

# ION – ATOM COLLISION IN SOLIDS

**J. Braziewicz<sup>1</sup>, S. Chojnacki<sup>2</sup>, I. Fijał<sup>3</sup>, M. Jaskóla<sup>3</sup>, A. Korman<sup>3</sup>, W. Kretschmer<sup>4</sup>, U. Majewska<sup>1</sup>, M. Polasik<sup>5</sup>, K. Słabkowska<sup>5</sup>**

<sup>1</sup>Institute of Physics, Świętokrzyska Academy, Kielce, Poland

<sup>2</sup>Heavy Ion Laboratory, Warsaw University, Warsaw, Poland

<sup>3</sup>The Andrzej Soltan Institute for Nuclear Studies, Otwock - Świerk, Poland

<sup>4</sup>Physikalisches Institut, Universität Erlangen - Nürnberg, Erlangen, Germany

<sup>5</sup>Faculty of Chemistry, Nicholas Copernicus University, Toruń, Poland

## Background

In collisions of heavy ions with target atoms the strong Coulomb field of one of the 'collision partners' can cause simultaneous ejection of several electrons of the second one. This process results in a reduction of the nuclear charge screening and increases the binding energy of remaining electrons [1]. Consequently, the energies of x-rays emitted from such multiply ionized atoms are shifted with respect to the corresponding x-ray energies of singly ionized atoms and reflect the actual configuration of electrons during x-ray emission. Finally, as a result of the multiple ionization, instead of a single-hole x-ray transition called the diagram line, the structure of x-ray satellites appears.

Collision processes have been studied extensively for many years but these studies were focused mainly on single or multiple K-, L-, M-shell ionization occurring in the target atoms. The satellite structure of K and L x-ray lines of the target atoms, observed mainly with high resolution spectrometers [2-5], was interpreted as the result of additional vacancies in outer shells of the atom. However, more complicated processes are experienced by a projectile as a second partner of the ion-atom collision. During the first collision K-vacancy and/or multiple L-, M-, N- or higher shell vacancies can be produced. Further, the ion can collide with the other target atoms and capture or loss of electrons can occur before its original vacancies are filled by outer electrons. The competition between ionization, excitation, electron capture, electron loss and decay processes leads to the establishment of an electron-vacancy equilibration in different shells of the moving projectile. Collision processes occurring between the swift heavy ions and target atoms have been investigated during last decades [6-13] but most systematic experiments were performed using high resolution measurements of K or L x-rays from different projectiles in solid targets [14-23]. A review of problems discussed and experimental works has been published by Beyer *et al.* [24].

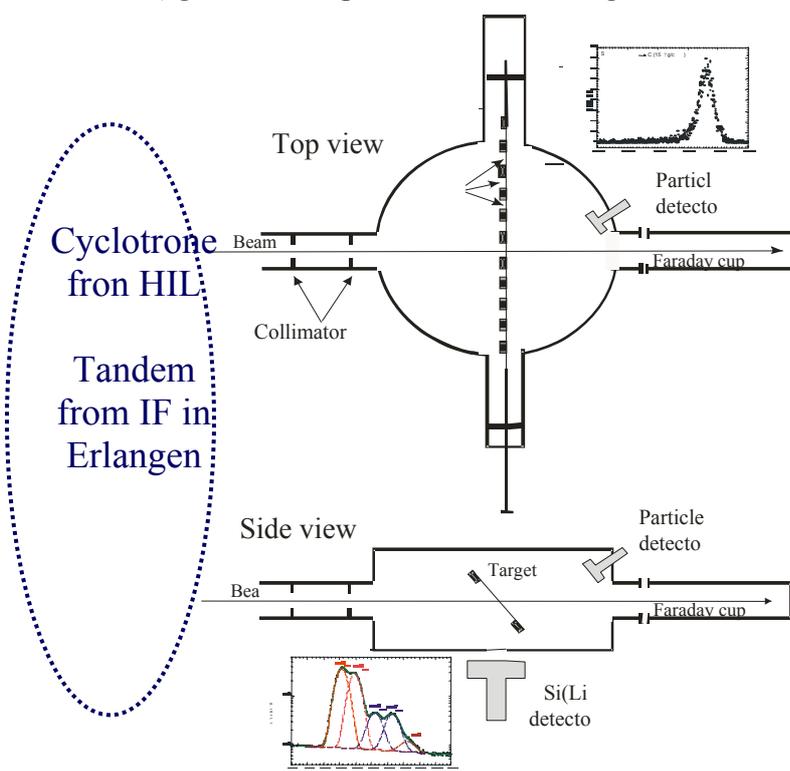
For low resolution x-ray spectrometers, such as a semiconductor Si(Li) detector, the satellite structure of induced x-rays cannot be resolved. Nevertheless, the measured x-ray spectra are strongly affected by multiple ionization, namely the x-ray lines are shifted towards higher energies and broadened [25,26] and thus information on probabilities of multiple ionization still can be extracted. The method of analysis of multiple ionization from low resolution x-ray spectra was used in our works to study the ionization probabilities for M, N and O shells of heavy atoms generated by projectiles [25,27-29] and for K, L and M shells of sulphur projectiles passing through a carbon foil [30]. We have used the fact that the x-ray satellite structure can be well approximated by the Gaussian profile, whose energy shift and

