Projectile charge-state dependence of transfer ionization to single capture ratio in collisions of multiply charged ions with He

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The ratios of the cross sections for the processes of transfer ionization (TI) and single capture (SC) were measured for 2 MeV/u $Cl^{7+,9+,13+,14+,15+}$ and $Ti^{15+,18+}$ projectile ions on He targets. The ratio was determined using a cold He gas target and measuring the coincidences between the projectile and the charge states of the emerging recoil ions. The measured σ_{TI}/σ_{SC} ratio shows a strong dependence with the projectile charge state that is well described by calculations based on the independent electron model. A model to take into account the effect of the electron screening of partially dressed projectiles in the target ionization is also presented. [S1050-2947(97)02603-6]

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I. INTRODUCTION

Collisions involving multiply charged ions and neutral atoms are characterized by the presence and the simultaneous action of several collision channels, resulting in multielectron transitions within and between the participating systems. In general, these many-electron processes can be as likely as the single-electron ones, a characteristic that renders difficult a comprehensive theoretical description of the collision system. On the other hand, single-electron processes are simpler to calculate than many-electron ones, a feature that makes potentially useful the class of simple models that are able to express many-electron probabilities in terms of the single-electron probabilities. This is the main merit of the independent electron model (IEM), which has been successfully used in several collisions systems and at different velocity regimes [1]. Although the IEM considerably simplifies the analysis of the collision dynamics, its practical use in describing collisions involving highly charged ions is narrowed by the fact that some single-electron probabilities cannot be determined in a straightforward way.

This difficulty arises because collisions involving multiply charged ions usually have a channel that cannot be treated within first-order theories. Even if all other participating channels have sufficiently small probabilities to be treated up to first order, this dominant channel affects the experimental outcome of all other channels. Consequently, it must be well known if the whole behavior of the colliding system is expected to be understood.

In the intermediate-to-high velocity regime, the collision channels that occur with higher probability are the ionization and excitation of the target. Target ionization and excitation cross sections can reach very high values in collisions with highly charged ions and, under these conditions, any attempt to describe the collision system within first-order theories fails. This means that simple questions such as, for example, what is the dependence of some particular process with the projectile charge state or velocity, cannot be obtained by simple extrapolation of the results obtained from a similar system working in the perturbative regime.

Collisions involving highly charged ions with He are basic to our understanding of multielectron processes, not only to verify if the IEM works properly but also to establish if and how the parametrizations given by first-order theories break down. For example, the projectile charge state q affects all collision channels and, if q increases, there is a strong deviation from some of the first-order predictions, such as the quadratic dependence of the ionization or excitation cross sections with q. In this work we report the dependence, on the projectile charge state, of the ratio (R) between transfer-ionization (TI) and single-capture (SC) processes in 2 MeV/u $Cl^{7+,9+,13+,14+,15+}$ and $Ti^{15+,18+}$ collisions with He. The TI involves the removal of two electrons from He while the SC restricts the action of the projectile mostly over one electron of He. For both processes the IEM should be invoked and, because both of them include electron capture, the ratio R is expected to have a weak connection with the capture channel, presenting the characteristic quadratic q dependence of the ionization channel, at least for small values of q. This behavior was observed previously for bare light ions (q=1–8) [2]. For large values of q this first-order insight cannot be used. Previous measurements by Datz *et al.* [7] with U^{27+} and U^{35+} projectiles show, in fact, a strong deviation from the q^2 law.

The present measurements are carried out using bare as well as dressed projectiles. The understanding of collisions with bare projectiles is essential to proceed towards dressed projectiles but, again, results obtained through the former

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cannot be used indiscriminately to predict the behavior of the latter. Although for distant collisions a dressed ion can be considered as a ''bare'' projectile with a net charge q, for close collisions the effective charge increases towards Z_p, the projectile nuclear charge. Thus, the effective projectile charge has an impact parameter dependence that modulates the probabilities associated with the bare ions and the corresponding cross sections cannot be inferred from bare ion measurements.

This paper is arranged as follows: in Sec. II the experimental setup and data analysis are described; in Sec. III the experimental results and the IEM are discussed; in Sec. IV the unitarization procedure including the role played by the excitation channel is discussed; in Sec. V the effect of the screening for the dressed projectile cases is shown; and, finally, in Sec. VI a summary of the work is presented.

