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K-shell ionization of heavy atoms by 40–110-MeV α particles

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Using intrinsic Ge as x-ray detector, K-shell ionization cross sections have been determined for Ho, Tm, Lu, Ta, W, Pt, Au, Pb, and Bi under bombardment with 40–110-MeV α particles. The target-atom K-shell ionization cross sections are compared with the predictions of different models. The agreement is considerably improved when relativistic effects are taken into account. Intensity ratios have also been measured and found to agree with previous determinations. X-ray satellite emission, following nuclear rearrangements, was also observed and a calculation made in the case of Ho to relate nuclear reactions and K x-ray production cross sections. Small peaks in the spectra of Au, Pb, and Bi have been tentatively attributed to double K-shell ionization, and hypersatellite energy is given for these atoms.

I. INTRODUCTION

The interaction of a charged particle beam with atoms results in the creation of inner-shell vacancies because of atomic Coulomb excitation (primary ionization),[1] and ionization following nuclear reactions and recoil.[2] Primary ionization can be followed by the emission of specific x rays of the target and nuclear reactions can be followed by the emission of satellite x rays. Both processes will be successively discussed in this paper.

The ionization of inner shells by the interaction of fast protons and alpha particles is a process now well described by different models. In addition, much experimental data on K x rays from medium-weight elements ($10 < Z < 50$) are now available, though very little has been carried out on K-shell ionization of heavy elements ($Z > 50$) by charged particles, the reasons being that the cross sections are very small at low incident velocity and that there is a problem in the high-resolution detection of hard x rays. Fortunately, high-resolution and high-efficiency intrinsic Ge detectors are now available, making possible the nondispersive detection of hard x rays. K-shell ionization cross sections for high-energy charged particles can now be measured on a large-energy scale (20–150 MeV) using variable energy cyclotrons.

Garcia *et al.*[3] presented an extensive review of the theoretical and experimental work relevant to the creation of atomic inner-shell vacancies in collisions of ions and atoms, showing that semiclassical models and approximate quantum-mechanical treatments yield universal curves which can be compared with experimental data on a large-energy scale for light and medium atom weight elements ($10 < Z < 50$). Experimental data coalesce remarkably onto a single curve when expressed in terms of parameters included in the models.[3,4] The situation is not so satisfactory for heavy atoms where very little data are available[5] and where nonrelativistic calculations underestimate seriously the cross sections.

Relativistic effects have been considered by Hansen[6] who introduced a simple relativistic correction on the average electron velocity, and by Amundsen[7] who made a semiclassical calculation using relativistic hydrogenlike wave functions. In both cases the calculated cross sections were found to be higher than in nonrelativistic calculations.

It is felt that there is a serious need for a coherent set of experimental data on high-Z atoms, and on a large velocity range in order to improve the relativistic models. One of the purposes of this paper is to measure K-shell ionization cross sections for ^4He ions in the 40–110 MeV energy range on nine elements from Ho ($Z = 67$) to Bi ($Z = 83$) and to compare the results with the relativistic predictions as calculated in Ref. 7. Double K-shell ionization during the bombardment of light atoms with heavy ions was also reported, and the emission of $K\alpha$ hypersatellites was observed.[8] In the case of ionization of heavy atoms by α particles, the predicted cross section[9] for double K-shell ionization is a factor of 10^4 lower than the single K-shell ionization cross section. However, an attempt will be made to identify hypersatellite peaks in the x-ray spectra of heavy elements bombarded with high-energy α particles. If primary ionization is the dominant process in light- and medium-weight atoms, for heavy elements the cross section is to a lower order of magnitude.

For high velocity He^{++} beams a competition arises from the interaction of the projectiles with the nucleus itself. Different processes are responsible for the creation of inner-shell vacancies during and after nuclear reactions, i.e., nuclear recoil, nuclear reactions followed by γ decay, shake-off processes.

It is well known that nuclear reactions of the (α, pxn) and (α, xn) type are very intense for high-energy α particles. If Z is the atomic number of the target atom, after reaction the residual nucleus has atomic number $(Z+1)$ or $(Z+2)$. If ionization occurs, additional x-ray lines will be observed during the bombardment. These are x rays from the residual atom; they are known as nuclear satellites.[2,10-12] The principal mode of ionization following nuclear reactions induced by α particles was assumed to be the internal conversion of cascade γ rays emitted during the deexcitation of the residual nucleus,[2] the contribution of other ionization mechanisms being negligible. In the second part of this paper, x-ray satellites following nuclear reactions will be identified and a tabulation of the ionization cross sections $\sigma_I(Z+1)$ and $\sigma_I(Z+2)$ will be given. An attempt will also be made to relate the cross section for satellite emission with the nuclear cross sections and to define average parameters for this interaction.

II. EXPERIMENTAL METHOD

A beam of He^{++} ions was extracted from the variable-energy cyclotron of Louvain-la-Neuve and focused on a target mounted at an angle of 45° with respect to the beam axis. After passing through the target, the α-particle beam continued on into a shielded Faraday cup. At the target position the beam diameter was approximately 4 mm wide. X rays from the target passed through a 1-mm-thick aluminum window before reaching a 5-mm-thick intrinsic germanium detector. This detector had a sensitive area of 28 mm² and a total resolution of 475 eV for 100-keV x rays. K x-ray yields from natural and separated isotopes of Ho, Tm, Lu, Ta, W, Pt, Au, Pb, and Bi were measured. The target thickness and backing are summarized in Table I. Absolute measurements were carried out at α-particle energies of 40, 50, 60, 80, and 110 MeV. The beam was collected in a Faraday cup and the current was digitized and integrated in order to monitor the x-ray measurements.

