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IMPACT PARAMETER DEPENDENCE OF K-SHELL VACANCY PRODUCTION IN COLLISIONS OF 1 MeV/a.m.u. Cu IONS WITH Cu, Ge AND Ag ATOMS

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1. INTRODUCTION

Recent results of the differential cross section measurements of the K-shell ionization[1-7] in the medium-Z and -energy region show large discrepancies between the experiment and the rotational coupling model[11-14], which has been widely used so far to explain the data for the low-Z and -energy collisions. A similar disagreement was observed recently even for collisions of Cl and Ar ions of relatively low energy with both gaseous and solid targets[8-10]. An alternative statistical model of inner-vacancy production[16,17] which treats the electron promotion through many non-isolated densely spaced level-crossings reproduces quite well the gross structure of the experimental data, especially for very asymmetric collision systems. The model, however, cannot exactly describe the shape of the K-shell vacancy production probability as a function of the impact parameter. Both the rotational coupling model and the statistical model explain the electron vacancy production in the K-shell of the lighter partner exclusively. The $1s\sigma$ molecular orbital (MO) correlating in the limit of separated atoms (SA) to the 1s atomic orbital of the heavier partner, is well separated over the whole range of internuclear distances from all other orbitals except the $2p\sigma$ MO - the 1s orbital of the lighter parner in the SA limit. Therefore, the K-shell vacancies in the heavier collision partner come from the sharing process between the $2p\sigma$ and $1s\sigma$ MO's in the outgoing part of the trajectory. The Demkov-Meyerhof model of this process[18,19] was extended by Briggs[20] to give the impact parameter dependence of the vacancy sharing ratio. The experimental results for the Cu-Ge collision system are compared in our work with the predictions of that model.

2. EXPERIMENTAL SET-UP

A beam of 63 MeV $^{63}Cu^{4+}$ ions was obtained from the U-300 cyclotron of JINR Laboratory of Nuclear Reactions in Dubna. It was collimated to a spot of diameter less than 1.5 mm on the target and to a maximum beam divergence of 0.4°. Self-supporting targets of 80 $\mu g/cm^2$ natural Cu and Ag and 80 $\mu g/cm^2$ Ge on

20 μg /cm^2 carbon backing were placed at 45° to the beam axis. An intrinsic germanium detector placed in a close geometry at 90° to the beam measured the emitted X-rays. The detector efficiency was calibrated with standard radioactive sources placed at the position of the target. The accuracy of the calibration amounts to ~20%. The height of the germanium K-absorption edge was estimated to be no more than 15% from a comparision of the intensity ratios in spectra measured with germanium and silicon detectors /27/. The scattered ions passing through an annular diaphragm were detected with a parallel-plate avalanche detector in the 1.9°-19.4° (lab.) scattering angle range. The accuracy of the definition of the angle varied from 3% to 7% depending on the angle value. Single spectra of X-rays and scattered ions recorded simultaneously with coincident events permitted a determination of both total and differential cross sections. A detailed description of the experimental procedure is given in our previous work /1/.

3. DATA ANALYSIS

The K-X-ray emission probability can be obtained for each of the partners of the collision according to the formula:

$$P_X\,(b) = \frac{N_X^c(\theta)}{\epsilon \cdot N_I(\theta)}\,, \qquad (1)$$

where $N_I(\theta)$ is the number of ions scattered into the angle θ in the lab. system, $N_X^c(\theta)$ is the number of coincident K-X-rays, ϵ is the X-ray detector efficiency in the geometry of the experiment and b is the impact parameter corresponding to the scattering angle θ.

The transformation of scattering angles into parameters was done using the Rutherford formula. For the measured range of scattering angles the uncertainties introduced by neglecting the screening are several times smaller than those from geometrical factors. The K-shell vacancy production probability is given by:

$$P_K(b) = P_X(b)\,/\omega_K\,, \qquad (2)$$

where ω_K is the K-X-ray fluorescence yield. We used the single-vacancy ("neutral atom") values of ω_K from the work of Langenberg and van Eck /21/. According to calculations of Bhalla /22/ and Larkins /23/ the fluorescence yield ω_K for differently ionized atoms differs significantly from a "neutral atom" value only for an almost completely stripped L-shell.