width depend on the ionization probability for the L and M shells at the moment of x-rays emission. In the present paper this method is also used.

The dependence of x-ray production cross sections of heavy ions on foil thickness has been demonstrated in several papers [6,9,23,31-35]. Generally, measured projectile x-ray yields are strongly dependent upon whether or not a K vacancy exists in the incoming ion. In some works it was found that K vacancy equilibrium is not reached until the ion passes through many atomic layers in the foil. Indeed, the works of Scharfer *et al.* [36] and Gray *et al.* [37] concerning sulphur ions show directly that the magnitude of K vacancy fractions of the ions are dependent on foil thickness. On the other hand, the charge equilibrium state for shells higher than K shell is reached very rapidly (in the first few atomic layers), as it was shown by Cocke *et al.* [38]. To describe these results quantitatively a model based on formulation of Allison [39] has been widely applied. Betz *et al.* [6] used a so called 'two component' version, in which the beam is considered to consist of two fractions of ions, those with and those without the K vacancy. In the case of a collision, when the fraction of ions with two K vacancies plays a significant role, Gardner *et al.* [11] have shown that it is necessary to consider a 'three-component' version, where the projectile may have zero, one or two K vacancies.

## Experimental details

Sulphur ion beams with incident energies of 9.6, 16.0, 22.4, 32.0 MeV and with initial charge states  $q=4^+$ ,  $6^+$  were obtained from the tandem accelerator at the Institute of Physics of the Erlangen Nürnberg University. Other energies 65, 79, 99 and 122 MeV and with incident charges of  $13^+$  and  $14^+$  were obtained from the U-200P cyclotron at the Heavy Ion Laboratory of Warsaw University. A schematic diagram of the experimental arrangement is shown in figure 1. Two collimators located at 24 and 38 cm in front of the target are used to define 2 mm in diameter beam spot on the target. Self-supporting carbon foils with effective thickness of  $15\text{-}210\mu\text{g}/\text{cm}^2$ , were positioned in the target holder at the center of vacuum chamber at an



angle of  $25^\circ$  to the direction of the beam. The geometry of the experimental arrangement used in this work means that the detector should register x-rays emitted by projectile inside the target as well as from distance up to 1.2 cm behind the target, so during such experiments x-rays with lifetimes up to  $10^{-12}$  sec are registered.

The carbon targets were prepared by vacuum evaporation and their absolute thickness was determined in the separate measurements of energy loss of 5.48 MeV  $\alpha$  particles from  $^{241}\text{Am}$  source. The stopping power values for  $\alpha$  particles in carbon needed to calculate the

Fig. 1. Schematic diagram of

foil thickness were obtained from Biersack and Maggmark [40] and the final target thickness was calculated by the computer code TRIM [41]. Additionally these thicknesses were checked using elastic scattered 2.0 MeV  $^4\text{He}^+$  ions from the Van de Graaff accelerator. Absolute target thicknesses were determined with the accuracy of about  $\pm 4\%$ . The targets could be considered as 'thin' because the ions passing through the foil did not lose energy appreciably ( $\Delta E$  was less than  $0.15E_0$  for the thickest target and for the lowest ion energy and decreased rapidly up to  $0.01E_0$  for the highest projectile energy). The effective ion energy was further used. The self-absorption of the measured x-rays in a target is also small (less than 4%). Independent measurements of target thickness enabled absolute normalization of the x-rays intensity on the incident number of projectiles obtained from elastically scattered sulphur ions detected in a silicon surface-barrier detector located inside the chamber at  $12.5^\circ$  to the beam direction.

