Dependence of the K-shell ionization probability on small impact parameters with 1- and 2-MeV protons

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The absolute impact-parameter dependence of K-shell ionization probability $I(p)$ has been measured in the range 10^{-12}–10^{-11} cm at incident proton energies of 1 and 2 MeV with Al, Cu, and Ti targets. In the first part we give a condensed survey of previous results showing that a detailed investigation of $I(p)$ behavior at very small impact parameters is required for a better understanding of this problem. Such a study provides a useful check on the accuracy of classical and semiclassical ionization theories in a region where they are more likely to fail. Our results, obtained by a coincidence method, are compared with the presently available theories: the binary-encounter approximation and the semiclassical approximation. A systematic deviation is found between experimental and theoretical data which is not accounted for when relativistic effects and the actual projectile trajectory are included in the binary-encounter–approximation model.

I. INTRODUCTION

Mainly as a result of progress in experimental techniques and applications for trace analysis, ionization of inner shells by heavy charged particles has been widely investigated in the last few years. In the following, we restrict ourselves to ionization of the K shell by projectiles with atomic number Z_1 much smaller than the target atomic number Z_2. In this case, the ionization process is described in terms of a direct Coulomb interaction between the projectile and the target electrons. A great number of total K-shell ionization cross-section data are now available[1] which generally agree with theoretical calculations. Three different theories have been developed, namely, the plane-wave Born approximation (PWBA),[2] the semiclassical approximation (SCA),[3] and the binary-encounter approximation (BEA).[4] Even at low projectile velocities, these theories agree with experimental total cross sections when account is taken of the Coulomb deflection of the incident particle in the field of the target nuclei and the increase in the K-shell binding energy.[5] However, the success of these oversimplified theories may appear to be somewhat surprising. For example, relativistic effects are usually neglected, and other important effects, when included, are taken into account in a too approximate way, such as the projectile retardation by the target nucleus,[6] the increase in the K-shell binding energy,[5] and the final-state interaction between the projectile and the outgoing electron. Furthermore, some of these effects might sometimes cancel and lead to a fortuitous agreement between simplified theories and experiments.

Thus, measurements of the K-shell ionization probability, as a function of the collision impact parameter $I(p)$, are of great importance since they can give a more detailed test of the ionization mechanism. Recently, experimental determinations of $I(p)$, using coincidence measurements between the diffused projectile and x rays, have been given in the range $10^{-10}<p<10^{-11}$.[7,8] The shape of the curve was found in relatively fair agreement with SCA or BEA predictions, but substantial discrepancies appear for large as well as small p values. This paper relates extension of $I(p)$ measurements for very small impact parameters ranging between 10^{-11} and 10^{-12} cm. For example, in the Cu case, the impact parameter corresponding to the angle 50° is only about three times the nuclear radius.

II. THEORETICAL CONSIDERATIONS

The first complete description of the ionization of inner shells by heavy particles was given by Merzbacher and Lewis using the PWBA.[2] This quantum theory does not give the ionization probability dependence on impact parameter. On the other hand, the SCA theory as developed by Bang and Hansteen[3] predicts the $I(p)$ dependence on p. Here, the projectile motion is treated classically and a quantum first-order time-dependent perturbation theory is used to derive the ionization probability per energy interval dE_f of the ejected electron as a function of p:

$$\frac{d\sigma}{dE_f}(p)=\frac{1}{\hbar^2}\left(\int_{-\infty}^{+\infty}e^{i\omega t}\langle f|V(t)|i\rangle dt\right)^2, \tag{1}$$

where $\langle i|$, $\langle f|$ are the hydrogenic nonrelativistic wave functions of the initial and final state of the electron, $\omega=(E_f+E_B)/\hbar$, E_B is the binding energy of the K-shell electron, and $V(t)$ is the perturbing Coulomb potential.

