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EXCITATION OF KX-RAYS BY BOMBARDMENT OF THICK SOLID TARGETS WITH 150 MEV Xe IONS

1. Introduction

The investigation of characteristic X-rays, excited by the interaction of heavy ions with matter, shows that even the K-shells of the colliding partners become ionized with high probability. The order of magnitude of the cross section values σ_K as well as its dependence on the ion energy, on the atomic numbers of the projectile (Z_1) and of the target (Z_2) raise the question about the mechanism of the K-shell ionization. For ions with the smallest atomic numbers Z_1, such as protons and a-particles, the experimentally determined values of the total cross section σ_K are in good agreement with that for direct Coulomb ionization[1-6]. For ion-target combinations, for which the relation $Z_1 \ll Z_2$ is not valid, the measured cross sections σ_K exceed by several orders of magnitude the values predicted for Coulomb ionization[2]. In these cases the dependence of the cross sections σ_K and σ_L on the experimental parameters is described in a qualitative way by the molecular orbital (MO) model[2,7-12]. Unfortunately, no quantitative calculations of the cross sections, which are to be expected from the MO model, are so far carried out, so that direct experimental tests are currently impossible.

Some recent papers indicate that multiple collisions of ions in a target material lead to an asymmetric charge distribution, which contains also very high charge states[13,14]. Such high charge states may mainly result from outer shell ionization, but K-shell vacancies are not to be excluded. The concepts advanced so far on the formation of charge distributions by the interaction of ions in solids, give no statements on the mechanism of an individual collision. The K-shell vacancies, necessary for the production of KX-rays may, therefore, originate from electron promotions as well as from any kind of direct process.

The investigations of K excitation cross sections, which are described in this paper, may contribute to clarify the mechanism of inner shell ionization in the case of high energies and heavy colliding ions.

2. Experimental Method

At the U-300 heavy ion cyclotron investigations of the emission of KX-radiation arising from the bombardment of different solid targets by $^{136}Xe^{9+}$ ions were carried out. The ion energy was 150 MeV. Currents of about 2×10^{10} ions per second were used. The principal experimental arrangement was described earlier /15/. By means of a somewhat better focusing system, a focus of 6 mm in diameter was achieved at the target location. The targets consisted of thick metallic foils, covered by a frame of pure aluminium with an opening of 15×15 mm^2. The parameters of the targets used are listed in table 1. Figure 1 shows the target-detector arrangement. The targets were exposed at an angle of 45° with respect to the beam direction, but the X-rays were measured perpendicularly to the beam direction. The grid of 0.1 mm tungsten wires, placed in front of the target, served to normalize the measured spectra to the equal quantities of incident ions.

For the detection of the KX-rays, a planar Ge(Li) detector of about 11 mm thickness and 2.8 keV energy resolution at 60 keV energy was used *. The measure-

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* The results of measurements with a Si(Li) detector of 440 eV energy resolution at 26 keV full energy are in preparation.

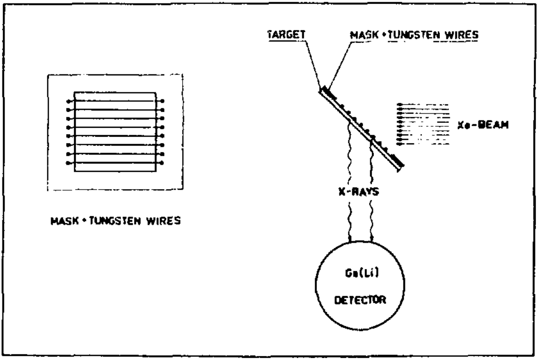

Fig. 1. Arrangement of target and detector.

ments were performed with a 1024-channel analyser. Figure 2 shows the energy spectrum obtained by bombardment of a *Bi* target.

