# ON THE UNIDENTIFIABLE PEAKS IN CHARACTERISTIC X-RAY SPECTRA

S. V. Adamenko, A. S. Adamenko, A. V. Andreev,\* I. A. Kossko,<sup>†</sup>

S. S. Ponomarev,<sup>†</sup> V. V. Kovylyaev,<sup>†</sup> and A. N. Zakusilo

Electrodynamics Laboratory Proton-21

14/1 Dovjenko Str., 03057, Kiyv, Ukraine<sup>‡</sup>

(Dated: May 18, 2004)

By X-ray spectroscopy with various sources of excitation of characteristic X-ray emission (CXE), we studied chemical inhomogeneities formed on the surface of accumulating screens as a result of the explosion-induced compression of solid targets. The goal of the work consisted in a purposeful search for the characteristic X-ray peaks which are unidentifiable in principle. The specificity of studies was in that the registered spectra included, as a rule, the characteristic X-ray peaks of a considerable number of chemical elements, which hampered somewhat their identification. The studies were performed on three different setups, in which CXE was excited by an electron beam, radiation of X-ray tubes with monochromator, and soft gamma-radiation from an isotope source with <sup>241</sup>Am, which allowed us to analyze a wide energy range 1...45 keV and to use various localities of the primary beam. While solving the problem of identification of the characteristic X-ray peaks in the registered spectra, we applied the method of long-term exposures with the purpose to reveal peaks with small intensity. In a number of cases, we registered additionally the spectra of relevant pure chemical elements or their simple compounds for the reliable identification of the characteristic X-ray peaks in complicated spectra. As artifacts of the analysis, we considered such phenomena as the formation of 'sums' in spectra and the appearance of peaks of elastically and inelastically scattered radiation from the very source. We established the presence of a number of unidentifiable peaks such that we failed to refer them, on the one hand, to any characteristic X-ray peaks of the known chemical elements or to consider them, on the other one, as artifacts. As one of the variants to interpret the registered peaks, we consider the assumption as for their affiliation to long-lived transuranium elements formed as a result of nuclear transformations of the target material.

Keywords: characteristic X-ray spectra, X-ray fluorescence analysis, X-ray electron probe microanalysis, unidentifiable X-ray peaks, long-lived transuranium elements

## INTRODUCTION

From 1999 till the present time, the staff of the Electrodynamics Laboratory Proton-21 carries on the experiments on the explosion-induced compression of a substance, which leads to the creation of superdense states. Till October 2003, over 5000 dynamical impact compressions of solid targets were performed at the Laboratory. The goal of these experiments is the verification of the preliminarily theoretically justified hypothesis of the possibility to initiate, by using the pulse coherent action on a solid substance, the self-organizing process of avalanchelike self-densification up to the state of collapse (electronnucleus plasma), in which the conditions for the running of collective many-particle nuclear reactions arise due to a decrease in the Coulomb barrier. For the verification of this hypothesis, the Laboratory's staff constructed the experimental setups which are able to transfer up to 1 kJ of energy for a pulse duration of about 10 ns with the help of the electron beam as a primary carrier of the concentrated energy. At the culmination stage of the process, a microvolume of the target substance was compressed up to a density of above  $10^{26}$  cm<sup>-3</sup>. In this case, the power density in the region of compression exceeded, by various estimations,  $10^{22}$  W·cm<sup>-3</sup>.

The impact compression of a target was realized in vacuum of  $\sim 10^{-3}$  Pa and led to its destruction by the explosion from inside. This process was usually accompanied by the radial dispersion of a target material with its deposition on a special accumulating disk-like screen of about 15 mm in diameter and 0.5 mm in thickness. Fig. 1 presents the micrographs of both a typical target after the explosion (a) and a typical accumulating screen with deposited products of the explosion (b) which were derived with a scanning electron microscope in the secondary electron mode. By using the method of scanning electron microscopy (SEM), we established that the products, which remained in the target crater and deposited on accumulating screens, formed a layer of irregularly distributed drops, splashes, films, particles, and other micro- and nanoobjects with complicated morphology. Because the explosion products are microobjects, we firstly studied them mainly with local methods. To investigate their element and isotope compositions, we used X-ray electron probe microanalysis, local Auger electron spectroscopy, and laser mass spectrometry [1, 2]. By secondary-ion mass spectrometry with various types of the excitation of secondary ions, we discovered deviations of the isotope composition from the natural abundance

<sup>\*</sup>Shevchenko Kyiv National University, 64 Volodymyrs'ka Str., Kyiv, 01033 Ukraine

<sup>&</sup>lt;sup>†</sup>Institute of Problems of Materials Science of the NAS of Ukraine, 3 Krzhizhanovsky Str., Kyiv, 03142 Ukraine

<sup>&</sup>lt;sup>‡</sup>Electronic address: enr30@enran.com.ua

of isotopes for a number of chemical elements [3, 4].