The total cross section, $\sigma_K = 2\pi \int_0^\infty P_K(b) \cdot b\,db$ was evaluated from the formula:

$$\sigma_K = \frac{N_X^s}{\epsilon \cdot N_I(\theta) \cdot \omega_K} \cdot \int \frac{d\sigma_R}{d\Omega}\, d\Omega, \qquad (3)$$

where N_X^s is the number of K–X-rays registered in the single spectrum, and the integration is made over the angle of acceptance of the particle detector and over the target area exposed to the beam (the latter having only minor effect on final result).

The targets used in the experiment were thick enough to ensure an equilibrium charge state of the projectile /2/. However, secondary small angle collisions in a target of finite thickness change the values of the ionization cross section σ_K and the probability $P_K(b)$ and (mainly through a kinematical shift in b) – the shape of the $P_K(b)$ distribution itself. As concerns the latter, Monte-Carlo calculations simulating a trajectory of a particle traversing the target revealed a dispersion and a small shift into higher values of the final angles of a scattered particle with respect to the single collision case. For example, for the Cu–Ge collision system the calculated shift was 0.18° with a dispersion of 0.05° for the scattering angle of 1.9° and 0.14° with a dispersion of 0.07° for the angle 5.6° (lab.). These values overestimate the effect because of the use of a non-screened Coulomb potential in the calculations. The increase of the $P_K(b)$ values due to secondary small angle collisions is roughly estimated to be of the order of $\Delta x \cdot \sigma_K$, where Δx is the target thickness in atoms per cm^2 (see Tserruya et al. /10/). The sum of $P_K(b)$ values of the lighter and heavier collision partners corrected in the above-shown way is presented in Fig. 1. The vertical bars in the Figure represent only statistical uncertainties. The correction in $P_K(b)$ integrated over the range of strong coupling gives 11%, 8% and 3% of the measured values of the total cross section σ_K for Cu-Cu, Cu–Ge and Cu-Ag collision systems, respectively.

4. RESULTS AND DISCUSSION

4.1. K -Shell Vacancy Production

The experimental values of $P_K(b)$ were compared with the predictions of the $2p\pi$–$2p\sigma$ rotational coupling model /11-14/. The theoretical curve was calculated using a computer code of Jäger /24/, written for homonuclear collisions, and a scaling according to Taulbjerg et al. /13/ for Cu–Ge and Cu–Ag

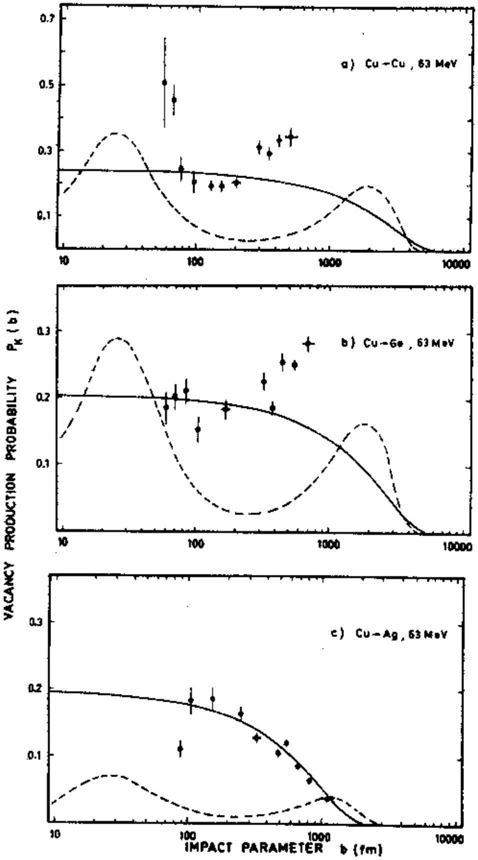

Fig. 1. Experimental results and theoretical predictions for the impact parameter dependence of the K-shell vacancy production in: a) Cu–Cu , b) Cu–Ge and c) Cu–Ag collisions. The broken lines refer to the rotational coupling and the solid lines to the statistical model predictions. The vertical error bars represent statistical uncertainties only. The values of the model parameters are shown in the Table.