The dead time of the electronic system was controlled and kept at less than 1% by using beam currents of low intensity. The total counting rate was maintained below 250 cps in order to reduce counting losses. The detector was positioned at

TABLE I. Targets used in K x-ray yield measurements.

Target	Thickness (mg/cm²)
$_{67}$Ho	5.07 (self-supporting)
$_{69}$Tm	5.02 (self-supporting)
$_{71}$Lu	5.05 (self-supporting)
$_{73}$Ta	3.88 (self-supporting)
$_{74}$W	9.98 (self-supporting)
$_{78}$Pt	2.14 (acrylic resin backing)
$_{79}$Au	9.32 (self-supporting)
$_{82}$Pb	6.06 (2.30-mg/cm² Al backing)
$_{83}$Bi	6.79 (2.30-mg/cm² Al backing)
$^{206}_{82}$Pb	0.10 (1.2-mg/cm² Al backing)
$^{207}_{82}$Pb	0.10 (1.2-mg/cm² Al backing)
$^{208}_{82}$Pb	0.10 (1.2-mg/cm² Al backing)

90° from the beam axis and at a distance of 20 cm from the beam spot on the target. The presence of heavy elements around the detector was avoided to prevent background from fluorescence on these elements; an iron shield was placed around both the Faraday cup and the detector.

The detection efficiency was determined experimentally by using a set of calibrated x-ray and γ-ray sources located at the beam spot position. The following sources were used: ^{241}Am (26.35 keV), ^{133}Ba (54 keV), ^{241}Am (59.6 keV), ^{203}Hg (72.1 keV), ^{133}Ba (79.5 and 81.0 keV), and ^{57}Co (122.0 and 136.3 keV). A smooth efficiency curve was drawn through the measurements. The accuracy on the absolute efficiency determination is estimated to be ±5% over the x-ray energy range of 45–95 keV. All the data were corrected for self-absorption and secondary fluorescence effects; in the less favorable cases both corrections amount to (2–3)%.

With the geometry used in our experiment the correction for self-absorption A in the target is given by

$$A = (1/\mu d)(1 - e^{-\mu d}), \qquad (1)$$

where μ is the mass absorption coefficient and d is the target thickness multiplied by a factor of $\sqrt{2}$.

This formula postulates that the production of x rays is uniform and isotropic in the sample. Since the projectiles progressively lose their energy in crossing the target, this assumption is only valid for thin samples bombarded with high-velocity projectiles. The energy loss is then very small and the cross section can be considered as constant.

III. DATA ANALYSIS

A typical spectrum of radiations detected during the bombardment of a target of heavy elements with fast α-particles is reproduced in Fig. 1. Besides large peaks from primary ionization of the target element with atomic number Z, other peaks are observed which originate from nuclear reaction effects.[2] In the case of Fig. 1, the target element was natural lead. Because of Pb(α, pxn) Bi and Pb(α, xn)Po nuclear reactions, Bi and Po K x rays are also observed. Unfortunately, the $K\alpha_2$ peak of Bi (74.805 keV) overlaps with the $K\alpha_1$ peak of Pb (74.957 keV); the same occurs for Po$K\alpha_2$ and Bi$K\alpha_1$, and for $K\beta$ lines. These overlappings necessitate a careful unfolding of the spectra. The $K\alpha_2$ radiation intensity from the target element Z is first determined; then by successive iterations the intensity of all the $K\alpha$ peaks is calculated from the spectrum after being corrected for detection efficiency, self-absorption, and fluorescence effects. In these iterations the $K\alpha_2/K\alpha_1$ ratio is not taken from the literature; only the variation of this ratio with atomic number is taken from Ref. 13. This variation is of the order of 0.5% between adjacent elements.

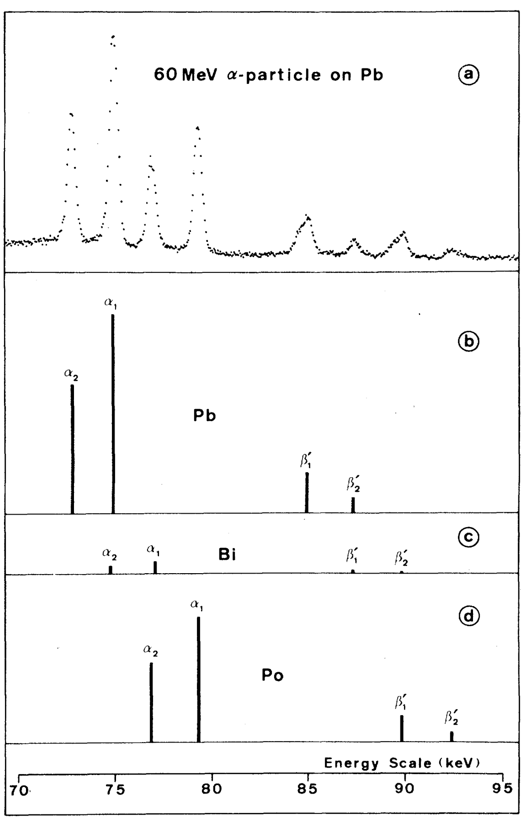

FIG. 1. K x-ray spectrum from 60-MeV α-particles on Pb. (a) Spectrum detected in a Ge detector during the bombardment; (b) amplitude of Pb x rays from primary ionization; (c) amplitude of Bi x rays following Pb(α, pxn) reactions; (d) amplitude of Po x rays following Pb (α, xn) reactions.