While studying the chemical composition of products of the explosion by the above-mentioned methods, we registered several tens of chemical elements in significant amounts in every analysis [including those cases where both a target and an accumulating screen were made of a single chemical element maximally purified from impurities, e.g., Cu (99.99 mass. %), Ag (99.99 mass. %), Pb (99.75 mass. %), and others]. These elements covered the range from small to large atomic numbers and were not found by highly sensitive experimental methods in the initial materials of targets and accumulating screens or were present in them as impurities but in quantities by 3...7 orders and more lower than those measured after the experiment [4]. Moreover, while studying them by Auger electron spectroscopy, we registered six Augerpeaks (one doublet) with energies 172, 527, 1095, 94, 560, and 130 (115) eV which cannot be referred, on the one hand, to artifacts of the analysis and, on the other one, be related to any Auger-peaks of the known chemical elements [1]. The mentioned Auger-peaks were referred by us to basically unidentifiable peaks in the scope of the known part of the Periodic table. As one of the variants of the interpretation of the discovered unidentifiable Auger-peaks, the assumption as for their affiliation to long-lived transuranium elements was advanced in [1]. The search for basically unidentifiable peaks in characteristic X-ray spectra is the main goal of the present work.

#### I. MATERIALS AND METHODS

The main objects of our study were accumulating screens made of Cu (see Fig. 1, b) both in the initial state and after the deposition of the explosion products on them. In fact, the accumulating screen served as a substrate, whose surface was covered by the studied material layer composed by the explosion products. The layer was characterized by a slightly pronounced relief and the axial symmetry. We used as-received accumulating screens, i.e., they were not undergone to cleaning procedures prior to the analysis, because such procedures could damage them or change their composition.

While studying the specimens by X-ray fluorescence analysis with the use of X-ray tubes, we took the standard specimens of metallic screens together with specially prepared substrates transparent for X-rays, on which we precipitated the deposit derived by dissolution of the upper layers of screens. The screens were treated by diluted (1:2) nitric acid (GOST 4461-77) in a teflon dish for 1 min. The precipitate derived by evaporation of the solution was applied on various substrates: aluminium foil, special powder (aerosil), various polymeric films.

As materials for targets (a wire of 0.5 mm in diameter), we used copper, lead, and silver with controlled purity.

We note that it was impossible to predict the distribution of possible traces of long-lived transuranium elements over the specimen surface or its bulk. Therefore, to solve the posed problem, we chose the methods of analysis which allow one to register the presence of chemical elements which are distributed over a macroscopic area of a specimen surface at low concentrations or are localized in the form of particles. In this case, the used methods must possess a high sensitivity because the expected concentrations of long-lived transuranium elements are small.

With the purpose to extend the energy range of analysis and to realize the action of an electron probe or an X-ray beam with various diameters, we used three analyzing setups.

To detect the characteristic X-ray spectra in the highenergy region, we used an installation for X-ray fluorescence analysis with a source of soft gamma-ray radiation on the base of isotope <sup>241</sup>Am which possesses the intense 59.6-keV line. The secondary emission was registered with an X-ray spectrometer on the base of a vertical semiconductor detector. The intense inelastically scattered radiation of the source, which was observed in the form of a wide asymmetric peak with a maximum near 49 keV and with a long low-energy tail, restricted the working range of studying the trace amounts of elements to the range of 20...35 keV.

The X-ray spectra in the energy range up to 30 keV were studied with the help of a setup for X-ray fluorescence analysis with an X-ray tube with molybdenum anode and with the use of a monochromator made of pyrolytic graphite. At the output of a monochromator, X-ray radiation has a maximum in the region near 36 keV with a half-width of about 3 keV. We used the accelerating voltage of 45 kV and carried out the measurements at a take-off angle of  $33^{\circ}$ , which corresponded to the depths of analysis up to one thousand  $\mu$ m. The size of an analyzed region of the specimen surface was usually of about 2 mm.

