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K- AND L-VACANCY PRODUCTION IN ASYMMETRIC 1 MeV/N COLLISIONS OF Cu AND Nb PROJECTILES WITH Au AND Pb TARGETS

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1. INTRODUCTION

In slow and very asymmetric collisions the ionization of the $3d\sigma$ diabatic state in a transiently formed quasimolecule leads to the creation of K- and L-vacancies, in the lighter and heavier collision partner , respectively (Fig.1). This ionization occurs at small internuclear distances between both colliding partners close to the united-atom (UA) configuration, where the $3d\sigma$ state is highly promoted. Meyerhof [1] proposed the extended UA-model of Briggs [2] to this situation. During the separation of the two collision partners the $3d\sigma$ vacancies are shared on the outgoing part of the trajectory, among the K- and L-shells of the lighter and heavier partners, respectively. The impact parameter dependence of the inner-shell vacancy-sharing process serves as a sensitive test of the molecular-orbital (MO) model applied in the description of the inner-shell ionization during slow ion-atom collisions. The K-L and L-K vacancy-sharing has recently been investigated, both experimentally [3-7] and theoretically [8]. The Demkov-Meyerhof [9,10] and Nikitin [11] models were exploited to explain experimental data. Fritsch and Wille have shown [8] the limitations of these two state models and presented the four-state treatment of K-L vacancy sharing, which seems to be more appropriate to explain the experimental results for light collision systems.

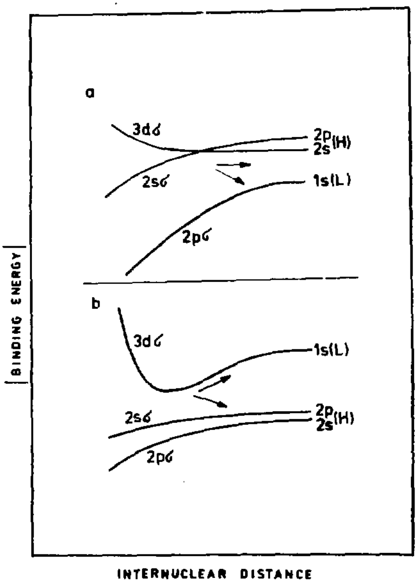

Fig.1. A schematic correlation diagram for (a) "unswapped" (Nb-Au) and (b) "swapped" (Cu-Au) collision systems.

To investigate the processes listed above for the heavier systems we carried out experiments in the region $108 \leq Z_1+Z_2 \leq 124$ (near the K-L level matching conditions) both for the "swapped" and "unswapped" cases (Fig.1).

2. EXPERIMENT AND DATA ANALYSIS

The U-300 cyclotron of the JINR Laboratory of Nuclear Reactions in Dubna provided a beam of $^{63}Cu^{4+}$ and $^{93}Nb^{6+}$ ions of energy 1 MeV/N. The experimental set-up consisted of a two-slit collimator, an intrinsic Ge X-ray detector and an avalanche gaseous particle detector. The details of the experimental set-up are described elsewhere[12]. Coincidences between scattered particles and X-rays were registered by a standard slow-fast coincidence circuit. The coincident events were processed at a TPAi minicomputer. The single spectra of X-rays recorded simultaneously with the single spectra of scattered particles and coincidence spectra enable us to obtain total and differential cross sections simultaneously. Thin 100 $\mu g/cm^2$ self-supporting foils of Au and Pb were used for targets. The measurements covered the scattering angles ranging from 2° to 16° in the laboratory system, which is equivalent to a 200-1500 fm impact parameter range (assuming unscreened Coulomb potential - an assumption which holds true in the scattering angles under consideration). To determine the energy dependence of the total $3d\sigma$ ionization cross section, the energy of the beam was varied by putting 0.77 mg/cm^2 nickel stopping foils into the beam. The degraded projectile energies were calculated with the help of Northcliffe and Schilling tables[13] to be 63, 47, 33 MeV and 93,71, 51 MeV for Cu and Nb ions, respectively. The estimated energy discrepancies are not higher than 10%[14]. The quantities measured in the experiment were put onto an absolute scale by X-ray detector efficiency calibration with standard absolute radiative sources placed at the position of the target. The effect of the Ge K-absorption edge, revealed as a drop in efficiency at 11.1 keV, was estimated to be of the order of 15%. It was determined from the difference in the L_β/L_α intensity ratio for Pb, measured with the Ge and Si(Li) detectors[15]. The estimated overall systematic uncertainty of the X-ray detector efficiency was about 20%. The fluorescence yields ω_K and ω_L used in the transformation of X-ray - into vacancy production cross sections were that for "neutral" atoms[16]. The accuracy of the absolute X-ray detector efficiency discussed previously is another source of uncertainty. The overall accuracy of the experimental values is therefore estimated to range from several tens percent to a factor of 2.

