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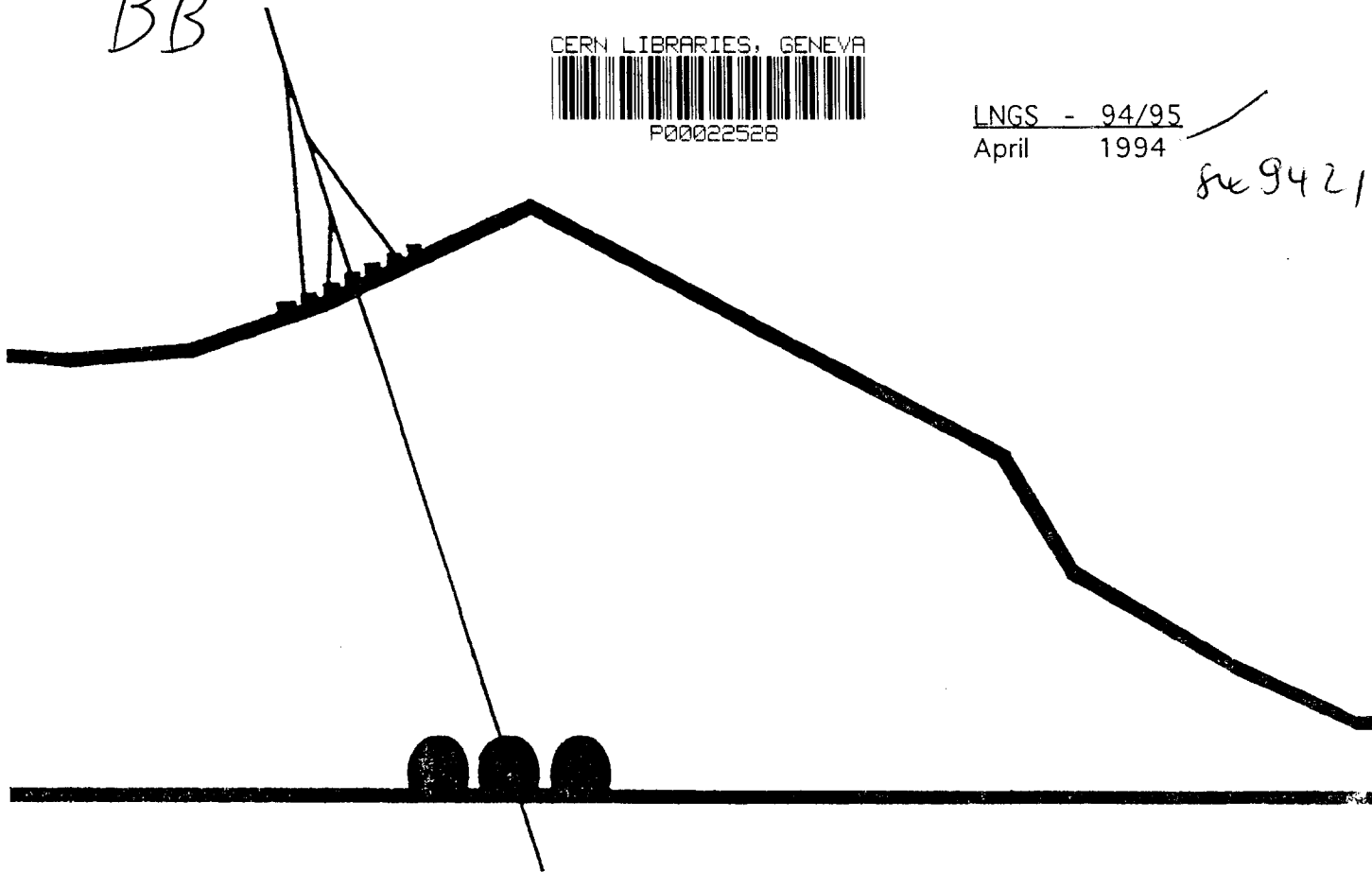


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Search for $\beta\beta$ decay of ^{96}Zr and ^{150}Nd to excited states of ^{96}Mo and ^{150}Sm

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Abstract. - New lower limits on the half-lives for double beta decay of ^{96}Zr and ^{150}Nd to excited states of ^{96}Mo and ^{150}Sm have been obtained. They range between $1 \cdot 10^{19}$ and $4 \cdot 10^{19}$ years (90% C.L.). The measurements were performed searching for the de-excitation gamma rays of the daughter nuclide with a germanium detector. In addition single beta decay of ^{96}Zr has also been studied, providing a half-life for this process longer than $3.8 \cdot 10^{19}\text{y}$ (90% C.L.).