The local analysis was performed on a REMMA-102 setup for X-ray electron probe microanalysis (the SELMI firm, Sumy, Ukraine). In the specimens under study, characteristic X-ray emission was excited by an electron 35-keV beam less than 50 Å in diameter. The analyzed region had a diameter of about  $3\,\mu\text{m}$ . The residual pressure of a gas in the working chamber was about  $3 \times 10^{-4}$  Pa. The surface chemical inhomogeneities, to which the probe was directed to register the spectrum, were chosen on a specimen surface image formed in the mode with atomic number contrast (the so-called COMPO mode). The chemical inhomogeneities were particles and localized regions with various sized and geometries.

In all three setups, a semiconductor energy-dispersion detector ensured the energy resolution of 160 eV.

At small concentrations of the elements we are interested in, a special attention was paid to the problem of separation of their characteristic peaks against the background. In order to accumulate the background signals and low intensity peaks in the amount sufficient for statistical analysis under conditions of a low counting rate

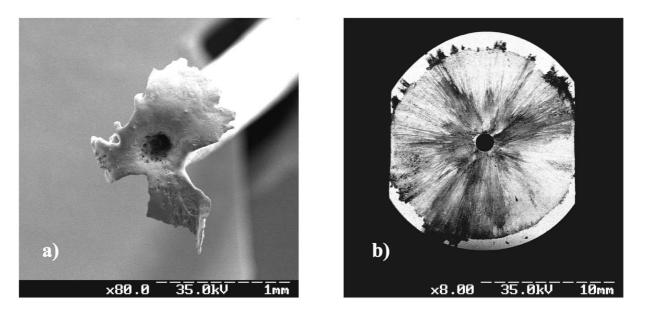


FIG. 1: Scanning electron micrographs (the secondary electron mode) of both a typical target after the explosion (a) and a typical accumulating screen with deposited products of the explosion (b).

(about 10 pulse/sec for film specimens), it was necessary to carry out the measurements with long-term exposures. However, on long-term exposures, parameters of the amplifying channel of an analyzer are usually undergone to a drift, which leads to a broadening of the peaks under study. To prevent this effect, we carried out measurements by series for 10...20 min. In each series, we calibrated the energy scale of spectra by means of the attachment to the reference points. As the latter, we took the most intense peaks, whose energies are known from the reference data [5]. Then all the spectra of a series were adjusted to a common energy scale and were summed.

As a criterion defining the fact of the presence or absence of a low intensity peak against fluctuations of the background, we took the statistical ' $3\sigma$ ' criterion [6]. A peak was considered reliable if its amplitude exceeded the triple value of a mean square background fluctuation  $\sigma$ . To determine the latter in the vicinity of a peak, we constructed a function approximating the dependence of the averaged background intensity on the energy channel number (we used a linear or square function). Then we calculated a mean square background fluctuation  $\sigma$  by the formula

$$\sigma = \sqrt{\frac{1}{N-1} \sum_{i=1}^{N} (I_i - \bar{I}_i)^2},$$
(1)

where N is the number of channels in the vicinity of a peak,  $I_i$  is the background intensity in the *i*-th channel,  $\bar{I}_i$  is the approximated mean background intensity in the *i*-th channel.

The choice of a function approximating the background is arbitrary in the general case. In addition, the calculation of the mean background level is significantly complicated if an unidentifiable peak is close to the characteristic peaks of chemical elements which are present in the composition of the specimen. In this connection, the result of calculations of a mean square background fluctuation had low accuracy and bore the estimating character.

To estimate the peaks reliability we performed the probability calculation for peaks of various width and height that may appear due to fluctuations of a background in neighboring channels. Let's assume that the level of a background in *i*-th channel of the detector is a random value obeying the normal distribution with the density of probability

$$\rho(x) = \frac{1}{\sqrt{2\pi\sigma}} e^{\frac{(x-s)^2}{2\sigma^2}},\tag{2}$$

where the parameter s is the mean value of a background in a given channel,  $\sigma$  is a root-mean-square deviation of a level of a background from the mean value. For simplicity let's assume that the parameters s and  $\sigma$  do not depend on a channel number. Actually, the mean background value and its fluctuations value depend on energy, hence, the channel number. However, these dependencies are rather weak and may be neglected while considering a small part of a spectrum.