A similar procedure was used for the determination of the intensity of the $K\beta$ radiations. After complete processing of a spectrum we obtain the three following intensity ratios: $K\alpha_2/K\alpha_1$, $K\beta/K\alpha$, and $K\beta_1'/K\alpha_1$, where $K\alpha = K\alpha_1 + K\alpha_2$, and $K\beta = K\beta_1' + K\beta_2'$. The $K\alpha_2/K\alpha_1$ ratios are obtained for x rays from the target atom and for reaction products as well. The cross sections are then calculated by relating the total number of incident particles to the intensity of the peaks using the fluorescence parameter ω_k. The following cross sections were obtained: $\sigma_I(Z)$ for K-shell ionization of the target element, and $\sigma_I(Z+1)$ and $\sigma_I(Z+2)$ for the K-shell ionization of the reaction products. The cross section for K-shell ionization is given by

$$\sigma_I = \sum_i \frac{N_x^i}{nN\omega_K\epsilon_i(1-T)}, \tag{2}$$

where the summation is extended to all the radiations emitted by a given atom (target atom Z for primary ionization or product atoms $Z+1$ and $Z+2$ from nuclear reactions). N_x^i is the number of counts in a peak for a given radiation after self-absorption and fluorescence corrections. n is the number per cm^2 of target atoms seen by the α particle, taking into account target tilt. N is the number of projectiles. ϵ_i is the total detection efficiency for isotropic emission of i-type radiation from the target, thus including all absorption and solid-angle effects. T is the counting-loss factor due to the dead time of the spectrometer.

The fluorescence factor ω_k represents the probability that a vacancy in the K shell is filled through radiative transition. This factor varies slowly with atomic number Z and can be calculated from the following relation[14]:

$$[\omega_K/(1-\omega_K)]^{1/4} = 0.015 + 0.0327Z - 0.64\times10^{-6}Z^3. \tag{3}$$

Experiments performed with crystal spectrometers have revealed a K satellite structure due to single-K, multiple-L-shell ionization.[8] In nondispersive detection the resolution is not sufficient to observe the fine structure; the only possible observation is a small energy shift and an asymmetry of the observed peak. The cross section for multiple ionization by α particles was calculated by McGuire[9] who found that this effect is small for heavy elements. We tested this experimentally by irradiating holmium with 59.6-keV γ rays from a ^{241}Am source. Fluorescence K x rays were detected and the $K\alpha_1$ peak was compared with a similar peak produced by ionization with 60-MeV α particles. No significant difference was observed in the position and the width of the peak, indicating that the emission of satellites does not appreciably alter our spectra.

IV. IONIZATION BY ATOMIC COLLISION

A. Data

Samples of natural elements (Ho, Tm, Lu, Ta, W, Pt, Au, Pb, Bi) and of separated isotopes (^{206}Pb, ^{207}Pb, ^{208}Pb) were bombarded with ^{4}He^{++} beams of 40–110 MeV and K x-ray spectra were recorded during irradiation. After unfolding the spectra, the following radiation intensities were obtained: $K\alpha_2$, $K\alpha_1$, $K\beta_1'$, and $K\beta_2'$; and the following intensity ratios were determined: $K\alpha_2/K\alpha_1, K\beta/K\alpha$, $K\beta_1'/K\alpha_1$.

We shall call $\sigma_I(Z)$ the cross section for primary ionization of the target atom Z, i.e., the K-shell ionization due to the interaction of the α projectile with the electron cloud. This cross section was calculated from the data using formulas (2) and (3). The cross section $\sigma_I(Z)$ is given in Table II for all the elements and all the bombarding energies.

The ionization cross sections $\sigma_I(Z+1)$ and $\sigma_I(Z+2)$ from nuclear interactions are also given in this table, they will be defined in Sec. III. Also given in this table are the intensity ratios: $K\alpha_2/K\alpha_1, K\beta/K\alpha, K\beta_1'/K\alpha_1$ which are compared with the values given by Salem.[13] The ω_K values adopted in the calculations are indicated in the first column below the target element symbol; these values were calculated according to formula (3).

B. Comparison with models

In a first approach, the ionization cross-section values are compared with the predictions of simple theoretical models in which the ionization is supposed to originate from direct Coulomb interaction between the incident α particle and a K-bound electron described by a nonrelativistic wave function. The impulse approximation was used in the binary encounter model (BEA) which fits well with experimental data on light atoms.[3] When hydrogenic velocity distributions are used for the K electrons, the product of the squared binding energy and the cross section is a universal function of the incident energy expressed in units of the binding energy, i.e.,[3]

$$u_K^2\sigma_I/Z_1^2=f(E_1/\lambda u_K)\,, \tag{4}$$

where Z_1 and λ are the charge and mass (in electron mass units) of the projectile. E_1 is the projectile energy and u_K is the K-shell binding energy. Our data on heavy atoms are compared in Fig. 2(a) with the values of the universal function and, as expected, the agreement observed for light atoms is not reproduced here. A similar observation is made when data are compared with PWBA calculations [Fig. 2(b)]. Here again, the theory largely underestimates the cross sections. The universal curve was drawn according to the following relation:

$$\theta_K\sigma_K/\sigma_{0K}=f(\eta_K/\theta_K^2)\,, \tag{5}$$

where σ_K is the K-shell ionization cross section σ_I; all the other parameters have the same signification as in Ref. 4. In this representation the abscissa η_K/θ_K^2 is proportional to the incident energy.