No *X* -rays of energies below 10 keV could be detected due to an electrically inactive layer inside the detector. Therefore the *LX* -radiation of the target materials could not systematically been investigated. As a result of the Coulomb excitation of the lowest excited states of the tungsten nucleides ^{182}W, ^{184}W and ^{186}W, strong γ -ray transitions at 100.1 keV, 111.2 keV and 122.6 keV were obtained together with the characteristic *KX* -radiation of the collision partners (fig. 2). By means of the intensity of the Coulomb-excited γ -ray transitions, the normalization of the *X* -ray spectra to equal charges was carried out.

In order to obtain the absolute cross section values, the charge was measured without target foil. The number of Coulomb-excited γ -rays of the tungsten grid was compared with the charge of the ions getting through the grid to the Faraday cup. In this way any falsification

Table 1

The types of targets used. The symbols designate the following: Z_1 and Z_2, the atomic numbers of projectile and target, respectively; d', the effective target thickness; R_0, the range of 150 MeV Xe ions /23/.

Target	Z_2	Z_1/Z_2	ρ g/ cm^3	d' mg/cm^2	R_0 mg/ cm^2
Rh	45	1.20	12.4	19.4	8.4
Ag	47	1.15	10.5	11.7	8.6
La	57	0.95	6.15	20.0	9.7
Gd	64	0.84	7.95	8.2	11.9
Lu	71	0.76	9.74	6.2	13.1
Au	79	0.68	19.3	20.9	14.2
Bi	83	0.65	9.7	139.6	14.8

of the charge measurement due to changes in the charge of the Xe ions inside the target was avoided.

The following corrections are to be made in the target arrangement in fig. 1.

1. Self-absorption of the X-rays within the range of the Xe ions in the target as well as absorption in penetrating the target dead layers *.
2. Absorption of the tungsten grid γ-radiation by the target. The whole target thickness is effective for this purpose.
3. Excitation of Xenon KX-radiation by the tungsten grid. Its portion was determined by a measurement without target, which was also normalized to the intensity of the Coulomb-excited γ-ray transitions of the tungsten grid.
4. In the case of the Gd and Lu targets, Coulomb-excited γ-ray transitions were obtained, the inner conversion of which contributes to the target KX-ray intensity. Its portion was calculated from the intensity of the respective γ-ray lines and the known conversion coefficients.

The rate of KX-radiation, measured with our target-detector arrangement, is given by the following relation:

$$N_m = \int n\sigma[E(r)] e^{-\mu(d'-r)} dr, \qquad (1)$$

where $E(r)$ is an ion energy dependent on the penetration depth along a stretched path inside the target; $\sigma[E(r)]$ is the cross section as a function of the ion energy. The value

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* In ref. /16/ it was supposed that the K_α - radiation of Cl ions was emitted only in a thin target layer of about 100 $\mu g/cm^2$. The account of this effect would considerably alter our absorption correction, and enlarge the cross section values $\sigma_{K_\alpha}(Xe)$ by more than one order of magnitude. Since this effect has not been explained, we assume that the target radiation as well as the Xe-radiation was homogeneously excited over the entire range of Xe ions.

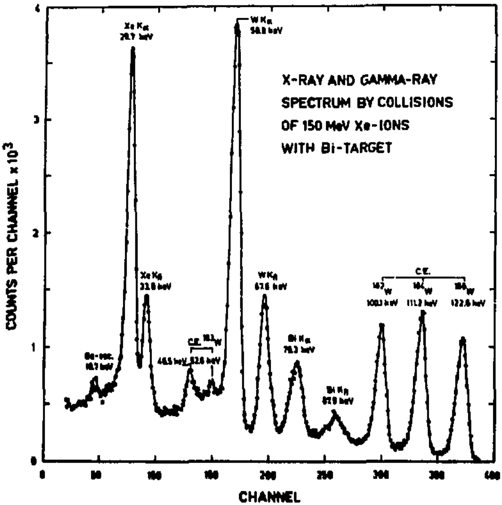

Fig. 2. The spectrum obtained by Xe ion bombardment of a bismuth target together with a tungsten grid.

