Enhanced Double Ionization of the $3d\sigma$ Molecular Orbital at Small Impact Parameters

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The impact-parameter–dependent probabilities for single- and double-vacancy production in the $3d\sigma$ molecular orbital have been determined for 1.5-MeV/nucleon ^{58}Ni on ^{208}Pb collisions from triple coincidences between Ni K and Pb L characteristic x rays and scattered particles. The observed double-ionization probability is enhanced relative to the predictions of the independent-particle model based on the measured single-ionization probabilities by factors varying between 2 and 3. The presence of correlated electron processes during close collisions, with probabilities as large as a few percent, could explain the observed enhancement.

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Currently, there is considerable interest in electron correlation effects which occur during a collision,[1,2] the so-called "scattering correlations."[3] These effects are produced by the mutual (Coulomb) interaction between two electrons whose description leads one beyond the independent-particle model (IPM). If one can obtain within this model the probability of a two-electron transition essentially by the product of the probabilities for the corresponding one-electron processes, then correlation effects can be verified directly by observing the probability for a two-electron process to be large in comparison with the related factorized probability. In this case, it is possible to compare exclusively experimental results, as has been done in the present work.

Significant electron correlation effects in ion-atom collisions are believed to occur in double ionization. In particular, two-electron processes in the collisions of fast protons (p) and antiprotons ($\bar{p}$)[4] with He atoms have been the subject of extensive research (see, e.g., Ref. 5 and the references quoted therein). The cross-section ratio for double to single ionization has been found systematically higher for $\bar{p}$ than for p impact, while the corresponding single-ionization cross sections are found to be identical within experimental uncertainties. In order to explain this p-$\bar{p}$ difference, McGuire[6] suggested that the double ionization of He is described by two different amplitudes, one involving only nucleus-electron interaction, the other, the electron-electron interaction. The two amplitudes can interfere, resulting in a term which depends on the sign of the projectile charge. *Ab initio* quantal calculations[7] have shown that to obtain quantitatively the experimentally observed p-$\bar{p}$ double-ionization difference, it is necessary to have (i) terms in the Born series past the first, (ii) electron correlation effects, and (iii) nondipole transitions. They also revealed that the effect is much more pronounced at small impact parameters.

Recently, differential cross sections for double and single ionization of He resulting from impact of fast protons have been measured as a function of both the scattering angle and the impact energy.[5,8] At small scattering angles (<0.55 mrad) the differential cross sections are dominated by the binary encounter of the projectile proton and the quasifree target electron, and the importance of electron correlation was made evident on the basis of a comparison with photoionization data.[8] In the transition region between small and large deflections, at proton energies below 1 MeV a distinct peak is observed near 0.9 mrad in the double- to single-ionization differential-cross-section ratio.[5] The peak values at each energy are considerably larger than the ratio of the integrated cross sections, implying that the enhancement occurs in an angular region beyond that of very small scattering angles which counts in the total cross section. Controversial explanations have been proposed[9] for this enhancement of the double ionization, running from successive projectile encounters with independent target electrons to higher-order processes and interferences among them.

The aim of the present work is to look for possible deviations from IPM, such as electron correlations in double ionization of heavier collision systems. To retain the relative simplicity of the two-electron, two-nuclei problem, we have chosen to study the ionization of a selected molecular orbital (MO), transiently formed in slow (asymmetric) ion-atom collisions. In particular, the electrons promoted into the $3d\sigma$ MO in the region of K-L level matching are easily ionized at small impact parameters, resulting in ionization probabilities of the order of 10^{-1} or larger.[10] Consequently, double-ionization probabilities of the order of 10^{-2} are expected based on IPM. A rough estimate shows[2] that a correlated process should be of the same order of magnitude as such an IPM double-ionization probability. Therefore it would be able to alter significantly the double-ionization probability relative to the IPM expectation. From the experimental point of view, the interpretation of the x-ray spectra in noninverted[11] "swapped"[12] collision systems near K-L level matching is simplified at low velocities by the concentration of the vacancies in the K level of the lighter collision partner and in the L_3 level of the heavier one. The reason is the dominance[11] of the direct excitation of the promoted electrons out of the $3d\sigma$ level near the united-atom limit,[13] followed by sharing of vacancies between the MO levels correlating to the separated-atom K and L_3 shells of the lighter and heavier collision partners, respectively.[12] The situation is illustrated in the qualitative correlation diagram of Fig. 1.

