INVESTIGATIONS ON NUCLEAR SPECTROSCOPY AT THE REACTOR AND THEIR APPLICATIONS[1]

I.A. Kondurov, E.M. Korotkikh, Yu.E. Loginov, V.V. Martynov

Introduction

Physical launch of the WWR-M reactor in the branch of Physical-Technical Institute of AS of the USSR in Gatchina took place on the 29th of December in 1959. However the first work on nuclear spectroscopy was carried out before the reactor was launched; namely, gamma-radiation was measured of short-lived isomeric nucleus with lifetime of some tens of minutes produced by irradiation in the reactor mock-up processed at a power of 1-2 Wt. The internal conversion coefficient measured from X- and γ-ray intensity ratio coincided with the one measured earlier and L.I. Rusinov delivered a report about the beginning of scientific activities at the WWR-M reactor [1].

This was a new level of work with short-lived isotopes, as earlier the most short-lived nucleus investigated in the laboratory of L.I. Rusinov was ^{180m}Hf with $T_{1/2}$=5.5 h. Earlier this isotope irradiated in the reactor of Kurchatov Institute in Moscow was brought to Physical-Technical Institute in Leningrad by plane. That work was undertaken in order to bring a contribution in a study of K-forbiddenness. Indeed, it was wonderful that the 57.6 keV transition of E1 type determines such a large half-life [2]. Launch of a new reactor in Gatchina has given a possibility to deal with very short-lived isotopes using various pneumatic and other posts in which the minimal time of delivery of sample to measuring installation amounted to 40 ms [3].

Generally speaking, the works on nuclear spectroscopy in Physical-Technical Institute began as early as in 1934, when an investigation of properties of ^{80m}Br decay was carried out with participation of L.I. Rusinov [4]. That work was a push for development of studies about nuclear isomerism. So, all the following works of Laboratory of nuclear isomerism in PhTI in some way or other were connected with this interesting phenomenon. The first works in this direction made at the WWR-M reactor under the leadership of D.M. Kaminker were developed to the measurements of lifetimes of nuclear levels of doubly odd nuclei after radiative capture of neutron. It should be noted that the excited states of doubly odd nuclei are excited in the decay of radioactive nuclei very seldom, therefore, one nucleon transfer reaction are nearly the only source of information about such nuclear states. In these works carried out by A.M. Berestovoj and Yu.E. Loginov eight new nuclear isomers were found and investigated [5, 6].

Nuclear spectroscopic technique at the WWR-M reactor

The beginning of investigations at the WWR-M reactor coincided with the development and manufacturing in Physical-Technical Institute of the first semiconductor detectors widely used at the reactor channels. As early as in 1964, the first systematic study of γ-spectra from the (n,γ) reaction [7] was made using these detectors. Application of Ge(Li)-detectors with a high energy resolution permitted to create γ-spectrometers for measuring spectra from the (n,γ) reaction in the energy range of 5 – 10000 keV (up to neutron binding energy). Two coupled spectrometers formed the setup for measurement of prompt and delayed γ-γ-coincidences. A combination of the fast scintillator and semiconductor detectors gave a possibility to carry out lifetime measurements of excited states in milli-, micro-, nano- and subnanosecond time range. These investigations

[1)] The text of the present review was written by Igor Andreevich Kondurov and was intended for 25 anniversary of foundation of the Institute. Drs. E.M.Korotkikh, Yu.E.Loginov, I.A.Mitropolsky, V.V.Martynov and A.A.Rodionov do not change essentially anything; by only adding the material reflecting recent evolution of the investigations laid down by him.

were made by Yu.E. Loginov, P.A. Sushkov and V.V. Martynov [8].

Soon after the WWR-M reactor was put in operation, Cauchois bent crystal γ-spectrometer GSK-2 was designed by O.I. Sumbaev and A.I. Smirnov [9]. Afterwards it was modernized by V.L. Alexeev [10]. This spectrometer permitted to measure γ-rays in the energy range of 10 – 1000 keV with an accuracy of few eV. The possibility of simultaneous measurements in five orders of reflection increased substantially reliability of results obtained.

Measurements of conversion electron spectra from the (n,e^-) reaction became possible after installation in 1968 by B.A. Emelianov, V.S. Gvozdev, Yu.L. Khazov and S.L. Sakharov of the sector magnetic β-spectrometer completed with focusing lens using the through channel. This channel was specially drilled for this purpose through concrete shield of the reactor. Then the lens was substituted by the inner-channel transport solenoid [11] that resulted in increasing sensitivity of the spectrometer by a factor of seven. This brought closer characteristics of our spectrometer to ones of the (n, e^-) spectrometer BILL at Institute Laue-Langevin, Grenoble, of which target are placed in a neutron flux 300 times larger.

The presence of all the spectrometers mentioned permitted to carry out complex measurements of γ-rays and conversion electrons from the neutron capture reaction for one and the same nucleus. Experimental data obtained were evaluated with the system of collective usage created in the Neutron Research Laboratory on the base of commercial computers, which were significantly modernized and were equipped with special processors [12].

Processing of information obtained was made with a set of computer codes developed by L.P. Kabina, I.M. Shesterneva, T.M. Tyukavina, T.K. Korotkova and E.I. Fedorova [13]. This set of data processing codes was modernized many times as our computers were being improved and at present the codes still operate effectively.

It should be added that as a result of our investigations on a field of nuclear spectroscopy in the (n,γ) reaction our activity was recognized by scientific community. Soon we collaborated with European and American groups working in analogous directions. This permitted us to use also their experimental installations.

Level structure of doubly odd nuclei studied at the WWR-M reactor

The complex of nuclear spectroscopy devices at the WWR-M reactor gives a possibility of nuclear structure investigations by means of measurement of the radiation from the neutron capture reaction.

This work was fulfilled in close collaboration with several nuclear structure groups from ILL (Grenoble), BNL and LANL (both USA), MPI Heidelberg, TU München, Imperial College Reactor Centre (London), University of Fribourg (Switzerland) and groups from Riga and Kiev.

