MEASUREMENTS OF INTERNAL RADIOACTIVE CONTAMINATION IN SAMPLES OF ROMAN LEAD TO BE USED IN EXPERIMENTS ON RARE EVENTS

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ABSTRACT

Improved results of a series of measurements carried out on two different types of Roman lead to be used in shields for experiments on rare events are reported. The chemical impurities in both samples have been determined by neutron activation. Underground measurements based on γ spectroscopy on large masses of the two types of lead, show the absence in both samples of radioactive contamination from ^{214}Bi and ^{232}Th, in secular equilibrium, with upper limits of a few tenths of a mBq kg^{-1}. The contamination from ^{40}K is less than a few mBq kg^{-1}. Much care has been addressed to the contamination due to ^{210}Pb which breaks secular equilibrium and which contributes to most of the background in experiments searching low energy events like direct interactions of Weakly Interacting Massive Particles (WIMPS). We have applied to this problem the technique of cryogenic detection and found for the two samples upper limits for contamination of ^{210}Pb of 4 and 7 mBq kg^{-1} , the lowest ever determined for any type of lead.

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1. INTRODUCTION

Great interest in the reduction of the background of spurious counting has been recently motivated by experiments on rare events like interactions by solar neutrinos , Weakly Interacting Massive Particles (WIMPs), axions, etc., and rare spontaneous processes like double beta decay. The background due to cosmic rays (mainly muons and neutrons) can be strongly reduced by operating the experiment deep underground. The effect of the intrinsic radioactivity of the set-up can be reduced by specifically choosing the materials immediately surrounding the detector or employed in the construction of the detector itself. In order to reduce the environmental background due to γ - rays from external radioactivity one has to adopt massive shields made of materials of large atomic number and low intrinsic activity.

Due to its high Z, reasonable cost , mechanical properties and low activation cross section for environmental neutrons, lead is an excellent shielding material. Measurements by γ spectroscopy , including the one reported in the present study, indicate a very low radioactive contamination from the nuclei of the ^{235}U , ^{238}U and ^{232}Th chains , *when in secular equilibrium,* in all samples of commercial lead. Secular equilibrium is however broken by ^{210}Pb which represents an important component of environmental radioactivity [1,2]. Its β decay , with a lifetime of 22.3 years and 63.5 keV transition energy [3] is followed by β decay of ^{210}Bi to ^{210}Po with half-lifetime and transition energy of 5.013 days and 1162.7 keV. This short chain is closed by α decay of ^{210}Po to ^{206}Pb with a lifetime of 138.376 days and a transition energy of 5407.46 keV.

The ^{210}Pb contamination in modern lead can be as high as 2500 Bq kg^{-1} [4] and reach values up to 50 000 Bq kg^{-1} [5] in lead for solder. In common lead used in underground experiments this contamination is normally around 200 Bq kg^{-1} . At somewhat higher , but reasonable, price one can obtain lead with a content of less than 50 Bq kg^{-1} [6] or 20 and even 5 Bq kg^{-1} [7] . The best , but very expensive, available lead has been obtained by Johnson and Mathey [8] by optimizing the process of production using control measurements based on α counting and, very likely, ore selection. Various measurements , including the one performed in this experiment , yield for this special modern lead values for ^{210}Pb

activity of few tenths of Bq kg^{-1}. ^{210}Pb is expected to be totally absent in ancient lead as the one found in wrecks of Roman ships sunk in the Mediterranean sea [9] or near Britain [10]. In addition, the overburden of water has prevented the lead, and even more its chemical contamination, to be activated by cosmic ray neutrons [11].

The presence of ^{210}Pb can severely affect the sensitivity of experiments searching for low energy rare events like interactions of WIMPS, due to the continuum background bremsstrahlung produced by β decay of this nucleus and especially of ^{210}Bi. As an example about half of the counting rate in the low region of the spectra obtained in the Heidelberg-Moscow experiment [12] is due to this contribution, even if a shield of Johnson and Mathey lead was used.

The presence of a tiny contamination of ^{210}Bi is hard to be revealed by γ spectroscopy. The only measurable γ rays due to this nucleus or to the nuclei produced by the further decays have energies of 46.54 (^{210}Pb) and of 803.1 keV (^{206}Pb). The former is strongly absorbed by the cup of the Germanium detectors, while the branching ratio of the latter is very low (around 10^{-5}). Measurements based on α and X-ray spectroscopy and on the bremsstrahlung are more promising and have been performed on samples of modern and ancient lead [9]. They can however hardly reach sensitivities definitely better than one Bq kg^{-1} .

We have shown in a previous experiment [13] that much better sensitivities on the presence of ^{210}Pb in lead can be reached with the technique of bolometric detection [14]. Low temperature thermal detectors have been recently developed in view of experiments on nuclear, subnuclear and astroparticle physics [15-17]. These bolometers take advantage of the fact that the heat capacity of a cold dielectric and diamagnetic crystal is proportional to the cube of the ratio between the operating and Debye temperatures and can therefore become extremely small. As a consequence even the tiny energy released by a particle can induce a sizable increase of the temperature which can be measured by a suitable thermometer. Our approach is the construction of a bolometer where the absorber is made with the same material to be investigated: it acts therefore as source and detector of its own contamination [13].