1.Introduction

Double beta decay is nowadays considered one of the most fascinating processes of Nature: the proposed [1], but yet-unseen neutrinoless decay mode has intrigued theoreticians and experimentalists since many years ago, because of its implications on the lepton number conservation and neutrino properties [2]. Neutrinoless double beta decay, which would violate by two the lepton number, can be the consequence of a finite neutrino mass and/or of the presence of right-handed weak currents. The principal contribution to this decay should come from the transition from the ground state of the candidate nucleus ($J^\pi = 0^+_{\text{gs}}$) to the ground state of the daughter nucleus ($J^\pi = 0^+_{\text{gs}}$), but transitions to excited states of the daughter nucleus are also possible, when kinetically allowed and not forbidden by some selection rule. The neutrinoless $\beta\beta$ decay to the 2^+ level of the daughter nucleus is very interesting because it can go only through right-handed currents. The detection of $\beta\beta$ decay to the 0^+_1 excited state gives similar information about $\beta\beta$ decay, as the observations of the decays to the ground 0^+_{gs} state.

Since the energy E released in $\beta\beta$ decay to the excited levels is less than for the ground state transition, the space phase is also substantially less. In first approximation, the rates for the lepton conserving 2ν and the lepton violating 0ν decay mode are proportional to a terms ranging from E^8 to E^{11} , and from E^5 to E^7 , respectively. In addition the first calculations of the $\beta\beta(2\nu)$ decay to the 2^+ excited states indicate rates strongly suppressed with respect to

the transition to 0^+_{gs} [3,4]. Only recently it has been shown [5] that this suppression factor may be not so large and that therefore even this decay could be in principle detectable in low background experiments. The study of $\beta\beta$ decay to the 0^+_{1} excited states of the daughter nuclei [6] has been recently suggested for ^{96}Zr , ^{100}Mo and ^{150}Nd [7]. These nuclei are attractive candidates because of their large transition energies to the 0^+_{1} excited states of the daughter nuclei (2202, 1903 and 2627 keV, respectively) from which measurable half-lives (10^{20} - 10^{21} years) can be calculated.

A possible signature for $\beta\beta$ decay to excited levels is the emission of a de-excitation gamma rays from the daughter nuclide. Since the energy of the gamma rays is well defined, a good background rejection can be achieved if the detector has a high energy resolution, as, for instance, germanium diodes. Few results exist for $\beta\beta$ ($0^+_{gs} \rightarrow 0^+_{1}$) decay [2], with the only evidence [8] for $\beta\beta$ decay of ^{100}Mo to the 0^+_{1} state at 1131 keV of ^{100}Ru with $T_{1/2}$ $(8.1 \pm 2.4_{-1.5}) \times 10^{20}$ years obtained using a low background HPGe detector. The nuclear matrix element for this decay has been calculated assuming a two neutrino $\beta\beta$ decay [9] and found to be similar to the corresponding one for $\beta\beta$ decay to the ground state. This is confirmed by the fact that the ratio of the measured rates for the ($0^+_{gs} \rightarrow 0^+_{gs}$) [2] and ($0^+_{gs} \rightarrow 0^+_{1}$) transitions is reasonably consistent with the ratio of the corresponding phase spaces, as expected theoretically if the two matrix elements are the same. We can apply the same procedure to predict the rate for the ($0^+_{gs} \rightarrow 0^+_{1}$) two neutrino $\beta\beta$ decay of ^{96}Zr to ^{96}Mo , from the recent evidence [10] obtained geochemically for $\beta\beta$ decay of ^{96}Zr with $T_{1/2} = (3.9 \pm 0.9) \times 10^{19}$ y. Assuming that this value refers only to two neutrino $\beta\beta$ decay to the ground state one can predict $T_{1/2}(2\nu; 0^+_{gs} \rightarrow 0^+_{1}) \sim 10^{21}$ y, somewhat larger than predicted in [7]. The reliability of the geochemical result should be checked however by new experiments. The half lifetime for ($0^+_{gs} \rightarrow 0^+_{gs}$) two neutrino decay of ^{150}Nd to ^{150}Sm has been recently measured to be $\sim 10^{19}$ [11]. On the basis of the expected dependence on the space phase one can predict an lifetime of $\sim 10^{20}$ y for $\beta\beta(2\nu)$ decay of ^{150}Nd to the first 0^+_{1} excited level of ^{150}Sm .