Here we report results on single- and double-inner-shell-vacancy production in 87-MeV ^{58}Ni+^{208}Pb collisions at small impact parameters. The measurement has been performed at the MP tandem accelerator of the Max-Planck-Institut für Kernphysik in Heidelberg, using an x-ray–x-ray–scattered-particle coincidence technique (X-X-P).[14] The details of the experiment and data analysis will be reported elsewhere[15] and will be only briefly discussed here. The x rays were detected by two Si(Li) detectors located at 104°, and about 15 mm distance relative to the beam axis. With several tens of pA current of Ni^{7+}, the x-ray count rate was kept below 2 kHz. The particles were detected in a parallel-plate avalanche detector (PPAD). Its anode was divided in concentric rings which defined, in our geometry, ten impact parameters in the range $b=175$–550 fm. The inclusive probability of one-vacancy production as a function of b has been measured separately with the same setup, and has been found in agreement with previous measurements.[10] In order to detect any contribution from vacancy production in two successive collisions, we used three target thicknesses, i.e., 10, 20, and 40 μg/cm^2 of Pb evaporated on 15-μg/cm^2 carbon backings. The target uniformity was checked with an optical microscope. The contribution of the C backing to the triple-coincidence yield has been measured in an independent run and found negligible.

Figure 2 shows the time spectrum of the X-X-P coincidences. The true triple coincidences form a peak, while the various ridges correspond to partly correlated events such as X_1-P true, X_2 random coincidences. They have been subtracted, correcting for the double counting of the completely uncorrelated events which form the overall background in Fig. 2. After normalizing to the scattered particles, no target-thickness dependence has been found within the statistical uncertainty, indicating a negligible contribution of the double collisions to the observed triple-coincidence yield. The energy spectrum of the x rays detected by one of the detectors in true coincidence with both the x rays detected by the other detector and the scattered particles shows hypersatellite components besides the Ni K_α and Pb L_α satellite lines. This confirms the existence of double vacancies in the projectile K and target L shells.

By normalizing to the scattered particles, and using fluorescence yields,[16] K_β/K_α ratios,[17] and the decay width[18] for singly ionized atoms, the X-P and X-X-P true coincidence rates were converted into single Ni-K, single Pb-L_3, double Ni-K, Ni-K and simultaneously Pb-L_3, and double Pb-L_3 shell ionization probabilities, P_K, P_{L_3}, P_{KK}, P_{KL_3}, and $P_{L_3L_3}$, respectively. Given the weak ionization of the L_1 and L_2 subshells as compared to L_3, the feeding of the latter by Coster-Kronig transitions has been neglected. Effects such as the contribution of Ni K x rays and scattered particles from the C backing were small, but have been accounted for. The triple coincidences were summed up for adjacent rings of the detector until the statistical fluctuations became less than 20%. Thus, the double-ionization data represent weighted averages over ranges of impact parameters. The uncertainties in the detection efficiency and in the normalization procedure introduce an additional sys-

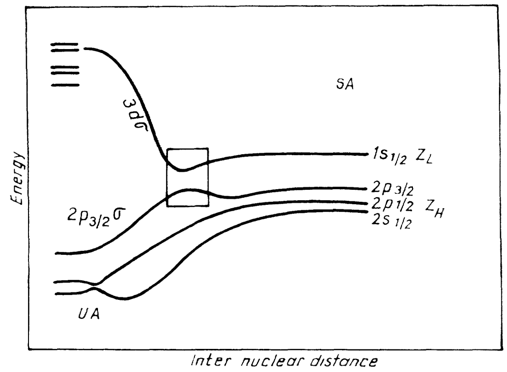

FIG. 1. Qualitative correlation diagram for a "swapped" collision system near the K-L level matching, connecting the united-atom (UA) levels to the separated-atom (SA) levels of the light (Z_L) and heavy (Z_H) collision partners, respectively.