of $\bar{\sigma} = \sigma(E)$ averaged over the range R_0 of the Xe ions and dependent only on incident energy E , is obtained from eq. (1) by making use of some correction factors

$$\bar{\sigma} = \sigma(E) = \frac{\mu N_{mc}}{n\epsilon_{tot} MD e^{-\mu(d'-R_0)}(1-e^{-\mu R_0})}, \quad \text{for} \quad R_0 < d'$$

or

$$\bar{\sigma} = \sigma(E) = \frac{\mu N_{mc}}{n\epsilon_{tot} MD (1-e^{-\mu d'})}, \quad \text{for } R_0 > d', \qquad (2)$$

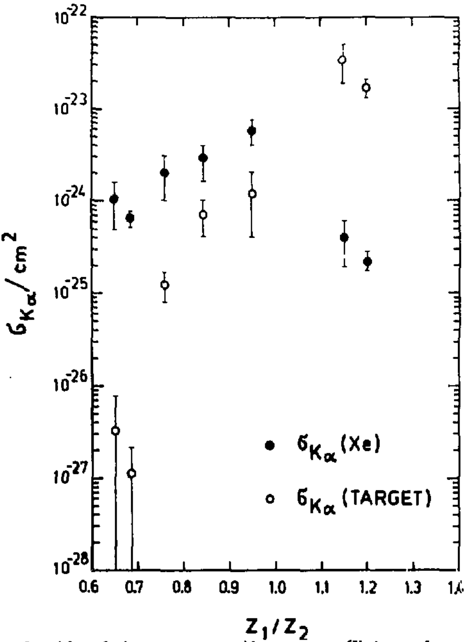

Fig. 3. Absolute cross sections σ_{K_α} *(Xe)* and σ_{K_α} (target) as a function of the ratio of the atomic numbers of projectile (Z_1) and target (Z_2). The abscissa allows to compare the obtained results with those of ref. /2/ which, in spite of the preferential use of *Ne* ions, shows in principle the similar dependence of σ_K on Z_1/Z_2.

where N_{mc} is the measured and corrected number of K_α X-ray quanta. In calculating the cross section σ_{K_α} *(Xe)*, the *Xe* K_α -radiation excited by the tungsten grid was subtracted. In calculating the cross sections σ_{K_α} (target) for *Gd* and *Lu*, the inner conversion of the

Coulomb-excited γ-ray transitions was taken into account;

μ is the target absorption coefficient for the K_α-radiation;

d' is the effective thickness of the target material;

n is the number of target atoms per cm^3 ;

ϵ_{tot} is the total efficiency of the Ge(Li) detector;

D is the number of Coulomb-excited γ-rays of the tungsten grid, acting as a normalization factor;

M is a calibration factor obtained from the absolute charge measurement for Xe^{9+} ions.

3. Experimental Results

In table 2, the measured cross section values σ_{K_α} for Xe and the investigated targets as well as the ratios σ_{K_α} (target)/ $\sigma_{K_\alpha}(Xe)$ are summarized. Figure 3 shows the dependence of these cross sections on the ratio of the atomic numbers Z_1/Z_2 for a constant incident energy of 150 MeV. In the calculations of the errors $\Delta\sigma_{K_\alpha}$, given in table 2 and fig. 3, only the errors in the peak areas $N_{K\alpha}$ and D, the target thicknesses d', the absorption coefficients μ and the ranges R_0 have been taken into account. These values are uncertain from target to target and affect the relative positions of several points in fig. 3. The neglected errors of the detector efficiency ϵ_{tot} and the calibration factor M influence to an equal extent all the points of fig. 3, i.e., only the absolute cross section values. The account of the systematical errors $\Delta\epsilon_{tot}$ and ΔM increases the relative errors $\Delta\sigma_{K_\alpha}/\sigma_{K_\alpha}$ by 25%.