The probability of a level of background in a given channel to be in some interval is equal to the integral of the density of probability (2). The deviation is in terms of  $\sigma$ . Thus, the probability of the excess of the deviation of the level of background above mean value by n rootmean-square fluctuations  $\sigma$ 

$$p\{x-s>n\sigma\} = \int_{s+n\sigma}^{\infty} \rho(x) \, dx = \frac{1}{2} \left(1 - \operatorname{Erf}\left[\frac{n}{\sqrt{2}}\right]\right), \quad (3)$$

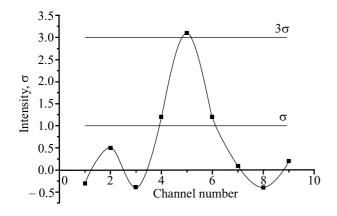


FIG. 2: Fluctuating peak of the form  $\{1,3,1\}$ 

where  $\operatorname{Erf}(z) \equiv \frac{2}{\sqrt{\pi}} \int_{0}^{z} e^{-t^{2}} dt.$ 

For calculation of the probability of the appearance of a fluctuating peak with width k in the given point of a spectrum that is concurrence of the excess of a level of background by  $n_1\sigma$  in the given channel a, by  $n_2\sigma$  in the following channel, etc. (see Fig. 2), assuming the values of background in channels to be independent, it is necessary to find the product of the probabilities determined by Eq. (3)

$$P_{\{n_0\dots n_{k-1}\}} = \prod_{i=0}^{k-1} p\{x_{i+a} - s > n_i\sigma\}.$$
 (4)

The probability of a fluctuating peak appearance in any point of a spectrum can be estimated by multiplication of the probability received via Eq. (4) by the number of possible positions of the peak in a spectrum, that is approximately equal to the number of channels (in our case 1024). Thus, the probability of appearance of the peak of a given form, determined by a set of numbers  $\{n_0, n_1, \ldots, n_{k-1}\}$ , anywhere in a spectrum is  $1000P_{\{n_0...n_{k-1}\}}$ . In Table I the resulting probabilities for various forms of peaks are given. The values of  $1/(1000P_{\{n_0...n_{k-1}\}})$  are also given, they represent estimated number of spectra which are required to detect the fluctuating peak.

Thus, while verifying peaks, the analysis of the probability of background fluctuations is required. The Table I shows that the width of a peak is the necessary criterion as well as the height.

#### **II. RESULTS AND DISCUSSION**

The analysis of a great number of specimens showed that the high-energy action products contained the majority of elements from the Periodic table. In most cases, the spectra contained the X-ray lines of light elements with atomic numbers up to 30. We established the appearance of basically unidentifiable low-intensity peaks

TABLE I: Probabilities of fluctuating peaks appearance

]

Peak form	P	1000P	1/(1000P)
$1,\!3,\!1$	$3.398\cdot10^{-5}$	$3.398 \cdot 10^{-2}$	29
$1,\!1,\!3,\!1$	$5.391 \cdot 10^{-6}$	$5.391 \cdot 10^{-3}$	186
$1,\!3,\!2$	$4.873 \cdot 10^{-6}$	$4.873 \cdot 10^{-3}$	205
$^{3,3}$	$1.822\cdot 10^{-6}$	$1.822 \cdot 10^{-3}$	549
2,3,2	$6.897\cdot10^{-7}$	$6.897\cdot10^{-4}$	1 431
$1,\!3,\!3$	$2.896 \cdot 10^{-7}$	$2.896 \cdot 10^{-4}$	$3\ 451$
1,2,3,2	$1.108 \cdot 10^{-7}$	$1.108\cdot 10^{-4}$	9025
$1,\!3,\!3,\!1$	$4.587\cdot10^{-8}$	$4.587 \cdot 10^{-5}$	21 800
1,2,3,2,1	$1.759\cdot 10^{-8}$	$1.759 \cdot 10^{-5}$	56 850
3,3,3	$2.460 \cdot 10^{-9}$	$2.460 \cdot 10^{-6}$	406 400

which revealed themselves against the background of a lot of peaks with known energies. The last fact hampered, on the one hand, the detection of the presence of unidentifiable peaks but also had a positive side, on the other one, because we were able to realize the rigid control of the energy scale.

The term 'unidentifiable peaks' means that they cannot be identified with the use of reference data on the characteristic X-ray peaks of known chemical elements [5]. In the complicated cases, the identification of spectra was hampered by artifacts of the analysis. As artifacts, we considered 'the peaks of sums,' which were formed on the simultaneous entry of two quanta to a detector which belong to one high intensity line or different high intensity lines in a spectrum (in this case, the registered energy is the sum of the energies of quanta), as well as the peaks of elastically or inelastically scattered quanta of the emission lines of the isotope source with  $^{241}$ Am in the cases where it was used. The source had a number of weak lines which were not documented due to their extremely low intensity.