In this paper the results of the search for $\beta\beta$ decay of the ^{96}Zr to excited states of ^{96}Mo , and also preliminary results on the ^{150}Nd $\beta\beta$ decay to excited states of ^{150}Sm are presented. In Fig.1 the decay schemes for triplets of ^{96}Zr - ^{96}Nb - ^{96}Mo and ^{150}Nd - ^{150}Pm - ^{150}Sm are shown. The search for these decays was carried out using a Ge- spectrometer to look for γ - rays with energies corresponding to these decay schemes. The results obtained with the Zirconium sample were also used to establish a limit to the single β - decay half -life for ^{96}Zr .

2. Experimental setup and methods

To detect the gamma-ray signature of double beta decay to an excited state, a 314 cm³ high-purity germanium detector was used. A sample of 18.74g of ZrO_2 powder, enriched in ^{96}Zr at the level of 57.3%, and contained in a disc-shaped plastic bag, was placed on the cap of the detector. A shielding of OFHC copper and low activity lead with 7 and 25 cm minimum thickness, respectively, was mounted all around the detector to minimize background radiation. To avoid Radon contamination, a plastic cover, into which nitrogen vapours were flushed, surrounded the shielding. The detector was placed in the Gran Sasso Underground Laboratories (3500 mwe) [12] to reduce cosmic-ray background. Data were accumulated for

2503 hours of effective running time using a 12bit-multichannel analyzer and were recorded for off-line analysis. Energy calibration was obtained using a Least-Square-Linear fit to 16 different background gamma lines between 122 keV and 2615 keV. In a similar way, the energy resolution at different energies was extracted from a fit to the different FWHM measured for those 16 background gamma lines. The actual detection efficiencies for our extended and rather thick sample were computed with a Monte Carlo code [13]. The program was experimentally tested with a calibrated point-like gamma-ray source placed at different locations on the top face of the detector. Unfortunately not all the dimensions and materials composing the detector are precisely known and an estimation on these unknown data could only manage to make the calculated efficiencies differ from the measured ones within a 10% uncertainty, which should be taken into account as a systematic error in our final results. The absolute efficiencies - here defined as the ratio of counts in the full-energy peak divided by the number of de-excitations from a particular state of the daughter nucleus - are evaluated by taking into account gamma-gamma correlations [14] in all the cases in which the deexcitation goes through the emission of more than one gamma. Table 1 reports the approximatedly weighted full energy efficiencies, taking into account γ branching ratios and possible γ - γ correlations.

A preliminary search was also carried out on $\beta\beta$ decay of ^{150}Nd to excited states of ^{150}Sm following the same technique and the same procedures for analysis as for ^{96}Zr . In this case single beta decay is energetically forbidden and the transition energies available for the decay to the first excited levels of ^{150}Sm (see fig.1) are particularly high (for instance $Q_{\beta\beta} = 2627$ keV for the transition to the 0^+_1 excited level of ^{150}Sm). ^{150}Nd is present in natural neodimium with 5.64 % atomic percentual abundance, which makes an experiment using natural neodimium meaningful. This preliminary measurement, totalling 1282 hour of effective running time, was carried out using two rods of natural metallic neodimium, 330g each, sealed in a thin plastic bag to avoid oxidation and placed on the end cap of a 414 cm³ High Purity Ge detector.