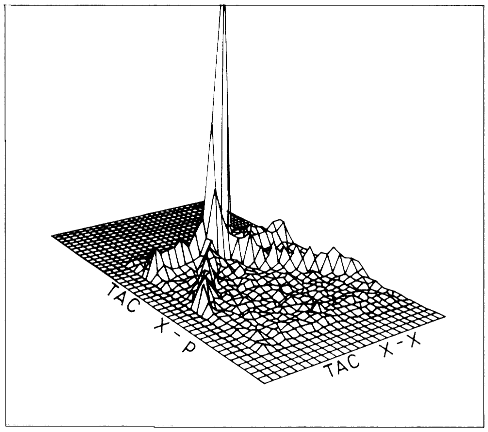

FIG. 2. The time spectrum of the X-X-P coincidences.

tematic error on the absolute scale of about 20% for single- and 30% for double-vacancy probabilities.

A detailed analysis[15] shows that both the magnitude and impact-parameter dependence[13] of the measured ionization probability, as well as the vacancy sharing probability,[12] are consistent with the MO excitation model. According to this (see also Fig. 1), only the $3d\sigma$ and $2p_{3/2}\sigma$ MO's contribute significantly to the observed K and L x-ray yields.[11,12] Since we have found[15] that the ionization probability for $2p_{3/2}\sigma$ is, on the average, only 10% of that for $3d\sigma$, we take

$$P_{KK}+P_{KL_3}+P_{L_3L_3}=P(2) \tag{1}$$

as the exclusive double-ionization probability, and

$$P_K+P_{L_3}=P(1)+2P(2)=Q(1) \tag{2}$$

as the inclusive single-vacancy probability for the $3d\sigma$ MO. The experimental results for $Q(1)$ and $P(2)$ are shown in Fig. 3.

The simplest independent-particle model for vacancy production results in a binomial distribution of the vacancies in $3d\sigma$ MO:

$$P(0)=(1-p)^2\,, \tag{3a}$$

$$P(1)=2p(1-p)\,, \tag{3b}$$

$$P(2)=p^2\,, \tag{3c}$$

where p is the vacancy production probability per electron. If its value p_{exp} is obtained from $P(1)=Q(1)-2P(2)$ according to Eq. (3b) and is introduced in Eq. (3c), one has constructed the experimental IPM expectation for the double-ionization probability p_{exp}^2. An analysis of Fig. 3 reveals that the measured $P(2)$ is systematically larger than p_{exp}^2. Defining an enhancement factor

$$F^2=P(2)/p_{\text{exp}}^2\,, \tag{4}$$

for the present data it reaches values as high as 3.2. Such a "discrepancy" is far beyond the experimental uncertainties. Also it cannot be ascribed to the use of fluorescence yields for singly ionized atoms. Indeed, an analysis[15] of the outer-shell multiple-ionization effects shows that, in spite of a reduction of $Q(1)$ by 35% and of $P(2)$ by almost 60%, the reported values of F^2 would only decrease by 16% on the average.

The enhancement factor is shown in Fig. 4 as a function of the reduced impact parameter $x=b/\bar{b}$. Here $\bar{b}=hv/\Delta E$ is the so-called "adiabatic radius," i.e., the impact parameter which characterizes the region with the greatest contribution to the ionization cross section,[20] v being the collision velocity and ΔE the energy transfer to the excited electron. The latter has been approximated by the binding energy of the $3d$ level in the united atom.[21]

Similar enhancement of $P(2)$ relative to the IPM is revealed in the U L-vacancy production in 1.4-MeV/nucleon U+Sn collisions,[19] when the data are reanalyzed as above. More specifically, since the authors of Ref. 19 have found that the subshell differentiation was of minor importance, denoting by i the probability of ejecting one U L-shell electron, one can write $P(1)=8i(1-i)^7\simeq P_L-2P(2)$, $P(2)=P_{LL}/2$; for $F^2=P(2)/28i^2(1-i)^6$ one obtains the results represented as open circles in Fig. 4. In calculating $\bar{b}$ for U+Sn collisions, a value $\Delta E\sim 12$ keV has been used, as obtained from the experiment.[19] In spite of a different correlation diagram, the results for U+Sn are in surprisingly good agreement with the enhancement found in Ni+Pb collisions at about the same velocity. Similar, or even larger, enhancement is found assuming that fewer than eight electrons can be ionized with non-negligible probability out of the U L shell. Thus, for the limiting case of only two electrons, F^2 varies between 3.1 and 5.6.