The Coulomb excitation of the atom can also be calculated using the first-order time-dependent perturbation theory, the interaction being integrated along the classical path of the projectile. A straight line version of this semiclassical approximation (SCA) was introduced and tabulated by Hansteen *et al.*[15] The data were plotted for each atom in Fig. 3 and compared with the SCA calculations. Again this approximation underestimates the observed cross sections for the elements heavier than Ta.

These conclusions are not completely unexpected since nonrelativistic wave functions are used in the calculations, and this approximation is very poor for heavy atoms. Recently, simple relativistic corrections were introduced by Hansen[6] in the BEA model. We used these corrections, but found that they cannot account for the differences observed between our data and the BEA model.

More extensive calculations have been published by Amundsen[7] using relativistic Coulomb wave functions for the electron in the scheme of the SCA model. We compared our data with the curves calculated for Au, Ta, and Pb in Ref. 7, and with more recent calculations on other elements and at other energies.[16] These calculations were made for proton projectiles. We extended them to α particles of the same velocities using the σ_I/Z_1^2 rule and then multiplied the energy scale and the cross section by a factor of 4. The comparison between our data and the available calculations is reproduced in Fig. 3 where we observe that, except for Au and W, the relativistic calculations always overestimate the cross sections.

Amundsen *et al.*[17] proposed simple corrections to the SCA model to take into account the relativistic effects in heavy atoms. A correction was calculated using formulas (4)–(6) of Ref. 17, and the resulting curves were plotted in Fig. 3. As pointed out by Amundsen *et al.*, it is found that this calculation yields results which are very similar to the results of the relativistic SCA theory. We consider that the error on the absolute value of

TABLE II. Ionization cross sections for the target element and for nuclear reaction products and K x-ray intensity ratios for 40–110-MeV α particles on heavy atoms. The errors are estimated at 10% for $\sigma_I(Z)$ and (10–12)% for $\sigma_I(Z+2)$, the errors on $\sigma_I(Z+1)$ and on the intensity ratios are indicated in the table.

			Target elements						Nuclear reactions products	
Element	He^{++} energy (MeV)	$\sigma_I(Z)$ (b)	$K\alpha_2/K\alpha_1$ (This work)	$K\alpha_2/K\alpha_1$ (Salem [a])	$K\beta/K\alpha$ (This work)	$K\beta/K\alpha$ (Salem [a])	$K\beta_1'/K\alpha_1$ (This work)	$K\beta_1'/K\alpha_1$ (Salem [a])	$\sigma_I(Z+1)$ (b)	$\sigma_I(Z+2)$ (b)
$_{67}$Ho ($\omega_K=0.943$)	40	9.10	0.576 ±0.019	0.562	0.252 ±0.015	0.258	0.329 ±0.016	0.320	0.09 ±0.07	2.70
	50	16.4							0.31 ±0.19	3.38
	60	18.8							0.39 ±0.20	3.00
	80	31.2							0.35 ±0.21	3.22
	110	45.1							0.94 ±0.28	2.27
$_{69}$Tm ($\omega_K=0.947$)	40	6.53	0.570 ±0.018	0.565	0.274 ±0.014	0.262	0.335 ±0.017	0.323	0.16 ±0.13	2.10
	50	10.5							0.28 ±0.11	1.92
	60	15.1							1.13 ±0.23	2.59
	80	24.6							0.78 ±0.23	2.31
	110	37.3							1.07 ±0.21	1.87
$_{71}$Lu ($\omega_K=0.952$)	40	5.30	0.585 ±0.023	0.569	0.276 ±0.014	0.265	0.336 ±0.017	0.327	0.19 ±0.10	2.58
	50	9.15							0.34 ±0.10	2.81
	60	13.0							0.24 ±0.13	3.71
	80	21.0							0.91 ±0.18	3.32
	110	33.3							1.18 ±0.24	2.83
$_{73}$Ta ($\omega_K=0.956$)	40	4.05		0.574	0.252 ±0.020	0.268		0.330	0.15 ±0.07	2.29
	60	9.16							1.53 ±0.23	3.34
	80	15.8							0.68 ±0.10	2.92
	110	25.6							1.36 ±0.20	2.19
$_{74}$W ($\omega_K=0.957$)	40	3.99	0.568 ±0.022	0.576	0.284 ±0.014	0.269	0.334 ±0.017	0.332	0.08 ±0.05	1.85
	50	7.75							0.25 ±0.12	2.49
	60	9.51							0.85 ±0.13	2.21
	80	15.6							1.37 ±0.18	1.85
	110	26.2							2.13 ±0.28	1.52
$_{78}$Pt ($\omega_K=0.963$)	40	2.05		0.583	0.265 ±0.021	0.275		0.339	0.13 ±0.02	0.34
	60	5.66							0.48 ±0.07	0.70
	80	9.60							0.79 ±0.13	0.78
	110	16.1							1.32 ±0.20	0.77
$_{79}$Au ($\omega_K=0.964$)	40	2.12	0.575 ±0.020	0.585	0.280 ±0.017	0.276	0.342 ±0.021	0.341	0.02 ±0.01	1.75
	50	3.71							0.26 ±0.09	3.05
	60	5.44							0.14 ±0.07	3.32
	80	8.82							0.40 ±0.14	3.74
	110	14.9							0.61 ±0.12	2.74

