SOVIET PHYSICS

JETP

A translation of the Journal of Experimental and Theoretical Physics of the USSR.

SOVIET PHYSICS—JETP VOL. 5, NO. 2, PP 157-343 SEPTEMBER, 1957

Emission of Negative Ions from Metallic Surfaces Bombarded with Positive Hydrogen Ions

I. M. MITROPAN AND V. S. GUMENIUK

Physico-Technical Institute, Academy of Sciences, Ukrainian SSR

(Submitted to JETP editor July 23, 1956)

J. Exptl. Theoret. Phys. (U.S.S.R.) 32, 214–222 (February, 1957)

Results are presented of an experimental study of secondary negative ion emission from aluminum, stainless steel and copper targets bombarded by 200–1000 kev hydrogen and deuterium ions.

INTRODUCTION

WHEN metallic surfaces were bombarded by positive ions secondary emission of negative ions was observed together with secondary emission of electrons.[1–4] The experiments of Sloane and Love[3] show that the negative ions are partially due to the primary positive ions being converted into negative ones at the surface of the target.

The ejection of negative ions whose nature is different from that of the primary ones was observed in the work of Arnot[5] and of Veksler and Shuppe.[2] Arnot explained the appearance of the true secondary ions as a result of the excitation by the incident ion of the atoms adsorbed on the target. These ions, in giving up their excitation energy to the metal, capture an electron. Veksler and Shuppe consider that decomposition of electronegative impurities takes place at the surface of the target cathode. However, it remains unclear as to which of these processes is the dominant one when the elements bombarded by ions are those whose atoms have an electron affinity.

The object of the present work was to determine the coefficient of emission of secondary negative ions as a function of the energy of the primary hydrogen ions from metals ordinarily used in laboratory practice for the construction of high-voltage accelerating tubes. In addition, a mass spectrometric analysis of the resulting negative ions was carried out, and the influences of outgassing the target on the value of the coefficient of ejection K^- was studied.

APPARATUS AND EXPERIMENTAL PROCEDURE

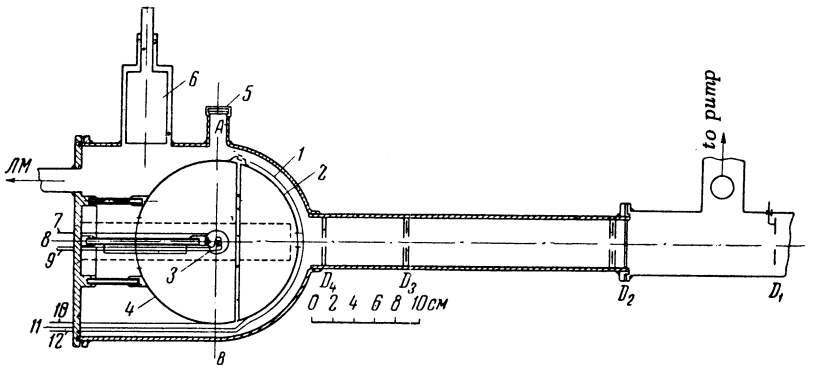
An electrostatic generator was used for the production of accelerated hydrogen ions, with a mass analyzer (magnetic prism deflecting the beam through 90°) placed at the exit. The beam of ions selected by the magnetic analyzer was collimated by means of a number of diaphragms situated between the valve and the experimental setup shown schematically in Fig. 1).

The diaphragm D_1 (Fig. 1) with an opening 3 mm in diameter had a lead which was connected to a microammeter and which served to record the current. The next two diaphragms with openings 2 mm in diameter defined a beam whose cross-section at the target amounted to 2.5–3 mm.[2] The diaphragm D_4 with an opening 4 mm in diameter was insulated from the body of the apparatus, and was placed at a potential of −90 V with respect to ground in order to suppress secondary electrons and ions scattered by the collimating diaphragms.

The pressure in the high vacuum part of the apparatus produced by oil diffusion pumps and liquid nitrogen traps was $\sim 10^{-6}$ mm of Hg. In this investigation, H_1^+, H_2^+, D_2^+ and D_1^+ ions were used.