The present data, and also the data of Ref. 19, indicate an important enhancement of the double ionization in a region of scattering angles beyond the region which dominates the total cross section ($x<1$). The result is based on a comparison of the experimental single- and double-ionization probabilities by means of the "product rule," Eq. (3c). This rule postulates that the IPM transition probability for a two-electron process is equal to the product of the probabilities for the associated one-electron processes.[2] The product rule has to be used with care when antisymmetrization effects are important. These effects, however, are expected to be negligible in the case of our experiment.[15] For the system investigated here the collision velocity was low enough so that the

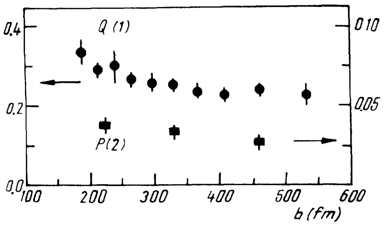

FIG. 3. The double-ionization (■, right scale) and inclusive single-ionization (●, left scale) probability of the Ni-K and Pb-L_3 manifold as a function of the impact parameter for 87-MeV ^{58}Ni on ^{208}Pb.

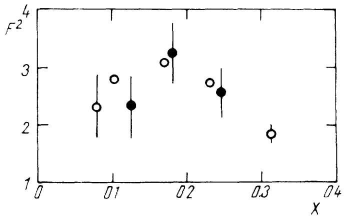

FIG. 4. The enhancement of the double ionization relative to the predictions of the independent-particle model vs the reduced impact parameter, for the $3d\sigma$ MO (●) in 1.5-MeV/nucleon ^{58}Ni on ^{208}Pb collisions (this work) and for the U L shell (○) in 1.4-MeV/nucleon U on Sn collisions (Ref. 19).

projectile 1*s* electrons were promoted on the incoming trajectory to form the 3$d\sigma$ MO and then were ionized in the vicinity of the closest approach between the projectile and target nuclei. Probing a state transiently formed during the collision, the present experiment is probing the *scattering* electron correlations, if these are indeed responsible for the observed enhancement of the double ionization. Near the united-atom configuration, the second-order probability for double ionization is likely to have the form[6]

$$P_{if}^{(1,2)} = |a_I^1 a_I^2 + a_I^1 A_I^e|^2 , \qquad (5)$$

where a_I^1 and a_I^2 are single-electron amplitudes for ionization due to the interaction of the two 3$d\sigma$ electrons (labeled 1 and 2) with the *two* nuclei[22] and A_I^e is the ionization amplitude due to electron-electron interaction. For $A_I^e=0$, Eq. (5) reduces to Eq. (3c). The amplitudes $a_I^{1,2}$ describe essentially monopole transitions.[23] If A_I^e also corresponds to a monopole transition, the two terms of Eq. (5) may interfere constructively, which would explain the observed enhancement of the double ionization. From the magnitude of this enhancement one may infer $|A_I^e|^2 \lesssim |a_I|^2 = p_{\text{exp}}$; i.e., the probability of a correlated process is, in the case investigated, of the order of several percent (we assume $|a_I^1| = |a_I^2| = |a_I|$). However, according to Eq. (5), in order to be observed, some action of the electron-nucleus interaction is required before the electron-electron interaction becomes effective.

In conclusion, a significant enhancement of the double ionization of the 3$d\sigma$ molecular orbital relative to the independent-particle-model predictions has been observed in 1.5-MeV/nucleon ^{58}Ni+^{208}Pb collisions well inside the adiabatic radius. A similar enhancement is exhibited also by the double ionization of the U *L* shell in collisions with Sn at comparable velocities.[19] Scattering correlations with probabilities in the 10^{-2} range are suggested as a possible explanation of these results. A definite conclusion, however, awaits more data and detailed calculations.

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