The level structure of doubly odd nuclei in A = 120 – 142 mass region was the subject of this research. Investigations resulted in construction of the excited level schemes of ^{104}Rh, ^{108,110}Ag, ^{114,116}In, ^{122,124}Sb, ^{128,130}I, ^{134}Cs and ^{142}Pr [14 – 20] up to excitation energies of about 1 MeV. The main feature of these level schemes is that almost all the levels excited in (n, γ) reaction are also seen in one nucleon transfer reactions. This permits to interpret these levels as members of p-n multiplets based on low-lying proton and neutron single particle states.

Correspondingly, the level schemes are divided into the level systems of positive and negative parities connected by few E1 transitions with retardation factors of $F^w \approx 10^5 - 10^6$. Intense intramultiplet transitions of M1 type proceed inside each level system with the fastest of them having $F^w < 10$. Branching ratios calculated for some multiplets of negative parity confirm the (d,p) and (d,t) reaction data about slight fragmentation of these multiplet states.

As noted above, the most of transitions in doubly odd nuclei are of M1 (sometimes E1) type. However, nine transitions of E2 type were found, with seven of them being enhanced (F^w ~

10 – 20). It is evident that the experimental half-lives cannot be reproduced in the framework of two-quasiparticle multiplets only. Really the fact that the measured B(E2) values are of the same order as the B(E2: $2^+_1 > 0^+_1$) ones for doubly even nuclei in the mass range A = 120 – 140 (see Fig. 1) shows that collective vibrational mode of excitation is as important in doubly odd nuclei as in doubly even ones.

In Fig.1 the B(E2) values are displayed in terms of the product of N_pN_n parameter, where N_p and N_n are the numbers of valence protons and neutrons, respectively, in the Z, N = 50 and 82 shells. Such a parameterization is chosen because the systematics of the B(E2: $2^+_1 > 0^+_1$) values for doubly even nuclei follows a smooth curve.

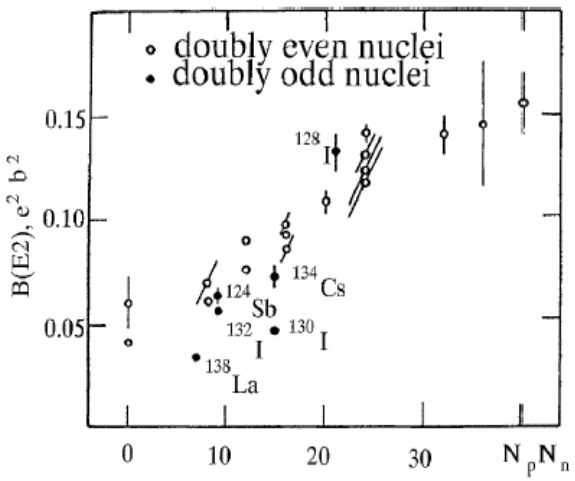

Fig. 1. The systematics of the B(E2: $2^+_1 \rightarrow 0^+_1$) values for doubly even nuclei and the B(E2) values for doubly odd nuclei in the A= 120–140 region in terms of the product of N_pN_n

New region of interest in collaborative works is doubly odd transitional nuclei ^{192,194}Ir. The level schemes up to 0.5 MeV were constructed for these two nuclei. Some properties of low-lying states can be explained assuming a small deformation and using Nilsson orbits expected for the proper nucleon numbers.

The found in ^{192}Ir isomeric nature of the 56.7 keV (1^-), 104.8 keV (1^-) and 118.8 keV(3^-) states indicated that these levels are heads of the corresponding rotational bands. The delayed γγ-coincidence experiment in ^{194}Ir aimed to search for γ-transitions populating the 32 ms isomeric state (E = 147 keV, 4^+) permitted to obtain the positive parity level system which cannot be detected in the (d, p) reaction.

Gamma transitions between the excited states of doubly even ^{118}Sn in the energy range of 0.1 – 10 MeV were measured in the framework of the experiment aimed to the investigation of P-odd effects in the output channels of the ^{117}Sn(n, γ)^{118}Sn reaction with thermal neutrons. Energy values and absolute intensities of γ-lines were determined. The level scheme of ^{118}Sn involving 46 excited states and 162 γ-transitions between them was constructed. Spin values and parities were assigned to all these states.

The absolute intensities of the 7.28 and 9.33 MeV γ-transitions were used for the interpretation of P-odd effect formerly observed for these γ-transitions [21]. The neutron binding energy in ^{118}Sn has been determined as 9326.12(4) keV [22, 23].

The photon strength function of twenty one M1 primary transitions does not follow punctually the E^{-3} dependence (see Fig.2).

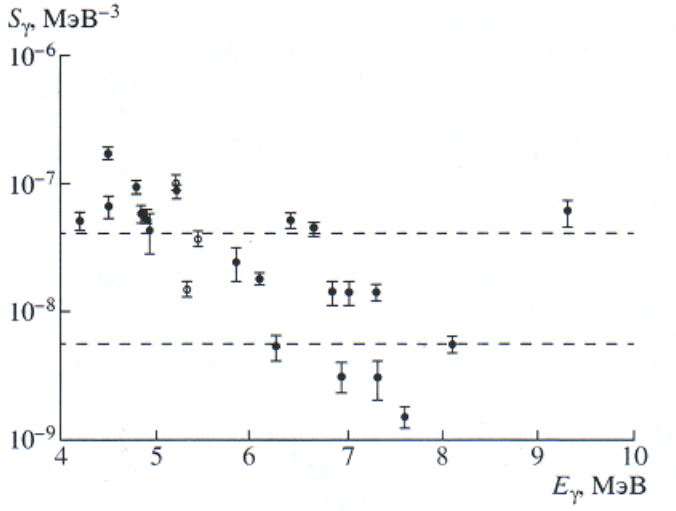

Fig. 2. The radiative strength function of primary M1 (black circles) and E1-transitions (open ones) in the ^{117}Sn(n,γ)^{118}Sn reaction.
The dotted lines show the upper and lower limits of M1 strength function values (see formula (2) in [23]).

