## **α-PARTICLE INDUCED FORWARD-BACKWARD ELECTRON** EMISSION FROM TITANIUM NITRIDE

V.P. Zhurenko 1, S.I. Kononenko 1, O.V. Kalantaryan 1, I.N. Mysiura, S.S. Avotin, N.Ja. Rokhmanov 1 V.N. Karazin Kharkiv National University, Kharkov, Ukraine;

2 Kharkiv National Agrarian University, «Dokuchaievske-2», Kharkov region, Ukraine E-mail: <u>v.zhurenko@gmail.com</u>

The electronic yields of forward and backward secondary ioninduced electron emission from titanium nitride bombarded by aparticles from a radioisotope source were experimentally measured. It was shown that the ratio of forward and backward electronic yields was approximately 1.79, which agrees well with the results obtained earlier for other materials and fast light ions. It was found that this ratio increased slightly with increasing specific ionization loss of the ion PACS: 78.00.00 INTRODUCTION The interaction of energetic ions with the matter leads to the deceleration of the incident particles because of their predominant interaction with electrons, which is commonly called electronic energy losses [1], along with nuclear energy losses, which is substantial at low ion velocity. For high ion velocity nuclear energy losses are negligible. Quantitatively, this process is characterized by the specific ionization losses of the ion in matter dE/dx, sometimes also referred to as the stopping power of a matter for a certain type of ion. A considerable part of the energy lost by the swift ion leads to the excitation and ionization of the atoms of the solid and, as a consequence, to the emission of electrons from the surface, i.e. to ion-induced secondary electron emission (IISEE). This phenomenon was discovered quite a long time ago, and in the literature one can find many reviews on this problem (see, for example [2 - 5]). It is well known that for light fast ions, the coefficient of IISEE is directly proportional to the average specific ionization loss of the ion in matter dE/dx [2, 3, 6]. For protons, this dependence is well satisfied in a wide range of energies of incident

ions of 5 keV  $\leq$  E  $\leq$  24 MeV [4]. However, for heavier ions, there is a slight deviation from simple proportionality, namely, a dependence on the velocity and atomic number of the ion appears in the proportionality coefficient  $\Lambda(Z1, v)$  [7]. Very often, emission experiments are performed with ion beams and thin foils as targets. Then, the emission of electrons from the front (from the side of the beam entrance - backward emission) and from the rear (from the exit side of the beam - forward emission) surfaces of the thin target are observed. Accordingly, the IISEE yields for backward emission  $\gamma B$  and forward emission  $\gamma F$  are distinguished. For the first time, the difference between the IISEE yields yB and yF was drawn to the attention by W. Meckbach, G. Braunstein and N. Arista [8]. Investigating the forward and backward emission from a thin carbon foil bombarded by the proton beam with energy of 25...250 keV, the authors showed that the integral energy spectrum of electrons in the case of forward emission had a greater intensity compared with that of backward emission [8]. The main reason for the differences in the IISEE forward/backward yields is explained by the fact that fast convoy [9] and  $\delta$ -electrons [10] move mainly in the direction of motion of the ion. Since some part of the energy of a fast ion can be transferred to convoy and *delectrons*, anisotropy in the energy losses of the projectile arises [11]. To characterize the anisotropy of the emission, the concept of the anisotropy coefficient, often called as Meckbach ratio, was first introduced in [8]:  $R = \gamma F / \gamma B$ , (1) for protons with energies above 150 keV, this ratio is 1.55 [8]. Since then, the problem of R-anisotropy has been of constant scientific interest [12, 13 -16]. For example, the authors of [12] carried out wide cycle of measurements of the thin carbon film IISEE yields induced by a series of ions  $(1 \le Z \le 32)$  with different ionization degree and with different velocities. The measurements performed from both target surfaces showed a significant difference in the IISEE yields for backward and forward emissions. In particular, for protons it was found that in the energy range of incident particles from 20 keV to 7.5 MeV the anisotropy ratio R varies slightly. However, the R independence on the velocity of the projectile is observed only for protons, and for heavier