The K x-rays emitted from the moving projectiles were measured in the beam incident side of the target at  $90^\circ$  to the beam direction by a Si(Li) detector ( $30\text{mm}^2$  active area, crystal thickness 5 mm and energy resolution of FWHM=170 eV for 6.4 keV) placed outside the target chamber. Projectile x-rays passed in their way to the detector through 10  $\mu\text{m}$  metallised Mylar chamber window, 25  $\mu\text{m}$  thick beryllium detector window and 5 mm air gap between both windows. Since the registered K x-rays of the sulphur projectile were attenuated due to transmission through these various absorbers the used x-ray spectrometer was carefully calibrated to obtain its detection efficiency. The calibration was performed in x-ray energy range of 1.5-120 keV using standard calibrating sources of  $^{57}\text{Co}$ ,  $^{133}\text{Ba}$ ,  $^{152}\text{Eu}$ ,  $^{241}\text{Am}$  and by PIXE measurements of x-rays from thin calibrating targets ( $Z_t=13-42$ ) according to the recipe of Pajek *et al.* [42]. For the relatively low energy region in this work (2-5 keV) the detector efficiency was determined within uncertainty less than 4%. The energetic calibration of the spectrometer (a second sensitive parameter at the present study) was checked several times during experimental runs by measurements of the x-rays emitted from standard radioactive sources ( $^{57}\text{Co}$ ,  $^{133}\text{Eu}$ ,  $^{152}\text{Ba}$  and  $^{241}\text{Am}$ ) and was determined with an uncertainty of 2-3 eV depending on the experimental run.

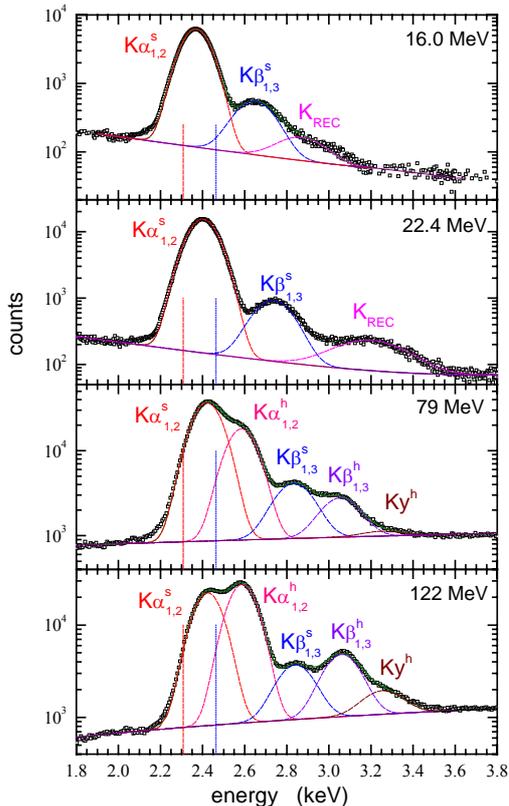


Fig. 2. X-ray spectra emitted by sulphur ions at energies 16.0, 22.4, 79 and 122 MeV passing through the carbon target.

## Spectra analysis procedure

Typical x-ray spectra recorded by Si(Li) detector for sulphur ions passing with energies of 16, 22.4, 79 and 122 MeV through a carbon target are presented in Fig. 2. The origin of all recorded peaks is described in detail in our previous paper [30]. The resolved  $K\alpha_{1,2}^s$ ,  $K\beta_{1,3}^s$  satellite and  $K\alpha_{1,2}^h$ ,  $K\beta_{1,3}^h$  hypersatellite peaks are the results of the overlapped contributions corresponding to transitions of the following types:  $1s^{-1} \rightarrow 2p^{-1}$ ,  $1s^{-1} \rightarrow 3p^{-1}$ ,  $1s^{-2} \rightarrow 1s^{-1}2p^{-1}$  and  $1s^{-2} \rightarrow 1s^{-1}3p^{-1}$ , respectively, from highly ionized sulphur projectiles. For sulphur ions with incident energies of 79-122 MeV an

additional highest-energy peak in the measured x-ray spectra labelled as  $K\gamma^h$  (see Fig. 2) has been detected. This peak corresponds to the hypersatellite transitions from the 4p and 5p subshells (i.e. the transitions of the following types:  $1s^2 \rightarrow 1s^1 4p^{-1}$  and  $1s^2 \rightarrow 1s^1 5p^{-1}$ ) and proves that the  $K\gamma^h$  satellite transitions (i.e. the transitions of the following types:  $1s^1 \rightarrow 4p^{-1}$  and  $1s^1 \rightarrow 5p^{-1}$ ) also take place. The lack of a separate  $K\gamma^s$  satellite peak in the observed spectra indicates [30] that the contribution of this type of transition must overlap with another peak. The  $K\gamma^s$  peak intensity has been calculated from  $K\beta_{1,3}^h$  one according to the procedure described by Majewska *et al.* [30].