3.Results

The relevant portions of the spectrum observed from the zirconium sample are shown in fig. 2. All background gamma lines have been identified to come from the decays of long-living isotopes such as ^{232}Th , ^{238}U , ^{235}U or ^{40}K , from fallout contaminants (^{125}Sb , ^{106}Rh and ^{137}Cs) and cosmogenic radioactive isotopes (^{57}Co , ^{58}Co , ^{60}Co , ^{54}Mn). The comparison of the spectrum with the background of the detector has shown that the ZrO_2 sample has only a tiny contamination in ^{40}K , which doesn't affect the total background level. No evidence is found for any of the de-excitation γ - lines expected for $\beta\beta$ decay of ^{96}Zr to excited states of ^{96}Mo . In correspondence of the principal deexcitation γ - line (778.2 keV), no background peak is present and the continuum is 1.3×10^{-2} counts/keV·h, which permits to reach a good sensitivity for the measurement. In fact, even if several excited states of ^{96}Mo can be populated in double beta decay of ^{96}Zr , they all decay with large branching ratios to the $J^\pi = 2^+$ level at 778.2 keV of ^{96}Mo and no relevant information is added by the study of the other involved

gammas. An exception can be made for the ($2^+_{2} \rightarrow 0^+$) transition, for which a combination of the results obtained on the three gamma lines gives the best limit.

The number of counts to be attributed to the transitions under study have been obtained applying the method of the Maximum Likelihood as stated by the Particle Data Group [15], using as free parameters the number of counts under the gaussian peak representing the gamma line and the two coefficients of the linear fit of the background in an energy window around the energy of interest. For each gamma peak the expected position and width have been fixed as stated in the previous section. The lower limits on the half-lives of ^{96}Zr established in this way are summarized in table 1 and compared with the previous published results [16]. Our limits are more than an order of magnitude higher than the earlier results and higher than the present limits with direct experiments on $\beta\beta(0\nu)$ and $\beta\beta(2\nu)$ decays of ^{96}Zr to the ground state in ^{96}Mo ($1.4 \cdot 10^{19}$ y [17] and $1 \cdot 10^{17}$ y [18], respectively). Our limit on the half life for the 0^+_{1} excited state transition ($T_{1/2} > 3.8 \times 10^{19}$ y) is roughly equivalent to the value obtained in the geochemical experiment [10].

To reach a sensitivity of $\sim 10^{21}$ y, as required to detect the $\beta\beta(2\nu; 0^+_{\text{gs}} - 0^+_{1})$ decay of ^{96}Zr , one would need one year of counting with ~ 150 g of ^{96}Zr . Practically the same sensitivity may be obtained by using 30-40 kg of high purity natural zirconium.

The results obtained from the present study can also be used to establish a limit on the single beta decay of ^{96}Zr . The product of this process is ^{96}Nb , which decays with a 23.4 h half-life to excited states of ^{96}Mo , that cascades through the $J^{\pi} = 2^+$ level at 778.2 keV in more than 96.7 % of the decays. Thus the absence of this peak in our spectrum allows us to set a limit of 3.8×10^{19} y (at 90 % C.L.) for the single beta decay of ^{96}Zr . This is more than an order of magnitude higher than the limit established by e.s. Norman and B.M. Meekhof [16]. It does not allow however to exclude that the result found geochemically be due to two successive single beta decay.

The spectrum obtained in the preliminary run with ^{150}Nd is reported in Fig.3. The comparison of this spectrum with the radioactive background of the detector itself shows small contaminants in the neodymium rods of ^{232}Th and U chains, not in secular equilibrium, and of Rare Earth isotopes, as ^{138}La and ^{176}Lu . All these contaminations are at a level of fractions of Bq/kg each. The present preliminary lower limits on the half-lives of the transitions to the first excited states of ^{150}Sm , reported in table 3, are more than an order of magnitude more stringent than in a previous work [6]. The technique used to extract these limits is the same as above.

4. Conclusions

The results reported here constitute the best lower limits up to date for $\beta\beta$ decay of ^{96}Zr and ^{150}Nd to the excited levels in daughter nuclei and also for single β decay of ^{96}Zr .

The test on natural neodymium has convinced us of the feasibility of an experiment on ^{150}Nd : we have already started a measurement with 6 kg of natural neodymium to further increase the limit presented in this paper. In parallel, studies for the purification of the material will be carried on. If a substantial improvement in the background will be reached, a further

measurement with a sensitivity more than an order of magnitude higher than the present one will be performed.

