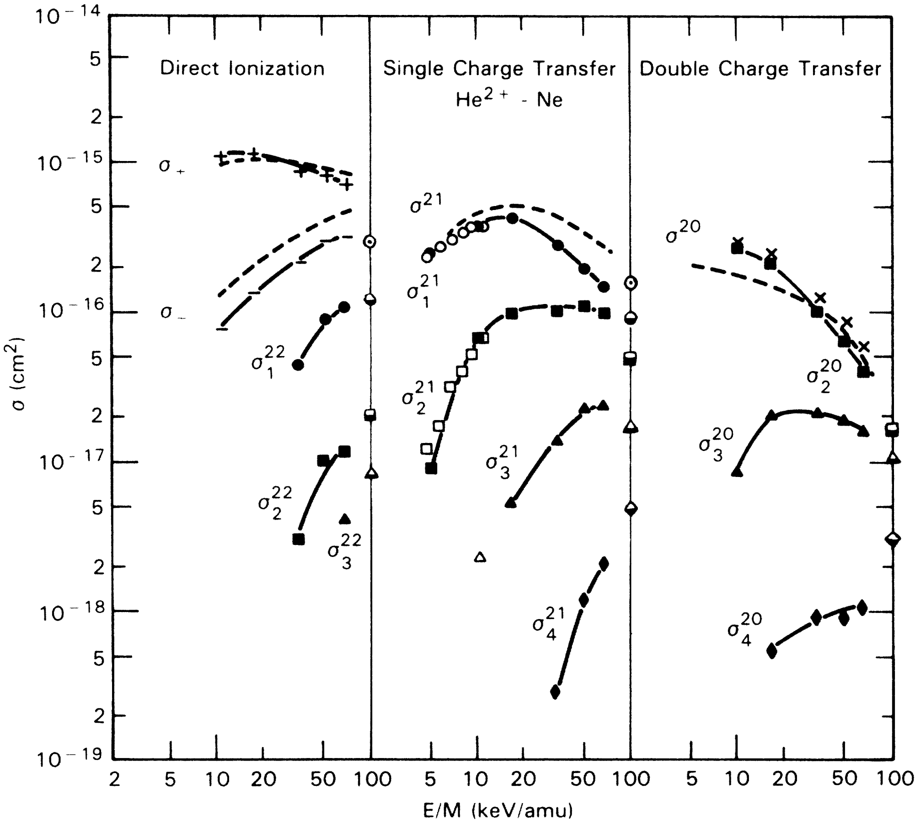

FIG. 2. Cross sections for ionization of neon by $^3He^{2+}$ impact. Total positive ion yields σ_+: +, present data; — — —, Ref. 15. Total free electron production σ_-: —, present data; — — —, Ref. 15; ⊙, Ref. 8. σ^{21}: — — —, Ref. 15, used for normalization. σ^{20}: ×, present data; — — —, Ref. 15. σ_q^{22}, σ_q^{21}, σ_q^{20}: $q=1$: ●, present data; ○, Ref. 7 norm to σ^{21}; ○, Ref. 8. $q=2$: ■, present data; □, Ref. 7 norm to σ^{21}; ⬓, Ref. 8. $q=3$: ▲, present data; △, Ref. 7 norm to σ^{21}, ◭, Ref. 8. $q=4$: ◆, present data; ⬙, Ref. 8. Solid curves through the present data are only to guide the eye.

were used for the present data.

The cross sections calculated by Becker *et al.*[8] for 0.1 MeV/amu He^{2+} impact are included in Fig. 2. As can be seen, the direct ionization cross sections σ_1^{22} and σ_2^{22} agree well with the extrapolated experimental cross sections although σ_3^{22} seems to be overestimated by theory. As stated previously, this same behavior was also observed for H^+-Ne collisions. In the single charge transfer channel, σ^{21} and σ_1^{21} agree well with experiment but theory underestimates both σ_2^{21} and σ_3^{21}. However, σ_4^{21} appears to be overestimated. The underestimation of σ_2^{21} and σ_3^{21} by theory results in a free-electron production cross section (σ_-) that is approximately 50% smaller than those measured experimentally.[16]

In the double-charge-transfer channel their calculated multiple ionization distributions were placed on an absolute scale by normalizing to a value of 3.3×10^{-17} cm^2 (obtained by extrapolating the present σ^{20} cross sections to 0.1 MeV/amu). The theoretical σ_2^{20} and σ_3^{20} cross sections are then in good agreement with experiment although σ_4^{20} is overestimated. Possibly the discrepancies between experiment and the theoretical calculations are due to the rather low energy where the comparisons are being made; however, the overall agreement between experiment and theory is encouraging and needs to be tested at higher impact energies.

