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# K-SHELL VACANCY PRODUCTION AND SHARING IN (0.2-1.75) MeV/u Fe, Co + Cr COLLISIONS

C. CIORTEA, I. PITICU, D. FLUERAŞU, D.E. DUMITRIU,

A. ENULESCU, M.M. GUGIU, A.T. RADU

*Nuclear Physics Department, National Institute for Physics and Nuclear Engineering "Horia Hulubei",*
*P.O.Box MG-6 Măgurele, 76900 Bucharest, Romania*

*E-mail address:* ccio@ifin.nipne.ro

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*Abstract.* K-shell ionization cross sections measured in the near-symmetric collision systems Fe and Co + Cr at 0.2-1.75 MeV/u energies are reported. The cross section values have been corrected for multiple ionization in the outer (L, M) shells by using the energy and yield shift method. Mean ionization probabilities per electron in the outer shells have been estimated. In the Fano-Lichten model of quasi-molecular excitation, 2pσ molecular orbital (MO) ionization cross sections and mean 2pσ-1sσ MO vacancy sharing probabilities were obtained.

The present 2pσ MO ionization cross sections could be explained at higher (≥ 1 MeV/u) bombarding energies by an one-step process (Briggs model using SCA calculations with relativistic hydrogenic wave functions and binding correction). The remaining discrepancy between experiment and direct ionization model at lower energies could not be reduced by considering the contribution of single collision two step or multiple collision processes.

The vacancy sharing results show that an exponential dependence of the vacancy sharing in function of reciprocal velocity is less fulfilled at higher (≥1.5 MeV/u) energies.

Concerning the present multiple ionization data, the Cr M-shell probability is comparable with that of the Cr L-shell for both collision systems, in contrast with the results for the projectile. In the latter case, a larger M-shell multiple ionization in comparison with that of the L-shell is found. The result for the target atom could be qualitatively explained by a rapid partially filling of the M-shell vacancies within the solid target.

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## 1. INTRODUCTION

The gross features of K-shell vacancy production mechanism in heavy ion collisions at low velocities (the projectile velocity $v_1$ small compared with the orbital velocity $v_e$ of the active electron, $v_1 << v_e$) in the near of K-K or K-L level matching are good known (see e.g. the papers of Meyerhof et al. [1-4]).

In slow collisions, a large enhancement of K-shell ionization cross sections over the predictions of the Coulomb excitation theory is explained in the electron promotion model (Fano and Lichten [5]). There are two main mechanisms: *(i)* the formation of vacancies in highly promoted molecular orbitals (MO) at small inter-nuclear separations, and *(ii)* the sharing of these vacancies on the outgoing part of the collision between the two partners (see Fig. 1 for schematic correlation diagrams).

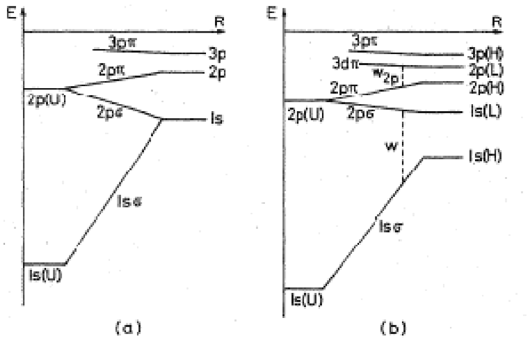

Figure 1.- Schematic correlation diagrams for (a) symmetric and (b) for asymmetric collisions [1]. Only levels relevant to the present experiment are shown. Vacancy sharing processes *w* and $w_{2p}$ are indicated in (b). The letters H, L and U refer to the higher-Z and lower-Z partners and to the united atom, respectively.

cy production in near-symmetric collisions is the ionization of the *2pσ* molecular orbital (MO) at small inter-nuclear distances followed by sharing of the primary vacancies among the K-shells of the partners [6].

In the present paper we report K-shell ionization cross sections for both collision partners, as well as *2pσ* MO vacancy production cross sections and mean *2pσ - 1sσ* MO vacancy sharing probabilities, for the near-symmetric collision systems Fe, Co + Cr at 0.2-1.75 MeV/u bombarding energy. The cross section values have been corrected for the effects of multiple ionization in the outer (L, M) shells by using the energy and yield shift method [7]. Mean ionization probabilities per electron have been estimated. Comparison of the experimental data with some model calculations are also presented.