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$\beta\beta$ transition	deexcitation γ [keV] of the daughter nucleus	B.R.	weighted effic.
$0^+ \rightarrow 2^+_1$	778.2	100 %	4.60 %
$0^+ \rightarrow 0^+_1$	369.7	100 %	5.68 %
	778.2 (following the first γ)		3.69 %
	1147.9 (sum of the 2 γ)		0.35 %
$0^+ \rightarrow 2^+_2$	719.5	71 %	2.79 %
	778.2 (following the first γ)		2.67 %
	1497.7	29 %	1.05 %
$0^+ \rightarrow 2^+_3$	847.6	95 %	3.27 %
	778.2 (following the first γ)	95 %	3.50 %
	1625.7	4 %	0.32 %

Table 1

$\beta\beta$ transition	$t_{1/2}$ (90 % C.L.) (this work)	$t_{1/2}$ (68 % C.L.) (this work)	$t_{1/2}$ (68 % C.L.) (previous work ^[16])
$0^+ \rightarrow 2^+_1$	$\geq 4.1 \times 10^{19}$ yr	$\geq 7.2 \times 10^{19}$ yr	$\geq 2.0 \times 10^{18}$ yr
$0^+ \rightarrow 0^+_1$	$\geq 3.3 \times 10^{19}$ yr	$\geq 5.8 \times 10^{19}$ yr	$\geq 1.8 \times 10^{18}$ yr
$0^+ \rightarrow 2^+_2$	$\geq 2.5 \times 10^{19}$ yr	$\geq 4.9 \times 10^{19}$ yr	$\geq 1.3 \times 10^{18}$ yr
$0^+ \rightarrow 2^+_3$	$\geq 3.1 \times 10^{19}$ yr	$\geq 5.5 \times 10^{19}$ yr	$\geq 1.7 \times 10^{18}$ yr

Table 2

$\beta\beta$ transition	deexcitation γ [keV]	B.R.	weighted effic.	$t_{1/2}$ (90% C.L.)
$0^+ \rightarrow 2^+_1$	333.96	100 %	0.01963	$\geq 8 \times 10^{18}$ y
$0^+ \rightarrow 0^+_1$	406.51	92 %	0.02019	$\geq 8.8 \times 10^{18}$ y
	333.96 (following the first γ)		0.01817	
$0^+ \rightarrow 4^+_1$	439.39	100 %	0.02086	$\geq 2 \times 10^{19}$ y
	333.96 (following the first γ)		0.01749	
$0^+ \rightarrow 2^+_2$	712.19	87 %	0.01685	$\geq 1.3 \times 10^{19}$ y
	333.96 (following the first γ)		0.01656	
	1046.15	7 %		

Table 3

FIGURE CAPTIONS

Fig. 1: Nuclear schemes for $\beta\beta$ decay of ^{96}Zr (a) and ^{150}Nd (b)

Fig.2 : Low energy region (a) and high energy region (b) of the spectrum obtained with the sample of ^{96}Zr