In contrast to the  $K\alpha_{1,2}^s$  and  $K\beta_{1,3}^s$  diagram lines in the x-ray spectrum of the singly ionized sulphur atom, the resolved (see 2)  $K\alpha_{1,2}^s$ ,  $K\beta_{1,3}^s$  satellite and  $K\alpha_{1,2}^h$ ,  $K\beta_{1,3}^h$ ,  $K\gamma^h$  hypersatellite peaks in the x-ray spectrum emitted by multiply ionized sulphur projectiles are broadened and shifted towards higher energies (the widths and energy shifts are characteristic for individual peaks). In this work all x-ray peaks recorded by Si(Li) detector were formed, in fact, by a convolution of a wide (150 eV) Gaussian response function of the semiconductor detector with the natural structure of the satellite or hypersatellite lines in the K x-ray spectrum, having typical energy spacing in the range of tens of eV. In our previous work by Banaś *et al.* [27] we have shown that assuming the binomial character of the intensity distribution of x-ray satellites and taking into account their natural widths and their Gaussian energy spread in the semiconductor detector, the measured x-ray peaks appear as the Gaussian profile which is shifted and broadened with respect to the diagram line. Moreover, we have demonstrated [27] that the energy shift and width of each measured x-ray peak can be expressed in terms of the multiple ionization probabilities and the energy shift per electron vacancy. In the present study we have adopted this method for analysis of the measured K x-ray spectra of multiply ionized sulphur projectiles passing through a carbon foil. The energies and intensities of resolved x-ray peaks were determined from a least-square analysis of the spectra using four or five (see Fig. 2) fitting Gaussian functions (with the characteristic width of Gaussian function for each peak) and a polynomial form of the background. The energy shift and width of each x-ray peak reflect the electronic configurations of highly ionized sulphur projectiles at the time of x-ray emission and the results of such studies have been published in our previous papers [30,43].

The measured energies of all of the x-rays emitted from the moving ions were corrected for the Doppler effect resulting in transformation of registered energies into the projectile rest frame. The stopping power of sulphur ions in the target was taken into account by using effective beam energy. The measured intensities of the x-ray peaks were corrected for the detector efficiency and for self-absorption of x-rays in the target.

## References

- [1] P. H. Mokler and F. Folkmann, in: I. A. Sellin (Ed.), *Structure and Collisions of Ions and Atoms*, Springer Verlag, Berlin, p. 201 (1978).
- [2] J. McWherter, J. Bolger, C. F. Moore, and P. Richard, *Z. Phys.* **263**, 283 (1973).
- [3] R. L. Kaufman, J. H. McGuire, P. Richard, and C. F. More, *Phys. Rev.* **A8**, 1233 (1973).
- [4] R. L. Watson, F. E. Jenson, and T. Chiao, *Phys. Rev.* **A10**, 1230 (1974).
- [5] P. Rymuza, Z. Sujkowski, M. Carlen, J.-Cl. Dousse, M. Gasser, J. Kern, B. Penry, and Ch. Rhome, *Z. Phys.* **D14**, 37 (1989).
- [6] H. D. Betz, F. Bell, H. Panke, G. Kalkoffen, M. Welz, and D. Evers, *Phys. Rev. Lett.* **33**, 807 (1974).
- [7] F. Hopkins, *Phys. Rev. Lett.* **35**, 270 (1975).
- [8] K. O. Groeneveld, B. Kolb, J. Schader, and K. D. Sevier, *Z. Phys.* **A277**, 13 (1976).