This theory can account for the deflection of the projectile in the field of the target nucleus, which effect is more pronounced in the low-energy case and for large deflection angles.[3] For adiabatic collisions, the monopole interaction is preponderant. For nonadiabatic collisions, the contribution of higher-order multipoles becomes important and the SCA can give their relative dependence on impact parameter.

Using the framework of the SCA, Brandt *et al.*[8] derived a simple analytical formula for $I(p)$, strictly valid in the adiabatic case. This formula leads to a useful scaling law which facilitates the comparison of results between projectiles of any atomic number and velocities impinging any targets. A universal curve is obtained by plotting the results as a function of universal variables W and y:

$$y=c\epsilon x, \tag{2}$$

where $x=pq_0$ introduces the projectile velocity v_1 with $q_0=E_B/\hbar v_1$, c accounts for the Coulomb deflection of the incident particle, and ϵ incorporates the change of binding energy of the K-shell electron due to the projectile. W is related to the ionization probability as a function of y, $I(y)$, as

$$W=I(y)/y^2. \tag{3}$$

Hereafter, this theory will be referred to as the low-energy SCA, while the Hansteen calculation will be referred to as the "full" SCA.

When applied to total-cross-section calculations, the BEA theory gives impressive agreement with experiment. Very recently, McGuire[9] and Hansen[10] have developed an impact-parameter formalism, in which the collision between the projectile and the electron is fully treated classically. $I(p)$ is evaluated as a function of the density distribution of the electron $\rho(r)$ and the collision cross section for two-body Coulomb interaction $\sigma(v_1, v_2)$ is evaluated as

$$I(p)=2\int_0^{\infty}\rho((p^2+z^2)^{1/2})\sigma(v_1, v_2)dz, \tag{4}$$

where the electron velocity v_2 is a function of the electron-nucleus distance.

For hydrogenic electron distribution simple scaling laws can be derived. McGuire[9] gives a table of $I(p)$ values for the proton-hydrogen case. Using the scaling laws it is easy to calculate $I(p)$ for arbitrary targets and projectiles of any velocity. The very simple expression of $I(p)$ in the BEA formalism gives the possibility for introducing second-order phenomena such as deflection, retardation, etc.[10]

III. SUMMARY OF PREVIOUS MEASUREMENTS

Laegsgaard, Andersen, and Feldman[7] first reported measurements of the ionization probability dependence on the impact parameter. They used 1- and 2-MeV protons as projectiles to create K-shell vacancies in Cu, Se, and Ag. At each angle, the scattered protons were detected in a magnetic spectrometer, the energy resolution of which was good enough to discriminate between protons which had produced a K-shell vacancy and those elastically scattered. The spectrum of inelastically scattered protons in coincidence with K x rays was recorded. The data are limited to a maximum angle $\theta=10°$, corresponding to p values ranging between 3×10^{-10} and 3×10^{-11} cm. These results were compared to SCA calculations[7] and later to BEA theory[9] assuming a straight-line trajectory of the projectile. Both theories reproduce approximately the experimental curve shapes. However, we feel that SCA gives somewhat better $I(p)$ absolute values especially for the smallest p values.

Later Brandt, Jones, and Kramer[8] measured a large number of $I(p)$ values. The diffused particle was detected at a fixed angle 5.1° in coincidence with K x rays emitted by several targets Al, Ca, Ni, Ag. The range energy projectile was 0.3–3 MeV. The K x rays were detected in a NaI (Tl) scintillator detector coupled with a low-noise photomultiplier. These data, as well as the previous, were shown on the universal low-energy SCA curve.[8] At low energy and large p values, the agreement is satisfactory. However, deviation from this scaling law begins to occur at values of $y=0.5$. Although the authors pointed out that the given expression can only be considered as a guide in the nonadiabatic case, this cannot explain the very large deviation of $I(p)$ for small p as measured with 2-MeV H^+ impinging upon an Al target.