3. EXPERIMENTAL RESULTS

3.1. Differential Cross Sections

The experimental values of the impact parameter dependence of the $3d\sigma$ vacancy production are presented in Fig.2 together with the theoretical model predictions. It can be seen that the shape of the experimental curve is roughly the same for both collision systems, and this suggests the same vacancy production mechanism irrespective of the collision partners. It seems reasonable, from the course of the experimental points, to apply, as a first guess, a simple Kessel model [17] to explain the data. The range of the P(b) dependence in the frame of that model, calculated from the total cross section, amounts roughly to 2000 fm for Cu–Au and to 1850 fm for Nb–Au systems. Therefore, the average impact parameter $\langle b \rangle$ for the $3d\sigma$ ionization equals 1300 fm and 1200 fm for these systems, respectively. The early Bang-Hansteen scaling rule [18] has been considered recently to be useful for heavier collision systems, too [19-21]. According to this rule, the average impact parameter $\langle b \rangle$ is related to the minimum energy transferred to the bound electron, E, by the equation $E = C \cdot h \cdot v / \langle b \rangle$, where v is the projectile velocity and C is an empirical constant of the order of 1 (ref. [19]). Applying this formula, one can obtain for the energy transfer the values of 6.7 keV and 7.3 keV for Cu-Au and Nb-Au collision systems, respectively. We can compare these values with the binding energy of the 3d electrons in the UA limit taken from ref. [22]. We obtain 5.9 keV and 7.7 keV for Cu–Au and Nb–Au, respectively. Therefore, it seems that the $3d\sigma$

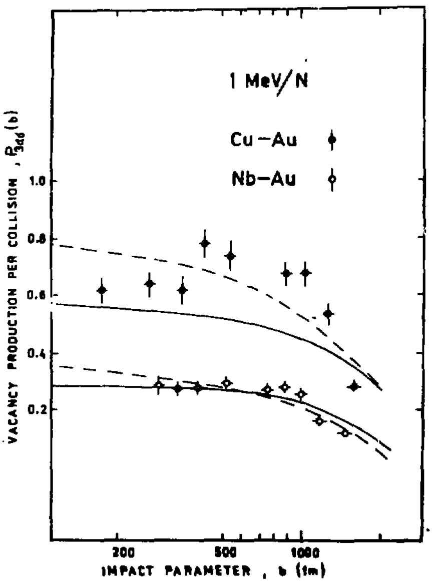

Fig.2. Experiment and model predictions for the impact parameter dependence of the $3d\sigma$ vacancy production; solid line – UA model of Briggs, dashed line – statistical model.

vacancy production is mainly determined by the direct ionization but not by couplings to other bound states. Moreover, we applied the UA-ionization model of Briggs [2] and Meyerhof [1] to our highly promoted $3d\sigma$ state. This model treats the amplitude of the ionization process as a coherent sum of two amplitudes for the ionization by each particle moving relative to the center of mass of the UA system. A comparison of the experimental data with this model shows that the experimental values are by a factor of 3÷5 larger than the model predictions. Assuming the Meyerhof's semiempirical formula [1] for the $3d\sigma$ total cross section dependence on energy and on the UA atomic number Z_{UA} we obtained the values of 29 kbarns and 11 kbarns for Cu-Au and Nb-Au collisions, respectively. The curves calculated within the UA-ionization model were normalized to these values (solid lines in Fig.2) by using our experimental total cross sections. One can see that after this normalization procedure the discrepancy between experiment and theory, however diminished, still exists. The main feature of our experimental curve is that it falls off more rapidly for large impact parameters than the predictions of the UA-ionization model do. An additional comparison of the experimental data with the statistical model [23,24] was done. This model has two parameters, R_0, an interaction radius, and D, a diffusion constant. The ejection of the electron by a diffusion through the energy level ladder may be treated independently for K and L shells. For the Cu-Au collision system we obtained two sets of parameters for Cu-K- and Au-L-vacancy production, independently. The curves obtained were summed later on to give the $3d\sigma$ ionization curve shown in Fig.2. For the Nb-Au collision system, practically only Au-LX-rays were measured. The values of the parameters for the two collision systems are listed in the Table.