Energy values and absolute γ-intensities of 359 γ-lines from the ^{183}W(n,γ)^{184}W reaction were measured in the range 0.03 – 7.5 MeV [24]. The comparison of energy values of γ-lines with $E_\gamma > 4.0$ MeV with energies of corresponding γ-lines determined with neutron energies about 2 keV [25] permitted to distinguish between primary and secondary γ-transitions. The neutron binding energy in the ^{184}W was obtained to be 7411.40(9) keV.

Acceleration of thermal neutrons by isomeric nuclei

One of the interesting works on the structure of nuclei is the investigation of the so-called acceleration effect of the thermal neutrons by isomeric nuclei performed at the WWR-M reactor. As predicted by Yu.V.Petrov [26] an inelastic scattering reaction with a noticeable cross section, in which the excitation energy is transferred to the scattered neutron, is possible as a result of the interaction of neutrons with nuclei in the excited state. Thus for ^{113m}In and ^{115m}In isomers with life-times of few hours the calculation based on the optical model and treatment of the experimental data for the reverse reaction give the value about 0.1 b for the cross section of inelastic acceleration in the energy region of ≈0.1 MeV. However, a direct reaction of neutron acceleration could not be observed in spite of several attempts. The first successful measurements, in which the acceleration of thermal neutrons was observed, were made by E.M. Korotkikh with ^{152m}Eu [27] (M3 transition, $I^\pi = 3^-$, $E_m = 48.5$ keV, $\tau_m = 13.42$ h).

The ^{152m}Eu isomer was obtained by irradiating 25 mg of ^{151}Eu samples for 20 h in the WWR-M reactor of PNPI. The thermal neutron flux measured with iron foils amounted to $(1–2)\cdot10^{14}$ neutr.·cm^{-2} s^{-1}.·After irradiation, the target containing tenths of milligrams of the isomeric nuclei was placed in the neutron beam. As the predicted effect is small, we modulated the thermal neutron beam by shutter in order to single out it. The accelerated neutrons were slowed down in plastic and were detected with proportional ^{3}He-counters. The counters were shielded against decay γ-rays, because the activity of the target after irradiation was nearly 1 kCi. The decay γ-ray intensity exceeded the predicted accelerated neutron intensity by a factor of 10^{12}. For shielding, a thick lead shield surrounded the target. The detector counting efficiency for fast neutrons was (6.6 ± 0.7) %. The efficiency was measured with a standard Sb-Be source. The counting rates, which were related with the corresponding shutter positions («beam open» and «beam closed»), were recorded separately step-by-step into the computer memory. The difference

of these counts contains the effect of interest. The component which decreases in time with τ_m = 13.42 h was selected by the least-square fit.

The cross section of the inelastic acceleration was determined by expression:

$$S = I^m/(N_{m0}\times\varphi_{th}) = \delta_r\times\delta_t\times\varepsilon\times\sigma_{in}$$

Here φ_{th} is the flux of thermal neutrons with the velocity of 2200 m·s^{-1}, which is incident on the target, δ_t is the ratio of average target flux $< \Phi_{th} >$ to Φ_{th}. The product $N_{m0} \times \delta_r$ gives the total number of the isomeric nuclei after irradiation in the reactor, N_{m0} is the number of the isomeric nuclei obtained disregarding the block-effect and δ_r consider this effect. Finally the quantity ε denotes the detector efficiency for the accelerated neutrons.

The weighted average of the value S from nine experiments was $< S > = (1.08 \pm 0.12) \times \times 10^{-26}$ cm^{-2} with $\chi^2 = 1.19$. This means that the effect of the acceleration of thermal neutrons was observed at the level of nine standard deviations.

The average neutron flux on the target was measured as the half-sum of its value at the edges: $\delta_t = 0.83 \pm 0.03$. The target self-shielding and blocking coefficient in the reactor during irradiation $\delta_r = 0.7 \pm 0.1$ was obtained by measuring the activation of the ground state of ^{152g}Eu. From these measurements of the cross section of the neutron acceleration at the velocity of 2200 m·s^{-1} we obtained the value $\sigma_{in} = (0.28 \pm 0.06)$ b.

The theoretical estimation made by means of the formulas from [26] gives $\sigma_{in} \approx 0.3$ b. Although theoretical value is valid with an accuracy of an order of magnitude only, its agreement with the cross section measured shows that the (n, n′) reaction offers no appreciable hindrance to the nuclear transition ^{152m}Eu > ^{152g}Eu, whereas the γ-transition is hindered by a factor of more than 106 ÷ 108.

For investigation of influence of the electromagnetic transition hindrance on the neutron acceleration process the analogous experiment [28] was performed with the ^{180m}Hf isomer which lives relatively for a long time (τ_m = 7.7 h) due to K-hindrance. The isomer nuclei were prepared by irradiation of the ^{179}Hf in the light-water trap of the PNPI WWR-M reactor. After irradiation, the target was placed in the modulated beam of thermal neutrons. The neutrons were transferred to the target by means of the logarithmic neutron guide. The accelerated neutrons were slowed down in a plastic environment and were detected with proportional ^{3}He-counters. The acceleration cross section measured for thermal neutrons turn out to be $\sigma_{in} = (52 \pm 13)$ b in a good agreement with theoretical estimation. This means that K-hindrance, which is about 10^{16} for the E1($8^- \rightarrow 8^+$)-transition, is absent for the neutron acceleration process. It is eliminated, when the (^{180}Hf + n) system goes through the stage of the highly excited compound nucleus, where the states with the different K-values are totally intermixed.

Hyperfine shift of K-lines of internal conversion electrons

Using radioactive sources produced at the WWR-M reactor a work is carried out by A.A.Rodionov on the investigation of hyperfine effects that are given rise to interactions of the nucleus magnetic moment with the internal K- and L-shells. It was also shown [29,30] that in K-capture as well as in K-conversion the nonstatistical population of subshells of hyperfine structure is possible that results in a shift of a level by a value of the order of the hyperfine interaction constant and that shift should be seen in the conversion spectra. The work is an attempt of reaching the accuracy of measurements of internal conversion line energy differences, necessary for discovering the effect of the hyperfine shift of conversion lines. Estimates show that value of the (relative) hyperfine shift of the K-line should be of the order of 10^{-6}.