One may wonder whether the direct Coulomb process still remains dominant for higher-Z projectiles in the nonadiabatic case. Coke and Randall[11] have measured $I(p)$ by a coincidence technique for oxygen beams of 25, 35, and 45 MeV (charge states 4^+, 6^+, and 7^+, respectively) bombarding a Cu target. The comparison of these results with SCA and BEA is much more difficult than in previous cases. While the shape of the curve $I(p)$ vs p, when normalized to total cross sections, is in general agreement with the SCA prediction, the experimental absolute values are three times larger than the calculated one. The discrepancy becomes even larger for small values of p.

Concluding this summary, we can say that the gross features of the experimental data are approximately accounted for by theory. However, the resulting calculations where compared to experimental data generally overestimate $I(p)$ for the largest p values and underestimate $I(p)$ for the smallest. The aim of the present paper is to investigate the region of very small impact parameter, where possible failures of classical and semiclassical theories are more likely to occur.

IV. RESULTS AND DISCUSSION

The K-shell ionization probability was measured by a coincidence technique for 2-MeV protons on Cu, 1-MeV protons on Ti, and 1- and 0.5-MeV protons on Al. The Cu and Al targets were self-supporting; Ti was evaporated onto very thin carbon backings. In the present set of experiments, the minimum angle at which protons were detected is 16° leading to impact-angle parameters ranging one order of magnitude lower than in the previous experiments. It must be noticed that because of the drastic decrease of the Rutherford cross section at large angles, these experiments are longer and more difficult as θ becomes larger. For example, in the case of 2-MeV protons scattered at 50° on Cu, 48 h of counting were necessary.

A. Experimental procedure

A well-collimated proton beam from the 4-MV Van de Graaff accelerator impinged on thin targets tilted at 45° relative to the beam directions. In order to make the effect of multiple scattering negligible, the target thicknesses were 100 $\mu g/cm^2$ for Cu and 20 $\mu g/cm^2$ for Ti and Al. To avoid target contamination with respect to long running time, liquid-nitrogen traps were mounted along the pipe. The x rays were detected by a 25-mm^2-active-area Si(Li) diode with a 12.5-μm Be window. In order to eliminate the L x rays (0.9 keV) in Cu experiments, we placed, in front of the detector, a Be foil 125 μm thick. The scattered protons were recorded in a 50-mm^2 surface-barrier detector. The total energy resolution, including target thickness and beam spread as measured with backscattered 2-MeV protons, was 16 keV.

Figure 1 gives the schematic drawing of the electronic system. Pulses from the x-ray and proton detectors were analyzed in a fast-slow coincidence system. The fast-coincidence resolving time was 15 nsec in the Cu case and 100 nsec with the Al target. One single-channel analyzer was used to select K_α-K_β x rays. In addition, two windows were selected in the time spectrum corresponding, respectively, to true and random events. A slow coincidence between these signals was used to trigger the ADC (analog-to-digital converter) so that true- and random-proton spectra were simultaneously recorded. Random events were then subtracted to give the actual number of true events. The coincidence x-ray spectrum was also obtained as well as direct x-ray and proton spectra.

B. Experimental results

At each angle, the ionization probability is obtained by the following relation:

$$I(p) = (N_c/N_p)(4\pi/\omega_K \epsilon), \quad (5)$$

where N_c is the number of true coincidence events, N_p is the number of protons in the direct spectrum during the same time, ω_K is the fluorescence yield of the neutral atom,[12] and ϵ is the efficiency of the x-ray detector including solid-angle and Be-window transmission. For very low x-ray energies any dead layer in front of the active area of the detector can induce a drastic change in the efficiency curve. Using thick Al, Ti, and Cu targets, we measured the x-ray yield per incident proton. The efficiency ϵ was deduced by comparing our thick-target yields at the right energy with the previous measurements of Basbas *et al.*,[5] Ogier