TABLE II. (*Continued*)

Element	He^{++} energy (MeV)	$\sigma_I(Z)$ (b)	Target elements $K\alpha_2/K\alpha_1$ (This work)	$K\alpha_2/K\alpha_1$ (Salem [a])	$K\beta/K\alpha$ (This work)	$K\beta/K\alpha$ (Salem [a])	$K\beta_1'/K\alpha_1$ (This work)	$K\beta_1'/K\alpha_1$ (Salem [a])	Nuclear reactions products $\sigma_I(Z+1)$ (b)	$\sigma_I(Z+2)$ (b)
$_{82}$Pb ($\omega_K=0.968$)	40	1.47		0.593	0.269±0.022	0.280		0.346	0.05±0.04	1.21
	60	3.37							0.20±0.07	2.28
	80	5.65							0.44±0.09	2.59
	110	10.0							1.20±0.14	2.02
$_{83}$Bi ($\omega_K=0.969$)	40	1.36		0.595	0.272±0.022	0.281		0.348	0.06±0.02	0.68
	60	3.14							0.24±0.07	1.59
	80	5.03							0.53±0.19	1.94
	110	9.01							0.92±0.11	1.41

[a] From Ref. 13.

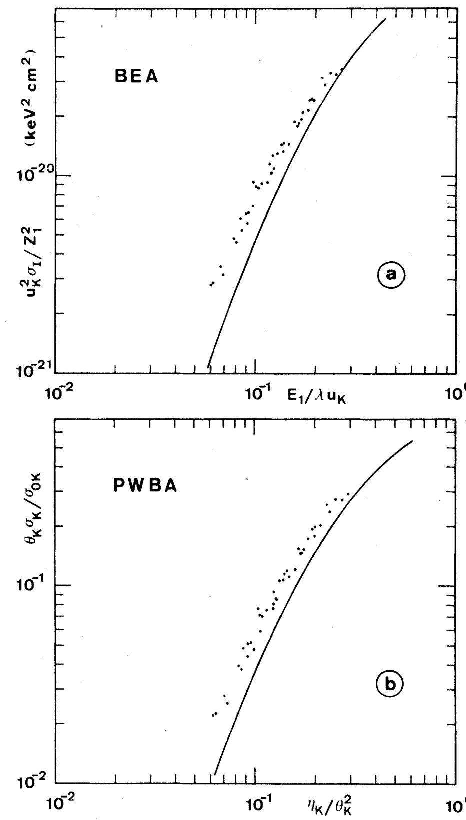

FIG. 2. Experimental data on K-shell ionization of Ho, Tm, Lu, Ta, W, Pt, Au, Pb, and Bi, by 40–110-MeV α particles. The solid lines represent universal curves calculated from nonrelativistic models. (a) binary encounter approximation (BEA); (b) plane-wave Born approximation (PWBA).

our data is about ±10%. Very few accurate data are available for comparison in the energy range considered here.[5] Hardt and Watson[18] published "smoothed" cross sections, obtained by fitting a curve through data at each energy in order to reduce the experimental fluctuations. These data are reproduced in Fig. 3; they are in good agreement with our measurements. Lark's data[19] on Ta, W, Pt, Au, Pb at 52 MeV are also reproduced and show a good agreement within the experimental errors. We then conclude that the relativistic calculations, which fit quite well on the proton data on Au,[7] also fit the α-particle cross sections on Au for the same projectile velocity. This indicates that the σ_I/Z_1^2 rule is valid; the difference in the Coulomb deflection effect between protons and α particles of same initial velocities is then negligible in this energy range.

For the other elements, only W gives a good fit, all the other cross sections being overestimated by this model. As stated by Amundsen,[7] further

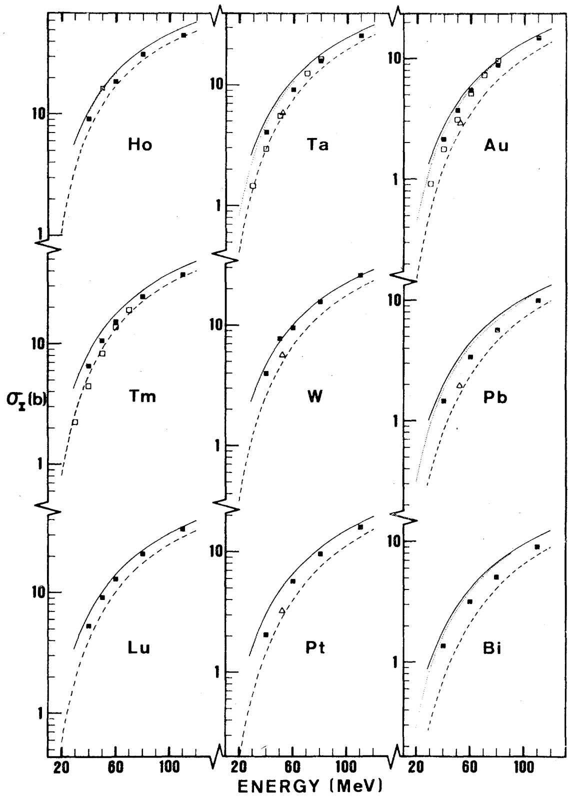

FIG. 3. Experimental data on primary K-shell ionization cross section [■, present work; □, Hardt and Watson (Ref. 18); △, Lark (Ref. 19)]. The dotted curve was drawn after the relativistic calculation of Amundsen (Refs. 7 and 16), the solid curve is the SCA curve corrected for relativistic effects (Ref. 17), the dashed curve is the result of a SCA nonrelativistic calculation (Ref. 15).

measurements were needed at high energy on heavy elements. Our results now cover a large energy range where relativistic models must be applied.