systems. The theoretical values should be multiplied by a factor ν, the number of vacancies in the $2p\pi_x$ orbital created prior to the collision or in the early stage of the same collision. It is to be obtained from the comparison of the experimental and theoretical cross sections:

$$\sigma_K^{exp} = \nu \cdot \sigma_K^{th} . \qquad (4)$$

The factors ν are similar for Cu–Cu and Cu–Ge collisions, but considerably smaller for the Cu–Ag collision (see the Table). The broken lines in Fig. 1 represent the theoretical predictions for $P_K(b)$. As is seen in the Figure, for Cu–Cu and Cu–Ge collisions the theory explains qualitatively the two-humped shape of the $P_K(b)$ distribution but fails to predict the magnitude of the effect. The experimental data suggest also that the increase of the $P_K(b)$ values for large impact parameters starts at smaller values than the "adiabatic peak" at $b \simeq 2r_K$ (united atom) in the rotational coupling picture. Concerning the Cu–Ag collision system a large shift of the experimental $P_K(b)$ distribution towards smaller values of impact parameter is observed as compared with the rotational coupling model. Due to this fact the rotational coupling fails completely to predict the magnitude of the effect.

Table

Experimental total cross sections and the parameters of the models (explanation in the text)

collision system	Cu - Cu	Cu -Ge	Cu - Ag
total cross section, σ_K, 10^3 barns	53.3±10.7	36.9±7.4	4.9±1.0
number of $2p\pi_x$ vacancies, ν	0.35	0.29	0.07
Thomas-Fermi screening length R_0, 10^{-9} cm	1.08	1.06	0.99
fitted radius of strong interaction R_0, 10^{-9} cm	1.15	1.06	0.40
expected values [a] of the diffusion constant D_K, cm^2/sec	27.0	29.9	46.5
fitted values of the diffusion constant D_K, cm^2/sec	21.3	22.4	8.6

a) according to the semiempirical formula
$D_K \simeq [\frac{1}{12}(Z_1+Z_2)]^2 \cdot \frac{\hbar}{m_e}$
from Ref. 4

In another approach the experimental results were compared with the ionization probability calculated from the statistical model /16,17/. In the model the probability of the K - shell ionization is described by the equation:

$$P_K(b) \simeq 1 - \frac{2}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{n+1/2}\cdot \exp[-(n+1/2)^2\cdot\pi^2\cdot s_K(b)], \qquad (5)$$

where

$$s_K(b) = \frac{2R_0\cdot v}{D_K}\{[1-(b/R_0)^2]^{1/2} - (b/R_0)\cdot \arccos(b/R_0)\} = w_K\cdot F(b/R_0) \quad (6)$$

with a notation $w_K = 2R_0 v/D_K$, where v is the relative velocity of the colliding ions, R_0 is an effective interaction range, usually taken as the Thomas-Fermi screening length in the combined atom, $R_0 = 0.885 a_0 (Z_1^{2/3} + Z_2^{2/3})^{-1/2}$, D_K is a factor describing the diffusion of electrons through the crossings of the energy levels. The function $F(b/R_0)$ is defined for $b \leq R_0$ and is equal to zero outside. The total cross section is equal to:

$$\sigma_K = 2\pi\cdot\int_0^\infty P_K(b)\cdot b\,db = S(w_K)\cdot\pi\cdot R_0^2, \quad (7)$$

where after the coordinate transformation: $\cos\Theta = b/R_0$

$$S(w_K) \simeq 1 - \frac{4}{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{n+1/2}\int_0^{\pi/2}\sin\Theta\cos\Theta\exp[-w_K\pi^2(n+1/2)^2 \times \times(\sin\Theta - \Theta\cos\Theta)]d\Theta. \quad (8)$$