At first measurements were carried out on H_1^+ and D_2^+ ions, which allowed us to eliminate impurities of D_1^+ and H_2^+ as a result of the contamination of the source by deuterium. The contamination of H_2^+ in the work with D_1^+ was small as could be indirectly inferred from the shape of the curve $K^- = f(E_{kin})$, and introduced an error into the measured value of the coefficient of emission which was within experimental error.

The main problem arising in the study of secondary negative ion emission is the separation of the electron and the ion components of secondary emission. This separation was achieved by subjecting the target-collector system, which was in the form of a spherical condenser, to a constant magnetic field. It may be shown that electrons originating on the surface of the inner sphere, with energies of several thousand ev and negative



cross-section of the vacuum chamber along the line A-B

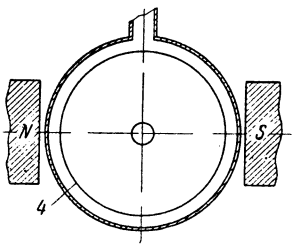


FIG. 1.

hydrogen ions of energies of a few ev may be separated in the magnetic field chosen for this work. Calculations show that the region in which the charged particles can move is bounded by a cylindrical surface whose axis coincides with the direction of the magnetic field. The radius of the cylindrical surface is given by

$$\rho = (v_\varphi / 2\omega) + \sqrt{(v_\varphi / 2\omega)^2 + 1}, \qquad (1)$$

where v_φ is the azimuthal component of the velocity, and $\omega = eH/mc$ (e and m are the charge and the mass of the particle, H is the intensity of the magnetic field, and c is the speed of light). For H = 100 oersted $\omega = 1.759 \times 10^9$ rad/sec for the electron and $\omega = 0.96 \times 10^6$ rad/sec for the negative hydrogen ion. If we assume that the electrons have a kinetic energy $E_{kin} \leq 3000$ ev, then the radius of the cylindrical surface which limits the region in which the electrons can move is $\rho_e \leq 1.7$ cm. For negative hydrogen ions of kinetic energy $E_{kin} \geq 10$ ev the radius of the cylindrical surface is given by $\rho \geq 5.75$ cm.

On the basis of the above calculations, we selected such parameters for our apparatus which allowed us definitely to separate secondary negative ions of energy $E \geq 10$ ev from secondary electrons of energy $E_{kin} < 3000$ ev. The spherical condenser used in our measurements consisted of an inner sphere *3* (Fig. 1) 20 mm in diameter, and an outer sphere R of diameter 160 mm. The inner sphere served as the target and was mounted on a porcelain insulator. A tungsten spiral was placed inside the inner sphere, was isulated from the target and was provided with insulated leads. By means of this spiral the target was heated to 1000° C during operation. The spherical segment *2* cut off

by the plane passing perpendicular to the incident beam at a distance of 10 mm from the surface of the target served as the collector for the negative ions. The spherical segment was insulated from the remainder of the sphere. The target-collector system was situated inside a cylindrical copper chamber outside of which was placed an electromagnet with plane pole tips 100 mm in diameter The spherical segment was screened on the outside by a hemisphere of aluminum foil 1. The hydrogen ion beam, after passing through the collimator passed through an opening 10 mm in diameter in the screening hemisphere, and then through an opening 11 mm in diameter in the spherical segment, after which it reached the target.

Such a choice of the apparatus geometry allowed us to prevent the arrival at the collector of scattered particles and of particles ejected from the diaphragms, which was experimentally checked by placing the target inside a screening cylinder (shown by dotted lines in Fig. 1) with a 10 mm diameter opening in its end and situated coaxially with the direction of the beam. By sending the beam through the apparatus we established that no current is observed from the collector to ground.

The targets utilized in this work made of copper, aluminum and stainless steel of the EI–1 type were in the form of polished spherical surfaces. Before being placed in the apparatus, the target was washed with acetone and was then conditioned at a temperature of 900° C in a vacuum for 20–30 min with a subsequent cooling of the target for 1.5–2 hours before each measurement. The carbon target was obtained by depositing a thick layer of aquadag on a spherical copper base.