particles, the value of the ratio increases significantly with increasing velocity [12, 17]. Experiments with various incident ions made it possible to reveal an increase in the anisotropy effect with an increase in the atomic number of the bombarding particle [17]. For thick targets, the value of the R-ratio varies from 1.2 for protons to 5 for very heavy ions such as, for example, uranium. In our earlier papers [11, 18] we already presented the results for IISEE from a number of metal targets (Al, Cu, Ni and Ti) for both cases: forward and backward emission induced by  $\alpha$ particles from a radioisotope source. Our experiments differed from the mentioned above one by the fact that forward and backward emission were caused by ions with exactly the same energy. It was shown that Rratio for these materials differed insignificantly, by no more than 10% of the mean value. In this paper we carried out similar studies of the IISEE yields for titanium nitride. TiN is widely used material in various industries. For example, TiN films ISSN 1562-6016. BAHT. 2018.  $N_{24}(116)$  294 are used as stable hard coatings [19] in various technological applications and as diffusion barriers in microelectronics [20]. TiN are of great interest because they exhibit a number of properties similar to metals (electrical conductivity, superconductivity etc.), while retaining characteristics found in insulating materials. This material has been well studied, for example, using Auger electron spectroscopy [21], X-ray photoelectron spectroscopy [22], UV photoelectron spectroscopy [23], electron energy loss spectroscopy [24], and several others. However, there are no data on TiN IISEE in the literature, in particular the IISEE yields for forward and backward cases. 1. EXPERIMENTAL SETUP Experiments on IISEE study with  $\alpha$ particle radioisotope source were done on the experimental setup, which described in detail in [11]. A source of  $\alpha$ - particles, a target and a massive collector were placed in the vacuum chamber. The flux of primary particles was irradiated by a radioisotope source with isotope Pu239 (initial flow intensity 4.64×106 particles per second). aparticles with energy of 5.15 MeV were isotropically radiated into the solid angle  $2\pi$ . The working part of the source had a diameter of 37 mm. The target was an aluminum thin foil with TiN layer. It was mounted on a metal

round holder. The diameter of the target was 50 mm. The foil was selected in such a way that its thickness was less than the mean free path of the  $\alpha$ -particles emitted by this radioisotope in the target material. The radioisotope source and the metal holder of the target were in the electrical contact. They were installed on a teflon substrate, which provided their electrical isolation and made it possible to apply potential to the target. In this case, the planes of the target and the active part of the source were parallel, and the distance between them was less than 1 mm. Fig. 1. Scheme of experiments The flux of  $\alpha$ -particles, passing through the target, caused kinetic IISEE from its outer surface (forward emission) (Fig. 1). Then  $\alpha$ -particles hit the massive collector and induced electron emission from its face surface (backward emission). To provide collection of all emitted electrons both in the case of forward emission from the target and backward emission from the collector, we applied an electric field in the space between target and collector. We applied a potential of amplitude from -300 to +300 V to the target using a constantcurrent source of the Keithley-6487 type. We measured the current of  $\alpha$ -particles and emission electrons in the collector electrical circuit. The value of the registered currents was of order of 10-13...10-12 A. Therefore, the measuring electric circuit of the collector had to satisfy the requirements imposed on the electrometric circuits. We used a Keithley-6487 picoammeter with a sensitivity of 10-14 A for precise current measurements. To adjust the measuring system, a movable flap between the source and the target was installed. It permitted to overlap the flow of  $\alpha$ -particles. The scan of the current-voltage characteristics was done in an automatic mode with PC control and data accumulation. The current-voltage characteristics measured in this way allowed to calculate IISEE yield  $\gamma$  from the saturation current, i.e., from the maximum current corresponding to the total collection of emission electrons. Taking into account that we performed current measurement in the experiment, it is possible to calculate  $\gamma$  by the expression:  $\gamma = 2$ (Ie)/ I $\alpha$ , where Ie is the corresponding saturation emission current, I $\alpha$  is the current of  $\alpha$ -particles that passed through the target and caused the emission; the coefficient 2 characterizes the fact that the charge of the  $\alpha$ - particle is twice the electron charge. All experiments were carried out in a vacuum chamber at a residual gas pressure no worse than  $2 \times 10-4$  Па. Thin layers of titanium nitride (thickness of the order of 0.5  $\mu$ m) were deposited on the outer surface of the target (Al foil) and on the collector face surface (the side of  $\alpha$ -particle incidence). A vacuum ion plasma technology was used to deposit titanium on the samples in a nitrogen atmosphere. 2. RESULTS AND DISCUSSION To simulate the passage of  $\alpha$ -particles through our target, we used the SRIM-TRIM software (Fig. 2) [25]. Fig. 2. Ion trajectories in the Al-TiN target for incidence angle of 0°(SRIM/TRIM simulation) As we have already mentioned, in the absence of the target, the particles emitted by the  $\alpha$ -source emitted isotropically into the solid angle  $2\pi$ . The solid angle into which the  $\alpha$ particles moved passing through the target is determined by the thickness of the foil. Indeed, at large exit angles with respect to the normal to the surface, the projectile path in a target foil becomes compa-ISSN 1562-6016. BAHT. 2018. №4(116) 295 rable to the range. As a result, if a certain angle is exceeded, the  $\alpha$ -particles cannot leave the target. Thus, initially the monochromatic flow of  $\alpha$ -particles, passing through the target, would be distributed by energy from 0 to Emax (Fig. 3). The maximum energy would correspond to the particles with incidence angle of 0°. In our case, Emax was approximately 4.02 MeV (~ 1 MeV/amu). 0 10 20 30 40 50 60 70 80 0 1000 2000 3000 4000 5000 E, KeV  $\Theta$ , deg Fig. 3. Dependence of  $\alpha$ -particle energy passed through the target (calculated by SRIM) on incident angle We measured current-voltage characteristics for target-collector system, calculated yF and yB for TiN layers 16.94 and 9.48, respectively. So, in our case Rratio was approximately 1.79. It's worth paying attention to the experimental results of A. Clouvas and coworkers [12]. They studied forward-backward emission induced by large number of ions from thin carbon foil. Among others they used Li2+ ions with energies  $0.86 \le E \le$ 1.15 MeV/amu. In spite of the fact that measured  $\gamma F$  and  $\gamma B$  from both sides of a thin foil, R-ratio obtained in these experiments were approximately equal to 1.73. The authors of [13] pointed out that for 1.2 MeV He ions and carbon targets the Meckbach factor R were