II. EXPERIMENT

The experiment was performed in the J.R. Macdonald laboratory at KSU. The high charge state beams ($q \geqslant 13$) were obtained through the LINAC while the lower charge states were obtained directly from the Tandem Van de Graaff accelerator. The collimated Cl^{q+} and Ti^{q+} beams, with typical currents of 50 pA, passed through a low density ($\sim 10^{11}$ atoms/cm^3) He gas jet target pointed transversely relative to the beam direction. Before and after the collision chamber there were a magnet and an electrostatic deflector, located at 0.5 and 0.05 m from the gas jet, respectively. The combination of electric and magnetic fields makes a clear charge state selection, eliminating undesirable contributions from beam contamination.

The He jet was collimated in such a way that the thermal momentum along the beam is smaller than 0.6 a.u. (see [3–5] for details). The He^+ and He^{2+} ions produced in the interaction region were extracted by a 5-V/cm uniform electric field and directed into a position-sensitive channel-plate detector. The projectiles emerging from the collision region were charge state analyzed by a magnet that directed them into another position-sensitive channel-plate detector placed about 4 m downstream, where the main beam (charge q) is blocked and the charge-changed beam (charge $q-1$) is detected. The capture projectiles (charge $q-1$) were measured in coincidence with the He recoil ions. A typical two-dimensional scatter plot of the recoil time of flight, which separates the He^{2+} from the He^+ recoils, versus the recoil position along the beam direction, which is proportional to the recoil longitudinal momentum, is shown in Fig. 1 for 2 MeV/u Ti^{15+} on He. The He^{2+} and the He^+ are readily visible at ~ 150 and ~ 210 on the time-of-flight axis. The recoil ions are thrown backwards in a capture event but not in an ionization event. Therefore, events involving a single capture will occur at a different location on the detector compared to an event involving only ionization. The events involving the capture of one electron, either TI or SC, occur at ~ 70 (arbitrary units) on the recoil longitudinal momentum axis and are well separated from the chance coincidences or events involving single ionization of He by $q-1$ impurity beam ions. Even though the impurity fraction is very small, typically 1% or less, the ratio of the cross section for pure ionization to that of pure capture at 2 MeV/u is >100. The

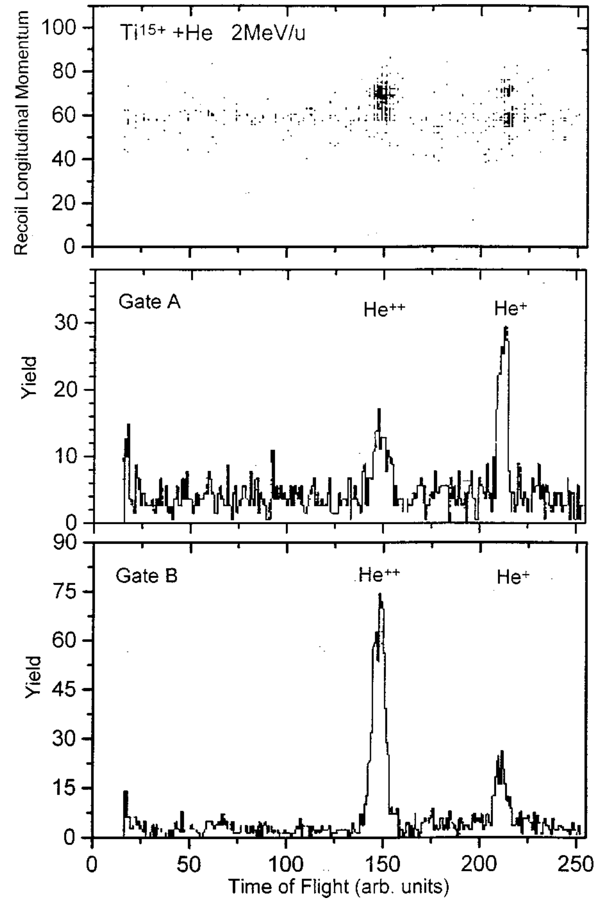

FIG. 1. Two-dimensional scatter plot of recoil time of flight (in arbitrary units) vs recoil longitudinal momentum (rlm, in arbitrary units) in coincidence with true-capture detected projectiles for 2 MeV/u Ti^{15+} incident ions. Gate A is the time-of-flight spectrum gated on random coincidences and ionization events (zero recoil longitudinal momentum). These events correspond to a window located at rlm ~ 60 in the scatter plot. Gate B is the time-of-flight spectrum gated on the true capture events. These events correspond to a window located at rlm ~ 70 in the scatter plot.