- [9] T. J. Gray, P. Richard, K. A. Jamison, and J. M. Hall, *Phys. Rev.* **A14**, 1333 (1976).
- [10] F. Hopkins, J. Sokolov, and A. Little, *Phys. Rev. A* **15**, 588 (1977).
- [11] R. K. Gardner, T. J. Gray, P. Richard, C. Schmiedekamp, K. A. Jamison, and J. M. Hall, *Phys. Rev.* **A15**, 2202 (1977).
- [12] J. A. Tanis, W. W. Jacobs, and S. M. Shafroth, *Phys. Rev.* **A22**, 483 (1980).
- [13] B. B. Dhal, H. C. Padhi, K. G. Prasad, P. N. Tandon, and M. Polasik, *J. Phys. B:* **31**, 1225 (1998).
- [14] P. H. Mokler, *Phys. Rev. Lett.* **26**, 811 (1971).
- [15] C. W. Woods, F. Hopkins, R. L. Kauffman, D. O. Elliott, K. A. Jamison, and P. Richard, *Phys. Rev. Lett.* **31**, 1 (1973).
- [16] R. L. Watson, J. R. White, and F. E. Jenson, *Phys. Lett.* **A67**, 269 (1978).
- [17] R. L. Watson, J. R. White, A. Langenberg, R. A. Kenefick, and C. C. Bahr, *Phys. Rev.* **A22**, 582 (1980).
- [18] Y. Awaya, T. Kambara, M. Kase, H. Shibata, H. Kumagai, K. Fujima, J. Urakawa, T. Matsuo, and J. Takahashi, *Nucl. Instrum. And Methods Phys. Res. B* **10/11**, 53 (1985).
- [19] K. Shima, K. Umetani, and T. Mikumo, *Nucl. Instrum. and Methods* **194**, 353 (1982).
- [20] H. J. Hay, L. F. Pender, and P. B. Treacy, *Aust. J. Phys.* **34**, 155 (1981).
- [21] H. J. Hay, L. F. Pender, and P. B. Treacy, *Nucl. Instrum. And Methods* **194**, 349 (1982).
- [22] Y. Zou, Y. Awaya, C. P. Bhalla, T. Kambara, Y. Kanai, M. Oura, Y. Nakai, K. Ando, A. Hitachi, and S. Kravis. *Phys. Rev. A* **51**, 3790 (1995).
- [23] T. Mizogawa, Y. Awaya, T. Kambara, Y. Kanai, M. Kase, H. Kumagai, P. H. Mokler, and K. Shima, *Phys. Rev. A* **42**, 1275 (1990).
- [24] H. F. Beyer, H. J. Kluge, and V. P. Shevelko, *X-Ray Radiation of Highly Charged Ions*, Springer Series on Atoms and Plasmas, Springer-Verlag, Berlin Heidelberg, New York (1997).
- [25] M. Pajek, D. Banaś, J. Braziewicz, U. Majewska, J. Semaniak, T. Czyżewski, M. Jaskóła, W. Kretschmer, T. Mukoyama, D. Trautmann, and G. Lapicki, *AIP Conf. Proc.* **475**, 32 (1999).
- [26] D. Banaś, M. Pajek, J. Semaniak, J. Braziewicz, A. Kubala-Kukuś, U. Majewska, T. Czyżewski, M. Jaskóła, W. Kretschmer, T. Mukoyama, and D. Trautmann, *Nucl. Instrum. and Methods Phys. Res. B* **195**, 233 (2002).
- [27] D. Banaś, J. Braziewicz, U. Majewska, M. Pajek, J. Semaniak, T. Czyżewski, M. Jaskóła, W. Kretschmer, and T. Mukoyama, *Nucl. Instrum. and Methods Phys. Res. B* **154**, 247 (1999).
- [28] D. Banaś, J. Braziewicz, A. Kubala-Kukuś, U. Majewska, M. Pajek, J. Semaniak, T. Czyżewski, M. Jaskóła, W. Kretschmer, and T. Mukoyama, *Nucl. Instrum. and Methods Phys. Res. B* **164-165**, 344 (2000).
- [29] D. Banaś, J. Braziewicz, U. Majewska, M. Pajek, J. Semaniak, T. Czyżewski, M. Jaskóła, W. Kretschmer, T. Mukoyama, and D. Trautmann, *J. Phys. B* **33**, L793 (2000).
- [30] U. Majewska, K. Słabkowska, M. Polasik, J. Braziewicz, D. Banaś, T. Czyżewski, I. Fijał, M. Jaskóła, A. Korman, and S. Chojnacki, *J. Phys. B* **35**, 1941 (2002).
- [31] R. L. Watson, J. M. Blackadar, and V. Horvat, *Phys. Rev. A* **60**, 2959 (1999).
- [32] R. L. Watson, V. Horvat, J. M. Blackadar, and K. E. Zaharakis, *Phys. Rev. A* **62**, 052709 (2000).
- [33] H. Panke, F. Bell, H. -D. Betz, H. Stehling, E. Spindler, and R. Laubert, *Phys. Lett.* **53A**, 457 (1975).
- [34] H. Panke, F. Bell, H. D. Betz, and H. Stehling, *Nucl. Instrum. and Methods* **132**, 25 (1976).
- [35] J. A. Tanis and S. M. Shafroth, *Phys. Rev Lett.* **40**, 1174 (1978).