Fig.3 : Spectrum obtained with the natural sample of neodymium

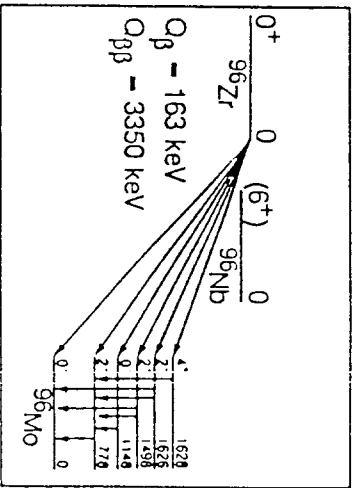


Fig. 1a

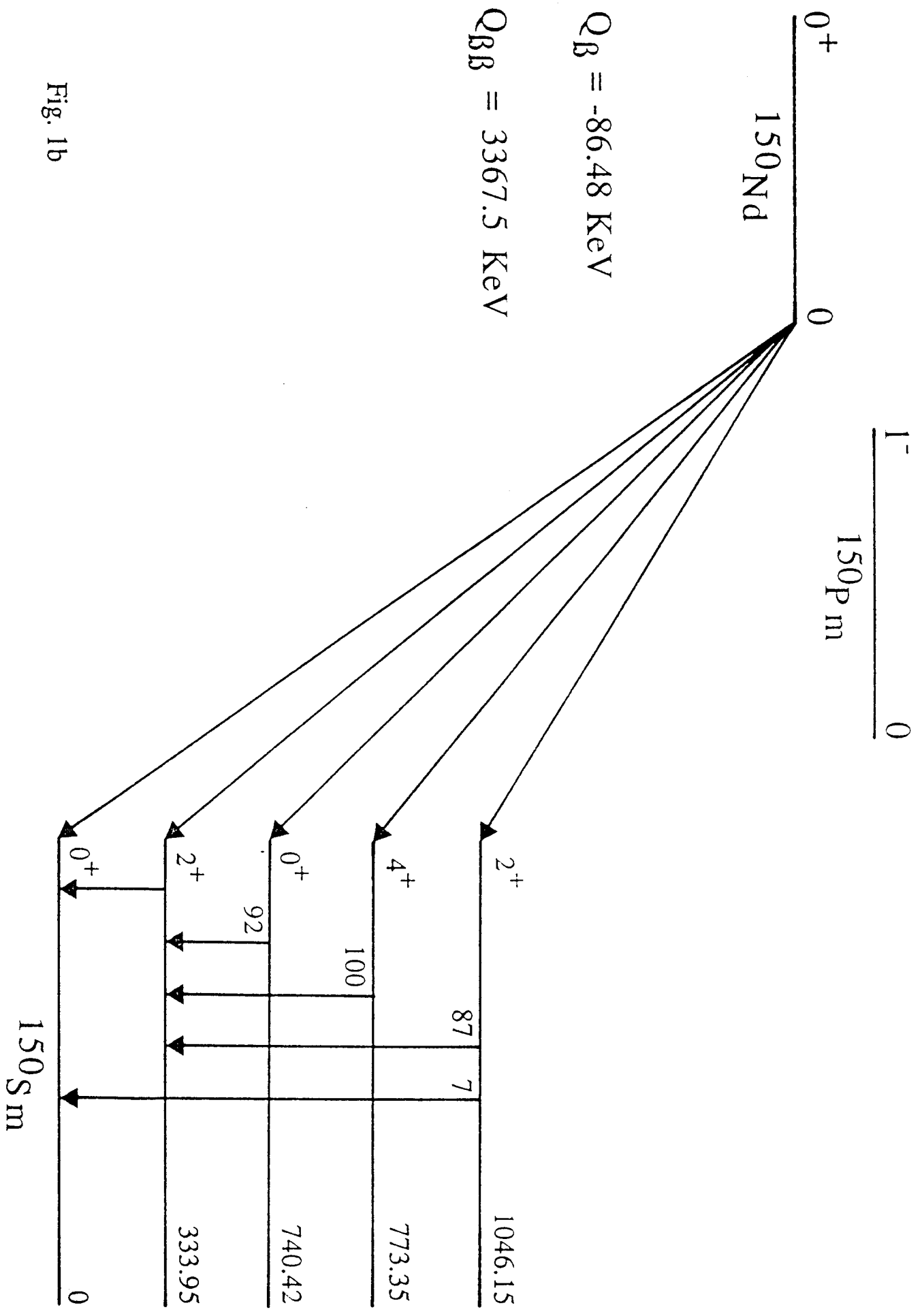


Fig. 1b

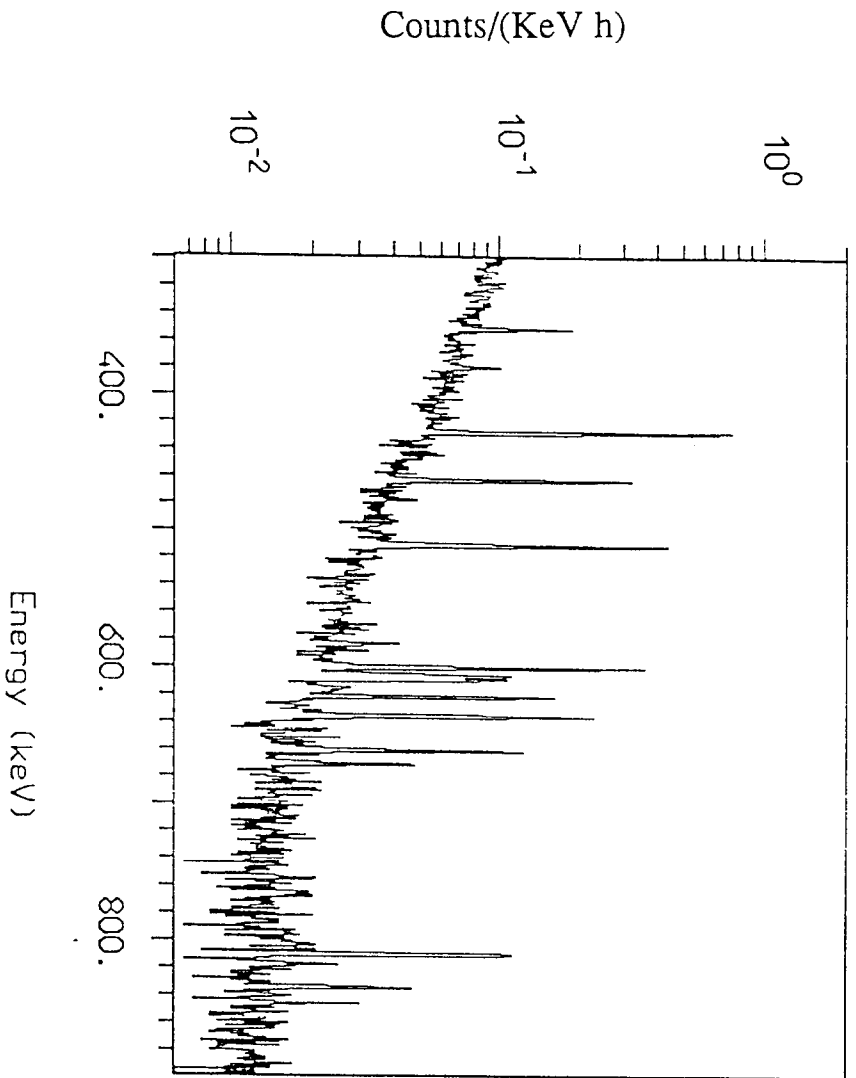