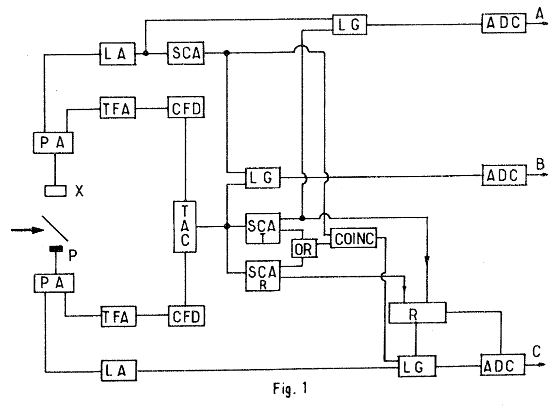

FIG. 1. Schematic drawing of the electronics setup. PA, preamplifier; TFA, timing-filter amplifier; CFD, constant-fraction discriminator; LG, linear gate; COINC, slow coincidence unit; R, router; LA, linear amplifier; ADC, analog-to-digital converter; SCA, single-channel analyzers for true (T) and random (R) events; A, B, and C outputs correspond, respectively, to the x-ray spectrum in coincidence with true time events, the time spectrum triggered by the K_α-K_β window signals, and the proton spectrum in coincidence with true and random events in two different parts of the memory.

et al.,[13] and Messelt,[14] in the Al, Ti, and Cu cases. From the good agreement between experimental and theoretical total cross sections found by these authors we estimate the error in the determination of ϵ within 10%.

If the trajectory of the incident particle is assumed to be classical, the ionization probability may be written

$$I(p)=\left(\frac{d\sigma_K/d\Omega}{d\sigma_R/d\Omega}\right)_\theta , \qquad (6)$$

where $d\sigma_R/d\Omega$ and $d\sigma_K/d\Omega$ are, respectively, the differential Rutherford and the K-shell differential ionization cross sections.

In order to ensure a valid comparison between theoretical and experimental $I(p)$, we have checked in every case that the projectile scattering follows a Rutherford law. This assumption was found to be satisfied within 5%. Experimental results are presented in Table I.

The parameter ξ introduced in Table I, defined as

$$\xi=[(1/1836.1)(E_1/E_B)]^{1/2},$$

represents the more or less adiabatic character of the collision. The reasons for rather large uncertainties in some measurements are given in the table caption.

Figure 2 shows our results on Cu. The incident-particle velocity is constant so that the adiabaticity of the collision does not change. We have plotted the results obtained from the BEA and SCA theories. We reported for comparison the data of Laegsgaard *et al.*[7] obtained at 6° and 3°. Our value at 16° is in very good agreement with the point at 6°, and in fact the theories predict that the relative variation between 6° and 16° must be very small.

In order to make a more comprehensive comparison between experimental and theoretical data, we have plotted our results and the smallest impact-parameter points from other authors in the reduced Brandt form in Fig. 3. The large discrepancy between experimental and theoretical low-energy SCA results at small impact parameters is clearly shown. This difference cannot be fully ascribed to the inadequacy of low-energy SCA to describe nonadiabatic interactions, since full SCA and BEA calculations presented in Table I are not in agreement with the data either. In the aluminum case, the large discrepancy found at 2-MeV proton energy[8] is confirmed for a similar impact parameter, but in different adiabaticity conditions.

At small impact parameters and large deflection angles, several effects usually neglected can be expected to be important, i.e., relativistic motion of the electrons and retardation and deflection of the particle in the nuclear Coulomb field. Such effects could be taken into account in a full SCA theory, but would lead to very intricate calculations even in the adiabatic case.[3] In the BEA, these effects can be included more easily to get an estimate of their importance.

The relativistic behavior of the electrons at small distances from the nucleus can lead to an increase of $I(p)$ for small impact parameters as suggested

TABLE I. Experimental values of the ionization probability of the K shell by protons on Cu, Al, and Ti targets. The theoretical values calculated from "simple" BEA and SCA are also reported when they were known. Large uncertainties on the Al points come from the rather poor time resolution (about 100 nsec) due to the low energy of the Al K x rays.