Because of fluctuations in intensity of the ion beam we used for the measurement both of the primary and the secondary ion current the method of accumulating charge on electrical capacitances during a time much smaller than the time constant for the discharge of these capacitances. The electrostatic field in the spherical condenser was set up by applying a potential to the target. The whole electrical supply system was insulated from ground. Both parts of the outer sphere were maintained at the same (zero) potential. Therefore the field of the spherical condenser was practically not distorted at all by the separation of the sphere into two parts insulated from each other.

The quantity of charge carried by the negative ions to the spherical segment (when the target was bombarded by primary ions) was measured by means of the electrometer *1*. The quantity of charge originating at the target and the quantity of charge carried by the secondary electrons to the capacitances part of the outer sphere were algebraically added by the circuit and were measured by means of the electrometer *2*.

The integrated coefficient of emission of negative ions was defined by the relation

$$K^- = q_1 / (q_2 - q_1), \qquad (2)$$

where q_1 and q_2 are the charges accumulated during the time of measurement in systems containing electrometers *1* and *2* respectively. The magnetic field set up by the electromagnet could be varied from 0 to 300 oersted and was practically homogeneous inside a cylinder 4 cm in diameter.

In order to determine the nature of the negative ions, a simplified mass-analyzer was used; it is schematically shown in Fig. 2. The plane surface of the target was placed at an angle of 45° to the direction of the beam and at an angle of 45° to the direction of the mass-analyzer axis. The target was cut out of copper foil of 0.1 mm thickness and was of oval shape. A tungsten spiral was mounted at the rear of the target in order to heat the target prior to measurement.

The ions emitted by the copper target bombarded by hydrogen ions were focussed by an electrostatic three-electrode lens. The focussing properties of the lens were checked by using electrons. The current focussed by the lens amounted to $\sim$ 2% of the total emission current of a tungsten cathode used in place of the target. The magnetic field of the mass-analyzer was limited to a sector with 60° apex angle. The distance between the pole tips was 20 mm, and the radius of curvature of the ion trajectory was 100 mm.

A Faraday cup with an entrance opening 6 mm in diameter served as the collector of negative ions. The Faraday cup was screened on the outside by a cylinder with an opening 5mm in diameter insulated from the detecting cylinder. The ions originating at the target were accelerated by a potential difference of 650 V applied between the target and the grounded first electrode of the lens. A preliminary calibration of the mass analyzer was carried out using the positive ions Na^+, K^+, Li^+, which were emitted by heating a target covered by a layer consisting of a mixture of the aluminum silicates of Na, K, Li.

The scale of the mass-analyzer was calibrated by means of these ions. The investigation of the negative ion spectrum was carried out by smoothly varying the intensity of the magnetic field. The total ion current to the target was measured by a string electrometer which was connected to a 50 μF capacitance. A second string electrometer with a capacitance of 1000 pF in parallel was connected to to the Faraday cup. The measurements were made

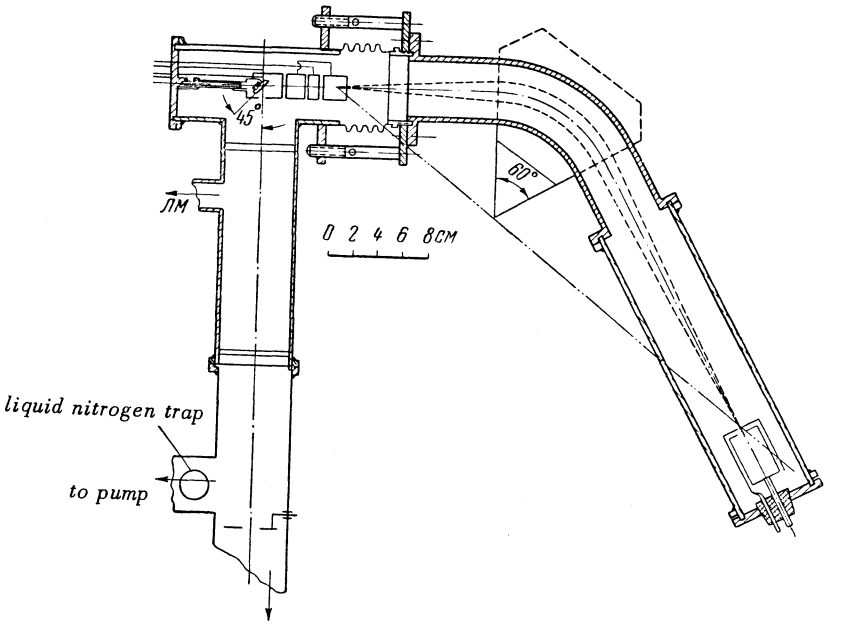


FIG. 2.