The statistical model gives the $P_K(b)$ and σ_K values calculated per one electron in the K-shell of the combined atom and therefore they must be multiplied by a factor of two before comparing them with the experimental ones. In our work the relevant parameters of the model: an interaction radius R_0 and a diffusion constant D_K were fitted in such a way that both the experimental total cross section σ_K and the value of $P_K(b)$ for b =200 fm were reproduced. The obtained values of R_0 and D_K are compared with the expected ones in the Table. The statistical model predictions are presented by solid lines in Fig. 1. It can be seen from the Figure and the Table that for Cu-Cu and Cu-Ge collision systems the magnitude of the $P_K(b)$ distribution can be well described by the model with reasonable values of the parameters. However, an increase of the $P_K(b)$ values for small and large impact parameters is not expected within the framework of the model. A good agreement between the experiment and the model for very asymmetric Cu-Ag system is noteworthy though it is achieved for the parameters different from those expected from the semiempirical formula of ref. /4/. In a quest for physical processes other than secondary small angle scattering which could affect the experimental σ_K and P_K (b) values, we calculated recoil effects and electron capture to the K-shell of the projectile from the target atoms. The correction for the recoil effects /14,15/ was about 0.3 barns for the Cu-Ge and 0.04 barns for the Cu-Ag

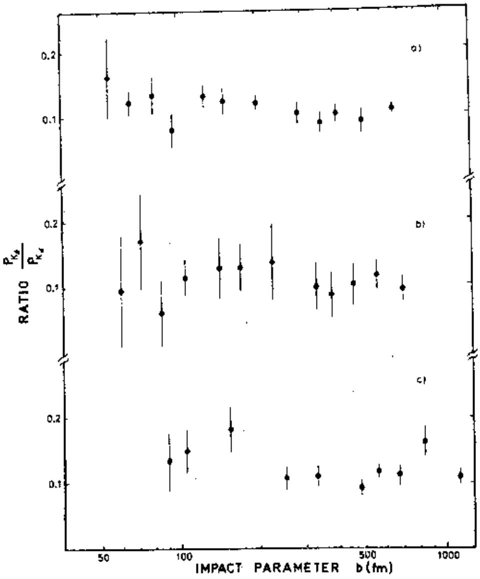

Fig. 2. The impact parameter dependence of the ratio K_β/K_α - X-rays in the collision: a) Cu-Cu, Cu K-X-rays; b) Cu-Ge, Ge K-X-rays; c) Cu-Ag, Cu K-X-rays. The error bars represent only statistical uncertainties.

collision system. The upper limit correction for electron capture /25/ gives about 7 barns and 50 barns for these systems, respectively. The contribution from both processes is evidently negligible. No attempt has been made to estimate the contribution to the K-shell vacancy production from direct Coulomb coupling of the $2p\sigma$ molecular state to the continuum or unoccupied bound states. The direct Coulomb excitation of the $2p\pi$ molecular state, if it occurs at large distances during the collision, increases the number of $2p\pi_x$ vacancies, ν, a parameter to be obtained from the experimental σ_K value. In order to check if the L-vacancies are created in the measured range of impact parameters not only at large distances, we investigated the ratio of K_β/K_α X-rays. This ratio depends on the relative abundance of the L- and M-shells. No impact parameter dependence of the ratio has been found (see Fig. 2), which suggests that most of the L-vacancies are created at large distances. The result is merely a suggestion, because of large statistical uncertainties and a weak dependence of the radiative transition widths on the number of L- and M-vacancies /22,23/.