In order to increase the accuracy of the conversion line energy measurement, a modernization of systems of power supply, of collection and information processing of the prism spectrometer [31] was fulfilled. The internal conversion electron processing codes which take *a priori* information into account were created. The measurements were taken with ^{169}Yb, as the exhausting information about its level scheme was available. Due to hyperfine interaction the K-conversion transitions from the 316 keV level to the 138 keV (μ = +1.39 n.m.) and 118 keV (μ = +0.76 n.m.) levels suffer a shift in energy relative to each other, estimated to be 0.21 eV. The L-conversion lines of these transitions are shifted too, but the shift is of the order of 0.034 eV.

Processing of many runs of internal conversion electron measurements shows that the accuracy of localization of line positions in each run amounts to about 0.2 eV. The energy differences δ((L198 - L177) - (K198 - K177)) obtained in each run with the accuracy of 0.5 – 0.8 eV were being determined, with the value of δ averaged over all the runs amounting to 0.32(15) eV. This value agrees with the theoretical estimate of energy difference within experimental uncertainty.

The investigation of the nucleus magnetic moments by the method of conversion lines hyperfine shift measuring is planned at the universal prism spectrometer, which is produced for use at the PIK reactor [32].

Nuclear physics methods for analytical chemistry

The experience from years of nuclear spectroscopy investigations led us to use of nuclear physics methods for analytical purposes. The methods are instrumental neutron activation analysis (INAA), neutron radiation analysis (NRA) and X-ray fluorescent analysis (XRF).

Neutron activation analysis of chemical element contents in a sample is based on measurement of decay γ-rays from radionuclides, which are produced in a neutron capture process during irradiation of a sample inside a nuclear reactor. The intensities of γ-rays are proportional to the element contents, which gives the minimal experimental uncertainties in element contents measured. In the INAA method, an influence of a matrix on results is negligible, because neutrons and γ-rays are used as exciting and characteristic radiation correspondingly. Moreover, the minimal pretreatment prevents samples from their contamination with outside elements. The sensitivity of the INAA varies to a considerable extent for different elements depending on the properties of atomic nuclei and reaches the value of a few $\mu g \times kg^{-1}$ (ppb).

The samples of interest and neutron flux monitors (pure metallic Fe and Zr and the standard reference materials, SRMs) of 10 ÷ 100 mg are packed into high-purity quartz ampoules and are irradiated for hours or days at a neutron flux of $(2 – 7)\cdot 10^{13}$neutr.$\cdot cm^{-2}\cdot s^{-1}$ depending on the task. After cooling off time (hours or days) the r-ray spectra are measured 3 times for each sample in the energy region of 30 – 2200 keV.

The first run of measurement is made to detect mainly Na, K, As, Br, Mo, Sb, Ho, Hg, Au and U. The planar HPGe detector with the energy resolution of 0.7 keV at 59 keV and 8192-cannel analyzer are used.

The second run of measurement is usually made after 8 days of cooling time with the 15 % coaxial HPGe detector with the energy resolution of 1.7 keV at 1333 keV. Sodium, Cr, Fe, As, Br, Rb, Sb, Ba, La, Ce, Nd, Sm, Tb, Dy, Yb, Lu, Hf, Ta, Au, Th and U are detected here.

The third run is made after 21 days of cooling time. The contents of Sc, Fe, Co, Zn, Se, Zr, Sr, Ag, Sb, Cs, Ce, Eu, Tb, Tm, Yb, Hf, Ta, Hg and Ir are obtained here.

The evaluations of the spectra measured are made with the original computer codes [13] designed in PNPI. These codes are based on using an *a priori* information about samples of interest and their γ-ray spectra. All the significant peaks (not only the analytical ones) are considered in a spectrum, when concentrations of elements are calculated in least squares fit. The

calculation is produced on the base of annually up-dated version of ENSDF (Evaluated Nuclear Structure Data File) [33] that is obtained from Brookhaven National Laboratory of the USA. The neutron cross sections and resonance integrals are taken from [34]. Remaining small discrepancies (experimentally possible) are eliminated using a SRM sample. The element contents for other (control) SRMs are always checked with this procedure and are usually in a good agreement (within error limits) with the reference data. When needed the spectrum of an empty quartz ampoule is taken into account as the background.

The results are the weighted averages of the element contents obtained for all the measurements made. The final uncertainties of the results include weighted errors of the experimental and reference data on the one-sigma level (68% confidence limit).

The detection limit for every element in one measurement is estimated as a three-times-square-root of the background level in the region of maximal sensitivity. The overall detection limit is the minimal value from all the experimental estimates for the sample.

The method was tested with many SRMs [35] and was practically used [36, 37]. However, everywhere one should keep in mind sample homogeneity (representativity) problems, when analyzing unique samples.

World-wide intercomparison exercise

The INAA group of the NRD of PNPI is one of the participants of the worldwide intercomparison exercise.

Quality assurance and quality control cover a very broad discipline; there are numerous factors and methods that must be employed to validate data and ensure adequate analytical performance. An intercomparison exercise serves as such independent external assessment. This should be a continuous and regular process, not only to maximize the confidence in a particular data set, but also to maximize the confidence in an analytical process.

For nearly thirty years the Marine Environmental Studies Laboratory (MESL) of IAEA-MEL has conducted intercomparison exercises that have resulted in the preparation of reference materials (RMs) for the analysis of trace elements and organic compounds as part of its contribution to IAEA's Analytical Quality Control Service.

In 2003, the sample material of marine sediments from the Algerian coast of the Mediterranean Sea was prepared and distributed to analytical laboratories worldwide. The data sets reported by laboratories and the technical and statistical evaluations of the results for each element and laboratory are given in [38]. In total, 103 laboratories from 47 counties reported results for 48 elements defined with 28 different analytical methods. The wide range of methods employed permits a statistically valid comparison of the principal method used, particularly the instrumental methods following sample digestion and the "non-destructive" techniques such as neutron activation analysis and X-ray fluorescence. For illustration, the reported values are graphically depicted in Fig. 3 (a – for Sm, b – for Sr and c – for Zn), where the results from the PNPI-INAA group for 28 elements were evaluated by IAEA-MEL.