## 2. EXPERIMENT

Beams of $^{56}$Fe and $^{59}$Co were produced at the Bucharest FN tandem Van de Graaff accelerator (see Fig. 2 for a schematic view of the apparatus). The beam was directed onto a self-supported target of $^{52}$Cr (approximately 1 mg/cm$^2$ thick), placed at 45$^0$ with respect to the beam direction. X-rays and scattered projectiles were detected. An hyper-pure Ge detector placed at 90$^0$ to the beam direction was used to detect the X-rays. The projectiles scattered at approximately 10.5$^0$ relative to the beam direction have been registered with a scintillation foil (50 μm thick).

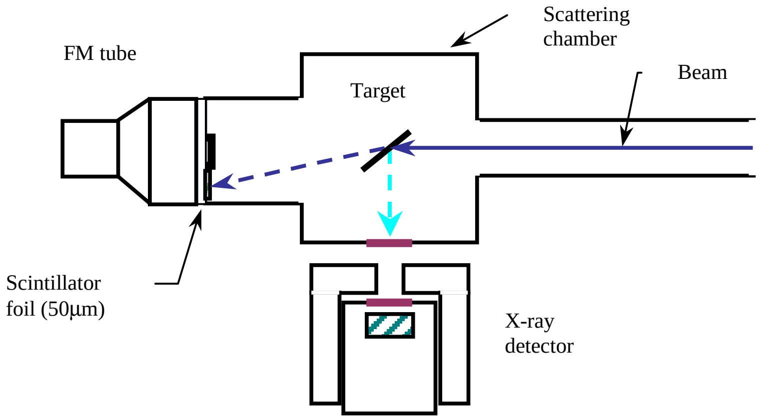

Figure 2. Schematic view of the experimental arrangement.

The detectors' absolute efficiency were determined by using standard X-ray and alpha particle sources ($^{241}$Am and $^{55}$Fe). Periodically, the energy calibration of the X-ray spectrometer was checked with radioactive sources.

## 3. RESULTS AND DISCUSSION

### 3.1. MULTIPLE IONIZATION

An inevitably ingredient of the inner-shell ionization studies in heavy ion - atom collisions is the need for an adequate treatment of the multiple ionization effects on the X-ray or Auger electron spectra. The presence of spectator vacancies in the outer shells during de-excitation of the inner-shell vacancies alter the decay rates and determines satellite lines emission. In X-ray spectra obtained with a relatively low energy resolution, as given by semiconductor detectors, the X-ray lines are shifted in energy and have enlarged widths, while their relative intensities are modified. In Fig. 3, the measured dependence in function of bombarding energy of the X-ray energy and yield shifts is given.