Fig. 2a

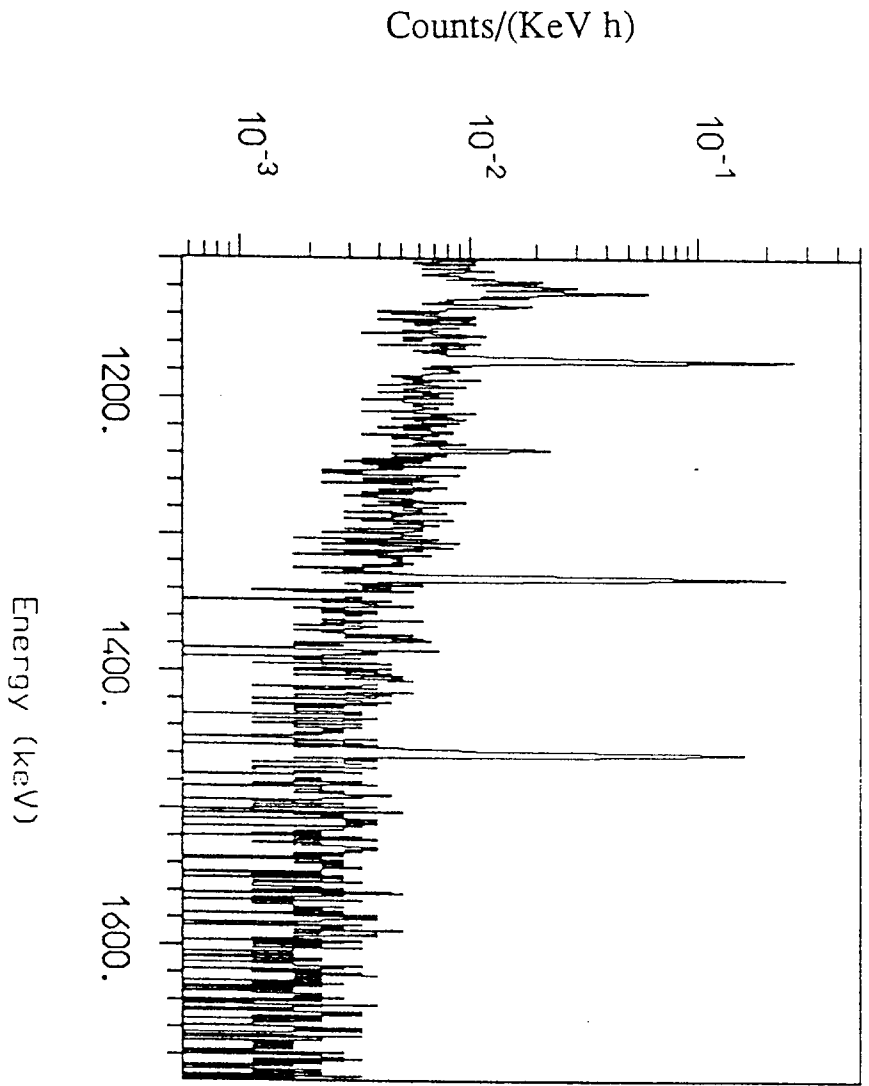


Fig. 2b

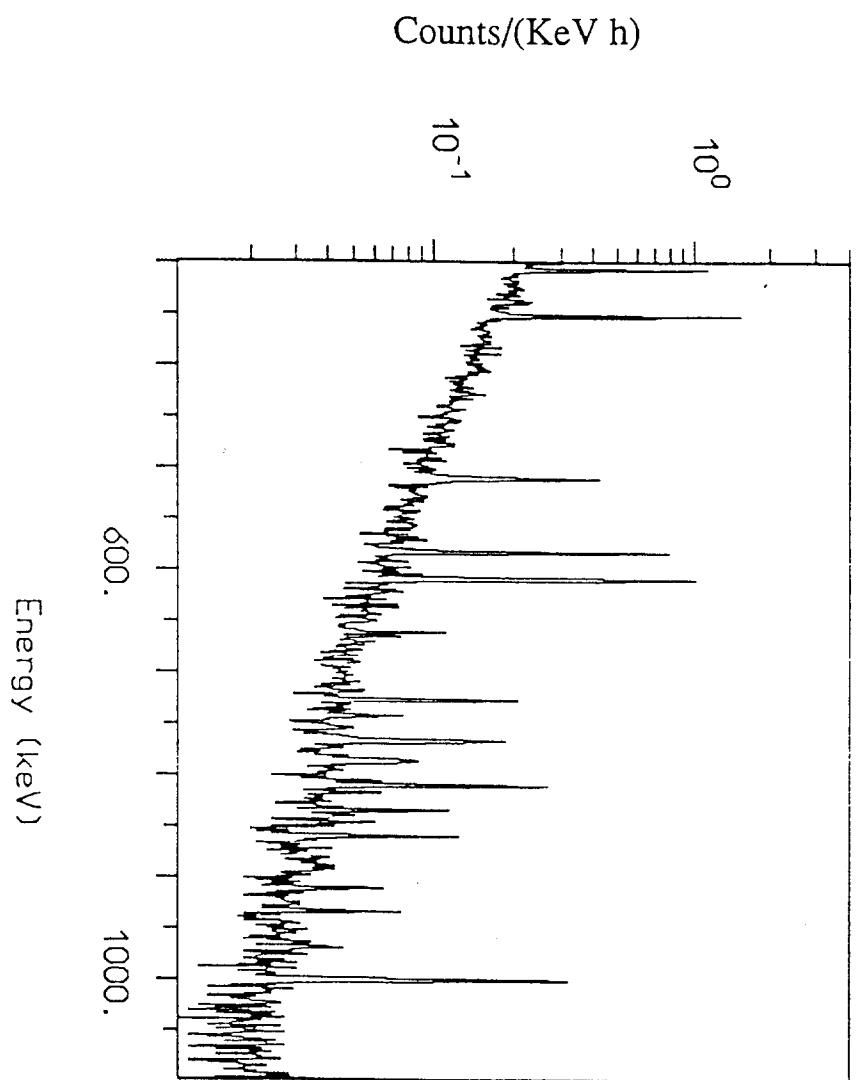


Fig. 3