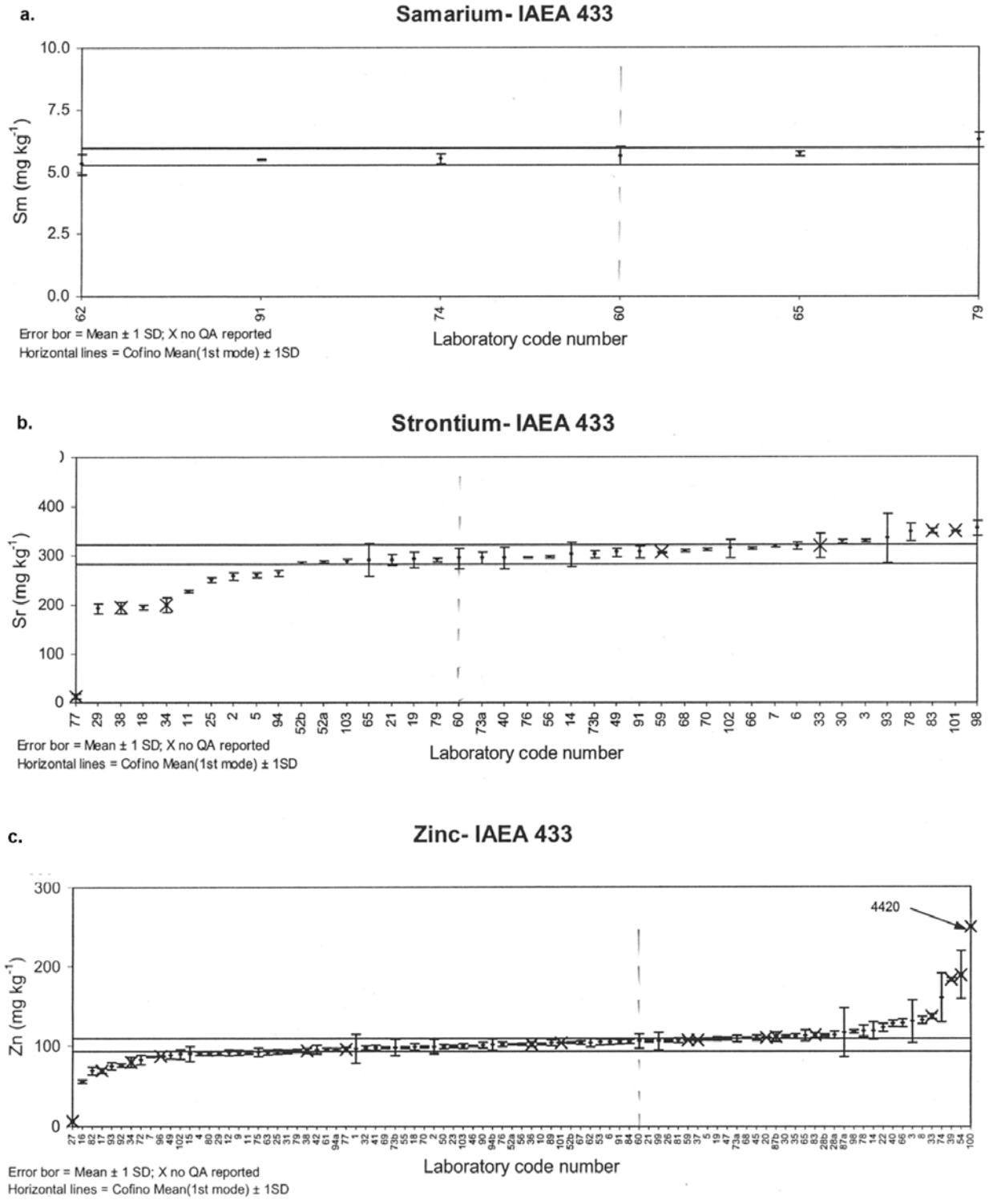

Fig. 3. The results from the PNPI-INAA group for 3 elements are presented:
a) for Sm, b) for Sr and c) for Zn.
The vertical axis shows the content of the element (in mg $kg^{-1)}$; in horizontal – the conventional number of a laboratory taking part in the exercises.
The INAA group of the NRD of PNPI has here the number 60.

Investigation of potentially toxic trace elements and their behaviour during coal combustion

The rising interest to main and trace element contents is connected with growing requirements of modern technological processes and environmental problems. There is a pressing necessity of producing more and more electricity by organic-fuel-fired power stations. Power generation from coal-fired power stations constitutes a potential mobilization mechanism for the potentially toxic trace elements (PTTEs), which are dangerous for a human health and an environment. For example a coal-fired power stations of 1000 MW of electric power consumes daily 12,000 t of a poor coal and thus mobilize a great amount of PTTE into the atmosphere.

In the work [39] the coal samples from the Teruel coal basin (NE Spain) were investigated. This coal is used to supply three power stations with the electric power of 1050, 175 and 143 MW and annual consumption of $5 \cdot 10^6$, $0.9 \cdot 10^6$ and $0.8 \cdot 10^6$ t, respectively. The aim of this study was an investigation of the behaviour of PTTEs during the coal combustion process. The elements of interest were As, B, Be, Ba, Cd, Co, Cr, Cu, Ge, Hg, Li, Mn, Mo, Ni, Pb, Se, Sr, Th, U, V and Zr. The measurements were made with INAA (in PNPI) and ICP-MS and/or ICP-AES (in Barcelona, Spain) methods. The results of these methods are in a good agreement. The sensitivities of the INAA for coal samples are shown in Table 1.