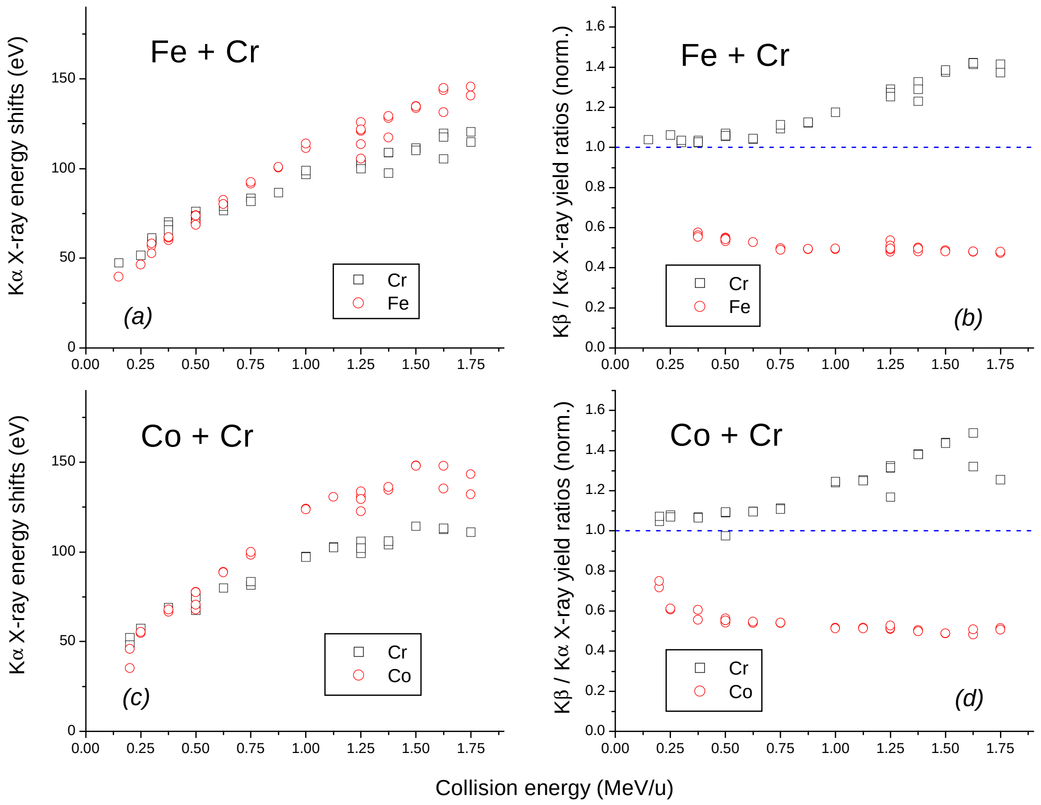

Figure 3.- Kα X-ray energy shifts (*a* and *b* frames in the figure) and Kβ / Kα X-ray yield ratios (*c* and *d*), measured for the collisions Fe, Co + Cr, in dependence of collision energy.

Being a many-body problem, an exact treatment of the multiple ionization is not possible. Still, an approximate treatment under reasonable assumptions could give valuable results. Such calculations in the independent electron approximation have been done in the present paper, and coupled with the measurements of energy and yield shifts of the X-ray lines, they were used: *(i)* to extract information about the average numbers of outer-shell spectator vacancies, or equivalently, the multiple ionization probabilities (defined as the ratio between the average number of vacancies created in the collision in the shell and the occupancy number of that shell, see Fig. 4a, c), and *(ii)* to correct approximately the inner-shell vacancy decay parameters (partial widths) (Fig. 4b, d).

Although this method does not have inherently a high precision and systematic errors could occur, it permitted us to obtain average charge states of 0.1 -1.5 MeV/u Mn, Fe, Co, Ni, and Cu ions in solid (Au and Bi) targets [8] in good agreement with other data obtained by more conventional methods. This success encouraged us in using these mean probabilities together with the inner-shell cross sections in an unified interpretation of inner shell vacancy production, as given by the statistical model [9].