relative contributions of these processes can be assessed in the plots labeled Gate A and Gate B in Fig. 1. Gate A is the time-of-flight spectrum gated on the chance and ionization events (zero recoil longitudinal momentum). The He^+ and He^{2+} peaks are thus the contributions from single and double ionization of He by the $q-1$ impurity beam. Gate B is the time-of-flight spectrum gated on the true capture events (recoils thrown backwards). The He^+ and He^{2+} peaks in this spectrum represent the SC and TI events, respectively. One other important piece of information can be gleaned from this type of data. It is clear from the scatter plot that TI is indeed single capture plus ionization and not double capture followed by autoionization. The latter would occur at a recoil momentum twice the former and therefore would appear as a peak at ~ 80 (arbitrary units) on the recoil longitudinal momentum scale.

III. EXPERIMENTAL RESULTS AND THE IEM

Figure 2 shows our present measurements of R for He as a function of q, together with the O^{7+} and $F^{8+,9+}$ measure-

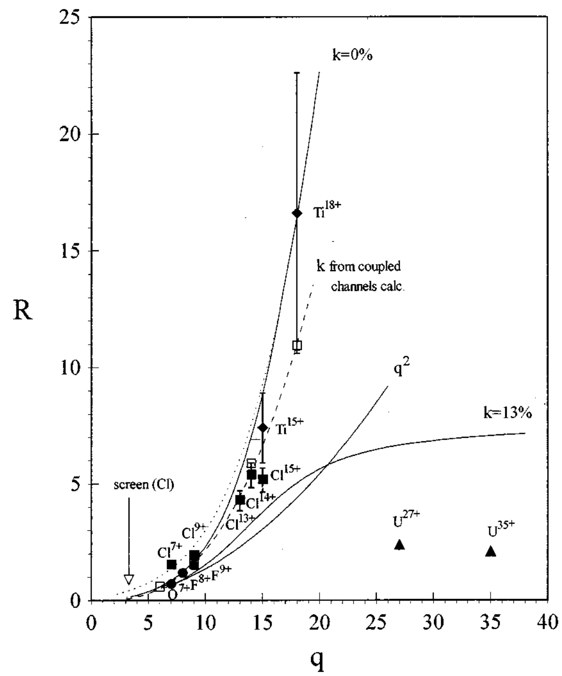

FIG. 2. Ratio of the cross sections of transfer ionization and single capture for 2 MeV/u projectile ions incident on He. Experiment (this work): solid squares are $Cl^{7+,9+,13+,14+,15+}$ data points; solid lozenges are $Ti^{15+,18+}$ data points. Experiment (Ref. [6]): solid circles are O^{7+} and $F^{7+,8+}$ data points; Experiment (Ref. [7]): solid triangles are 1 MeV/u $U^{27+,35+}$ data points. Theory: the thin solid curve indicated by q^2 is the square law relative to the He^{2+} projectiles; the thin solid curves indicated by $k=0\%$ and $k=13\%$ are the present model calculations (see text) considering 0% and 13% contributions of the excitation channel relative to ionization, at small impact parameters; the open squares (with the dashed curve to guide the eye) are the results of our model calculations obtained through the excitation to ionization ratio, k, at small impact parameters, given by coupled channel calculations; the dotted curve is the screening-corrected $k=0\%$ curve for Cl projectiles (see text).

ments of Shinpaugh *et al.* [6] and the $U^{27+,35+}$ results of Datz *et al.* [7] for 1-MeV/u projectiles. The figure also shows the q^2 law, obtained through normalization to the He^{2+} case, and which is clearly not followed by the present highly charged ions data. It is also apparent from this figure that the present measurements point towards a different trend from that shown by the previous measurements of Datz *et al.* for U^{q+} projectiles. However, it should be recalled that the U^{q+} data were obtained for 1-MeV/u projectiles. Although R shows a weak energy dependence for low q projectiles [6], this behavior might be not true for the high q cases.