Table

Statistical model parameters

Collision system		Cu-Au	Nb-Au
Total cross section $\sigma_{3d\sigma}$, kbarns		99	27
K-shell of the lighter partner	R_{0K}, fm	7000	-
	D_K, cm^2/s	9	-
L-shell of the heavier partner	R_{0L}, fm	10000	8000
	D_L, cm^2/s	29	23

It can be seen that the statistical model describes the experiment a little better than the UA-ionization model, but discrepancies at large impact parameters are still observed (Fig.2).

3.2. Total Cross Sections

Near the K–L level matching the $3d\sigma$ vacancy production is commonly accepted to be merely a sum of K- and L-vacancy production in the lighter and heavier collision partners, respectively. In Fig.3 the $3d\sigma$ cross sections are plotted versus projectile energy for the investigated collision systems: Cu-Au, Cu–Pb, Nb–Au, Nb–Pb. As found recently by Meyerhof [1], the $3d\sigma$ total cross section depends on the projectile velocity v and on the nuclear charge sum $Z_{UA} = Z_1 + Z_2$ in the following way:

$$\sigma(3d\sigma) = F(v)\exp[-g(v)Z_{UA}]. \qquad (1)$$

The calculations performed by Meyerhof in terms of the UA-ionization model [1] give the following dependence of F(v) and g(v) on v:

$$F(v) \sim \frac{1}{v}, \quad g(v) \sim \frac{1}{\sqrt{v}}. \qquad (2)$$

A similar slope of the energy dependence of $\sigma(3d\sigma)$ for all systems investigated (Fig.3) suggests that our data can be interpreted by means of this universal formula. The least-squares fits of expression (1) to our experimental data are plotted in Fig.3. The fit gives

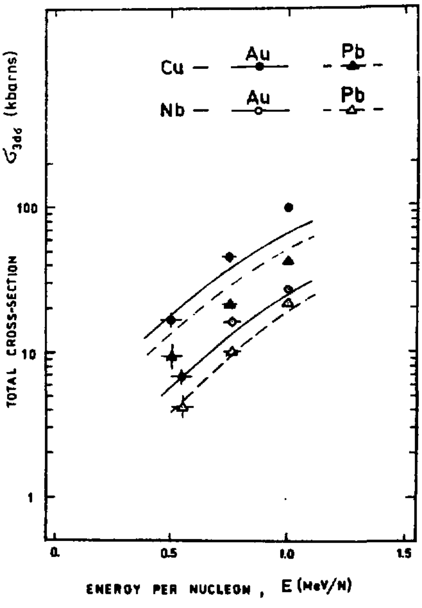

Fig.3. Energy dependence of the vacancy production cross section: solid line – fit to the semiempirical formula from [14] for Au target, dashed line – the same for Pb target.

$\sigma(3d\sigma)$ in the form:

$$\sigma(3d\sigma) = \frac{A}{\sqrt{E}} \exp\left(-\frac{BZ_{UA}}{\sqrt{E}}\right) \text{ kbarn}, \qquad (3)$$

where E - projectile energy in MeV/N, $A = (4.4 \pm 1.8) \times 10^5$, and $B = (8.2 \pm 0.3) \times 10^{-2}$. One can state that the general behaviour of the experimental total cross sections may be explained by expression (1). However, some discrepancies exist, especially for Cu projectiles.