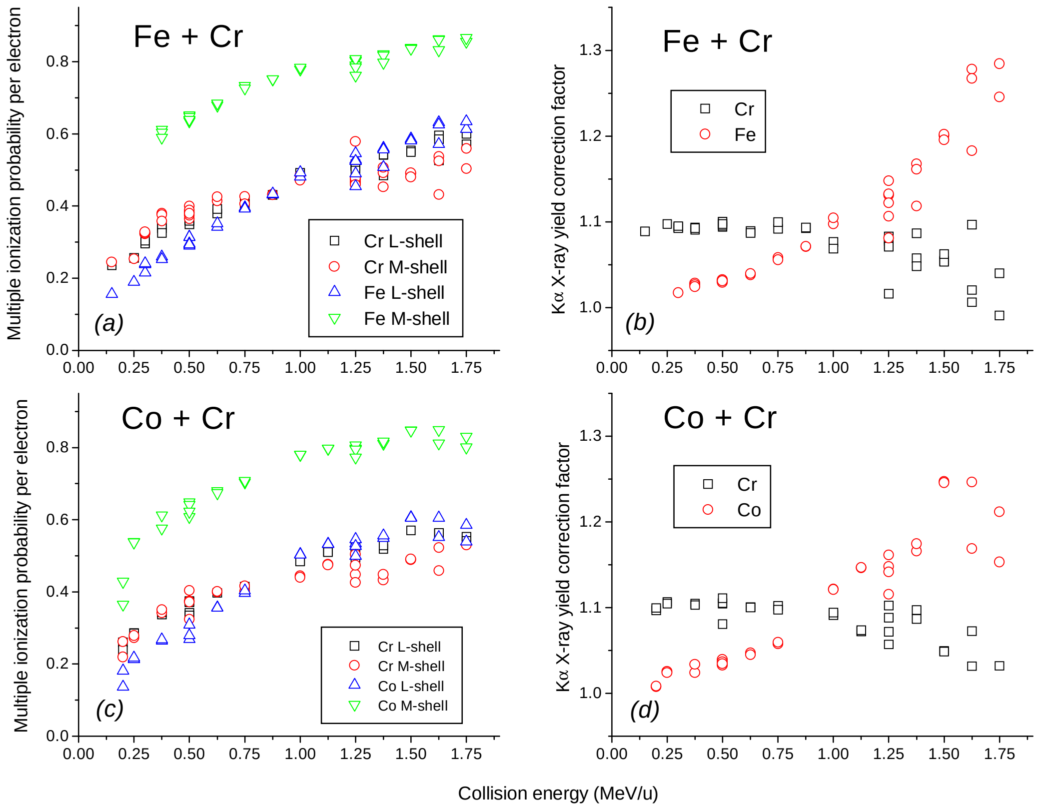

Figure 4.- Mean multiple ionization probabilities per electron (*a* and *c* frames in the figure) and the Kα X-ray yield correction factors (*b* and *d*), determined for the collisions Fe, Co + Cr, in dependence of collision energy.

Concerning the present multiple ionization data, it is interesting that the multiple ionization probability of the Cr M-shell is comparable with that of the Cr L-shell for both collision systems, in contrast with the results for the projectile. In the latter case, a larger

M-shell multiple ionization in comparison with that of the L-shell is found. The result for the target atom could be qualitatively explained by a rapid partially filling of the M-shell vacancies within the solid target.

### 3.2. IONIZATION CROSS SECTIONS

By normalizing the X-ray yields to the elastically scattered projectiles, the absolute ionization cross sections of the K-shells of both collision partners were determined (Fig. 5a, c). Typical experimental errors of these data are in the range 15% - 30%.

