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МЕТОДИКА ФИЗИЧЕСКОГО ЭКСПЕРИМЕНТА

PRECISE DETERMINATION OF NEUTRON BINDING ENERGY OF ^{64}Cu

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The neutron binding energy in ^{64}Cu has been accurately measured in thermal neutron capture. A composite target of natural Cu and NaCl was used on a high-flux neutron beam using a large measuring time. The γ-ray spectrum emitted in the (n,γ) reaction was measured with a HPGe detector in large statistics (up to 10^6 events per channel). Intrinsic limitations of HPGe detectors, which restrict the accuracy of energy calibration, were determined. The value B_n of ^{64}Cu was determined as 7915.867(24) keV.

Энергия связи нейтрона в ^{64}Cu аккуратно измерена при захвате тепловых нейтронов. На высокопоточном нейтронном пучке была использована составная мишень из естественного Cu и NaCl в течение длительного времени измерений. Спектр гамма-лучей, испускаемых в (n,γ)-реакции, измерялся детектором HPGe до получения большой статистики (до 10^6 событий на канал). Определены внутренние свойства HPGe-детекторов, которые ограничивают точность энергетической калибровки. Получено значение B_n ^{64}Cu, равное 7915,867(24) кэВ.

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INTRODUCTION

Modern intrinsic germanium detector systems enable us to measure γ rays with energy up to 10 MeV with precision of $\leqslant 0.1$ keV relative to a standard set of energies. In the work devoted to determination of the neutron binding energy in ^{118}Sn [1], B_n in ^{64}Cu was determined as well with a value of 7915.52(8) keV using a HPGe detector. This value differs from the adopted value [2] of 7915.96(11) keV which was obtained in mass measurements [3]. From this discrepancy we decided to carry out a devoted measurement of B_n for ^{64}Cu using very large statistics. Furthermore, in order to improve the referential lines in the calibration, we make use of accurate value of B_n for ^{36}Cl obtained on high-resolution crystal spectrometer in the (n,γ) reaction [4]. Well-established γ rays in the reaction ^{35}Cl$(n,\gamma)^{36}$Cl were used as a standard set of energies. Making use of a pure thermal neutron beam and a long measuring time, we undertook to study neutron capture by copper and chlorine with large statistics. In processing the resulting γ spectrum some difficulties must be resolved. The precision of energy measurement became limited not by statistics, but by the characteristics of the HPGe detector. Two main aims were pursued in this work: to achieve energy accuracy as best as possible and to determine the limitations on energy precision in measurements by HPGe detectors.

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1. EXPERIMENTAL

The experiment was carried out at the light water reactor LWR-15 in Řež near Prague. The facility was described in [5]. It includes a curved neutron guide through which thermal neutrons are transported from the reactor core to the target, a detector system with associated shielding and the electronics. The guide is formed by a mirror-type tube of rectangular cross section, circularly bent in the vertical direction. The internal cross section of the guide is 150×4 mm. The overall length of the guide is 5.63 m, the curvature radius being 825 m. At the target position the neutron flux at reactor power of 8 MW is $(2.8 \pm 0.5) \cdot 10^6$ cm$^{-2} \cdot$ s^{-1}. At the entrance to the target area, dimensions of the incoming neutron beam are additionally reduced to 20×2 mm by a $^6Li_2CO_3$ plate.

A polyethylene bag with sizes $20 \times 20 \times 2$ mm with NaCl and a copper wire with 2-mm diameter and 20-mm length were used as a target. Captured γ rays were detected by a 28% HPGe detector (Canberra). Overall time of measurement was about 12 h. The spectrum obtained in this measurement is shown in Fig. 1.