approximately 1.6. As it can be seen, the above mentioned results for Rratio are in agreement with our results. In the Table we present the results of our previous studies for Al, Cu, Ni [11], Ti [18] and H. Rothard's He ion data [13] for comparison reasons. Material Al Cu Ni Ti TiN C R-ratio 1.57 1.69 1.82 1.61 1.79 1.6 The energy that goes to IISEE is delivered by a fast ion. Since it is now theoretically and experimentally found that IISEE yield is directly proportional to the ionization losses of the fast light ion, it would be reasonable to assume that the Mekbach factor also depends on the losses. We analyzed R-ratio data depending on electronic stopping power for He ions and different targets shown in the table (Fig. 4). We found that R-ratio slightly depend on electronic stopping power for the materials under study. Since this anisotropy of the electronic yields is associated with delta and convoy electrons, it is expected that the specific fraction of fast electrons increases with increasing energy loss by the ion in the substance. 15 20 25 30 35 40 45 50 1.4 1.5 1.6 1.7 1.8 1.9 2.0 C TiN Ti Ni Cu Al R-ratio dE/dx, eV/A Fig. 4. Dependence of R-ratio on electronic stopping power for He ions and different targets CONCLUSIONS An experimental study of forward-backward anisotropy of IISEE for titanium nitride was carried out. The results obtained make it possible to conclude that the generation of convoy and delta electrons increases with the increase in the specific energy losses of fast ions. In addition, R-ratio (the Mekbach factor) for conductive substances (metals, carbon) and the substance that exhibits the properties of both a conductor and a dielectric (titanium nitride) differ insignificantly. REFERENCES 1. N.P. Kalashnikov, V.S. Remizovich, and M.I. Ryazanov. Collisions of Fast Charged Particles in Solids. M.: "Atomizdat", 1980 (in Russian). 2. E.J. Sternglass. Theory of secondary electron emission by high-speed ions // Phys. Rev. 1957, v. 108, № 1, p. 1-12. 3. J. Schou. Transport theory for kinetic emission of secondary electrons from solids // Phys. Rev. B. 1980, v. 22, p. 2141-2173. 4. D. Hasselkamp. Secondary emission of electrons by ion impact on surfaces // Comments At. Mol. Phys. 1988, v. 21, p. 241-255. 5. H. Rothard, K. Kroneberger, A. Clouvas, et al. Secondary-electron yields from thin foils: A possible probe for the electronic stopping power of heavy ions // Phys. Rev. A. 1990, v. 41, p. 2521-2536. 6. A. Clouvas, A. Katsanos, B. Farison-Mazuy, et al. Projectile dependence of ion-induced electron emission from thin carbon foils // Phys. Rev. B. 1993, v. 48, p. 6832-6838. 7. H. Rothard, M. Jung, J.-P. Grandin, et al. Heavy ion track potentials in solids probed by electron yield measurements // Nucl. Instr. and Meth. B. 1997, v. 125, p. 35-40. 8. W. Meckbach, G. Braunstein, N. Arista. Secondaryelectron emission in the backward and forward directions from thin carbon foils traversed by 25...250 keV proton beams // J. Phys. B. 1975, v. 8, № 14, p. L344-L349. 9. R. Latz, J. Schader, H.J. Frischkorn, et al. The relation between convoy electron velocity and ion velocity of ions penetrating solids // Nucl. Instr. and Meth. B. 1984, v. 2, p. 245-247. ISSN 1562-6016. BAHT. 2018. №4(116) 296 10. H.Rothard. Electron ejection from solids by heavy ions at high energies (1...100 MeV/ u) // Nucl. Instr. and Meth. B. 1998, v. 146, p. 1-10. 11. V.P. Zhurenko, S.I. Kononenko, V.I. Karas', and V.I. Muratov. Dissipation of the Energy of a Fast Charged Particle in a Solid-State Plasma // Plasma Physics Reports. 2003, v. 29, № 2, p. 130-136. 12. A. Clouvas, C. Potiriadis, H. Rothard, et al. Role of projectile electrons in secondary electron emission from solid surfaces under fastion bombardment // Phys. Rev. B. 1997, v. 55, p. 12086-12098. 13. H. Rothard, K. Kroneberger, A. Clouvas, et al. Secondary-electron yields from thin foils: A possible probe for the electronic stopping power of heavy ions // Phys. Rev. A. 1990, v. 41, p. 2521-2536. 14. H. Rothard. Electron ejection from solids by heavy ions at high energies (1...100 MeV/ u) // Nucl. Instr. and Meth. B. 1998, v. 146, p. 1-10. 15. H. Rothard, J. Schou, K.O. Groeneveld. Projectileand charge- statedependent electron yields from ion penetration of solids as a probe of preequilibrium stopping power // Phys. Rev. A. 1992, v. 45, p. 1701-1710. 16. Y. Sato, A. Higashi, D. Ohsawa, et al. Projectile charge dependence of electron emission from foils // Phys. Rev. A. 2000, v. 61, p. 1-16. 17. H. Rothard, M. Jung, M. Caron, et al. Strong projectiledependent forward-backward asymmetry of electron ejection by swift heavy ions in solids // Phys. Rev. A. 1998, v. 57, p. 3660-3664. 18. S.I. Kononenko, V.P. Zhurenko, O.V. Kalantaryan, A.A. Semerenskiy.