4.2. Vacancy Sharing

In the outgoing part of the collision trajectory the $2p\sigma$ vacancies are shared between the $2p\sigma$ and $1s\sigma$- MO's by the radial coupling. According to the Demkov-Meyerhof model of this process /18,19/ the vacancy sharing ratio is:

$$r = P_K^H(b=0)/P_K^L(b=0) = \exp(-2x), \qquad (9)$$

with

$$2x = 2\pi(I_H^{1/2} - I_L^{1/2})/(2m_e v_1^2)^{1/2}\,, \qquad (10)$$

where I is the neutral atom binding energy of the K-shell, v_1 is the velocity of the projectile and m_e is the rest mass of the electron. The letters H and L refer to the heavier and lighter collision partner, respectively. In an extension of this model Briggs/20/ gives an impact parameter dependence of the vacancy sharing ratio $r(b) = P_K^H(b)/P_K^L(b)$ for "one- and two-passage" cases. These terms signify the situation when the vacancies are born in the collision and shared in the outgoing part of the ion trajectory in contradiction to the situation when they are brought into the collision and shared later on. The model calculations of Briggs for the O-Ne collision system/20/ give essentially the same result as the more accurate one-electron two-state MO calculations/26/, but the latter show a bit more complex behaviour of the vacancy sharing ratio versus the impact parameter.
In the present work we investigated the vacancy sharing ratio for the Cu-Ge and Cu-Ag collision system. The values obtained from the total cross section measurements, 0.263 ± 0.0002 and $(2.6 \pm 0.6) \times 10^{-4}$ respectively, are in good agreement with the values of 0.241 and 1.64×10^{-4} obtained according to Meyerhof's formula. The indicated errors are only statistical uncertainties. The vacancy sharing ratio versus the impact parameter for the Cu-Ge collision system is shown in Fig 3 together

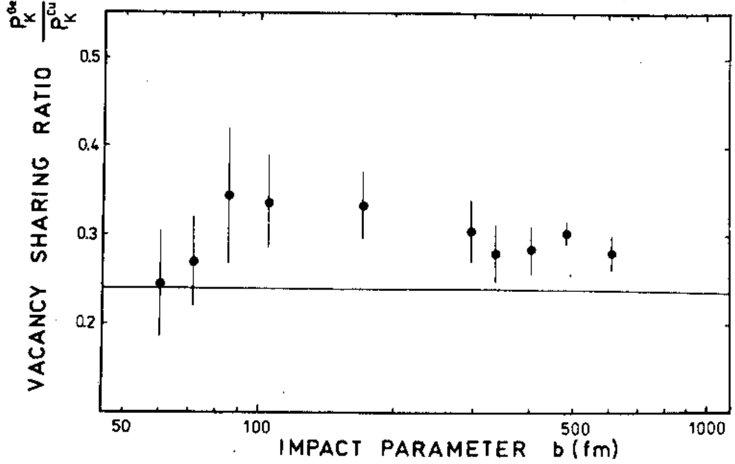

Fig.3. The vacancy sharing ratio for the Cu-Ge collision system versus the impact parameter b. The solid line represents the "one passage" calculation following Briggs/20/. The error bars represent only statistical uncertainties.

with the "one passage" calculations following Briggs. The overall agreement between them is fairly good.
However, despite large uncertainties there seems to be an indication for some structure in the experimental curve. A similar but more pronounced behaviour has been recently found by Bethge et al./7/.

5. CONCLUSIONS

In attempt to explain the magnitude of the vacancy production probability $P_K(b)$ the statistical model gives good results. This fact points out to the necessity of including coupling to higher orbitals in the description of the effect. On the other hand, the shape of the $P_K(b)$ distribution suggest that the rotational $2p\pi - 2p\sigma$ coupling is still an important mechanism of the K-shell vacancy production in symmetric and near-symmetric Cu-Cu and Cu-Ge collisions. The vacancy sharing the collision partners is well described by the Briggs-Meyerhof-Demkov model even for such asymmetric as Cu-Ag system. However, for Cu-Ge system a slight difference is observed between the experiment and the "one passage" calculations according to Briggs. The latter predict the ratio to be nearly constant in the measured range of impact parameters.

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