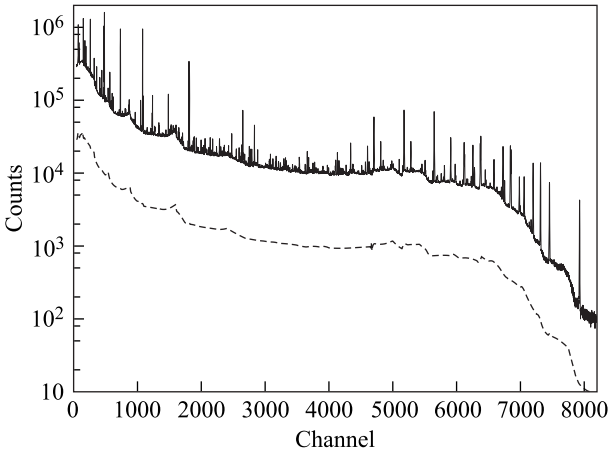

Fig. 1. Spectrum Cu, Cl (n, γ). Fitting background is shown shifted one order down

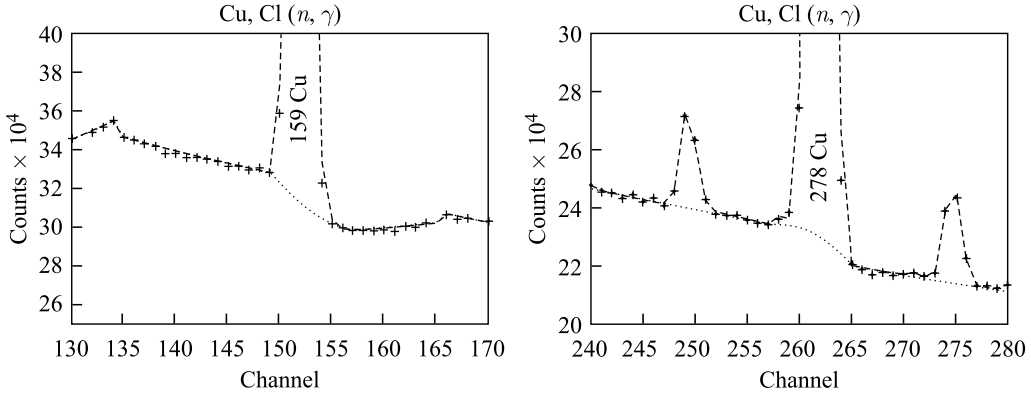

Fig. 2. Peaks with obvious step from scattering γ's

It can be seen that this spectrum is very complicated not only owing to a great number of peaks, but also because of very complicated feature of background. There are the Compton steps from intense transitions, there are "tails" on the right sides of intense double escape peaks. There is one else source of step on the left side of very strong peaks. This step is attributed to small-angle scattering of the γ rays by the target and entrance windows [6]. The peaks at 159 and 278 keV exhibit a marked step in the spectrum as shown in Fig. 2.

2. PEAK FITTING

The spectrum was fitted using the FORTRAN computer code SPANAL [7]. Gaussian integrated in limits of each channel is used to form a peak. Gaussian can be spread by exponential for obtaining asymmetric peak. Initial values of the full width on a half of maximum (FWHM) and asymmetry must be given. Code gives the best values of position, area, FWHM and asymmetry of each found peak. Background is described by segments of parabolas which have the same derivative in the points of junction. In view of this, points of break of background (see Fig. 1) must be chosen as the ends of intervals in adjustment.

Initial values of FWHM and asymmetry can be given as polynomial functions and can be fixed, but it was found that on spectrum with large statistics the fixation cannot be used, because FWHM has not smooth behavior. It is known that FWHM of annihilation peak is always greater than that of peaks from reaction, besides, these secondary γ transitions have individual FWHM from the Doppler broadening which is determined by lifetime of level from which this transition decays, and this broadening is used to measure the lifetimes of levels [8–10]. Furthermore, single escape (S) peaks and double escape (D) peaks are much broader than full energy (F) peaks at the same region of spectrum. Values of FWHM as a function of energy of γ transitions for hard part of spectrum are shown in Fig. 3. (Information about E_γ from ^{36}Cl was taken from [11, 12] and about E_γ from ^{64}Cu — from [2].) At last,