by the method described above. The coefficients for the emission of negative ions K^- were determined for potential differences of –100 and –400 V applied to the spherical condenser and with the intensity of the magnetic field varying from 100 to 300 oersted.

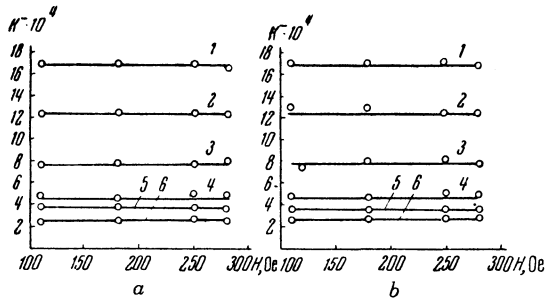


FIG. 3. Dependence of the coefficient of ejection of negative ions K^- on the intensity of the magnetic field frr various values of potential difference applied to the spherical condenser: *a*–200, *b*–400 V for the following energies of the H^+ ion: *1*–125, *2*–205, *3*–300, *4*–500, *5*–600, *6*–700 kev.

Figures 3*a* and *b* show the dependence of the coefficient K^- on the intensity of the magnetic field for a stainless steel target bombarded by protons of various energies. As may be seen from those Figures, the value of K^- for target potentials ranging from –200 to –400 V and for magnetic field intensity lying between 100 and 300 oersted, does not depend on the potential of the target and on the intensity of the magnetic field.

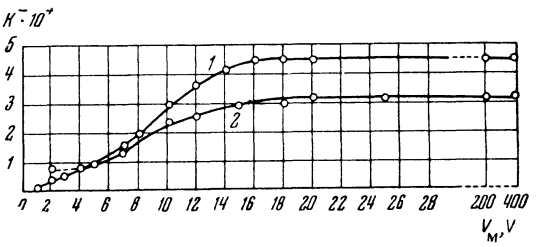


FIG. 4.

Measurements of secondary ion current were also carried out for lower values of target potential. Typical curves showing the results of such measurements for an aluminum target for a primary H_1^+ ion beam energy equal to 550 kev (curve *1*), 660 kev (curve *2*) and for a magnetic field intensity equal to 110 oersted are shown in Fig. 4.

The decrease in the coefficient K^- as the negative target potential is varied between 20 and 0V is explained by the presence of secondary positive ions of energy lying between 0 and 20 ev. By applying a positive potential to the target which retards the negative ions, we obtain the characteristic curves shown in Fig. 5. From these curves it may be seen that the energy of the negative ions does not exceed 10 ev. However, it is not possible to obtain

from these curves the energy distribution of the ions at low energies since it may be seen from expression (1) that hydrogen ions of energies 2–3 ev may be partially held up by the magnetic field (H = 110 oersted). From the curves which show the manner in which the negative ions are retarded by the electric field (Fig. 5) we see that for the targets which we have investigated positive emission at energies higher than 150 kev is of the same order of magnitude as the emission of negative ions. The value of K^- which characterizes a given target for normal incidence of the beam at a given energy of bombarding ions was determined as the arithmetic mean of 8–10 measurements. The relative probable error amounts to ±3.7%.