To exclude similar artifacts, we carried out the registration of spectra in the specimens taken from the pure materials of a substrate, which did not participate in the experiments including the high-energy action, under the same conditions. In addition, we performed the careful examinations of the spectrum of the source radiation which was scattered by the light matrix (organic glass). In this case, all peaks of the elastically scattered radiation possessed the high intensity 'twins' of inelastic scattering peaks. The energy shift of inelastic scattering peaks relative to the relevant elastic scattering peaks was calculated by the Compton formula and was compared with that observed in the spectrum. Because the cross section of inelastic scattering of X-rays on light matrices exceeds considerably that of elastic scattering, a similar trick facilitated the detection and identification of weak source-related peaks elastically scattered by specimens with heavy matrices.

To exclude the hypothetical possibility for the appearance of other instrumental artifacts, we performed the examination of spectra of the initial materials of a target and an accumulating screen, which did not participate in the experiments, in each case of the registration of an unidentifiable peak.

We also excluded all peaks, whose amplitudes did not satisfy the ' $3\sigma$ ' criterion, as possible statistical fluctuations of the background.

The peaks which remained after the exclusion of possible artifacts of the analysis were referred to basically unidentifiable peaks. Information about the registered unidentifiable peaks is given in Table II.

The 21300-eV peak was detected on studying the specimen, being a polymeric film of about 20  $\mu$ m in thickness. It served as an accumulating screen, on which the products of the explosion of a target were deposited in the experiment on the initiation of the process of nuclear transformations. We studied the specimen on an installation for X-ray fluorescence analysis with excitation by the radiation of an X-ray tube. Due to a small thickness of the specimen, the counting rate of pulses was low and varied in the limits of 8...15 pulse/sec. The total exposure of one series of measurements of 20 min in duration was 10...17 h. One of the derived spectra with the unidentifiable 21300-eV peak is presented in Fig. 3.

This peak was registered at two points of the specimen. At one of the points, its appearance was confirmed by the repeated measurement performed in several days after the first finding. However, two subsequent measurements at the same point failed to detect this peak.

On the statistical analysis, it turned out that these peaks do not satisfy the statistical ' $3\sigma$ ' criterion. However, in the summary spectrum of two registrations performed at a single point, the peak amplitude was equal to 2.8 in units of the mean square background fluctuation. This fact together with the appearance of the peak in three spectra allow us to consider these peaks to be reliable with high probability.

The possible positions of 'the peaks of sums' nearest to the unidentifiable peak [21102 eV (Pb  $L_{\alpha 1}$  + Pb  $L_{\alpha 1}$ ) and 21516 eV (Cu  $K_{\beta 1}$  + Pb  $L_{\beta 1}$ )] are separated from the 21300-eV peak by the interval exceeding the energy resolution of a detector by several times. In addition, the appearance of 'the peaks of sums' is extremely improbable at a counting rate of the order of 10 pulse/sec.

We also found the 23600-eV unidentifiable peak in the spectrum derived on an installation for X-ray electron probe microanalysis on studying the metallic specimen No. 8545, being an accumulating screen with the products of the explosion of a target. The spectrum was registered on the surface of a spherical particle of about 2  $\mu$ m in diameter. The particle image derived by a scanning electron microscope is given in Fig. 4, and its spectrum is presented in Fig. 5.

The unidentifiable peak was observed in the spectrum near the theoretical 'peak of sums' of Cu  $K_{\beta 1}$  + Pb  $L_{\gamma 1}$ (23665 eV). However, the spectrum does not contain the peaks corresponding to the sums and duplications of the significantly more intense peaks of Cu  $K_{\alpha 1}$  and Pb  $L_{\alpha 1}$   $(16088\,{\rm eV},\,18595\,{\rm eV},\,{\rm and}\,\,21102\,{\rm eV}).$  Thus, we established that the 23600-eV peak is not a 'peak of sums.'

According to the above-described procedure of registration, the given spectrum was composed from the series of spectra subsequently saved in the computer memory. Thus, by summing the different close groups of individual spectra into series, it was possible to monitor the variations in the relative intensities of the characteristic peaks of chemical elements in time. The results of evolution of the content of some chemical elements are given in Table III.

As seen in Table III, the concentrations of elements at the analyzed point changed remarkably during the implementation of the series of registration. We explain this fact, on the one hand, by a slight drift of the electron beam and, on the other one, by the processes of diffusion initiated by an electron probe. In addition, the long-term action of an electron probe on a local region of the specimen caused the formation of a carbon deposit on the particle surface due to the presence of the residual atmosphere in the specimen chamber and, as a consequence, the selective screening action of the formed film of contaminations on the characteristic X-ray signals from various chemical elements. From our viewpoint, the above-mentioned reasons for the disappearance of the unidentifiable peak on repeated registrations are not sufficient. The assumption comes to mind that just the damaging action of the beam leads to the disappearance of the unidentifiable peak. We note that, with lower energies of electrons affecting the surface, we succeeded in making a repeated registration of unidentifiable peaks in Auger-spectra [1].