C. Intensity ratios

The intensity ratios $K\alpha_2/K\alpha_1, K\beta/K\alpha, K\beta_1'/K\alpha_1$ were determined from the data. No significant variation was observed with the energy. The data given in Table II are average values calculated from the values found at different energies. They agree with the values given by Salem[13] and obtained from other fluorescence data.

D. Double K-shell ionization

The double K-shell ionization is depicted by the observation of K hypersatellites (K^h); these are $K\alpha$ and $K\beta$ peaks which are shifted towards high energy. These shifts amount to 1280 ± 60 eV in Pb and 1160 ± 70 eV in Tl as observed in β-decay

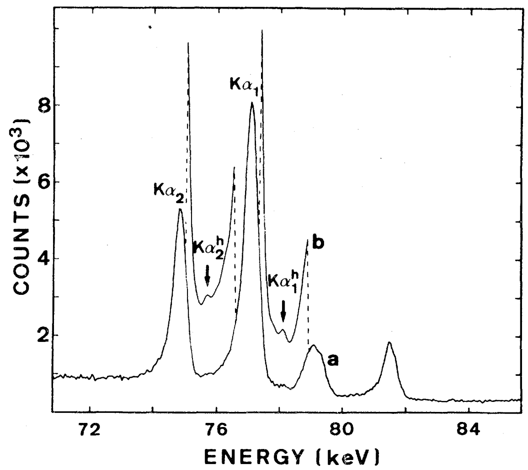

FIG. 4. $K\alpha$-x-ray spectrum detected during the bombardment of Bi with 110-MeV α particles. (a) A direct spectrum measured with a Ge detector. (b) A spectrum obtained by smoothing the data and magnified by a factor of 3. The arrows indicate the small peaks which are tentatively attributed to hypersatellite emission.

measurements.[20,21] The detection of K hypersatellites emitted during the bombardment of heavy atoms by charged particles is very difficult, the expected cross section σ_{2K} being very small,[9] a factor of 10^4 less than for the single ionization σ_{1K}. However, our spectra from Au, Pb, and Bi bombardment all show the presence of two additional peaks which can be tentatively assigned to the double K-shell ionization. Although relatively small, these peaks were observed systematically in repeated measurements, including the three lead isotopes. For lighter elements (Ho, Tm, Lu, Ta, W, Pt) the $K\alpha_1$ and $K\alpha_2$ peaks are so close that hypersatellite peaks could hardly be resolved. A typical spectrum is reproduced in Fig. 4. It corresponds to the bombardment of Bismuth with 110-MeV α particles; the direct and the smoothed curve are given. The observed shift is 980 eV for Au, 1050 eV for Pb, and 915 eV for Bi with an error estimated to about 120 eV. These values are slightly smaller than the above given data for Pb and Tl and than the result of a calculation made by Desclaux which gives 1290.5 eV for Bismuth.[22] If this assessment is correct, the cross section for double K-shell ionization σ_{2K} is about $10^{-2}\ \sigma_{1K}$. This measured value is in disagreement with McGuire's predictions[9] which are a factor of 100 lower.

V. IONIZATION FOLLOWING NUCLEAR REACTIONS

The ionization of product atoms from nuclear reactions is responsible for the emission of characteristic x rays or nuclear satellites. This phenomenon can easily be observed during the bombardment of heavy atoms. For light atoms the primary K x-ray peaks are very high and so close to the nuclear satellite peaks that the latter can hardly be extracted from the background. The origin of the ionization has been discussed previously.[2] It was essentially attributed to the internal conversion in the decay of highly excited nuclear products creating K-shell vacancies. Indeed, (α, pxn) and (α, xn) reactions have high cross sections below 110 MeV, and these reactions leave nuclear products of atomic number $Z+1$ and $Z+2$. Above this energy the competition of fission becomes important and the (α, pxn) and (α, xn) cross sections decrease. Above 50 MeV, the total cross section for these reactions is practically the geometrical cross section of the nucleus, i.e., 2–3 b for heavy nuclei. The residual nucleus is in a highly excited state which decays by neutron and γ-ray cascades, γ rays being predominantly produced in E2 and M1 transitions. These are relatively low-energy γ rays and the conversion coefficients are important for such transitions. In principle it should be possible to calculate the intensity of nuclear satellites if all the parameters of the nuclear decay were known: γ-ray energy, transition multipolarity, and nuclear reactions responsible for γ-ray emission $(\alpha, \alpha n)$, (α, pxn), (α, xn). The general formulas relating the ionization cross sections to the nuclear cross sections are

$$\sigma_I(Z+1) = \sum_x \sigma(\alpha, pxn)\left(\sum_i I_i \alpha_K^i\right), \tag{6}$$

$$\sigma_I(Z+2) = \sum_x \sigma(\alpha, xn)\left(\sum_i I_i \alpha_K^i\right), \tag{7}$$

where σ_I is the ionization cross section of the residual atom after nuclear reaction, $\sigma(\alpha, pxn)$ and $\sigma(\alpha, xn)$ are the nuclear reaction cross sections, I_i is the probability that a given γ ray is emitted, and α_K^i is the K-shell conversion coefficient corresponding to this γ-ray energy and multipolarity. The second summation is extended to all γ rays (i index) following a given value of x, i.e., a given (α, pxn) or (α, xn) reaction. In these formulas it is supposed that when cascade γ rays are emitted, each γ ray will find two electrons in the K shell. This in turn postulates that the lifetime of a K-shell vacancy is much shorter than nuclear level lifetimes. Typically, the lifetime of a K hole in a heavy atom is 10^{-17} sec,[23] and the nuclear lifetime ranges between 10^{-9} and 10^{-13} sec in rotational bands of deformed nuclei; this condition is then always fulfilled.