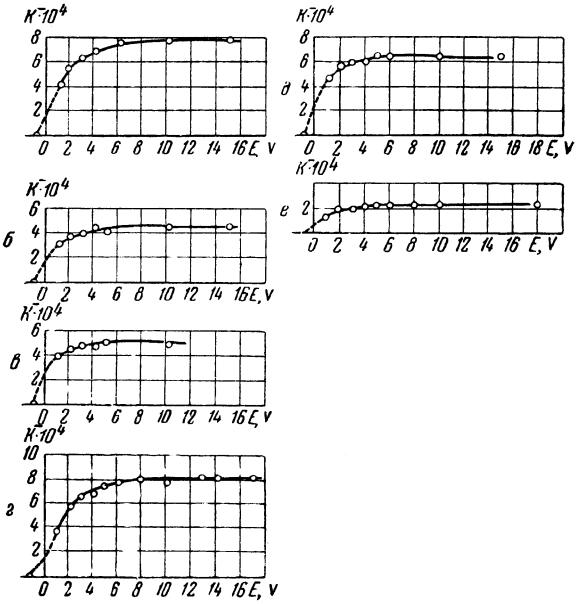


FIG. 5. Curves showing the retardation of negative ions for a magnetic field intensity of 110 oersted. *a*—aluminum target bombarded by H_1^+ ions of energy 175 kev; *b*—the same at an energy of 550 kev; *c*—the same at an energy of 660 kev; *d*—aluminum target bombarded by D^+ ions of energy 500 kev; *e*—stainless steel target bombarded by H^+ ions of energy 500 kev; *f*—carbon target bombarded by H^+ ions of energy 500 kev.

We have made a comparison of the coefficients for the ejection of negative ions by atomic and molecular ions of hydrogen and deuterium. We have compared the coefficients K^- obtained for molecular ions at energies of 1000 and 800 kev and for atomic H_1^+ and D_1^+ at energies of 500 and 400 kev respectively. The K^- measured for molecular ions of energy E_1 is twice as large as the K^- measured for atomic loss of energy $E_1/2$. From this we can conclude that the effectiveness of molecular hydrogen and deuterium ions from the point of view of secondary negative ion emission is equal to the effectiveness of two protons of the same speed. This enabled us to "join" the curves obtained for K^-, using atomic ions in the energy interval of 500 to 1000 kev, to curves obtained using molecular ions in the energy interval of ~ 150 to 500 kev and recalculated per atom.

The energy of the bombarding particles was determined by means of the magnetic analyzer. The radius of curvature of the trajectory in the magnetic field was 100 cm, the angle of deflection was 90°, as determined by the entrance and exit diaphragms with diameters of the openings given above. The magnetic poles were beveled at an angle of 45° at the points where the beam entered and left the magnetic field. The energy of the particles was measured with an accuracy of ± 20 kev.

EXPERIMENTAL RESULTS AND DISCUSSION

Figure 6 shows curves obtained for the dependence of K^- on the kinetic energy of the bombarding hydrogen and deuterium ions using targets made of copper, stainless steel of type EIa–1, carbon and aluminum. Before the copper target was subjected to bombardment by primary ions it was conditioned over a long period of time in high vacuum at a temperature of 900° C and a pressure of ~ 10^{-6} mm of Hg in the absence of a beam during 1.5–2 hours. The measurements of the magnitude of K^- were made on a cold target free of oxides.

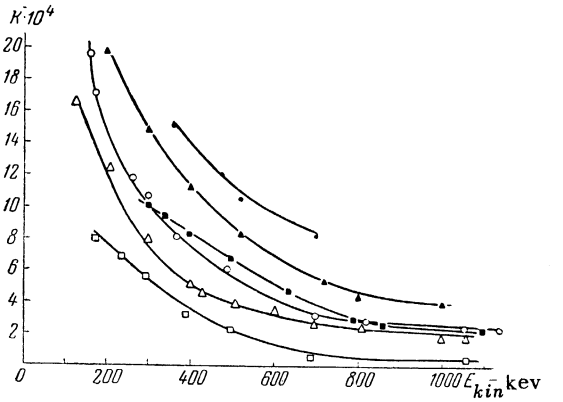


FIG. 6. Curves showing the dependence of the yield of negative ions on the kinetic energy of bombarding ions. Aluminum target: o–H_1^+, and o–D_1^+; carbon and stainless steel targets: Δ–H_1^+, ▲–D_1^+; copper target: □ –H_1^+, ■–D_1^+.