There is a tendency of R to follow the q^2 law for small q. This trend was observed by Shinpaugh *et al.* [6] for $q \lesssim 8$ and can be explained with the aid of the IEM as follows. If the single-electron ionization and capture probabilities of He, $P_I(b)$ and $P_C(b)$, respectively, are much less than unity, R can be written within the IEM as

$$R=\frac{\sigma_{\mathrm{TI}}}{\sigma_{\mathrm{SC}}}=\frac{2\pi\int_0^\infty db\ b[2\,P_C(b)P_I(b)]}{2\pi\int_0^\infty b\ db[2\,P_C(b)]}. \tag{1}$$

Because $P_I(b)$ is approximately constant over the range of impact parameters where $P_C(b)$ is relatively large [8], one can make $P_I(b) \simeq P_I(0)$ in Eq. (1) to obtain $R \simeq P_I(0) \simeq q^2 p(0)$, with $p(0)$ being the ionization probability by protons with the same velocity at zero impact parameter. This simple reasoning shows not only the dependence of R with q^2 but also the weak dependence of R to the details of the capture process. However, as shown in Fig. 2, the validity of the quadratic law is restricted to $q \lesssim 8$.

For large values of q, the approximation $P_I(0) \simeq q^2 p(0)$ breaks down because the collision goes outside of the scope of the perturbative regime. Furthermore, for large q, the ionization and excitation probabilities can be near unity. Under these conditions, according to the IEM, the *single* capture probability is no longer given by $2P_C(b)$, but by $2P_C(b)[P_0(b)+P_E(b)]$, where $P_0(b)$ and $P_E(b)$ are the probabilities that the second electron of He either stays in the ground state or is excited, respectively. Assuming $P_0(b)+P_E(b)+P_I(b)+P_C(b)=1$, the ratio R is given by

$$R=\frac{\sigma_{\mathrm{TI}}}{\sigma_{\mathrm{SC}}}=\frac{2\pi\int_0^\infty db\ b[2\,P_C(b)P_I(b)]}{2\pi\int_0^\infty b\ db\{2\,P_C(b)[1-P_C(b)-P_I(b)]\}}. \tag{2}$$

The term $[1-P_C(b)-P_I(b)]$ in the denominator of Eq. (2) gives an additional dependence on q, preventing R from following the simple q^2 law as suggested by Eq. (1).

Another important point that can be seen from Fig. 2 is the systematic difference between the measured values of the dressed and undressed ions having the same charge state. This feature is more prominent for small values of q and is due to the decrease of the electron screening in close collisions. This point will be recalled in Sec. V.

IV. MODEL CALCULATIONS: UNITARIZATION AND THE ROLE OF THE EXCITATION CHANNEL

As mentioned before, the theoretical estimates of the probabilities corresponding to the inelastic channels in collisions between swift highly charged ions and neutral atoms are difficult to be obtained. This difficulty comes from different sources, such as the presence of strongly dominant channels, like ionization and excitation, which have very large cross sections [9], the strong nonperturbative character of the collision, and the presence of multielectron transitions that can occur through either single- or multiple-step processes.

Within this scenario, simple models based on first-order calculations that incorporate some *non-ad-hoc* procedure to force the necessary unitarization for the set of participating channels can be useful. In fact, in some complex cases, this can be the only available methodology able to give some guidance in interpreting the existing experimental data. In this paper we use an extended version of the unitarization procedure given by Sidorovich *et al.* [10], which conveniently allows the use of probability distributions obtained independently for the various competing channels.

Let us denote by $p_I(b)$, $p_C(b)$, and $p_E(b)$ the probabilities for a target electron to be ionized, captured, or excited, respectively. As mentioned above, these probabilities are calculated independently, and usually, although not necessarily, through first-order theories. Following Ref. [10] and including the excitation channel, the unitarized probabilities $P_\alpha(b)$, where α denotes I, C, or E, is given by

$$P_\alpha(b)=\frac{p_\alpha(b)}{p_I(b)+p_C(b)+p_E(b)}[1-e^{-(p_I(b)+p_C(b)+p_E(b))}]. \quad (3)$$

In this way, although the excitation channel does not appear explicitly in Eq. (2), it is implicitly included in the calculation of the probabilities $P_I(b)$ and $P_C(b)$. This inclusion is necessary because excitation is one of the dominant channels in these highly charged ion collisions. The question that can arise, however, is whether the excitation channel plays an important role in the numerical value of the TI/SC ratio.

It can be seen from Eq. (2) that $P_C(b)$ appears as a multiplying factor to both integrands in the numerator and in the denominator. Because $P_C(b)$ is important only at relatively small impact parameters (compared to the other collision channels), one should expect that the significant contribution from $P_E(b)$ and $P_I(b)$ to R should also come from the small impact parameter part of the corresponding probability distributions.