3.3. K-L Vacancy Sharing

The K-L vacancy sharing is generally a four - state process dominated by radial couplings between the σ states. To make this process tractable, it is possible to break down the four-state problem into a set of two-state problems [3,4]. This approach requires, from an experimental point of view, a good energy resolution of the L-spectrum of the heavier collision partner, and sufficient statistics in all L lines describing vacancy population in L subshells. Since our L spectra do not fulfil all these requirements satisfactorily, we decided to treat the K-L vacancy sharing as a two-state process, only. This is partially justified because the L-vacancy production is dominated by the $2p_{3/2}$ ionization, in particular, for the "swapped" collision systems. The K-L vacancy sharing occurs at internuclear distances far from the region of $3d\sigma$ vacancy creation. Assuming that only vacancies created in the $3d\sigma$ state participate in this process, the K-L vacancy sharing ratio should be independent of the impact parameter in the impact parameter range under consideration. It turns out from the data presented in Fig.4 that the vacancy sharing ratio for the Cu-Au collision system increases slightly for impact parameters $b \lesssim 600$ fm. This increase (dashed line in Fig.4, only to guide the eye) may indicate that an additional process, besides the $3d\sigma$ ionization, creates vacancies influencing the K-L vacancy sharing ratio, e.g., the direct ionization from molecular orbitals correlated to the L shell in the UA limit. At the moment no further conclusions are justified. This problem should be tested carefully with a variety of collision systems and at smaller impact parameters, i.e., in the region where this additional ionization should play a considerably greater role. Nevertheless, we should note that this eventual UA L shell ionization does not influence our total $3d\sigma$ cross section considerations given above, because of the much higher UA L-binding energy than that for the $3d\sigma$ state. As support

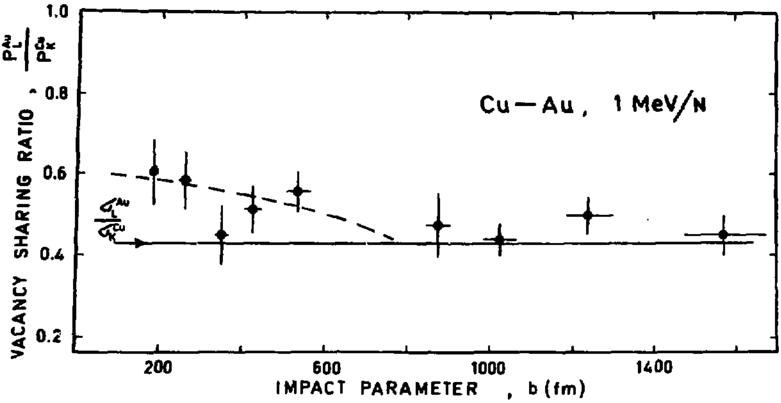

Fig.4. Impact parameter dependence of the vacancy sharing ratio for Cu-Au collision system: solid line - from the total cross sections.

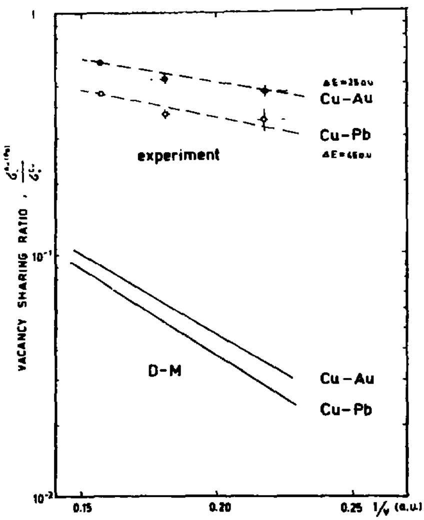

Fig.5. Energy dependence for the vacancy sharing ratio (see the text for explanations).

to this statement serves the behaviour of the K-L sharing ratio for b>600 fm, Fig.4. In this impact parameter region, which mainly contributes to the total $3d\sigma$ vacancy production, the sharing ratio is constant, and coincides with the value taken from the total Cu-K and Au-L vacancy production ratio (solid line in Fig.4).

The energy dependence of the K-L sharing ratio taken from the total vacancy production is presented for the Cu-Au and Cu-Pb collision systems in Fig.5. The exponential fall of the sharing ratio with 1/v, v being the projectile velocity, is a common feature of the Demkov-Meyerhof [9,10] and Nikitin [11] model predictions.