The dependence of the cross section σ_{K_α} on Z_1/Z_2 is characterized by the following:

1. the cross section σ_{K_α} (target) decreases with growing atomic number Z_2, whereas $\sigma_{K_\alpha}(Xe)$ shows a resonancelike behaviour with a maximum at $Z_2 = Z_1$.
2. the cross section ratio σ_{K_α}(target)/ $\sigma_{K_\alpha}(Xe)$ shows that the K-shell of the colliding partner having the smaller atomic number is preferably ionized.

4. Discussion

The experimental facts, described in section 3, may be explained qualitatively by means of the molecular orbital (MO) model [9,11]. This model handles the limiting case of an adiabatic collision, in which the relative velocity v of the colliding partners is small compared to the velocity u of the atomic shell electrons. The approach of both atoms causes a strong perturbation in the Coulomb potential and consequently in the electron motion. Under the condition $v \ll u$, the electron configuration is able to follow the perturbation. For final distances R between the colliding particles a "quasimolecule" with common electron shells is formed; but in the limiting case $R \to 0$ we can speak of a "quasiatom * ". The energy eigenvalues of the wave functions of the quasimolecule may be given as a function of the distance R. In the frame of the MO model this dependence is discussed on the basis of the hydrogen molecule theory [17,18]. For $R \to 0$ and $R \to \infty$, the atomic levels are characterized by the main quantum numbers n and the quantum numbers l of the orbital momentum. For small distances R the levels split into states, which are described by the additional quantum number λ representing the projection of the orbital momentum l on the axis of the molecule $(0 \leq \lambda \leq \ell)$. The notations σ , π , δ , ϕ are used for λ = 0,1,2,3, respectively.

The spin-orbital coupling of the electrons has been neglected. By drawing the principal behaviour of the energy states $E(n\ell\lambda)$ as a function of R, a correlation scheme is obtained, which provides information on the occupation of the quasimolecular states by the electrons of both atoms. In fig. 4 correlation schemes are drawn for the systems $Rh+Xe$ and $Gd+Xe$. On the basis of the MO model,

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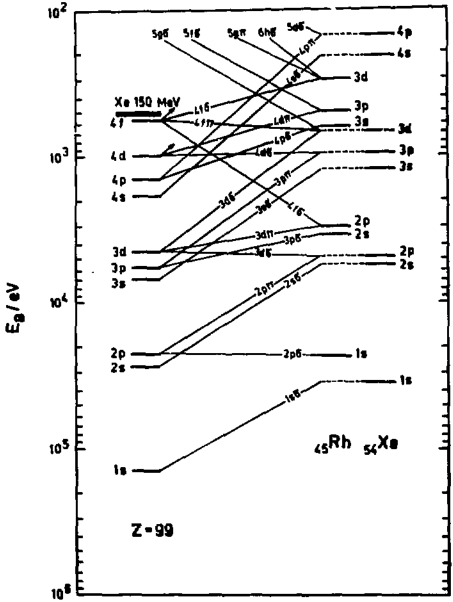
* Nuclear reactions may be neglected, because the ion energy used is far below the Coulomb barrier.

the origin of characteristic X-rays may be understood in the following way:

a) the level splitting leads to changes in the main quantum numbers of electrons (promotion) and sometimes to drastic changes of the electron binding energies during the formation of a quasimolecule or a quasiatom;

b) at the crossing points of molecular levels a certain probability exists for transitions of electrons from one state to another. Such a transition probability exists mainly for the asymptotic crossings $R \to 0$ and $R \to \infty$, where the molecular levels join the atomic ones;



c) Vacancies are produced by electron transitions from an occupied to an unoccupied level. If the lifetime of a vacancy exceeds the collision time, it can be transferred to the other collision partner and give rise to its characteristic X -radiation.