Although the total cross sections for He excitation and ionization by highly charged ions are both large, they have quite different impact-parameter distributions. The distribution $P_I(b)$ is maximum around $b=0$, decreasing with b, an indication of the predominance of close collisions in the ionization process. On the contrary, $P_E(b)$ is a much flatter distribution, indicating that soft collisions are relatively more important in the excitation process. These features can be seen in Fig. 3, which shows the coupled-channel atomic-basis calculations for $q=6$ and 14 bare projectiles on He, for these two processes, as well for their ratio. The general theory of the semiclassical close coupling method can be found in the review by Fritsch and Lin [11]. Briefly, target-centered closed-coupling expansions were used to obtain the single ionization and single excitation probabilities for He. For the collision velocity used in this work, the use of single centered atomic orbital expansion should be valid for describing the dominant ionization and excitation channels. The two-electron He target was described by a quasi-one-electron model in which the active electron moves in an effective potential due to the He nucleus and the passive electron. The single centered atomic basis used in this work is a Slater-type orbital. We found a good convergence by including target states (bound and continuum) up to $l=5$. As the dashed lines in Fig. 3 show, the relative importance of the excitation with respect to ionization increases with b, after being approximately constant for small b. This behavior allows us to set $P_E(b)/P_I(b)\simeq k$, where k is independent of b in the region where capture is more important, i.e., for small values of b. With this approximation, Eq. (3) becomes

$$P_\alpha(b)=\frac{p_\alpha(b)}{(1+k)p_I(b)+p_C(b)}[1-e^{-((1+k)p_I(b)+p_C(b))}]. \quad (4)$$

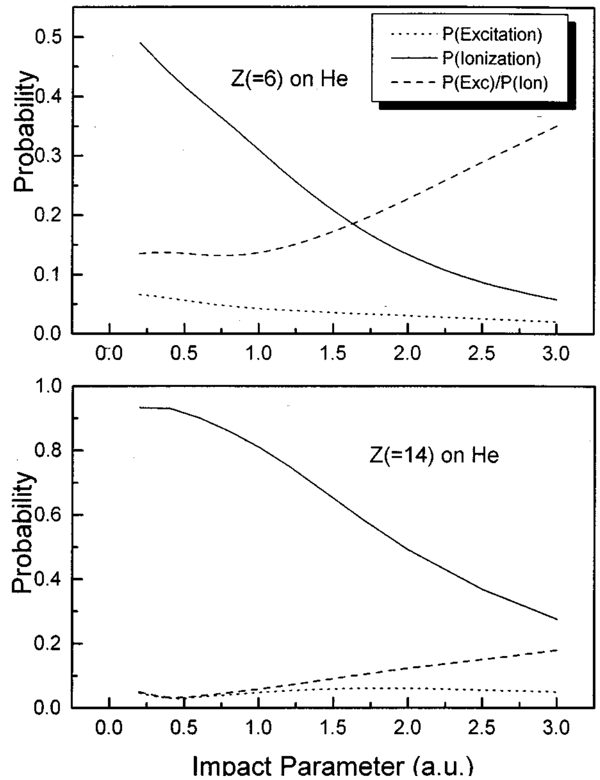

FIG. 3. Coupled-state calculations of target ionization and excitation probabilities as a function of the impact parameter for bare projectiles with (a) $Z_p=6$ and (b) $Z_p=14$ on He. Solid line, target ionization; dotted line, target excitation (to all single-electron states); dashed line, ratio between the excitation and the ionization probabilities.

The probability $P_C(b)$ is calculated using the model of Ben-Itzhak *et al.* [12], which follows the lines introduced by Bohr and Lindhard [13]. The probability $p_C(b)$, used to obtain unitarized probabilities for the other collision channels [Eq. (4)], is defined through the equation $P_C(b)=1-e^{-p_C(b)}$. In this way, if one sets $p_I(b)=0$ in Eq. (4), the capture probability given by Ref. [12] is recalled. On the other hand, if we set $p_C(b)=0$ in Eq. 4 we obtain $P_I(b)=[1-e^{-(1+k)p_I(b)}]/(1+k)$, which reduces to the unitarized ionization probability of Ref. [10], if $k=0\%$. The ionization probability by a projectile with charge q is calculated through the scaling law $p_I(b)=q^2p_{\rm sca}(b)$, where $p_{\rm sca}(b)=|a_{\rm sca}(b)|^2$ is the semiclassical calculation of the ionization probability by protons, taken from Ref. [14].