Table 1

The sensitivities of the INAA method for main and trace element contents in coals from Teruel basin in mg/kg (ppm)

Element	ppm	Element	Ppm	Element	ppm	Element	ppm	Element	ppm
Na	80	As	20	Sb	0.05	Gd	10	W	0.3
K	400	Se	1	Cs	0.2	Tb	0.1	Re	0.03
Ca	2000	Br	0.2	Ba	20	Dy	0.2	Ir	0.005
Sc	0.3	Rb	0.5	La	0.5	Ho	0.1	Au	0.02
Cr	2	Sr	100	Ce	2	Yb	0.01	Th	2
Fe	600	Mo	2	Nd	3	Lu	0.05	U	0.2
Co	0.5	Ag	2	Sm	0.2	Hf	0.1		
Zn	20	Cd	1	Eu	0.5	Ta	0.1		

Another example of using INAA results is the investigation of the trace element contents in atmospheric suspended particles. This study was focused on the determination of trace elements by INAA in two different areas in NE Spain (areas influenced by the emission of a large coal-fired power station and the urban and industrial areas of Castellión). The total suspended particles were sampled using cellulose membrane filters at the distances of 14, 26 and 54 km from the Teruel power station. The sensitivity of the measurements were at the level of ng.m-3 for Cr, Fe, Zn, As and Zr and of $fg \cdot m^{-3}$ for Sc, Co, Sb, Cs, Hf, Th and U. For details one can refer to [34].

These investigations were made by V.V. Martynov, L.P. Kabina, G.I. Shulyak, P.I. Piven and P.A. Sushkov.

Total reflection XRF in multielement analysis of microsamples

The total reflection on a highly polished backing is characterized by a high reflectivity, which leads to a drastic reduction of the spectral background. The sample to be analyzed is prepared on the backing as a residue of small quantities by the evaporation from solution or fine-grained suspensions. The microanalysis can be performed because < 1 mg of a solid sample or < 10 ml of a solution are required. Instrumental detection limits of few pg are state of the art for

this method. Besides the high detection power, the internal standardization is another important feature of TXRF, enabling very simple quantification of the detected elements.

TXRF analysis requires the excitation with a very narrow beam with a low angular divergence. When the beam impinges on the sample backing under a glancing angle below the critical angle of the total reflection, it is reflected passing twice through the sample. The X-ray tube radiation contains the high-energy bremmstrahlung, which requires a very small angle to be reflected. A quartz mirror, which serves as a high-energy cut-off filter, is used for eliminating these high-energy photons. That is why a very small part of X-ray tube radiation is actually used for preparing the necessary excitation beam. For increasing the intensity of the excitation beam one can use more powerful X-ray tubes.

Another approach for increasing the intensity of the source is to use X-ray optics for concentration of the excitation beam at the sample position. When the curved mirror is used as a high-energy cut-off filter, the most suitable shape of a beam is the logarithmic spiral due to its inherent property that the incidence angle for X-rays emitting from a source placed in the origin of the spiral is constant relative to the curve (logarithmic spiral). We used an optical glass plate coated with the Nb layer of a 110 nm thick bent into the logarithmic spiral as a cut-off filter [40]. The detection limits in the region of 10^{-12} g are obtained for some elements (see Fig. 4).

The application of focusing optics is always associated with an increase in the angular divergence, which is tolerable for TXRF to only a limited extent. The possibility to improve the divergence by means of an additional curved mirror has been studied theoretically [41]. These results are used in the X-ray optical device for preconditioning a parallel beam [42, 43]. The experimental set-up is schematically shown in Fig. 5. The measurements show that the intensity of the excitation beam at the sample position increases 4 times and further improvements are also possible [44].

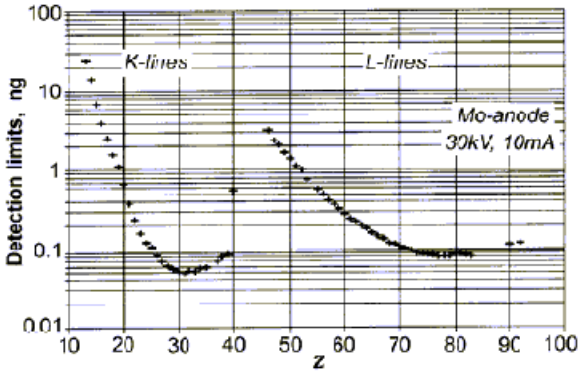

Fig. 4. The detection limits obtained for some elements

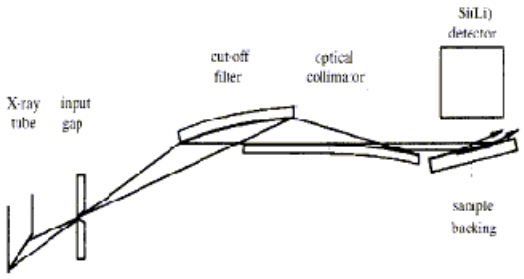

Fig. 5. Schematic view of the X-ray spectrometer with a parallel beam

The scheme of logarithmic neutron guide with "white" parallel beam of thermal neutrons was elaborated as application of focusing optic methods for neutrons [45]. The analytic expressions which described the configurations and the sizes of bent mirrors, the width of the beam and intensity distribution on the width were suggested. The estimates show that this neutron guide can produce 5 – 9 times intense beam of thermal neutrons compared to the conventional collimating.

The total reflection XRF method was used for quantitative determination of the ability of microorganisms to bind ions of heavy metals, e.g. of Sr and Cs [46].

The samples of yeast Pichia farinosa No 46 grown in the medium containing SrO and $CsCl_2$ taken for investigation are irradiated by X-rays and spectrum of secondary X-rays was measured by Si(Li)-detector.

The quantities of Sr and Cs bonded by yeast were obtained with method of inner standard. As standard was used gallium in concentration 10 milligram/liter introduced into the sample under investigation. Gallium was chosen because of it does not occur in natural and biological samples. The lower limits of the detectable concentrations of Sr and Cs were obtained to be 1.3 and 3 milligram/liter correspondingly.

The results of measurements show that the nature yeast strain could be used for bioremediation of industrial wastes solutions polluted by salts of heavy metals. The results could be a foundation for the developing a new technology for the concentration of heavy metals from the nature and industrial wastes.