In principle, the MO model is applicable to our experiment, because the condition $v \ll u$ is fulfilled for every target and the principal quantum numbers n = 1,2 and 3. For the M -shell of $_{45}Rh$ the condition $v/u \sim$ 0.3 is

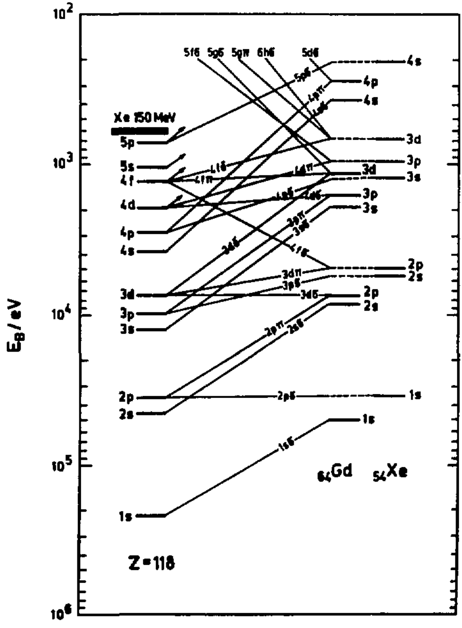


Fig. 4. MO correlation schemes for the asymmetric collision systems $Rh + Xe(a)$ and $Gd + Xe(b)$.

valid. This ratio decreases further with increasing Z of the target and decreasing quantum number n. The time required for penetration of the K- and L- shells of the collision partners is of the order of 10^{-18}s, whereas for solid targets the time between two collisions amounts to about 10^{-17} s. Vacancies in the M-, L- and K-shells of Xe have lifetimes of 10^{-14}, 10^{-15} and 10^{-16} s respectively[19]. They are, therefore, able to survive several collisions and to give rise to the emission of characteristic X-rays after the particles have been separated.

The microscopic processes, which lead to the formation of inner shell vacancies, may be described in the following way. Energetic ions entering a target undergo a further loss of shell electrons. On the other hand, they capture electrons from the target atoms. Already after the ions have penetrated a very thin target layer, a new equilibrium charge distribution is formed [14], which allows to define a mean charge state $\bar{q}$ of the ions. It may be expected that, after penetration of thick layers, the mean charge $\bar{q}$ decreases again due to the energy loss of the ions. For solid targets, $\bar{q}$ shows only a slight dependence on the target materials [14,20]. After penetration of gold foils by Xe^{8+} ions with an energy $E_{\ell ab}$ = 130 MeV a mean value $\bar{q}$ = 26 has been found [21]. This value is satisfactorily reproduced by a formula given in ref. [20], which for 130MeV Xe ions gives $\bar{q}$ = 26.9 and for 150 MeV Xe ions gives $\bar{q}$ = 28. It may, therefore, be assumed, that after some collisions with the outermost target layer a large number of Xe ions shows vacancies in the 3d-shell. As is shown in fig4a for a target with $Z_2 < 54$, according to the MO model, vacancies may get across the $3d\delta$ -term to the $3d$ -term of the quasiatom with Z = 99 and therefrom into the $2p$-shell of the Xe ion. Filling these vacancies by outer electrons causes LX -rays of Xe. If, however, during the lifetime of the $2p$ vacancy a further collision takes place, the vacancy may be transferred into the $1s$ -shell of the Rh atom. Multiple collisions lead to the excitation of KX -radiation of Rh. If the atomic number Z_2 nearly agrees with that of Xe, i.e., both target and projectile have almost equal K binding energies, also the $1s$ -electrons of Xe may be promoted

Table 2

Cross sections σ_K for the excitation of $K_\alpha X$ -rays by bombardment with 150 MeV Xe ions