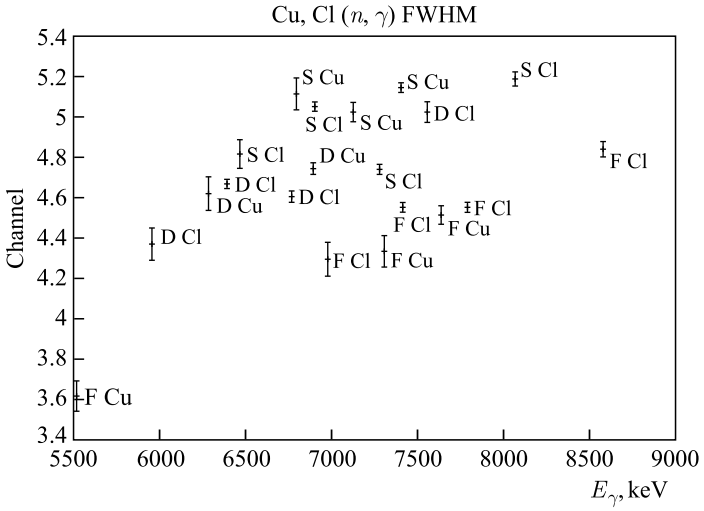

Fig. 3. Dependence of FWHM on E_γ. Single escape (S), double escape (D) and full energy (F) peaks are indicated

in view of big density of peaks on spectrum, probability of superposition of peaks is high. This can increase the value of FWHM.

In fitting of the spectrum, attention was paid to the values of χ^2 for each interval. This value was considered as acceptable if it was less than $2N$, where the degree of freedom N is the number of channels in the interval. From a total of 80 intervals, only 14 have large values of χ^2. All intervals in channels 3000–8000 have acceptable χ^2. As long as for the determination of the ^{64}Cu binding energy, we used positions of some peaks, only six peaks, which were used further, are from intervals with large values of χ^2. It was found that large values of χ^2 are in the intervals with peaks which have large statistics and non-Gaussian form. For some intervals, if one big peak is fitted as a sum of two or three peaks, the value of χ^2 becomes acceptable. Examples of these intervals are shown in Table 1.

Table 1. **Comparing of the values of χ^2 for the same intervals in dependence on the number of peaks found in the interval. (P — position of peak, S — area)**

P	ΔP	S	ΔS	FWHM	ΔFWHM	χ^2
Interval 257–265 channels						
258.341	0.109	4331.1	728.2	1.006	0.173	2155.2
261.715	0.001	2150308.0	1714.9	1.489	0.002	
259.833	0.105	16042.4	1027.3	2.297	0.278	
261.628	0.001	1682613.1	1589.0	1.301	0.002	2.3
262.646	0.003	436142.4	1230.6	1.268	0.007	
Interval 480–486 channels						
482.497	0.000	3659295.5	2020.9	1.632	0.001	5736.8
478.725	0.106	581368.1	3419.3	1.604	0.059	
482.503	0.000	3408197.3	1974.3	1.604	0.001	0.2
483.940	0.004	243463.1	932.3	1.046	0.005	
Interval 710–738 channels						
714.195	0.020	30178.0	490.5	1.748	0.055	
725.797	0.091	6845.4	440.3	1.884	0.900	
728.757	0.105	12882.4	580.7	2.835	0.225	289.3
731.192	0.001	993296.3	1273.9	1.588	0.003	
733.285	0.001	1944156.4	1587.3	1.946	0.002	
714.360	0.020	29036.5	479.7	1.996	0.049	
720.738	0.269	2223.9	453.8	2.077	0.780	
723.536	0.175	3806.9	472.3	2.249	0.426	
726.495	0.076	9212.5	488.2	2.356	0.187	
728.843	0.072	5955.4	407.8	1.408	0.180	21.4
731.304	0.001	941844.1	1201.1	1.698	0.003	
733.216	0.001	1725429.1	1557.5	1.810	0.002	
734.304	0.004	270329.0	1082.3	1.762	0.009	
739.746	0.158	2471.1	392.5	1.502	0.455	