The application of formulas (6) and (7) to practical cases is rendered difficult because of the lack of experimental data on γ decay during α-induced reactions. We found that holmium was a good case since it is a single isotope (^{165}Ho) and since very extensive information can be found on this nucleus for $(\alpha, 4n)$ reaction at 50 MeV incident energy.[24]

If satellites from (α, pxn) and (α, xn) reactions are easily separated in the x-ray spectrum because they correspond to atomic numbers $Z+1$ and $Z+2$, respectively, the energy spectrum from the γ-ray radiation emitted during the bombardment is much more complex. About 140 peaks are seen between 60 and 620 keV in the γ-ray spectrum from bombardment of holmium with 50-MeV α particles. Most of these can be attributed to nuclear decay following (α, pxn) and (α, xn) reactions[24] but other reactions can also contribute in small proportions: (α, α'), $(\alpha, 2pxn)$, ... In what follows we shall indicate how to calculate the ionization cross section $\sigma_I(Z+2)$ for ^{165}Ho. The ionization following $(\alpha, 4n)$ reactions, for which complete data on γ decay are available, is first calculated, then average parameters are extracted allowing the extrapolation of the calculations to other (α, xn) reactions, namely $(\alpha, 2n)$, $(\alpha, 3n)$, and $(\alpha, 5n)$. This will be done in two different ways: either by defining an average conversion coefficient $\overline{\alpha}_K$, or by calculating the average number $\overline{n}_K$ of K-shell ionization per nuclear reaction. The $(\alpha, 4n)$ reaction initiated on holmium ($Z=67$) leaves thulium ($Z+2=69$) as a residual atom following the equation

$$^{165}_{67}\text{Ho} + \alpha \rightarrow {}^{165}_{69}\text{Tm} + 4n\,.$$

The partial K-shell ionization cross section due to $(\alpha, 4n)$ reactions is given by

$$\sigma_I^4(Z+2) = \sigma(\alpha, 4n)\sum_i I_i \alpha_K^i \tag{8}$$

or

$$\sigma_I^4(Z+2) = \sum_i \sigma_{\gamma_i} \alpha_K^i, \tag{9}$$

where σ_{γ_i} is the cross section in barns for the production of a given γ ray. Sixty-four γ-ray energies from the Ho $(\alpha, 4n)$ reaction are reported in Ref. 24 with their relative intensities and with the spin sequence of the corresponding nuclear levels. The most probable multipolarity was calculated for each transition, using the selection rules for γ-ray emission and the transition probabilities given in Ref. 25. The conversion coefficient α_K^i corresponding to each γ-ray energy and multipolarity was also obtained from Ref. 25. The data of Ref. 24 had to be normalized since they are given in relative units. For this purpose, a γ-ray spectrum was recorded during the bombardment of a holmium target of 5.07 mg/cm^2 and for a total collected charge of 2 μC. The absolute value σ_{γ_i} was calculated from this spectrum. Using all the data on cross sections and conversion coefficients, by application of (9), we obtain

$$\sigma_I^4(Z+2)=1.98\text{ b}. \quad (10)$$

Let us now define an average conversion coefficient $\overline{\alpha}_K$ in the following way:

$$\overline{\alpha}_K=\sigma_I^4(Z+2)\Big/\Big(\sum_i \sigma_{\gamma_i}\Big)=1.98\Big/\Big(\sum_i \sigma_{\gamma_i}\Big). \quad (11)$$

We obtain the value

$$\overline{\alpha}_K=0.403,$$

which is the average conversion coefficient for the γ rays following Ho(α, $4n$) reactions.

We now postulate that the average conversion coefficient $\overline{\alpha}_K$ is the same for the other (α, xn) reactions, a statement which is true only if the residual nuclei have similar structures. With this definition of $\overline{\alpha}_K$ we can write by extension

$$\sigma_I^x(Z+2)=\overline{\alpha}_K\sum_i \sigma_{\gamma_i}, \quad (12)$$

where x can be 2–5.

Since

$$\sigma_I(Z+2)=\sum_x \sigma_I^x(Z+2), \quad (13)$$

we obtain

$$\sigma_I(Z+2)=\overline{\alpha}_K\sum_i \sigma_{\gamma_i}, \quad (14)$$

where the summation extends to γ rays from all (α, xn) reactions. A calculation made on the basis of formula (14) gives the following result:

$$\sigma_I(Z+2)=3.01\text{ b},$$

a figure which has to be considered as an order of magnitude with regard to the hypothesis made in this calculation (average coefficient $\overline{\alpha}_K$, multipolarity assignments). However, it compares nicely with the observed magnitude which is 3.38 b for 50-MeV α particles (Table II).