For specimen No. 8545, we registered a spectrum at its other point chosen on the surface of a 6- $\mu$ m particle, in which we found an unidentifiable peak in the same energy region. The energies of these peaks are about 100 eV, which is of the order of the detector resolution. This allows us to consider these two peaks to be the manifestation of a single peak in two spectra.

For the same specimen, we registered the spectrum with the 28900-eV unidentifiable peak on the surface of a spherical particle of  $15 \,\mu\text{m}$  in diameter. The spectral region containing the peak is shown in Fig. 6. At this point, we carried out two series from 9 measurements for 400 sec (the total exposure was 2 h). The unidentifiable peak is most clearly observed in the summarized spectrum of the first series derived for 1 h of registrations. In the summarized spectrum of two series, its contrast is somewhat lower. We note that, in the 28900-eV region, there are no possible 'peaks of sums' of the high intensity characteristic X-ray lines registered in this spectrum.

After the processing of metallic accumulating screens and internal surfaces of the chamber of the high-energy action setup with nitric acid, we derived a substance from the solution and precipitated it on the powder of aerosil. On the series of specimens made in this way, we found two unidentifiable peaks in the spectra derived on the installation containing the isotope source with <sup>241</sup>Am.

Peak energy, eV	Method of analysis	Characteristic element environment	Specimen		ed atomic the element By $M_{\alpha}$	Number of detections	Peak height <sup><math>a</math></sup>
20000	XEPMA	Pb, Cu, Au, Fe, Ni	No. 3 (deposit on organic glass)	109–110	198–202	1	4.6
21300	XFA with X-ray tube	Cu, Pb, Ag, Cd, Sn, In	Polymeric screen (deposit on the screen)	112 - 113	204 - 208	3	2.8
22600	XEPMA	Pb, Cu, Fe, Ti, Ca, K, Si, Zn	No. 43 (Cu screen)	115 - 117	209-214	1	4.7
23600	XEPMA	Al, Cu, Pb, Ag	No. 8545, 41 (Cu screens)	117–118	213-218	2	4.2
24600	XEPMA	Al, Si, Ca, Ti, Fe, Cu, Pb	No. 43 (Cu screen)	119 - 121	218-222	1	3.1
26300	XEPMA	Cu, Ag	No. 41 (Cu screen)	123 - 125	224 - 229	1	3.9
26700	XEPMA	Cu, Al	No. 8545 (Cu screen)	124 - 125	226-231	1	3.5
28900	XEPMA	Ag, Pb, Bi, Sn, Cu	No. 8545, 41, 43, 47 (Cu screens), No. 8858 (target)	128-129	234-239	5	5.6
36600	$\stackrel{ m XFA with}{^{241} m Am}$	Ag, Ba, Nd	No. 2 (deposit on aerosil)	141 - 143	260 - 267	1	3.2
38100	$\stackrel{ m XFA with}{^{241} m Am}$	Ag, Ba, Nd	No. 1 (deposit on aerosil)	144 - 145	265 - 272	1	5.1
51950	${}^{XFA}_{241}{}^{With}_{Am}$	Pb, Cu, Au, Fe, Ni	Polymeric screen (deposit on the screen)	162–164	306-314	1	5.9

TABLE II: Data on unidentifiable peaks registered in X-ray spectra.

<sup>*a*</sup>maximum, in units of the mean square background fluctuation

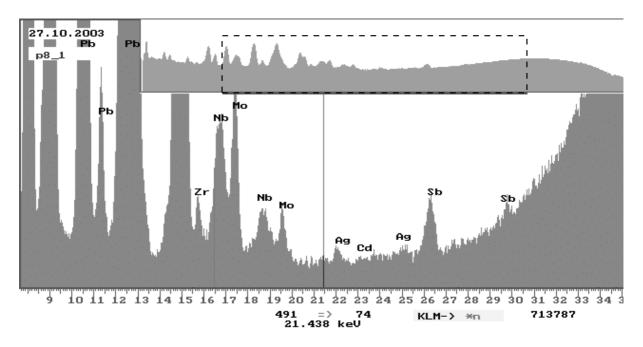


FIG. 3: High-energy region of the spectrum of a film specimen with the unidentifiable 21.3-keV peak.