However, deviations from this exponential character are found for light collision systems both experimentally [7] and theoretically [8]. Our results seem to establish the exponential behaviour of the sharing ratio (Fig.5), but in a very small interval of the 1/v parameter and for high projectile velocities. The predictions of the Demkov-Meyerhof model [10] used in the original form underestimate our data by a factor of ten (Fig.5), in agreement with previous findings [5,6] (solid line in Fig.5). The K–L vacancy sharing ratio S is given within this model by the formula:

$$S = \frac{\sigma_L}{\sigma_K} = \exp\left(-\frac{\pi \Delta E}{a v}\right), \qquad (4)$$

where v is the projectile velocity, ΔE is the energy separation between both interaction states assumed to be a constant in the coupling region, $a=(\sqrt{I_1}+\sqrt{I_2})/\sqrt{2}$, where I_1 and I_2 are the binding energies of both coupled states taken as for separated atoms. All quantities are expressed in atomic units. The solid lines in Fig.5 are drawn by setting $\Delta E = I_1 - I_2$ into formula (4). These ΔE values are 140 and 190 a.u. for Cu-Au and Cu-Pb collision systems, respectively. Such a ΔE value is obviously not correct for the K–L vacancy sharing and leads to discrepancies with experiment. For the "swapped" collision systems, the energy separation ΔE in the region of interaction is generally smaller than $I_2 - I_1$ [25]. As proposed by Stolterfoht [26], it should be possible to readjust the ΔE parameter in expression (4) to describe the K–L vacancy sharing by means of the simple Demkov-Meyerhof model. The broken lines in Fig.5 are drawn according to (4) by using ΔE as a free parameter. The values of this parameter are found to be 28 a.u. and 46 a.u. for Cu–Au and Cu–Pb, respectively. ΔE may be interpreted as a mean energy separation extended over the interaction region $1/a$. At the moment we are unable to compare this ΔE value with the real energy separation taken from the correlation diagrams. However, we should point out that the slope of the experimental vacancy sharing ratio is reproduced by the Demkov-Meyerhof formula very well (see Fig.5).

The Nikitin model takes into account the variation of ΔE with the internuclear distance. According to this model, the K–L vacancy sharing ratio S is given by:

$$S = \frac{\exp[2\pi\Delta\epsilon \cos^2(\theta/2)/(a v)] - 1}{\exp[2\pi\Delta\epsilon/(a v)] - \exp[2\pi\Delta\epsilon \cos^2(\theta/2)/(a v)]}, \qquad (5)$$

where $\Delta\epsilon, a, \theta$ are the parameters describing the shape of the energy separation $\Delta E(R)$ between two interacting states.

This formula was exploited to fit the experimental data by using different sets of the introduced parameters $\Delta\epsilon, a, \theta$ (refs. [5-7]). This method seems to work quite well for light collision systems. For heavier systems, the sets of parameters used were uncapable of describing the experimental data satisfactorily [4]. As is mentioned in [4], the discrepancies increase as the separated-atom energy difference $I_2 - I_1$ becomes smaller. Moreover, as discussed recently by Fritsch and Wille [8], the range of applicability of this two-state model is limited due to differences between the real shape of the dynamic coupling and the approximated symmetric shape of the coupling involved in the Nikitin consideration [11]. In our case it was impossible to fit the experimental data within the Nikitin model by use of realistic values for the parameters $\Delta\epsilon$, a, θ. It was not possible either to get simultaneously the correct absolute values and the correct slope of the K-L vacancy sharing ratio S.

In the "unswapped" Nb-Au collision at 1 MeV/N we extracted the L-K vacancy sharing ratio of $(5.9 \pm 1.0) \times 10^{-3}$. This value is almost by a factor of ten smaller than the Demkov-Meyerhof model predictions (4.1×10^{-2}) calculated with $\Delta E = I_2 - I_1$, being 230 a.u. for that system. This discrepancy can be understood if we remind that the real energy separation ΔE in the interaction region is for "unswapped" collision systems higher than $I_2 - I_1$ [25]. After readjusting the ΔE value the Demkov-Meyerhof formula (4) should still hold true. From our single value of the L-K sharing ratio for the Nb-Au system, we obtain a mean energy separation in the interaction region of $\Delta E = 355$ a.u.

4. CONCLUSIONS

The existing models are uncapable of providing a proper explanation to all our experimental data. The experimental P(b) curves fall off rapidly with increasing impact parameter, as opposed to the statistical and UA-ionization model predictions. The reason for that is not clear at the moment. For the Nb-Au collision system, the agreement between the experiment and the UA-ionization model of Briggs is better. This can be seen in the behaviour of the total cross section vs. energy, too (Fig.3). The energy dependence of the vacancy sharing ratio seems to follow an exponential form of Demkov type. It would be proved better in an experiment at higher energy. The simple Demkov-Meyerhof formula seems to be capable to describe the behaviour of the K-L vacancy sharing ratio by readjusting properly the ΔE parameter.

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