Applied in-beam researches of elemental and isotopic analysis at the WWR-M reactor

The method of precision in-beam γ-spectroscopy is based on the measurement of prompt γ-rays from the (n, γ) reaction. This method can successfully be used for quantitative determination of elemental and isotopic contents. It has the best results for elements with large neutron capture cross sections (such as B, Cd, Sm, Gd, etc.). However, it can also be used for investigation of different elements, when other analytical methods (INAA, XRF, ICP-MS, etc.) are inconvenient.

The method is based on a comparison of γ-lines intensities in γ-ray spectra from the sample of interest and the sample of well known element contents (SRM). The samples in the special holder are placed one by one in a narrow neutron beam formed by neutron-guide. Capture γ-rays are registered with the semiconductor detector. The possibility of such a comparison is provided by the identity of volumes of samples under investigation.

The example of an application of this method is the element content determination in the synthetic LaBaMnO3 monocrystal. The artificial mixture of compounds with the equal amounts of La, Ba and Mn was used as the standard. The measurements of two specimens annealed in vacuum and oxygen resulted in the following chemical formula: $La_{0.708(6)}Ba_{0.30(2)}Mn_1O_3$ for the first sample and $La_{0.705(8)}Ba_{0.315(24)}Mn_1O_3$ for the second one.

The control of the antimony contents in the Sb doped sample of high-temperature-super-conductive (HTSC) ceramics after two stages of the annealing can be presented as another example [47]. Here the initial mixture of compounds for HTSC synthesis was used as the SRM, with antimony contents being known precisely. The measurements resulted in C(Sb) = 0.89(3)% after the first stage of the annealing and C(Sb) = 0.90(2)% after the second one, which means that the annealing process does not change the antimony contents in the mixture.

This method was also applied for the boron content determination in the river sediments from Argentina [48] and in the Chinese geochemical standards [49]. Our neutron capture γ-ray facility has the following sensitivity (in ppm) for trace elements: B is 1, Gd is 0.01, Sm is 0.3 and Cl is 20.

If each isotope of the element captures neutrons individually, the isotopic content measurements are possible. The objects of investigations are commonly isotopic changed samples. The targets with natural isotopic contents, which are known very well, are usually used as standards. The results of the isotopic content determination for a «nickel-62» sample can be presented as the next example. Natural Ni was used as the standard. The contents measured are as follows: ^{58}Ni is 1.03(22)%; ^{60}Ni is 0.70(35)%; ^{62}Ni is 91(6)%. The appropriate data of the certificate are 1.07%, 0.65% and 98.1%, respectively.

The method of a quantitative determination of the ^{6}Li contents in several lithium compounds is based on the neutron transmission through lithium containing samples. The measurements have been developed both with reactor neutrons and with neutrons from a Pu-Be neutron source [50]. The ^{6}Li contents, which can be determined, are ranged from 0.1 to 96 %.

Finally, the in-beam γ-spectroscopy method can be used for the determination of thermal neutron capture cross sections or the absolute intensities of capture γ-rays. The measurements of absolute γ-intensities from the (n,γ)-reaction are necessary both at the applied searching for elemental or isotopic content and in nuclear structure investigations with neutrons. Up to now, these data according to many isotopes are absent.

This method was applied for the determination of absolute γ-intensities of 18 γ-lines from the ^{35}Cl(n,γ)^{36}Cl reaction in the energy range 0.4 – 2.7 MeV with the target consisting of the mixture of $PbCl_2$ and H_3BO_3 [51]. The γ-line with the energy 478 keV from the ^{10}B(n,αγ)^{7}Li reaction served as γ-intensity standard. Obtained results have the accuracy twice as better as known before.

The absolute intensities of 9 γ-lines from the ^{55}Mn(n,γ)^{56}Mn reaction in the range 0.1-0.5 MeV were measured with the aim to refine these values which are used in the quantitative elemental analysis of Mn-containing samples [52]. As inner γ-intensity standard was used γ-line with the energy 845 keV from the decay of ground state of ^{56}Mn ($T_{1/2}$ ~2.6 h,) which is observed in the measured γ-spectrum in equilibrium with the capture radiation at the measuring time ~24 hours.

The thermal neutron cross sections of the ^{14}N and ^{19}F were measured with targets consisted of mixtures of $Pb(NO_3)_2$, CaF_2, LiF and Teflon with Al_2O_3 [53]. Thermal neutron cross section of Al is known well (0.231(3)b). The γ-line with the energy 1779 keV from decay of ^{28}Al ground state ($T_{1/2}$ ~3 min) which was used as γ-intensity standard, has the 100% branching ratio (I_γ = 100%). This γ-line is observed in the measured γ-spectra in equilibrium with the capture radiations.

As result, the thermal neutron radiative cross sections of ^{14}N and ^{19}F were obtained to be 0.0795(13)b and 0.00950(15)b, respectively. These values are significantly more precise than formerly known ones – 0.0750(75)b and 0.0096(5)b, respectively.

PNPI Centre of data on nuclear structure and nuclear reactions

One of the applications of our experience of investigations and developments in the field of nuclear spectroscopy was the development of works on nuclear data. The Data Centre of PNPI established in 1972 carried out the work on collection, analytical estimate and spreading of data on nuclear structure and on nuclear reactions. The Data Centre organizes its activities in the framework of the international network NSDD (Nuclear Structure and Decay Data Network), which comprises 15 laboratories and groups, that represent interests of 10 countries.

In accordance with its obligations the Data Centre is responsible for estimates of properties of mass chains A = 130 . 135. The estimation involves the collection of data about the structure of nuclei entering in chain of nuclei produced in decay and in nuclear reactions, and receiving of estimated data about the levels of nuclei studied and about their properties. Estimates obtained are written down in the ENSDF format (Evaluated Nuclear Structure Data File) That permits directly reproduced the edition «Nuclear Data Sheets» [54]. Twice a year the Data Centre receives the updated version of ENSDF file distributed by NSDD, including the estimates of all the known nuclei. A number of codes for dealing with ENSDF files developed that permitted to obtain systematics of data about any nuclear properties.