One of the spectra with the 38.1-keV peak is presented in Fig. 7. In the spectrum of other specimen derived by the same processing of the same accumulating screens and fragments of the chamber of the experimental setup, we registered the 36.6-keV unidentifiable peak. The energies of the possible 'peaks of sums' nearest to the unidentifiable peaks are, respectively, 38866 eV(Sb  $K_{\alpha 1}$  + Pb  $L_{\beta 1}$ ) and 36905 eV (Sb  $K_{\alpha 1}$  + Pb  $L_{\alpha 1}$ ), which allows us to assert that the unidentifiable peaks are not 'peaks of sums.'

© S.V. Adamenko, 2004

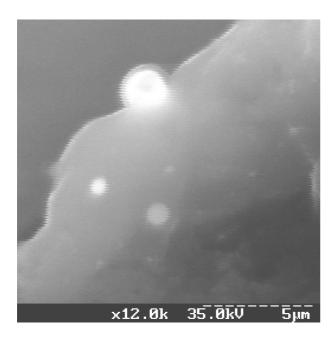


FIG. 4: Scanning electron micrograph of a particle, which is positioned on specimen No. 8545, derived with an scanning electron microscope.

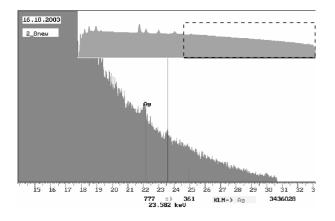


FIG. 5: Fragment of the X-ray spectrum registered from the particle on specimen No. 8545 which contains the 23.6-keV unidentifiable peak.

The unidentifiable peak with an energy of 20.0 keV was registered in the spectrum derived on an installation for X-ray electron probe microanalysis from the surface of a metallic particle of 1  $\mu$ m in diameter which was found on the organic glass substrate. In Fig. 8, we present the spectral region with this peak. Similarly to specimen No. 8545, we fixed an essential change in the chemical composition at the point under study in the course of time. On the repeated observation in 30 min, we registered a decrease in the intensity of the peaks from Ca and Pb and an increase in the intensity of the peak from Cu. In this case, the unidentifiable peak disappeared. The change in the chemical composition at the point under study and the disappearance of the unidentifiable peak

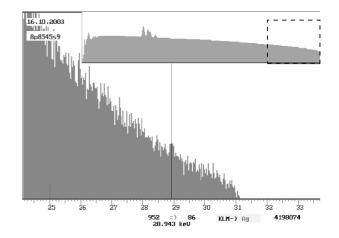


FIG. 6: Fragment of the spectrum of a point of specimen No. 8545 which contains the 28.9-keV unidentifiable peak.

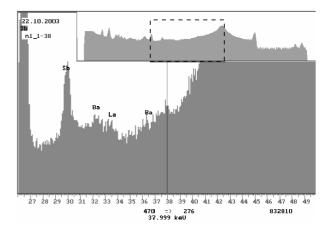


FIG. 7: Fragment of the spectrum of specimen No. 1 which contains the 38.1-keV unidentifiable peak.

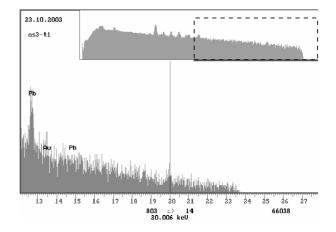


FIG. 8: Fragment of the spectrum of a particle on specimen 'organic glass No. 3' which contains the 20-keV unidentifiable peak.

TABLE III: Relative values of the areas of the most intense characteristic peaks of elements contained in specimen No. 8545 for various exposures, in % of the total area of peaks.

Exposure,		Chemical element				
sec	Al	Cu	Ag	$\mathbf{Pb}$		
400	6.95	45.71	3.46	43.88		
3600	7.60	48.52	3.52	40.36		
7200	11.33	59.83	2.19	26.65		

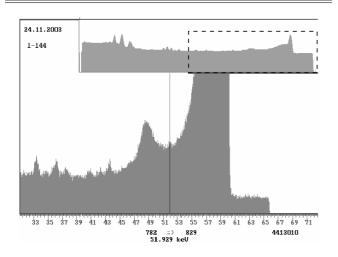


FIG. 9: Fragment of the spectrum of film which contains the 51.95-keV unidentifiable peak.

can be explained by melting of organic glass under the action of the electron beam, which led to 'the falling' of the studied particle into depth.

In the vicinity of the 20.0-keV peak, we could hypothetically observe 'the peaks of sums' equal to 20178 eV (Pb  $L_{\alpha 1}$  + Au  $L_{\alpha 2}$ ) and 20263 eV (Pb  $L_{\alpha 1}$  + Au  $L_{\alpha 1}$ ). However, the spectrum does not contain 'the peak of sums' corresponding to the duplicate value of the energy of the most intense Cu  $K_{\alpha 1}$  peak, which equals 16088 eV. This allows us to conclude that the 20.0-keV peak cannot be 'a peak of sums.'