Energy (MeV)	Angle (deg)	p 10^{-11} cm	$I(p)$	ξ	$I(p)$ BEA[a]	$I(p)$ SCA[b]
			Cu target			
2	16	0.74	$(1.6\pm0.2)\times10^{-3}$	0.35	8.2×10^{-4}	1.1×10^{-3}
	26	0.45	$(1.6\pm0.2)\times10^{-3}$		8.4×10^{-4}	
	36	0.32	$(2.2\pm0.2)\times10^{-3}$		8.4×10^{-4}	
	50	0.22	$(2.6\pm0.4)\times10^{-3}$		8.3×10^{-4}	
			Al target			
0.5	16	1.3	$(6\pm2)\times10^{-2}$	0.418	7.1×10^{-3}	1.1×10^{-2}
0.97	18	0.68	$(13\pm6)\times10^{-2}$	0.58	1.3×10^{-2}	2.0×10^{-2}
			Ti target			
1	18	1	$(2.8\pm0.5)\times10^{-3}$	0.33	1.4×10^{-3}	
1	25	0.7	$(2.3\pm0.5)\times10^{-3}$		1.4×10^{-3}	

[a] BEA, straight line, unrelativistic.

[b] SCA, straight line, unrelativistic (Ref. 15).

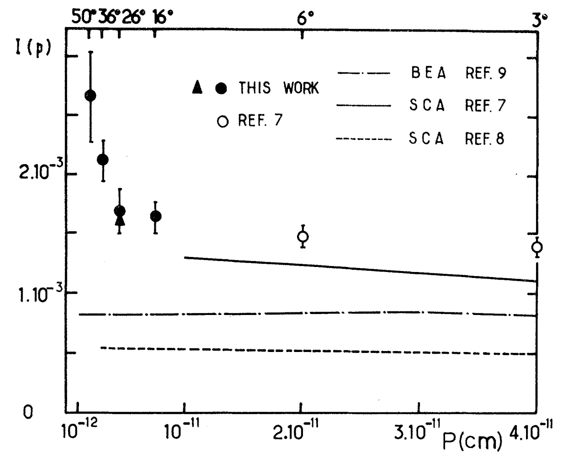

FIG. 2. K-shell ionization probability vs impact parameter in the case of 2-MeV H^+ on Cu. Dark point and triangle at 26° represent the results from two independent measurements, during different runs, using different targets.

by Hansteen.[16] An approximate correction has been introduced in the BEA by using the relativistic mass and velocity in the expression of $\sigma(v_1, v_2)$ [Eq. (4)]. For 2-MeV protons on Cu, a 12% $I(p)$ increase is found in the considered impact-parameter range compared with nonrelativistic BEA calculations. The results have been plotted in Fig. 4. When the same correction is applied to $I(p)$ for 1-MeV protons bombarding a silver target we find a factor of 2 difference between relativistic and

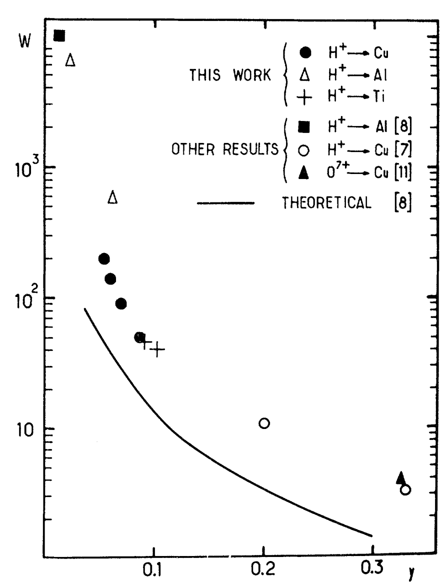

FIG. 3. Experimental data in the reduced form proposed by Brandt (Ref. 8): this work and results from other authors (Refs. 7, 8, and 11).

nonrelativistic calculations for zero impact parameters. This indicates that even for medium-mass targets it is necessary to take into account relativistic effects and that the simple scaling laws derived from hydrogen calculations fail for small impact parameters.