Target	$\sigma_{K_\alpha}(\text{target}) \pm \Delta\sigma_{K_\alpha}$ / cm^2	$\sigma_{K_\alpha}(Xe) \pm \Delta\sigma_{K_\alpha}$ / cm^2	$\sigma_{K_\alpha}(\text{target})$ / $\sigma_{K_\alpha}(Xe)$
Rh	$(1.7 \pm 0.4) \times 10^{-23}$	$(2.2 \pm 0.5) \times 10^{-25}$	77.3 ± 35.7
Ag	$(3.4 \pm 1.6) \times 10^{-23}$	$(4.0 \pm 2.1) \times 10^{-25}$	85.0 ± 84.6
La	$(1.2 \pm 0.8) \times 10^{-24}$	$(5.8 \pm 1.8) \times 10^{-24}$	$(2.1 \pm 2.1) \times 10^{-1}$
Gd	$(7.2 \pm 2.8) \times 10^{-25}$	$(2.8 \pm 1.2) \times 10^{-24}$	$(2.6 \pm 2.1) \times 10^{-1}$
Lu	$(1.2 \pm 0.4) \times 10^{-25}$	$(2.0 \pm 1.0) \times 10^{-24}$	$(6.0 \pm 5.0) \times 10^{-2}$
Au	$(1.1 \pm 1.0) \times 10^{-27}$	$(6.4 \pm 1.1) \times 10^{-25}$	$(1.7 \pm 1.8) \times 10^{-3}$
Bi	$(3.2 \pm 4.5) \times 10^{-27}$	$(1.0 \pm 0.5) \times 10^{-24}$	$(3.2 \pm 6.1) \times 10^{-3}$

into the higher states with high probability, resulting in a big cross section $\sigma_K(Xe)$ (fig. 3). This mechanism explains qualitatively that for $Z_2<54$ preferably the lighter collision partner becomes K ionized.

In fig. 4b the correlation scheme is drawn for the system $Gd+Xe$, which is in principle also valid for all other targets with $Z_2>54$. In spite of the increasing electron binding energies with increasing Z_2, the alternating shell sequence $K_T-K_{Xe}-L_T-L_{Xe}-M_T-M_{Xe}$ is preserved up to the system $Bi+Xe$. According to the already described mechanism, vacancies of the $3d$-shell of Xe ions may be transferred during further collisions into the $2p$-shell of Xe and therefrom into the $2p$-shell of Gd. A strong L ionization of both partners is to be expected.

In order to explain the origin of KX-radiation of Xe, the following processes might be discussed:

a) collisions of Xe ions with L ionized Gd atoms, and
b) collisions of L ionized Xe ions with Xe atoms, implanted into the target lattice.

Similar arguments are used in paper /22/ to explain the KX-radiation of Ne ions, originating from bombardment of an Ar gas target. For both types of collisions, only a very low rate determined by the ratio of the particle densities is to be expected. Neglecting the back diffusion of the ions, in the collisions of Xe with Xe an accumulation effect should be observed. At the end of a 30 min. irradiation, the ion current leads to one $Xe+Xe$ collision, compared with 10^5 $Gd+Xe$ collisions. Experimentally no alteration of the intensity ratio of the target and Xenon radiation has been found with an accuracy of a few percent for an irradiation period of 1.5 hours.

The origin of the KX-radiation of Gd could be explained as being due to recoils of L ionized Gd target atoms. During the formation of quasimolecules with other target atoms, K-shell vacancies may be produced in the symmetric colliding system. In this way the formation of target KX-rays may in principle be understood also for targets with $Z_2>54$.

For the minimum distance between the colliding particles the relation

$$R_{min} = \frac{\epsilon^2 Z_1 Z_2}{E_{lab}} \cdot \frac{M_1 + M_2}{M_2} \qquad (3)$$

is valid. Provided that the energy E_{lab} of the Xe ions is constant, R_{min} increases with increasing Z_2. In addition, with increasing $Z = Z_1 + Z_2$ the crossings of the molecule terms are shifted /11/ in the direction of $R \rightarrow 0$. Both effects lead to a decrease in the following ionization cross sections:

a) σ_L (Xe) for L ionization of the Xe ions in collisions with target atoms.

b) σ_K (target) for K ionization of recoiled target atoms in collisions with target atoms.