Forward and backward electron emission in binary cell of radioisotope current source // Problems of Atomic Science and Technology. Series "Plasma Electronics and New Methods of Acceleration". 2015, № 4, p. 331-334. 19. H.G. Tompkins // J. Appl. Phys. 1991, v. 70, p. 3876. 20. M. Eizenberg // MRS Bull. 1995, v. 20, p. 38. 21. P.T. Dawson, K.K. Tzatsov // Surf. Sci. 1985, v. 149, p. 105. 22.J. Card, A.L. Testoni. UA. Le Tarte // Surf. Interface Anal. 1995, v. 23, p. 495. 23. H. Hochst, R.D. Bringans, P. Steiner. Th. Wolf // Phys. Rev. B. 1982, v. 25, p. 7183. 24. P. Prieto, R.E. Kirby // J. Vac. Sci. Technol. A. 1995, v. 13, p. 2819. 25.J.F. Ziegler, J.P. Biersack, U. Littmark. SRIM – The stopping and range of ions in matter // Nucl. Instrum. Methods Phys. Res. 2010, B268 p. 1818-1823. http://dx. doi.org/10.1016/j.nimb.2010.02.091 Article received 06.06.2018

## ЭЛЕКТРОННАЯ ЭМИССИЯ НА ПРОСТРЕЛ И НА ОТРАЖЕНИЕ, ИНДУЦИРОВАННАЯ α-ЧАСТИЦАМИ ИЗ НИТРИДА ТИТАНА

В.П. Журенко, С.И. Кононенко, О.В. Калантарьян, И.Н. Мисюра, С.С. Авотин, Н.Я. Рохманов

Экспериментально измерены электронные выходы вторичной ионно-электронной эмиссии из нитрида титана на прострел и на отражение при бомбардировке α-частицами от радиоизотопного источника. Показано, что отношение электронных выходов на прострел и на отражение составляет примерно 1,79, что хорошо согласуется с результатами, полученными ранее для других материалов и быстрых легких ионов. Найдено, что указанное отношение слегка возрастает с увеличением удельных ионизационных потерь иона.

## ЕЛЕКТРОННА ЕМІСІЯ НА ПРОСТРІЛ І НА ВІДБИТТЯ, ІНДУКОВАНА α-ЧАСТИНКАМИ З НІТРИДУ ТИТАНУ

В.Р. Журенко, С.І. Кононенко, О.В. Калантар'ян, І.М. Мисюра, С.С. Авотін, М.Я. Рохманов Експериментально виміряно електронні виходи вторинної іонно-електронної емісії з нітриду титану на простріл і на відбиття під час бомбардування α-частинками від радіоізотопного джерела. Показано, що відношення електронних виходів на простріл і на відбиття становить приблизно 1,79, що добре узгоджується з результатами, отриманими раніше для інших матеріалів і швидких легких іонів. Знайдено, що зазначене відношення помірно зростає зі збільшенням питомих іонізаційних втрат іона.