As may be seen from the curves of Fig. 6, K^- decreases as the energy of the bombarding ions is increased. At the same energy of the primary ions K^- is larger when a given target is bombarded by D_1^+ ions than when it is bombarded by H_1^+ ions. Experiments carried out with stainless steel and carbon targets did not show any difference in the values of K^- and therefore the curves showing $K^- = f(E_{kin})$ obtained for a stainless steel target are also characteristic of a carbon target.

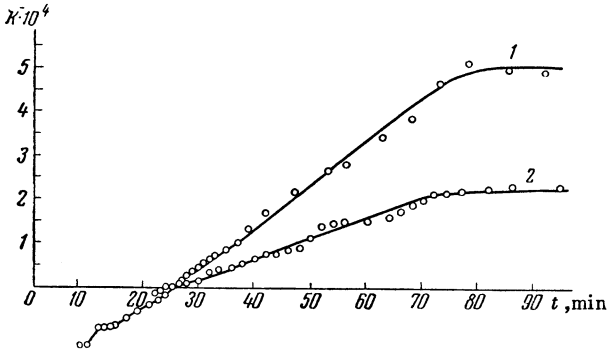


FIG. 7. Curve *1* refers to the bombardment by H_1^+ ions of kinetic energy 300 kev, curve *2* refers to the same at an energy of 500 kev.

An aluminum target was also subjected to bombardment by the positive ions H_1^+ and D_1^+. As may be seen from Fig. 6, the shape of the curve $K^- = f(E_{kin})$ is the same as for the other targets. However, these experiments show that the value of K^- depends on the target material. Of the targets investigated by us the lowest emission of negative ions is exhibited by a copper surface which is free of oxides and the largest emission is exhibited by aluminum.

The emission of negative ions depends markedly on the state of the surface of the target. It was found that a copper target subjected to preliminary heating for 8–10 min at a temperature of 900° K and then cooled for 10 min shows no emission of negative ions when bombarded by positive ions. But at the beginning of the bombardment of such a target, emission of positive ions is observed. By applying a retarding field, we found that the positive ions have an energy greater than 400 ev and therefore we may suppose that these are primary ions scattered through large angles in the Coulomb field of the target nuclei.

The measurement of the current to the collector (from a cold target which had been previously heated) over a certain period of time shows that the positive current gradually decreases, passes through zero and a negative current begins to grow reaching a constant value after a time of the order of 70 min measured from the moment of stopping the heating of the target. Figure 7 shows the change with time of the emission of secondary ions from a copper target which was heated in a vacuum of 10^{-6} mm of Hg and which was subsequently allowed to cool in the absence of the ion beam for 8–10 min. A measurement of the coefficient of secondary electron emission γ for a copper target bombarded by hydrogen ions both before the target is heated in vacuum and after such heating does not exhibit at any time any difference in the value of γ. This experiment shows that the method used by us for outgassing the metal surface does not affect the value of γ.

The yield of secondary negative ions from a cold copper target, which had been previously subjected to heating, is shown in Fig. 8 for the case of bombardment by deuterium ions of 300 kev energy. The dependence of K^- on the time (Figs. 7 and 8) may be related to the formation of a surface layer of atoms adsorbed on the target which is apparently due to the adsorption from the surrounding gas medium and to the diffusion of hydrogen from the body of the metal.

Measurements carried out using H_1^+ and D_1^+ ions showed that in the energy range from 200 to 1000 kev the deuterium ions liberate considerably greater numbers of secondary negative ions from the target than do ions of atomic hydrogen. If one plots the experimental results for K^- as a function of the velocity of the ion then the experimental points for the ions of both isotopes fall on the same curves (Fig. 9).

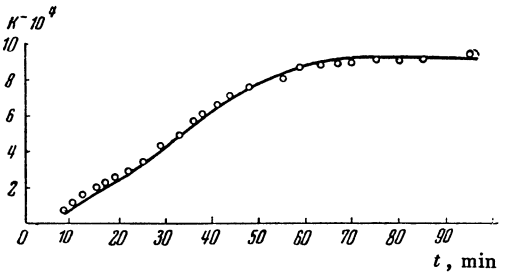


FIG. 8.