It can be ascertained from the results in Table 1 that in spectrum with large statistics (to 10^6 in one channel) fitting with acceptable values of χ^2 is possible. Hence, there is no big differential nonlinearity in amplitude-digital converter and other effects which can disturb

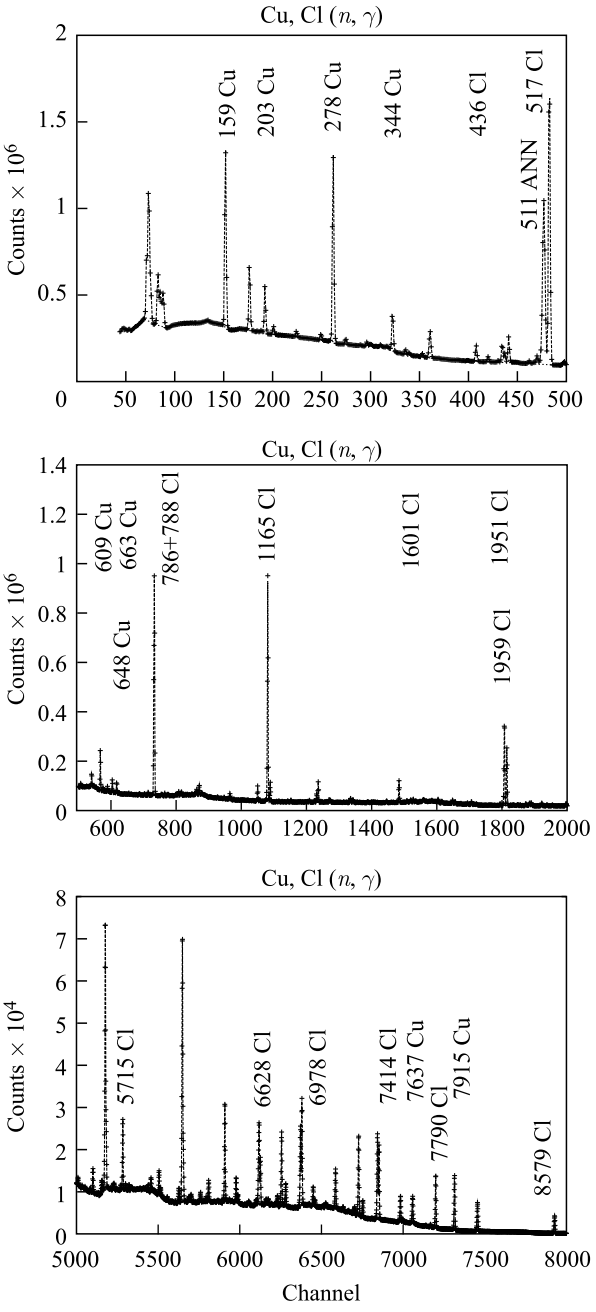

Fig. 4. Three parts of spectrum Cu, Cl (n, γ) with peaks which were used in calibration procedure

statistical properties of the spectrum. Sensitiveness of finding peaks was consciously reduced in intervals in which one or two big peaks must exist. One can hope that in spite of the large value of χ^2, positions of centroids of peaks are well.

Three parts of spectrum with peaks which were used for the determination of B_n are shown in Fig. 4.

What precision do we expect to obtain? An accurate calibration in 5–8 MeV region is needed for the determination of B_n for ^{64}Cu. There are 24 peaks in this region with error of position less than 30 eV, but precision of result will depend on how many peaks can be used in calibration procedure. It is known that energies of single escape (S) and double escape (D) peaks are not precisely 511 and 1022 keV less than full energy (F) peaks [13]. We tested these differences in our spectrum and the result is illustrated in Fig. 5, *a* and *b*. In this figure, the values $E_{\rm F}-E_{\rm S}-511$ keV and $E_{\rm F}-E_{\rm D}-1022$ keV are shown as a function of energy of F peak. The mean value of $E_{\rm F}-E_{\rm S}-511$ keV is 164 eV and of $E_{\rm F}-E_{\rm D}-1022$ keV is 144 eV. In view of this, S and D peaks were not used in calibration procedure.