Figure 2 shows the theoretical estimates of the ratio R, calculated from Eqs. (3) and (4) as described above, and using $k=0\%$, $k=13\%$, and $k=3.2\%$ obtained from coupled-states calculations. These estimates were obtained for bare ions. An important point emerging from these calculations is that R is very sensitive to the ratio k. Furthermore, it is clear from this figure that the value $k=0$ gives good agreement with the experimental data, a result that clearly indicates that excitation is very small at small impact parameters, even for highly charged projectiles. This conclusion is corroborated by our coupled-channel calculations (Fig. 3), and is consistent with the fact that the use of values of k obtained from these calculations also gives a good description of the trend of the experimental data.

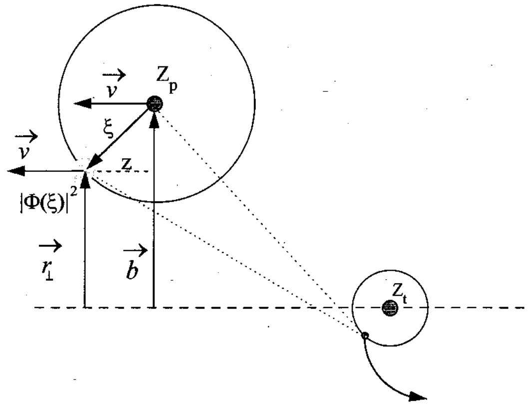

FIG. 4. Sketch of the target (Z_t) ionization caused by a dressed projectile Z_p. The ionization is due to a coherent superposition of the amplitude corresponding to the target nucleus, with an impact parameter b, and the amplitudes corresponding to the electron cloud. Each element of the electron cloud, located at ξ with respect to the projectile nucleus, has a local density $|\Phi(\xi)|^2$ and an effective impact parameter $r_\perp$.

V. SCREENING

If one compares the $Cl^{7+,9+}$ with the O^{7+} and F^{9+} data points in Fig. 2, it can be seen that the Cl data is systematically above the corresponding lighter ion data points with the same charge. Although these differences are not large, they are greater than the experimental uncertainties and indicate that different results are obtained if dressed projectiles are used, instead of bare, in this kind of experiment.

The deviation of the $Cl^{7+,9+}$ points from the curve calculated for bare ions, with $k=0\%$ in Fig. 2, is due to the partial electron screening, which occurs in close collisions. The contribution from the partial screening of a dressed projectile in the ionization process can be estimated using the semiclassical approach of Montenegro and Meyerhof as described in Refs. [15–17]. Following this model, the ionization probability by a dressed projectile can be viewed as a coherent superposition of two amplitudes: an amplitude related to the projectile nucleus and an amplitude related to the electronic cloud of the projectile. Figure 4 illustrates these two contributions. An impact parameter b and an amplitude $a_{\text{sca}}(b)$ are associated to the point nuclear charge Z_p. An electronic charge density $|\Phi(\boldsymbol{\xi})|^2$, an impact parameter $r_\perp$, and an amplitude $a_{\text{sca}}(r_\perp)$ are associated to each element $d\boldsymbol{\xi}$ of the electronic cloud, located at $\boldsymbol{\xi}$ with respect to the projectile nucleus. The resulting ionization probability is given by

$$P_I=\left|Z_p a_{\text{sca}}(b)-\int |\Phi(\xi)|^2 e^{i\omega z/v} a_{\text{sca}}(r_\perp)\, d\boldsymbol{\xi}\right|^2, \tag{5}$$

subject to the constraint

$$N_p=\int |\Phi(\xi)|^2 d\boldsymbol{\xi}, \tag{6}$$

where N_p is the number of projectile electrons.

It is instructive to note that if $N_p \ll Z_p$ and the collision is distant enough, we can make the approximation $r_\perp \simeq b$ in Eq. (5) (see Fig. 4). Neglecting the phase factor in the exponential appearing in this equation and using Eq. (6), we obtain the ''bare'' approximation, $P_I=q^2|a_{\text{sca}}(b)|^2$, with $q=Z_p-N_p$. For close collisions, however, these approximations cannot be done and the effective charge of the projectile is larger than q.