Concurrently with the study of the coefficient of emission of negative ions, we have also investigated their nature by analyzing their masses by means of the apparatus shown in Fig. 2. In the case of a copper target, we observed a large number of negative ions differing in their values of e/m

(Fig. 10). Under the same experimental conditions, the first peak has an intensity two orders of magnitude larger than the other groups, and is due to H^-_1 ions. The second group of peaks corresponds to masses 12, 16, 18. They correspond to C^{12-}, O^{16-}, and OH^- ions. The third group of negative ions apparently contained C_2^-, CH_2^-, $C_2H_2^-$ ions. When the target was heated by the method previously described, it was found that the hydrogen peak disappeared and was then reestablished during a time $\sim$ 40 min.

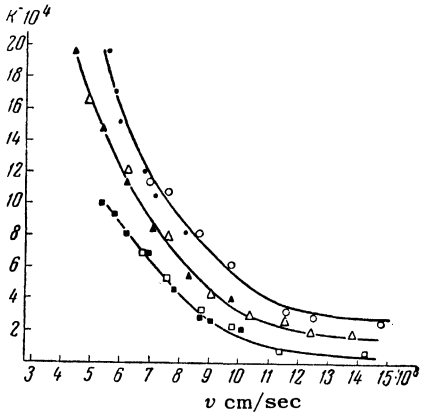


FIG. 9.

It would be of interest to investigate the dependence of K^- on the angle of incidence.

CONCLUSIONS

1. The coefficient of ejection of negative ions decreases monotonically as the energy of the primary hydrogen ions is increased. Within the investigated range of energies the maximum value of K^- at an energy near 200 kev is $\sim 10^{-3}$ and falls to $\sim 10^{-4}$ at an energy of 1000 kev.

2. The probability of forming negative ions by bombardment of metallic surfaces is determined by the speed of the primary ions and apparently does not depend on their mass.

3. The secondary emission of negative ions depends on the nature of the target. The value of the emission coefficient K^- progressively increases for the targets investigated in the following order: copper, stainless steel of type EIa–1, aluminum. No difference was observed in the values of K^- obtained for a stainless steel target add for a carbon target made by depositing a thick layer of aquadag on a copper support.

4. Outgassing the target decreases the value of K^- and the number of emitted negative ions may become less than the number of fast primary positive ions scattered by the Coulomb field. At the same time the coefficient γ remains constant.

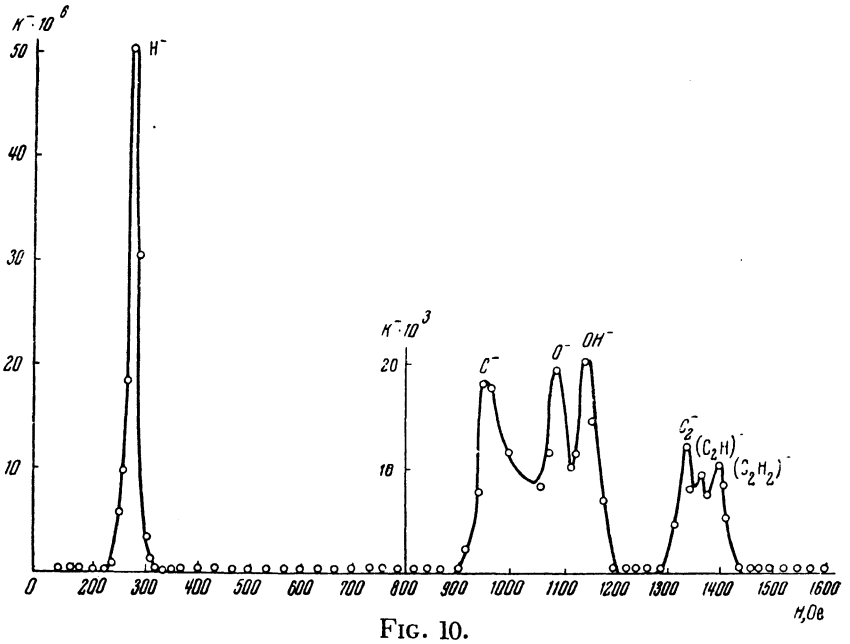


FIG. 10.