This leads to a decrease in σ_K (target) with decreasing Z_1/Z_2 (fig. 3).

Summary

1. For the bombardment of several targets with 150 MeV Xe ions the absolute cross sections $\sigma_K(Xe)$ and σ_K (target) have been measured.

2. The discussion of the experimental results shows that for $Z_2 < 54$ the MO model is able to explain the cross section ratios σ_{K_α} (target)/ $\sigma_{K_\alpha}(Xe)$ in a qualitative way. Moreover, from this model it may be expected that the cross section σ_{K_α} of the projectile shows a maximum in the case of a symmetric collision system $Z_1 = Z_2$. For Xe ions the height and position of this maximum may be interpolated from the measured cross section curve $\sigma_{K_\alpha}(Xe)$.

3. For $Z_2 > 54$, the KX -radiation of Xe and the target materials are no longer understood on the basis of multiple collisions of Xe ions with target atoms. An explanation of the Xe radiation with the help of the collisions of Xe ions with implanted Xe atoms is improbable. For a possible explanation of the target radiation, use was made of target recoil collisions. The Z_2 dependence of $\sigma_K(Xe)$ and σ_K (target) for $Z_2 > 54$ gives a hint at the formation of quasimolecules in this case too.

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References

1. E.Merzbacher and H.W.Lewis. Handbuch der Physik, ed. by E.Flügge, Springer-Verlag Berlin, 1958, Vol.34, p. 166.
2. W.Brandt and R.Laubert. Phys. Rev. Lett., 24, 1037 (1970).
3. J.M.Hansteen and O.P.Mosebekk. Z.Phys., 234, 281 (1970).
4. R.L.Watson, C.W.Lewis and J.B.Natowitz. Nucl. Phys., A 154, 561 (1970).
5. J.M.Hansteen and O.P.Mosebekk. Contr. to the Proceedings of the International Conf. on Inner Shell Ionization Phenomena, Atlanta 1972.
6. O.N.Jarvis and C.Whitehead. Phys. Rev., A5, 1198 (1972).
7. W.Lichten. Phys. Rev., 164, 131 (1965).
8. H.J.Specht. Z.Phys., 185 301 (1965).
9. U.Fano and W.Lichten. Phys. Rev. Lett., 14, 627 (1965).
10. J.Macek. Phys. Rev. Lett., 28, 1298 (1972).
11. M.Barat and W.Lichten. Phys. Rev., A 6, 211 (1972).
12. H.O.Lutz, J.Stein, S.Datz and C.D.Moak. Phys. Rev. Lett., 28, 8, (1972).
13. G.A.Ryding, A.B.Wittkower and P.H.Rose. Phys. Rev., A3, 1658 (1971).
14. H.D.Betz. Rev. Mod. Phys., 44, 465 (1972).
15. U.Hagemann, W.Neubert, W.Schulze and F.Stary. Nucl. Instr. Meth., 90, 415 (1971).
16. H.W.Schnopper, A.R.Sohval, H.D.Betz, J.P.Delvaille, K.Kalata, K.W.Jones and H.E.Wegner. Contr. to the Proceedings of the International Conf. on Inner Shell Ionization Phenomena, Altanta 1972.
17. P.H.Morse and E.C.G.Stückelberg. Phys. Rev., 33, 932 (1929).
18. H.Bethe. Handbuch der Physik, ed. by H.Geiger, Berlin 1933, Vol. XXIV, part I, p. 530.
19. H.R.Rosner and C.P.Bhalla. Z.Phys., 231, 347 (1970).
20. V.S.Nikolajew and I.S.Dmitriev. Phys. Lett., 28A, 277 (1968).
21. И.А.Шелаев и др. ОИЯИ, Р9-6062, Дубна, 1971.

22. H.Tawara and J.Kistemaker. Phys. Lett., 41A, 287 (1972).
23. L.C.Northcliff and R.F.Schilling. Nucl. Data Tables, A 7, 233 (1970).

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