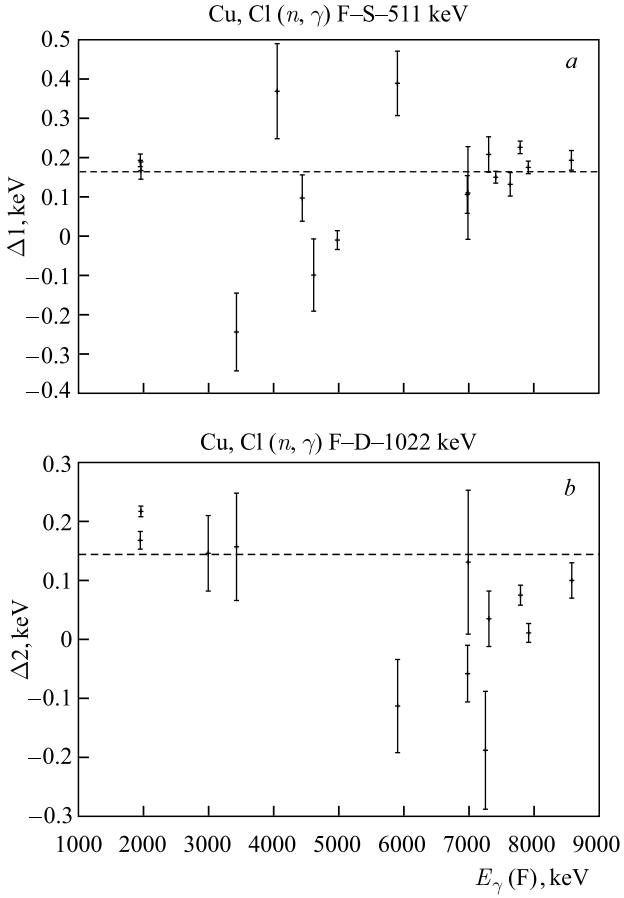

Fig. 5. Dependence of distances $\Delta 1 = E_{\rm F}-E_{\rm S}-511$ keV (*a*) and $\Delta 2 = E_{\rm F}-E_{\rm D}-1022$ keV (*b*) on E_γ(F). The mean values are shown by lines

3. ENERGY CALIBRATION

Calibration procedure described in [1] was used for the determination of B_n for ^{64}Cu. This procedure is a mathematically simple minimization function consisting only of terms having as parameters the coefficients of the energy calibration curve (polynomial). A calibration procedure is used for finding a correspondence between the peak positions on a spectrum and the energies. The Least-Squares Method (LSM) is used to determine coefficients of the calibration polynomial. This procedure is intended to determine one value, namely B_n. In the LSM the best value of one parameter gives minimal value of the chi-square function Q, and the second derivative in this point is connected with the standard deviation (SD) of the parameter. In the procedure the value of parameter is changed by steps and each value of function Q is calculated. From interpolation of all these values by parabola, the values of B_n and ΔB_n are determined. In order to take into account the errors of energies, which are common for some peaks, the Monte Carlo simulation is used in which right connection among all energies is reproduced in each simulation.

Input data for the calibration procedure may be divided into two groups:

(1) The variables, corresponding to some well-known standard energies. Part of them can have independent errors and other part are points, energies of which are connected.

As independent variables in group (1) intense secondary transitions in ^{64}Cu and ^{36}Cl can be used because their energies are known with high precision. Primary transitions in ^{36}Cl are connected to each other through the value of B_n. As B_n for ^{36}Cl the value (8579.7945 ± 0.0048) keV [4] was used.

(2) The variables which present peaks from isotope, B_n of which is determined. All energies of these peaks are connected to the value of B_n.