5. The coefficient of emission for negative

ions whose energy does not exceed 10 ev is of the same order as for slow positive ions.

The authors express their sincere gratitude to academician A. K. Val'ter and to Ia.M. Fogel' for valuable advice and for their continuing interest in this work.

1 R. Arnal. Ann. de Phys. 10, 830 (1955).

2 V.I. Veksler and G.N. Shuppe, J.Tech. Phys. (U.S.S.R.) 23, 1573 (1953).

3 R.H. Sloane and H.M. Love. Nature 1599, 302 (1947).

4 E. Ia. Zandberg, J.Tech. Phys. (U.S.S.R.) 25, 1386 (1955).

5 F. L. Arnot and C. Beckett. Proc.Roy. Soc. (London) A168, 103 (1938).

Translated by G.M. Volkoff
51

SOVIET PHYSICS JETP VOLUME 5, NUMBER 2 SEPTEMBER, 1957

Some Cases of Very Small Life Times of Low Nuclear Levels

E. E. BERLOVICH AND G. V. DUBINKIN
Leningrad Physico-Technical Institute,
Academy of Sciecnes, USSR

(Submitted to JETP editor August 14,1956)

J. Exptl. Theoret. Phys. (U.S.S.R.) 32, 223-226 (February, 1957)

The life times of short-lived excited states of nuclei were studied by the delayed coincidence method. The life times of the lower levels of the Ti^{46} and Mo^{95} nuclei were found to be less than 10^{-10} sec.

The experimental half-life of the excited state of the Tl^{203} nucleus was found to be $(2.9 \pm 0.3) .10^{-10}$ sec., a result which does not agree with that of de-Waard. The method applied by de-Waard is discussed. The partial half-times for magnetic dipole and electric quadrupole transitions are compared with the values predicted by the formulas of the single-particle model.

BERLOVICH[1] has demonstrated the influence of the particle energy and of the pulse front shape associated with it on the results of measuring the life-time of excited states by the method of delayed coicidences. This effect which is particularly important in the case of short-lived states (10^{-10} – 10^{-9} sec) was eliminated by introduction into both side channels of triple coincidence circuit amplitude analyzers by means of which it was possible to select the coincidences of only those pulses which lie in equivalent amplitude intervals.

In this article we present the results of investigating short-lived states of Ti^{46}, Mo^{95} and Tl^{203} nuclei. Before the measurements were made, the following control experiment was carried out: by means of investigating the curves of $\gamma\beta$–$\gamma\beta$–coincidences for the $Co^{60} \rightarrow Ni^{60}$ decay for which the limiting values of the energies of the β–spectrum and of the Compton distribution differ by a factor of more than 3½, we satisfied ourselves that when coincidences of pulses in energy–equivalent intervals were recorded no mutual displacement of the centers of gravity of the two curves occurred. This agrees with the results of Bay[2] according to which the life times for both excited states of the Ni^{60} nucleus are less than 10^{-11} sec.

Figure 1 shows the experimental results for the $Sc^{46} \rightarrow Ti^{46}$ decay. The maximum energy of the β-spectrum is 0.340 mev, the energies of the two γ-rays in cascade accompanying the β-decay are 0.880 and 1.120 mev. According to the data of Nag, Sen, and Chatterjee[3] the mean life of these levels is $\tau = 1.3 \times 10^{-5}$ sec. However, it follows from the measurements of Koicki, Ballini, and Chaminade[4] that this life-time must be less than 2×10^{-6} sec.

From the measurement of conversion coefficients,[5] and also from experiments on angular $\gamma\gamma$–correlation[6] and on angular correlation of the directions of polarization[7], it follows that both γ-transitions are of the E2 type. The life times of nuclear states for electric quadrupole transitions with energies of about one mev must (in accordance with the single particle formulas of Weisskopf[8] and of Moszkowsky[9] be smaller than the values given above by six or seven orders of magnitude.