During close encounters with the nucleus, the projectile follows a hyperbolic path. Furthermore, the velocity of the projectile decreases in the Coulomb field of the nucleus. These two effects have been included in the calculations assuming a straight-line path tangential to the actual trajectory at the distance of closest approach. This method has been demonstrated by Hansteen[3] to give a rather good approximation. The velocity of the incident particle at each point of the trajectory defined above is determined from the conservation-of-energy law. These two effects lead to a rather small decrease of the $I(p)$ values as shown in Fig. 4.

Finally, the dot-dashed line in Fig. 4 shows the results obtained when the three above-mentioned corrections are all included. The difference with the "simple" BEA is small, and involved phenomena do not explain the large discrepancy between experimental and theoretical results in the case of protons on Cu, and all the more in the Al case. It must be observed that a discrepancy of a factor up to 10 at such values of p does not contribute in a significant way on the total ionization cross section; so the total cross-section measurements can be in good agreement with BEA predictions even if the details of the $I(p)$ curve are not well reproduced.

During close encounters one may consider the capture of a target K-shell electron by the projec-

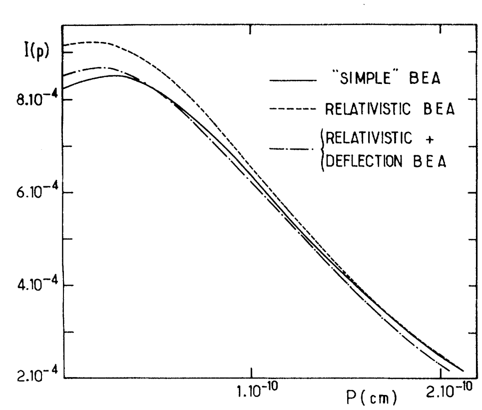

FIG. 4. Probability for K-shell ionization of Cu by 2-MeV protons calculated from the BEA model: the results from "simple" BEA (Ref. 9) are represented by the full curve; relativistic effects have been included in the dashed line as indicated in the text. Fully corrected results are shown by the dot-dashed line.

tile which has been shown to be important.[17] Addition of such a process may lead to an increase of $I(p)$. Theoretical calculations[18] confirmed by recent measurements[19] have shown that the contribution of electron capture to the total K-shell ionization cross section is less than 1% in our cases. The contribution of this phenomenon to $I(p)$ at small impact parameters is not expected to be very different from this value.

One might question whether the classical description of the projectile trajectory remains valid for these very small impact parameters; the condition for a classical treatment of the projectile motion in the considered process is[3]

$$\frac{Z_1 Z_2 e^2}{\hbar v_1} \frac{1+\sin(\theta/2)}{\sin(\theta/2)} \gg 1 .$$

For example, in the Cu case, for $\theta = 50°$, this ratio is about 10. Considering that the limits for the applicability of the above criteria are not well defined, it is difficult to assess the necessity of a fully quantal treatment in the present case.

Up to this point the ionization probability has been extracted from x-ray measurements assuming a fluorescence yield corresponding to the neutral atom.[12] However, recent experiments have shown that the fluorescence yield of the inner shell can present drastic changes with experimental conditions leading to possible erroneous interpretations of experiments. This change is usually ascribed to a multiple-ionization process. An estimate of the importance of simultaneous excitation of K- and L-shell electrons, at a given impact parameter, has been evaluated using the following formula derived by Merzbacher[20]:

$$I_{KL}(p)/I_K(p) = I_L(p)/[1 - I_L(p)] .$$

$I_L(p)$ is roughly evaluated, using the BEA model, to be 2%, so that the ratio from double to single ionization is about 2%. Such a value cannot give a large change in the mean fluorescence yield used.