Argon

In Fig. 3 cross sections for argon are given. The total double charge transfer channel is becoming less important and thus double ionization of argon is via the single charge transfer plus ionization channel. The charge transfer channels provide some very interesting and unexpected results. Previously Groh *et al.*[7] observed that in very low energy He^{2+}-Ar single-electron transfer collisions the production of Ar^{2+} exceeded that of Ar^+. They used a molecular-orbital (MO) analysis and showed that channels were available leading to double ionization that were not available for single ionization. The reason for the discrepancies between the present data and that of Groh *et al.* at 5 keV/amu is unknown but could be a result of the difficulty in obtaining such a low energy in the present experiment. At the highest energy measured, however, note that σ_2^{21} again equals σ_1^{21} and probably exceeds it at still higher energies. At these energies the MO argument used previously may be invalid since the projectile possesses sufficient kinetic energy to produce additional outer shell ionization. Even more interesting is the double charge transfer channel where σ_3^{20} is larger than σ_2^{20} throughout the entire energy range. Again the energies should be large enough to preclude using an MO analysis.

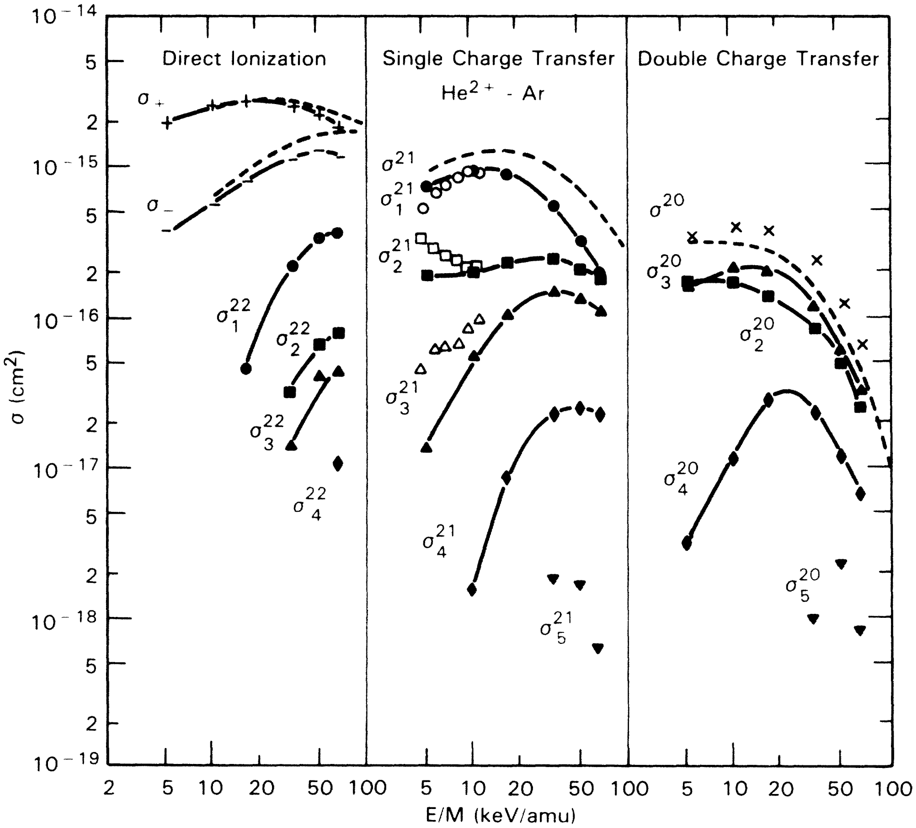

FIG. 3. Cross sections for ionization of argon by $^3He^{2+}$ impact. Total positive ion yields σ_+: +, present data; — — —, Ref. 15. Total free electron production σ_-: —, present data; — — —, Ref. 15. σ^{21}: — — —, Ref. 15 used for normalization. σ^{20}: ×, present data; — — —, Ref. 15. σ_q^{22}, σ_q^{21}, σ_q^{20}: $q=1$: ●, present data; ○, Ref. 7 norm to σ^{21}. $q=2$: ■, present data; □, Ref. 7 norm to σ^{21}. $q=3$: ▲, present data; △, Ref. 7 norm to σ^{21}. $q=4$: ◆, present data. $q=5$: ▼, present data. Solid curves through the present data are only to guide the eye.

The total positive ion production is again primarily due to the single charge transfer channel although now multiple ionization processes are becoming important. Agreement with the total yield measurements is quite good. Free-electron production is still dominated by the single-electron transfer channel although now higher order processes are increasing in importance. In addition the σ_3^{20} channel contributes significantly at low energies and the direct ionization channels are responsible for approximately 50% of the electron production at the highest energies. As before the total double charge transfer cross section appears to be approximately 30% too large although the total electron production is now only about 20% smaller than the data of Itoh and Rudd.[16]

Krypton

Results for krypton, shown in Fig. 4, demonstrate how higher orders of ionization are becoming increasingly more important. The total single charge transfer channel σ^{21} is now considerably larger than the total double charge transfer channel σ^{20}. Thus double ionization of krypton is mainly due to single charge transfer plus ionization σ_2^{21}. However, triple ionization of krypton at low energies is almost entirely via the double charge transfer channel σ_3^{20}. For krypton the present data are generally in good agreement with the data of Groh *et al.*[7] Although σ_2^{21} is smaller than σ_1^{21} at lower energies the opposite is true for larger energies.