The 51.95-keV peak (Fig. 9) was registered by X-ray fluorescence analysis with <sup>241</sup>Am source in the spectrum between intensive peaks of inelastic and elastic scattering while studying one of the accumulating screens.

This unidentifiable peak was reliably registered in the summarized spectrum after 24-hour exposure. The analysis of the spectra revealed that the peak was not registered in the spectra during the first 8 hours of the measurements. This peak appeared in the summarized spectrum of the next 8 hours along with very low-intensity 50.9-eV and 52.8-keV peaks that finally were not registered in the summarized spectrum of the summarized spectrum of the last 8 hours.

We consider the impossibility to establish the affiliation of the registered peaks to the characteristic lines of known chemical elements, on the one hand, and to artifacts of the analysis, on the other one, as the evidence for the manifestation of long-lived transuranium elements in the characteristic X-ray spectra. The values of energies of the unidentifiable peaks allow us to suppose that the peaks belong to the *L*-series of long-lived transuranium elements with atomic numbers in the range 100...150 or to the *M*-series of elements with those in the range 200...270. Generally saying, the peaks can be related to lines of the N-, O-, P-, etc. series of elements with arbitrarily large atomic numbers. The estimations of the atomic numbers of elements made under the assumption

the extrapolation of the energies of known characteristic lines to the region of superheavy elements are shown in Table II. The registered unidentifiable peaks are at the detection limit of the analyzing equipment used by us. However, such available methods as, e.g., the method of full external reflection in X-ray fluorescence analysis and the focusing of acting X-rays with lenses in order to enhance a dose of irradiation of an object, will allow one to radically increase the detection limit. In addition, we are busy now in solving the problem of separation of longlived transuranium elements with the purpose to derive

as to their affiliation to the lines  $L_{\alpha}$  or  $M_{\alpha}$  by means of

### **III. CONCLUSIONS**

enriched samples.

Based on the results of the study of the composition of the products of laboratory nucleosynthesis by X-ray electron probe microanalysis and X-ray fluorescence analysis, we drew the following conclusions.

- 1. We consider that the regular appearance of the peaks in characteristic X-ray spectra, which cannot be referred to the characteristic lines of known chemical elements or to artifacts of the analysis, is the evidence for the fact of registration of long-lived transuranium elements.
- 2. The appearance of the characteristic X-ray peaks supposedly related to the generation of long-lived transuranium elements is the fact testifying to the running of the reactions of nucleosynthesis and to the realized possibility of a controlled creation of conditions for their running in laboratory setups.
- 3. We established the fact of disappearance of unidentifiable peaks from characteristic X-ray spectra under the action of an electron probe or X-rays. In the former case, the disappearance of unidentifiable peaks depends directly on the energy of primary electrons. We also found that unidentifiable peaks disappeared significantly later in the latter case as compared to the former one.
- 4. We discovered a transformation of the chemical composition of the chemical environment of unidentifiable peaks under the action of an analyzing beam in the course of time.

- S. V. Adamenko, A. S. Adamenko, S. S. Ponomarev, and I. A. Kossko, To be published (2003).
- [2] S. V. Adamenko, A. S. Adamenko, G. A. Zykov, I. A. Kossko, and S. S. Ponomarev, To be published (2003).
- [3] S. Adamenko, Visnyk NANU (Bulletin of National Academy of Sciences of Ukraine) pp. 23–26 (2003).
- [4] S. Adamenko and A. Adamenko, in Proceedings of Int. Symp. New Projects and Lines of Research in Nuclear Physics (World Scientific, 2002), pp. 33–44.
- [5] M. A. Blokhin, Physics of X-Rays (GITTL, Moscow,

1957).

- [6] V. Scott and G. Love, Quantitative Electron-Probe Microanalysis (Wiley, New York, 1983).
- [7] M. A. Blokhin and I. G. Shveitser, *Reference Book on X-Ray Spectra* (Nauka, Moscow, 1982).
- [8] P. E. D. C. J. C. F. Joseph I. Goldstein, Dale E. Newbury and E. Lifshin, eds., *Scanning Electron Microscopy and X-Ray Microanalysis* (Plenum, New York, 1981).
- [9] J. I. Goldstein and H. Yakowitz, eds., Practical Scanning Electron Microscopy (Plenum, New York, 1975).