The second method consists in deriving the average number $\overline{n}_K$ of K-shell ionization in a given (α, xn) reaction:

$$\overline{n}_K=\sigma_I^x(Z+2)/\sigma(\alpha, xn), \quad (15)$$

which again will be supposed to be independent of x. The calculation was made for the (α, $4n$) reaction using the value 1.98 b obtained in formula (10) for $\sigma_I^4(Z+2)$ and the cross section (0.65 b) given in Ref. 26 for $\sigma(\alpha, 4n)$ on holmium, hence the result

$$\overline{n}_K=1.98/0.65=3.05.$$

Let us now extrapolate this result to all (α, xn) reactions; the nuclear cross section $\sigma(\alpha, xn)$ can be obtained from the data of Ref. 26 and Ref. 27. The total ionization cross section is given by

$$\sigma_I(Z+2)=\overline{n}_K\sum_x \sigma(\alpha, xn), \quad (16)$$

with

$$\sum_{x=2}^{5}\sigma(\alpha, xn)=1.21\text{ b}.$$

Then

$$\sigma_I(Z+2)=3.69\text{ b},$$

which again is in accordance with the observed value of 3.38 b. In these calculations we used parameters having a statistical meaning, i.e., $\overline{\alpha}_K$ the average conversion coefficient and $\overline{n}_K$ the average number of ionizations per nuclear reaction. We believe that $\overline{\alpha}_K$ and $\overline{n}_K$ are strongly dependent on the nuclear structure of the residual nuclei involved. In principle, the calculation made for holmium could be extended to all other target element in order to correlate the parameter $\overline{\alpha}_K$ with nuclear structure; however more data are needed on (α, xn) reactions.

The average number of ionizations $\overline{n}_K$ is easily accessible to experiment and can be deduced immediately from the x-ray spectrum using (16):

$$\overline{n}_K=\sigma_I(Z+2)\Big(\sum_x \sigma(\alpha, xn)\Big)^{-1}. \quad (17)$$

This was carried out on Au, Pb, and Bi for which nuclear cross sections $\sigma(\alpha, xn)$ are available[26-28]; the ionization cross section $\sigma_I(Z+2)$ was calculated from the satellite yield using (2). The results are reported in Fig. 5. A smooth variation is observed with the incident energy and the results are similar for neighboring atoms (Pb and Bi) but this trend must be confirmed by other data before drawing further conclusions.

A similar treatment was adopted by Röhl *et al.*[11] for the interaction of α particles with ^{208}Pb which is a double-magic nucleus. The ionization cross section can be explained quantitatively by a calculation based on internal conversion.

VI. CONCLUSION

Two kinds of information have been obtained in this paper. One is the primary ionization of heavy atoms by atomic collision, the other is the ionization following nuclear reactions.

A set of data on K-shell ionization cross sec-

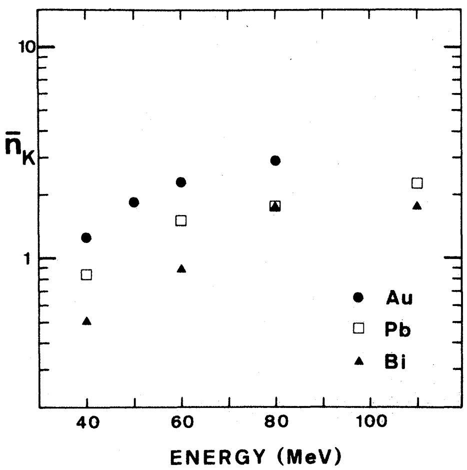

FIG. 5. Average number $\bar{n}_K$ of K-shell vacancies produced during the prompt nuclear decay of the atom following (α, xn) reactions on three heavy elements Au, Pb, and Bi and at different α-particle energies.

tions for 40–110-MeV α particles and the intensity ratios for the different radiations emitted after ionization have been tabulated for nine heavy atoms. When compared with universal curves from semiclassical models, the experimental results on all the elements and at all the energies gather on a smooth curve indicating a good coherence of the data. These results, however, do not agree with the predictions of any of the three models proposed (BEA, PWBA, and SCA) which considerably underestimate the data. The relativistic semiclassical model of Amundsen was then used. In the case of W and Au the agreement is excellent; for the other elements these calculations represent a real improvement on the classical models, but they overestimate the data by about (10–15)%. It is hoped that the measurements presented here will help to improve the relativistic calculations.

In the case of ionization following nuclear reactions, a quantitative justification of the hypothesis of the internal conversion mechanism was given for Ho, and an attempt was made to introduce average parameters $\bar{\alpha}_K$ and $\bar{n}_K$. More data on γ decay following (α, xn) reactions are needed to confirm the calculations made on holmium, and to calculate the average conversion coefficient $\bar{\alpha}_K$ for the other elements. The average ionization number $\bar{n}_K$ is more easily accessible; it can be extracted from the x-ray spectra and from total (α, xn) cross sections. We suggest that a correlation be established between these parameters and other nuclear spectroscopic parameters such as ellipsoidal deformation, moment of inertia, etc.

Unexpected x-ray peaks were observed and tentatively assigned to double K-shell ionization. The peaks were observed for Au, Pb, and Bi, and the corresponding hypersatellite energies were measured. However, the values found for these energies are slightly smaller than the data from β-decay measurements, and the cross sections are two orders of magnitude larger than predicted by semiclassical calculations. If this hypothesis is not correct, the question arises on the nature of those peaks.

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