- [36] U. Scharfer, C. Henrichs, J. D. Fox, P. Von Brentano, L. Degener, J. C. Sens, and A. Pape, Nucl. Instrum. and Methods **146**, 573 (1977).
- [37] T. J. Gray, C. L. Cocke, and R. K. Gardner, Phys. Rev. A **16**, 1907 (1977).
- [38] C. L. Cocke, S. L. Varghese, and B. Curnutte, Phys. Rev. A **15**, 874 (1977).
- [39] S. K. Allison, Rev. Mod. Phys. **30**, 1137 (1958).
- [40] J. P. Biersack and L. G. Maggmark, Nucl. Instrum. and Methods **174**, 257 (1980).
- [41] J. Ziegler, [http\ www.srim.org](http://www.srim.org)
- [42] M. Pajek, A. P. Kobzev, R. Sandrik, R. A. Ilkhamov, and S. A. Khusmorodov, Nucl. Instrum. and Methods Phys. Res. B **42**, 346 (1989).
- [43] U. Majewska, J. Braziewicz, M. Polasik, K. Słabkowska, I. Fijał, M. Jaskóła, A. Korman, S. Chojnacki, and W. Kretschmer, Nucl. Instrum. and Methods Phys. Res. B **205**, 799 (2003).

## Results

1. A new theoretical model based on the results of the single-configuration DF calculations (and using equilibrium charge state distribution and fluorescence yields) has been proposed in order to estimate the role of various types of electronic configurations and to evaluate the average population of different shells in fast sulphur ions passing through the carbon foils from low resolution K x-ray spectra.

[U. Majewska et al., *Configurations of highly ionized fast sulphur projectiles passing through a carbon foil evaluated from low-resolution K x-ray spectra*. J. Phys. B: At. Mol. Opt. Phys. **35** (2002) 1941].

2. A new interpretation of low resolution K x-ray spectra parameters of highly ionized sulphur projectiles passing through the carbon foils has been proposed using theoretical model based on the single-configuration Dirac-Fock calculations, equilibrium charge state distribution and fluorescence yields for multiply ionized sulphur ions, the population of L and M shells (in the case of 9.6 - 32.0 MeV beam energy), the probability of K-hole creation and the average population of L-shell, 3p and 4p subshells (in the case of 65 - 122 MeV) of sulphur ions have been estimated. Additionally, the lifetimes of discussed highly excited states of sulphur ions have been obtained from the studying of dynamics of formation of K-hole fractions of sulphur projectiles inside a carbon foil.

[U. Majewska, et al., *Highly excited states of sulphur projectiles inside a carbon target*, Nucl. Instrum. and Meth. in Phys. Res. B **205** (2003) 799].

3. The dependence of the satellite and hypersatellite sulphur K x-ray production cross sections on the target thickness has been examined, for the first time separately for each line recorded in spectra of characteristic x-rays of the projectiles at energy range of 65-122 MeV. The three component model, which expresses the probability of an ion charge-changing collisions by the cross sections  $\sigma_{ij}$  was used. For each sulphur projectile energy the values of the K-shell vacancy production cross sections and the K-shell vacancy loss cross sections (independently for the electron capture and for the radiative and Auger decays) have been fitted. The obtained experimental values of all  $\sigma_{ij}$  cross sections have been used later to determine the dynamics of formation of the K-vacancy fractions of the sulphur projectiles passing through a carbon foil.

[J. Braziewicz, et al., *Dynamics of formation of K-hole fractions of sulphur projectiles inside a carbon foil*, Phys. Rev. A **69** (2004) 062705]

4. It has been shown that the energies of K X-ray lines emitted from sulphur projectiles are very sensitive to the degree of ionization of their L and M shells. Therefore probably for the first time, the projectile low-resolution K X-ray spectra have been used for evaluation of a mean equilibrium charge,  $\bar{q}$ , for sulphur ions passing with energies of 9.6, 16.0, 22.4 and 32.0 MeV through Al, Ti and Fe targets.

[J. Braziewicz, et al., *Sulphur ion charge states inside solids from low-resolution K X-ray spectra*, Nucl. Instrum. and Meth. in Phys. Res. B (2004) accepted for publication]