Recoil for hard γ rays was taken into account.

One else correction was included in data values: to all transitions with error less than 10 eV, the value of 10 eV was attributed, otherwise there was large value of χ^2 for calibration function. This correction concerns the transitions with energy less than 2 MeV.

The most complicated part of this work was selection transitions as data for calibration procedure. There is a great number of peaks and if all peaks are included as data in the procedure, good calibration curve cannot be obtained. It is necessary to find some number of peaks with small errors which form a curve. In view of big density of peaks in spectrum, reason for excluding points with big deviation from expected curve almost always can be found since a small contamination to tested peak can visibly shift its position. Three variants of selected points and obtained calibration curves are shown in Fig. 6.

A special system of coordinates is used in calibration procedure and in Fig. 6 — deviation of energy calibration function from a linear function fixed at the first and the last points. It is more convenient to analyze results of calibration in this system. Only two points from ^{63}Cu(n, γ) reaction were used in calibration procedure. It seems that points below 3000 channel must not affect B_n of ^{64}Cu. But there are two reasons to use points from all spectrum: calibration procedure is needed for energy calibration of all spectrum and calibration curve in wide range gives more confidence to obtained results. Results of the determination of B_n for ^{64}Cu are shown in Table 2.

The behavior of the values χ^2 of fit of seven peaks which give the main influence in B_n was tested and results are shown in Table 3. It can be seen that sum of χ^2 of the last three

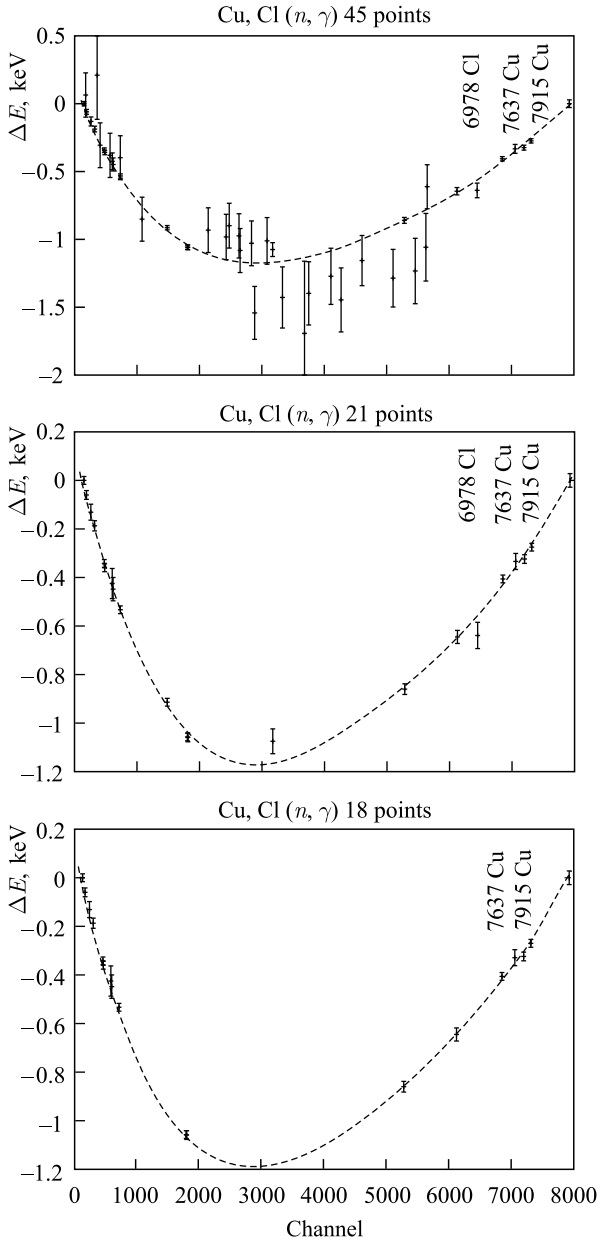

Fig. 6. Deviation of the energy calibration function from a linear one. Three variants of selected points and obtained calibration curves are given

peaks is somewhat high, but there are no suspicions that peaks have more complicated form than that used in fitting.