The electronic distribution of the projectile can be estimated through a Bohr-like potential

$$V(\xi)=\frac{Z_p-N_p}{\xi}+\frac{N_p}{\xi}\sum_i A_i e^{-\alpha_i \xi}, \tag{7}$$

which is related to $|\Phi(\boldsymbol{\xi})|^2$ through the Poisson equation,

$$\boldsymbol{\nabla}^2 V(\xi)=-4\pi[Z_p\delta(\xi)-|\Phi(\xi)|^2]. \tag{8}$$

Combining Eqs. (7) and (8) we obtain

$$|\Phi(\boldsymbol{\xi})|^2=\frac{N_p}{4\pi\xi}\sum_i \alpha_i A_i e^{-\alpha_i \xi}. \tag{9}$$

Substituting Eq. (9) into (5) and noting that $\xi=(|\boldsymbol{r}_\perp-\boldsymbol{b}|^2+z^2)^{1/2}$, we have, after some calculations:

$$P_I=\left|Z_p a_{\text{sca}}(b)-N_p\sum_i A_i\alpha_i^2\left[K_0(\gamma_i b)\int_0^b I_0(\gamma_i x)a_{\text{sca}}(x)x\,dx+I_0(\gamma_i b)\int_b^\infty K_0(\gamma_i x)a_{\text{sca}}(x)x\,dx\right]\right|^2, \tag{10}$$

with $\gamma_i=[(\omega/v)^2+\alpha_i^2]^{1/2}$ and K_0 and I_0 being modified Bessel functions. Equation (10) was evaluated numerically with the approximation $a_{\text{sca}}\simeq(|a_{\text{sca}}|^2)^{1/2}$. In these calculations the effective charge of the He target electrons was taken as 1.7 and the parameters A_i and α_i ($i=1$–3) were taken from the Hartree-Fock results of Ref. [18], calculated for neutral atoms, and considered the same for all projectile charge states.

The resulting screening contribution of TI/SC ratio for Cl ions with charge states $2<q<17$ is shown by the dotted curve in Fig. 2. The deviation from the bare ($k=0$) curve increases with N_p, as expected, and its magnitude is in good agreement with that given by the experiment. The effect of the screening is large enough to produce measurable deviations from the bare data but it is not sufficient to make significant changes in the general trend of the TI/SC ratio.

VI. SUMMARY AND CONCLUSIONS

The purpose of this paper is threefold. The first purpose is to obtain new and improved experimental data of the TI/SC ratio of highly charged projectiles on He. This was achieved through the combination of a cold-gas-jet and a recoil-ion detector in a system that is able to record both the position and the time to flight of the recoiling ions. This combination allows a clear separation, from the background, of the single-capture events. The capture channel has low intensity for high projectile charge states, and contributes to the errors in the TI/SC ratio measurements. In fact, because the SC cross section is much less than the target ionization cross section for the cases studied in this work, small beam impurities can be a major source of error in this kind of measurement if not properly separated.

The second purpose is to verify the adequacy of the IEM in a collision system that has several channels that cannot be treated perturbatively. The calculation of the various channel probabilities through the use of a simple unitarization procedure, based on first-order calculations, presents a good agreement with the present experiment. This agreement seems not to be fortuitous. Recent studies, based on the direct measurement of the impact-parameter dependence of SC and TI processes, have shown that such model calculations give a good agreement with the experiment at the impact-parameter level [8]. This behavior, together with the fact that the measured longitudinal momenta for TI and SC are the same, points towards the conclusion that TI is not due to the transfer of two electrons followed by autoionization. A conclusion pointing in the same direction was reached by Datz *et al.* [7] through the analysis of the emitted electron spectra. These authors concluded that TI is not due to the capture of one electron in a bound state accompanied by a capture of a second electron in the continuum.

The third purpose is to make a quantitative comparison between dressed and undressed projectiles with the same charge state. The proposed model gives a good estimate of the observed differences between these two kinds of projectiles. However, because highly charged ions have a great ability to capture or ionize target electrons at large internuclear distances, the effect of the partial screening, which occurs essentially at close collisions, does not have a large influence on the measured TI/SC ratio.

Finally we should comment that, within our present understanding of the behavior of the TI/SC ratio, we do not see how to conciliate the trend presented in this work with the measured U^{q+} data from Ref. [7]. However, we should point out that there is still a large gap between the projectile charge states used in these two sets of data, and the projectile energy used in these measurements. As the collision systems shown in Fig. 2 are highly nonperturbative, any attempt to make simple extrapolations in these collision regimes is not fully justified.

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