The total double charge transfer cross sections are again approximately 25% too large. As was observed for argon, σ_3^{20} is larger than σ_2^{20} throughout the entire energy range; in addition σ_4^{20} exceeds σ_2^{20} at the higher energies.

The total positive ion production is still dominated by the charge transfer channels. The total electron production is still dominated by the charge transfer channels with direct ionization accounting for only 50% of the electron production at the highest energies. Agreement with the measurements of Itoh and Rudd is again quite good in both cases.

CONCLUSIONS

Cross sections for all channels making significant contributions to the positive ion and free-electron production in He^{2+}—rare-gas collisions have been presented for the energy range where the collision is changing from charge transfer to direct ionization domination. This is precisely the region where multiple ionization effects are largest and where theoretical treatments are scarce. The cross

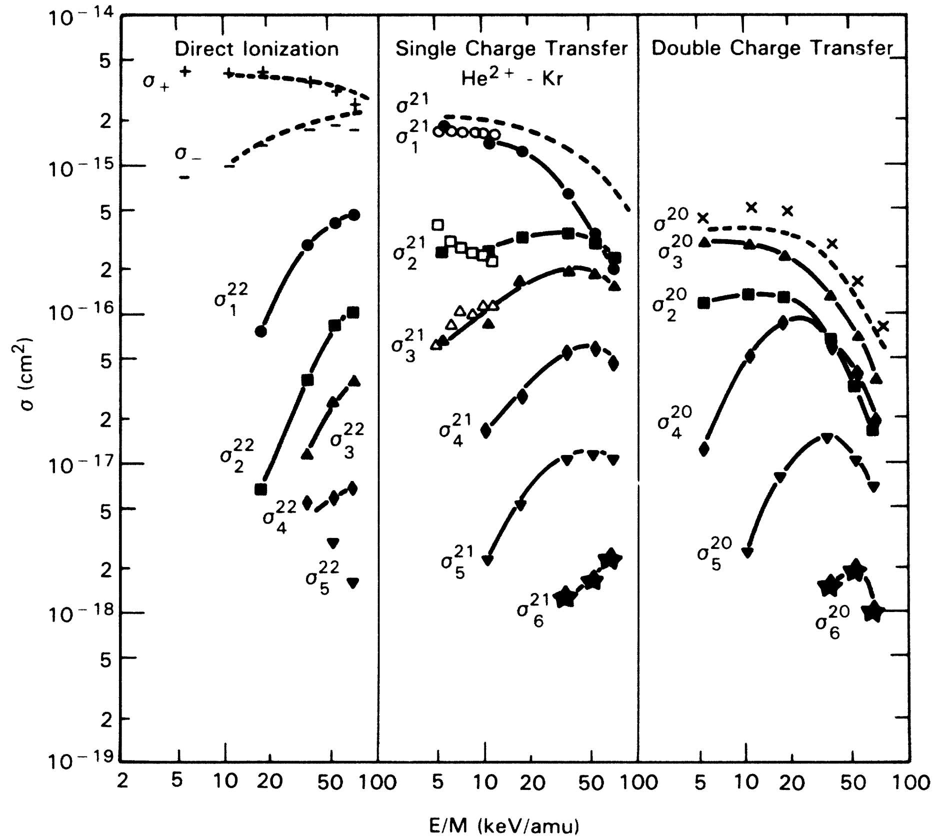

FIG. 4. Cross sections for ionization of krypton by ^{3}He^{2+} impact. Total positive ion yields σ_+: +, present data; — — —, Ref. 15. Total free electron production σ_-: —, present data; — — —, Ref. 15. σ^{21}: — — —, Ref. 15 used for normalization. σ^{20}: ×, present data; — — —, Ref. 15. σ_q^{22}, σ_q^{21}, σ_q^{20}: $q=1$: ●, present data; ○, Ref. 7 norm to σ^{21}. $q=2$: ■, present data; □, Ref. 7 norm to σ^{21}. $q=3$: ▲, present data; △, Ref. 7 norm to σ^{21}. $q=4$: ◆, present data. $q=5$: ▼, present data. $q=6$: ★, present data. Solid curves through the present data are only to guide the eye.

sections presented were made absolute by normalization to the single-electron transfer cross sections of Itoh and Rudd. However, in all cases the present measurements of the total double charge transfer cross section are approximately 30% larger than those of Itoh and Rudd. This could imply an efficiency problem for the different ionization channels which could possibly effect the magnitude of the direct ionization cross sections. However, such problems could not be experimentally identified.

It was shown that the electron production is dominated by the charge transfer plus ionization channels even for the helium target. A previous study of H^+ and He^+ impact found similar results—but only for heavier targets.[9] In addition it was shown that for argon and krypton, the single charge transfer plus ionization channel σ_2^{21} could equal or exceed the lower order pure single charge transfer channel σ_1^{21} not only at very low energies but also at much larger energies. Even more dramatic was that σ_3^{20} is larger than σ_2^{20} for the same targets throughout the entire energy range investigated. A detailed theoretical analysis may be required in order to understand these observations.

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