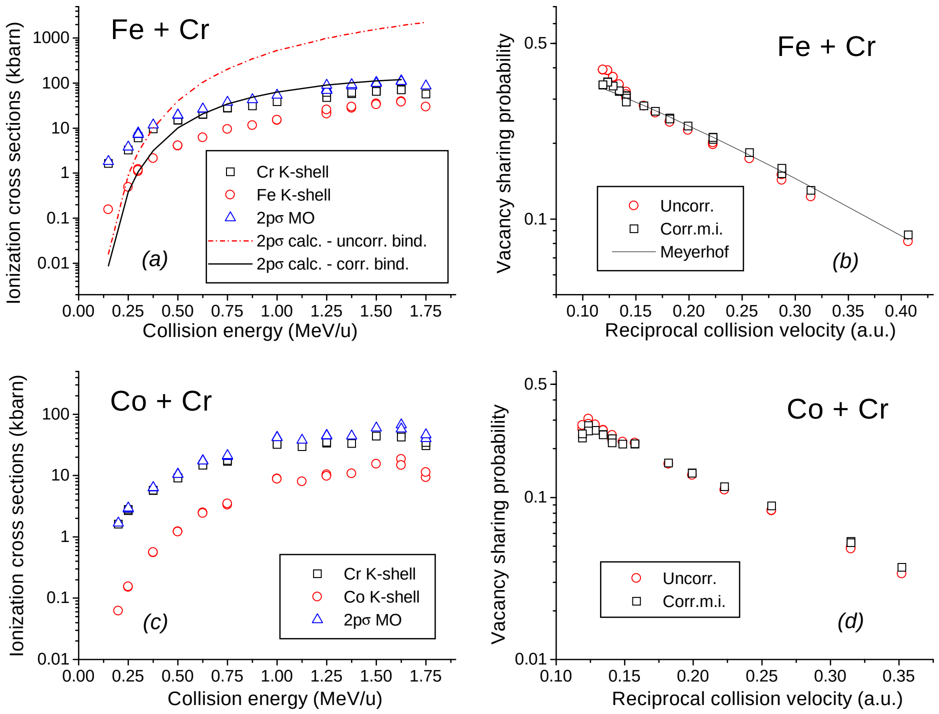

Figure 5. Ionization cross sections of the K-shell of both collision partners and of the 2pσ MO, in dependence of collision energy (frames *a* and *c* in the figure), as well as the vacancy sharing probability between the 2pσ-1sσ MOs (*b* and *d* frames) in function of the reciprocal collision velocity, determined for the collisions Fe, Co + Cr.

### 3.3. 2pσ - VACANCY PRODUCTION AND SHARING

The molecular model [1-6] has been successfully applied to the inner-shell vacancy production of both collision partners in symmetric or near symmetric collisions for which $v_{proj}/v_e < 1$. Since we are examining K-shell vacancy formation in collisions not far from symmetry, for which $v_{proj}/v_e < 0.35$, the molecular model should be appropriate. Within this model, the K-vacancy cross sections of the higher-Z (H) and the lower-Z (L) collision partner can be obtained by reference to the schematic diagram sketched in Fig. 1(b) for asymmetric collisions. One finds [2]:

$$\sigma(H) = (1-w)\,\sigma(1s\sigma) + w\,\sigma(2p\sigma), \tag{1}$$

$$\sigma(L) = w\,\sigma(1s\sigma) + (1-w)\,\sigma(2p\sigma) + \sigma_{K\text{-}L}\,. \tag{2}$$

Here $\sigma(1s\sigma)$ and $\sigma(2p\sigma)$ are the cross sections for exciting $1s\sigma$ and $2p\sigma$ molecular orbital vacancies, respectively. These vacancies are shared with the branching ratio $w/(1-w)$. In practice, one finds $\sigma(1s\sigma) << \sigma(2p\sigma)$, and the K-shell vacancies arise principally from $2p\sigma$ MO vacancy formation. The cross section $\sigma_{K\text{-}L}$ arises from K-L level matching effects, which are not important here. In these conditions, the summed K-shell cross section $\sigma_K^{sum} = \sigma_K(L) + \sigma_K(H)$ gives the $2p\sigma$ vacancy production cross section (Fig. 5a, c). The lines in the Fig. 5a represent some model predictions for $2p\sigma$ MO ionization which are discussed below.

A simple *two-state* model of sharing (Meyerhof-Demkov model [6]) gives the following expression for this sharing ratio:

$$\frac{w}{1-w} = e^{-2x} \tag{3}$$

$$2x = \frac{2^{1/2}\pi\left(I_H{}^{1/2} - I_L{}^{1/2}\right)}{m^{1/2} v_1} \tag{4}$$

Here $I_L$ and $I_H$ are the K-binding energies of the lower- and higher-Z collision partners, respectively, $v_1$ is the projectile velocity; tabulated, non-perturbed binding energies were used in calculation. The present experimental vacancy sharing probabilities $w$ are presented in Fig. 5b, d and the above prediction for the collision Fe + Cr is fairly good at lower energies. The results show that an exponential dependence of the vacancy sharing in function of reciprocal velocity [6] is less fulfilled at higher (≥1.5 MeV/u) energies.

In collisions with $Z_1$, $Z_2 \leq 10$, the $2p\sigma$ - $2p\pi$ electron promotion process (by rotational-coupling) dominates $2p\sigma$ vacancy formation. For $Z_1$, $Z_2 > 10$, which is the case here, the $2p\pi$ MO is generally filled. Nevertheless, electron promotion could operate by means either of one-collision two-step process or by a multiple-collision process.

The *one step* direct ionization of $2p\sigma$ MO has been estimated in the present work by using the Briggs united-atom (UA) ionization model [10]. In an approximation due to Meyerhof [11], and using the UA semi-classical calculations (SCA) [12], we can write:

$$\sigma(2p\sigma, v) = \left(\sqrt{\sigma_1(\alpha v)}/\alpha + \sqrt{\sigma_2(\beta v)}/\beta\right)^2 \tag{5}$$

where $\sigma_1(\alpha v)$ and $\sigma_2(\beta v)$ are corresponding to excitation of an UA state by the nuclei $Z_1$ and $Z_2$ (projectile and target), respectively. Here $\alpha = Z_1/(Z_1+Z_2)$ , $\beta = 1-\alpha$ .