Furthermore, the fluorescence yield used to extract $I(p)$ was calculated assuming a statistical population of the multiplet states arising from coupling open shells. Recent calculations have shown[21] that the fluorescence yield can depend strongly on the multiplet states. On the other hand, recent high-resolution Auger-electron measurements prove that the relative population of the multiplet states of neon is significantly different from the statistical value in the case of the excitation by proton or alpha particles.[22] The same phenomenon might be present in our cases and we cannot exclude that the relative population of the multiplet states should be dependent on the impact parameter.

Finally, it appears that the various theories in their present states are able to give roughly the right shape of the $I(p)$ curve, but do not give a good agreement with experimental values of $I(p)$ at very small impact parameters. More elaborate calculations in the SCA model involving relativistic electron wave functions and a hyperbolic path for the projectile would be very desirable in spite of the mathematical complexity. Furthermore, distortion of initial and final electronic wave functions induced by the projectile should also be included in the calculations. We have tried to improve in a simple way the agreement between experimental and theoretical results introducing in the BEA-model relativistic corrections for the electrons and an actual path; in spite of these modifications the discrepancy remains. Thus we show that the BEA model, which is useful for predicting total cross sections, appears too simple to provide a correct detailed description of the ionization process as investigated by differential measurements.

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[1] C. H. Rutledge and R. L. Watson, At. Data Nucl. Tables 12, 195 (1973).
[2] E. Merzbacher and H. W. Lewis, *Encyclopedia of Physics* (Springer, Berlin, 1958), Vol. 34, p. 166.
[3] J. Bang and J. M. Hansteen, Mat. Fis. Medd. Dan. Vid. Selk. 31, 13 (1959); J. M. Hansteen and O. P. Mosebekk, Nucl. Phys. A 201, 541 (1973).
[4] J. D. Garcia, Phys. Rev. A 1, 1402 (1970).
[5] G. Basbas, W. Brandt, and R. Laubert, Phys. Rev. A 7, 983 (1973).
[6] J. D. Garcia, Phys. Rev. A 1, 280 (1970).
[7] E. Laegsgaard, J. U. Andersen, L. C. Feldman, Phys. Rev. Lett. 29, 1206 (1972).
[8] W. Brandt, K. W. Jones, and H. V. Kramer, Phys. Rev. Lett. 30, 351 (1973).
[9] J. H. McGuire, Phys. Rev. A 9, 286 (1974).
[10] J. S. Hansen, Phys. Rev. A 8, 822 (1973).
[11] C. L. Coke and R. Randall, Phys. Rev. Lett. 30, 1016 (1973).
[12] W. Bambyneck *et al.*, Rev. Mod. Phys. 44, 716 (1972).
[13] W. T. Ogier, G. Y. Lucas, J. S. Murray, and T. E. Holzer, Phys. Rev. A 134, 1070 (1964).
[14] S. Messelt, Nucl. Phys. 5, 435 (1958).
[15] J. M. Hansteen (private communication).

[16]J. M. Hansteen, in *Arbeitstagung Uber Spezielle Probleme der Schwerionenstrevong-Atomphysik und Atomphysikalische Experimente und Schwerionenbeschleunigern*, edited by B. Fricke (Armbruster, Darmstad, 1973), p. 61.
[17]J. R. Macdonald, L. M. Winters, M. D. Brown, L. D. Ellsworth, T. Chio, and E. W. Pettus, Phys. Rev. Lett. 30, 251 (1973).
[18]A. Salin (private communication).
[19]J. R. Macdonald, C. L. Coke, and W. W. Eidson, Phys. Rev. Lett. 32, 648 (1974).
[20]E. Merzbacher, in Proceedings of Heavy-Ion Summer Study, Oak Ridge, Tenn., 1972 (unpublished).
[21]C. P. Bhalla, Phys. Lett. 46A, 185 (1973).
[22]C. P. Bhalla, D. L. Matthews, and C. F. Moore, Phys. Lett. 46A, 336 (1974).