In the frame of the international network the PNPI Data Centre also carried out refereeing of papers on nuclear structure and on nuclear reactions, published in Russian and soviet editions. Refereeing in keywords is carried out in NSR format (Nuclear Structure References). More than 10000 references of Soviet and Russian papers were directed to the network. Afterwards they are published in journal «Nuclear Data Sheets» (Recent References). Twice a year NSDD sends references added to NSR library to participants of the network. The total NSR library in the Centre contains more than 100 000 references of papers on nuclear structure and nuclear reactions, beginning from 1910.

Raw experimental data obtained directly in the experiment are the most valuable, as they did not contain any model considerations and can always be reprocessed taking into account new models and concepts. This information together with codes for handling spectra forms database of primary spectroscopic information of the Data Centre of PNPI. The ENSDF data can be used for energy calibration of gamma, electron and other spectrometers as well as in the final stage of nuclear spectroscopic studies, i.e., in construction and analysis of schemes of excited states of nuclei. Created in the Data Centre is a set of codes which permits one to calculate transition energies and intensities in the level scheme, to calculate all the possible internal conversion coefficients, probabilities of β-transitions, multipolarity mixing, coefficients of angular correlations and so on [55].

The investigations of nuclear monopole excitation were historically consolidated in PNPI. For that time the most full compilation of experimental data on 0^+ states and E0-transitions in doubly even nuclei were presented in paper [56]. It also contains a survey of all the nuclear models describing monopole excitations. The most exhausting compilation of properties of monopole excitations of doubly even nuclei (more than 1000 levels) was created on the basis of ENSDF in 2000. Electronic tables involve the known energies of 0^+ levels and their lifetimes.

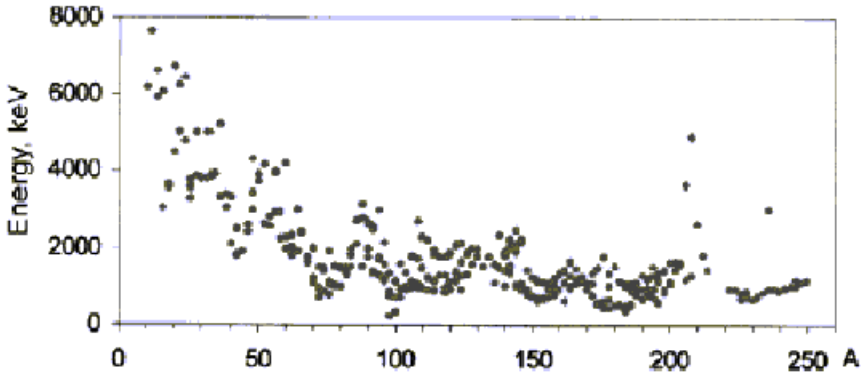

Fig. 6. Excitation energies of the first 0^+-levels in double even nuclei depending on mass number A

For example, a dependence of energies of the first excited 0+ levels E(0+) in even-even nuclei on the mass number A is presented on the Fig.6. The transition energies, relative intensity, type of transition and its reduced probability are listed for each decay channel. Given for E0 transitions also are values of amplitudes ρ(E0) and the ratio of X(E0/E2) [57].

Study of non-statistical features in spectra of low-lying nuclear excitations is new and apparently very advanced direction of investigations in the theory of nucleus. Here, interesting results [58,59] has already been obtained in the Data Centre of PNPI. The tuning effect was found in energy values of nuclear excitations and in binding energies that makes more probable an

appearance of values multiple to electromagnetic difference of nucleon, pion and lepton masses. In this field proceeds an accumulation of experimental facts and search for theoretical concepts, that demonstrate the most clearly the role of quantum chaos and dynamic stability in producing nuclear spectra.

At last, the activities on generalization of traditional models and their modifications to conditions of systematic analysis of experimental data. In particular, generalization of the model of variable moment of inertia for describing rotational bands of odd nuclei was achieved [60]. On the basis of the ENSDF file in 2000 the most full compilation of rotational bands in odd nuclei (more than 2000 bands) including spins, parities and energies of rotational levels (in total more than 16000 levels) were created. Distribution of the quantity of rotational bands on mass number A presented in Fig.5 reveals regions of deformed nuclei. These experimental data are systematized on the basis of the model of variable moment of inertia [61]. A good description of rotational energies is achieved. The systematic approach permitted us to remove a large number of ambiguities in primary experimental data and to eliminate unavoidable mistakes arising when introducing such large volumes of information. Last years due to progress in computer networks the character of exchange of information in NSDD is sharply changed. The exchange of operative information proceeds now via e-mail, the exchange of files does through INTERNET. This leaves

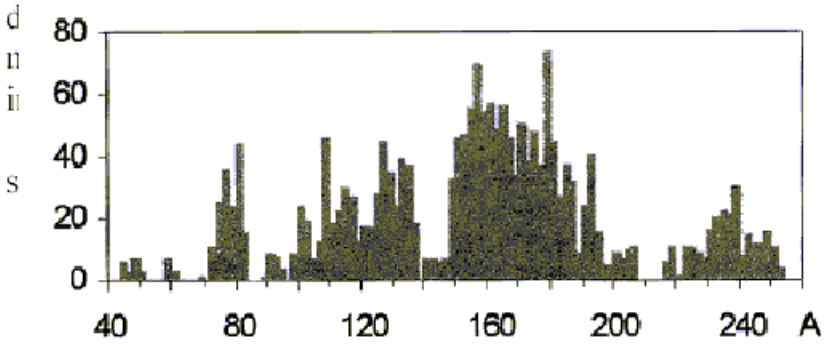

Fig. 7. Mass dependence of the rotational bands quantity in odd nuclei

an imprint to the activities of the Data Centre. In particular the Data Centre now has its Web-page **http://nrd.pnpi.spb.ru/topic/gr_mitr.html**, where the information is placed about the group itself as well as about its current activities. Use of standard program sets for the storage of information and for control of databases (Excel, Access) makes it easy to transfer information to users and to visualize systematic obtained in the form of graphs, tables and so on.

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