One can compare results from variants (2) and (3) (see Table 2). It is seen that removing of point 6978 keV (near 6500 channel) shifts result on 4 eV. Variant (4) in which only points

Table 2. **Results of the determination of B_n for ^{64}Cu**

Variant	N points	χ^2 pdf*	B_n, keV	ΔB_n, keV
1	45	1.249	7915.864	0.020
2	21	1.074	7915.863	0.020
3	18	0.527	7915.867	0.020
4	8	1.247	7915.871	0.020

* Per degree of freedom.

Table 3. **The test of fit of seven peaks — sum of χ^2 of N channels on intervals (from — to)**

E_γ	P	ΔP	From	To	N channels	$\sum \chi^2$
5715	5284.150	0.018	5278	5288	11	6.16
6628	6126.829	0.024	6123	6131	9	4.91
7414	6852.695	0.015	6849	6858	10	2.62
7637	7058.612	0.028	7050	7063	14	6.98
7790	7200.183	0.016	7192	7205	14	16.09
7915	7315.484	0.015	7307	7320	14	13.06
8579	7927.955	0.026	7921	7932	12	13.30

higher than 5700 keV were used gives also shift on 4 eV. It is not an error of statistical nature. In view of this, for final result the value of 4 eV was added to 20 eV linearly. The final result of this measurement for B_n of ^{64}Cu is (7915.867 ± 0.024) keV.

4. DISCUSSION

From spectrum of captured neutrons by copper and chlorine which was measured with large statistics (to 10^6 in one channel), B_n of ^{64}Cu was obtained as (7915.867 ± 0.024) keV. It is evident that precision of energy calibration is limited not by statistics, but characteristics of the HPGe detector. Value B_n can be obtained with precision of 10–20 eV. For achieving of such a result some problems must be taken into account:

(1) In fitting a spectrum from the HPGe detector the value of FWHM of peaks on spectrum has not smooth behavior in dependence on number of channels.

(2) It is reasonable to use the value of 10 eV for errors of positions of secondary transitions despite of high statistical precision (lower than 10 eV).

(3) One needs to be in luck to find some number of peaks with small errors which form a curve in calibration procedure.

One else point can be discussed now in more detail: the use of SE and DE peaks in calibration procedure. In our work these peaks were not used, because their energies do not differ from energies of FE peaks on m_0c^2 and $2m_0c^2$, respectively (see Fig. 5): energies of SE and DE peaks are below the expected value on about 170 and 140 eV, respectively. There was accurate work [14] in which it was demonstrated that in measurements of spectra from (n, γ) reaction by the Ge detector the spacing between escape peaks is m_0c^2 within an error of 15 eV.

On the other hand, the mechanism for obtaining the DE peak energy higher than E(FE)–1022 keV was suggested in [13]. It is the annihilation of the positrons on bound electrons. In such a process a small part of the annihilation energy must be used to overcome the electron binding energy which results in electron-hole pairs. In addition, the energy of DE peak may further be increased due to the Compton scattering of the annihilation photons to forward angles in the detector active volume.

In our case, we have more simple mechanism to obtain energies of DE and SE peaks, which are lower than expected. Our detector was relatively old and it was used in many experiments on neutron beam and its resolution became worse from radiation damage. This effect can be detected, first of all, in the peaks from the most higher energy. It is demonstrated in Fig. 3 that SE and DE peaks are wider than FE peaks in the same energy interval. From the bad resolution the mean position of peak is shifted down and we have the energy, which is lower than expected.

Now one may compare value obtained in this work with the previous results. Value of B_n for ^{64}Cu in [2] was 7915.96(11) keV and that one in [1] was 7915.52(8) keV. Value of 7915.867(24) keV obtained in this work is more close to the value from [2] and on about four standard deviations differs from that in [1].

Preliminary results of this work were presented in [15].

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