In Fig. 5a, the experimental $\sigma(2p\sigma)$ for the collision system Fe + Cr is compared with these calculations, taking into account the tabulated binding energies for the UA-2*p* state (dash-dot line). As expected, the one-step process – direct ionization - could not explain the experimental cross sections. A much better estimation at higher (≥ 1 MeV/u) energies could be obtained if we take into consideration the correction to the binding energy, obtained from non-relativistic calculations [13] (the straight line in the Fig. 5a). But at lower energies, as can be seen in the figure, the predictions are much lower than the experiment, and the ratio experiment/calculation is increasing when the bombarding energy is decreasing. Qualitative considerations of other promotion processes are given below.

Thus, in the *one-collision two-step process*, the $2p\pi$ MO could couple to vacant projectile or target states, e.g., via the $3d\pi$ MO (see Fig. 1); the resulting $2p\pi$ vacancies could be transferred in $2p\sigma$ MO by electron promotion. One might write for this process:

$$\sigma(2p\sigma) = N(v_1)\ \sigma_{rot}\ . \tag{6}$$

Here $\sigma_{rot}$ is the $2p\sigma$ - $2p\pi$ rotational-coupling cross section per incident $2p\pi$ vacancy [14], and $N(v_1)$ is a factor which describes the long-range coupling. One finds that an expression like

$$N(v_1) \cong C\ e^{-\alpha/v1} \tag{7}$$

could be used, where $C \cong 1$ and $\alpha$ is proportional to $\Delta E$, the energy splitting; this expression is typical for a large class of non-adiabatic transitions [15]. In the two-step process, the $2p\pi$ orbital would be radially coupled to many vacant $\pi$ orbitals. Because this mean binding energy would be considerably smaller than $2p$(H) binding, in first approximation we could neglect it, and set [6]

$$\alpha / v_1 \cong 2^{1/2}\ \pi\ [I_{2p}(H)]^{1/2}/m^{1/2}\ v_1\ , \tag{8}$$

where $I_{2p}$(H) is the $2p$ binding energy of the higher-Z partner. Estimations using eqs. (6-8) should give increasing contribution when the bombarding energy increase. Therefore, the one-collision two-step process could not explain the large $2p\sigma$ MO ionization cross section obtained at low energies.

The *multiple-collision process* is due to projectile $2p$ vacancies, made in some collision, which live long enough to be carried into a second collision, where the $2p\sigma$ - $2p\pi$ promotion can operate. If target recoil processes are negligible, only projectile $2p$ vacancies need to be considered. Multiple collision effects are important in solid targets, because the repeated excitation of the projectile electrons could produce an appreciable steady-state vacancy distribution in the projectile inner-shells. The multiple contribution to $\sigma(2p\sigma)$ due to steady-state projectile $2p$ vacancies is found to be

$$\sigma_{mc}(2p\sigma,v) = \frac{1}{3} n(v) w_{2p} \sigma_{rot} \tag{9}$$

Here $n(v)$ is the number of $2p$ vacancies carried by the projectiles moving with velocity $v$ inside the target material; $w_{2p}$ is the $2p$-vacancy sharing probability, given by an expression as before [6], by considering the $2p$ ionization energies of the projectile and target atoms. The equilibrium fraction of projectiles carrying one $2p$-vacancy $f_1^{eq}$ dominates, then $n(v) \cong f_1^{eq}$ .

Hence we can write [2]:

$$\sigma(2p\sigma) = \sigma_{sc}(2p\sigma) + \sigma_{mc}(2p\sigma)\ , \tag{10}$$

where both one- and two-step processes contribute to single-collision cross section $\sigma_{sc}(2p\sigma)$ .

However, estimations using eqs. (6-10) give increasing contribution to the 2pσ MO ionization when the bombarding energy increases. Therefore, the one-collision two-step process and the multiple collision process seem not to be important for 2pσ ionization in the present heavy ion – atom collisions.

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