We report in the present article a series of measurements performed on samples of two archeological finds of Roman lead in the Mediterranean sea. One

sample comes from the lead recovered from the wreck of a roman ship sunk near the Mal di Ventre island in the Sardinian province of Oristano at a depth of 28 meters. Some archeometric information and preliminary measurements on this lead have been already reported [9,18]. A second smaller quantity of lead has been found at a depth of 95 meters in the Silvia bay near Biserta at 12 miles from the African coast. These types of Roman lead will be defined as *Oristano* and *Silvia*, respectively.

We will report in the present article the following measurements on these types of lead:

a. neutron activation analysis in order to ascertain their chemical impurities
b. γ ray spectroscopy to determine their radioactive impurities due to ^{40}K and to the chains of Uranium and Thorium *in secular equilibrium*
c. cryogenic measurements to evaluate the contamination of ^{210}Pb. These measurements have also been carried out , for comparison, on a sample of Johnson and Mathey lead.

Determinations of ^{210}Pb contamination based on α and X-ray spectroscopy and on brernsstrahlung have not been performed, being definitely less sensitive than the cryogenic ones.

2. NEUTRON ACTIVATION ANALYSIS

Impurity concentration in the two types of lead have been determined with instrumental neutron activation analysis, by irradiating samples at the TRIGA MARK 2 reactor at the LENA laboratory of the University of Pavia . The irradiating position in the reactor was the rotating specimen rack facility, where the measured flux was 1.8×10^{12} cm^{-2} s^{-1}. Two different procedures have been employed:

a. samples of different sizes , ranging from 10 to 100 mg, carved from the two different types of Roman lead , were irradiated together with Standard Reference Materials , certified for trace element content. NBS1632 Coal and NBS 1633 Fly ash materials were chosen, due to their different concentration in

trace elements. Two different irradiation-counting procedures were adopted: one of two hours irradiation and 2,000 second counting for the determination of activated nuclei with intermediate half lives (Cu, As etc and possibly other elements) and another of 10 hours irradiation and 20,000 counting for long living isotopes. Longer counting runs were also performed after a cooling period of 20 days.

b. Discs of both types of lead with masses from about 10 to 100 mg were packed in polythene containers and irradiated together with two specially prepared standards of a similar shape obtained by centrifugating in vacuum calibrated solutions of the most relevant metals. One standard contained 10 μg of Sn, 10 μg of Sb, 6 μg of Au in HCl ; the other 50 μg of Ag , 10 μg of As, 10 μg of Cu, 50 μg of Zn, 50 μg of Se and 50 μg of Ni in HNO_3. After an irradiation of 8 hours, the γ activity of samples and standards were measured with Ge detectors at several decay intervals. The measurements on the low mass samples were carried out immediately after irradiation , those with more massive samples later to avoid excessive pile-up.

The results on the various impurity concentrations are reported in Table 1, where the errors include the contribution from statistics and from systematic uncertainties on the concentration in the standards, and on the slightly geometrical differences among lead samples and standards. The agreement between the measurements with the two procedures is reasonable for both types of lead. We note that, with the exception of Cu and Se, the content metals other than Pb is much lower in the Oristano lead , which , as pointed out in a previous paper [9], was probably de-silvered. The impurities in the Oristano lead appear to be homogeneously distributed, while some dishomogeneity seems to be present in the samples from the Silvia bay.

3. MEASUREMENTS OF RADIOACTIVE CONTAMINATION BY γ - RAY SPECTROSCOPY

Measurements of γ - spectroscopy were performed in the Gran Sasso Underground Laboratory where the muon and neutron background is reduced by

six and four orders of magnitude, respectively [19]. An intrinsic Ge detector of 113% efficiency made with specially chosen low radioactivity materials (Oxigen Free High Conductivity copper for the cap and gold instead of indium for the gaskets) was used. It was shielded with layers of OFHC copper and low radioactivity lead of 10 and 20 cm minimum thickness each. We have molded in a Marinelli shape and placed around the detector samples of Roman lead from Oristano and Silvia bay and , for comparison, samples of modern lead. The masses of the samples were of 26 kg each with the exception of the Silvia Roman lead for which only 5 kg were available. Each spectrum was collected with at least 300 hours of effective running time, and compared with a blank spectrum obtained by substituting the lead samples with OFHC copper (1280 hours of running time). We report in Fig.1 the spectra in the low energy region corresponding to

1. common lead produced without any attempt to reduce the content of ^{210}Pb
2. common lead with less than 20 Bq kg^{-1} ^{210}Pb
3. Roman lead from Oristano

The spectrum corresponding to the Roman lead from the Silvia bay is not distinguishable from the one from Oristano. The effect of the bremsstrahlung for samples containing ^{210}Pb can be clearly seen. We would like to note that the continuum counting rates for the blank are *above* (Fig.1b) those for Roman lead, as a consequence of the less effective shielding.

The counting rates in the peaks where statistics is significant are reported in Table 2. The peak at 46.4 keV is due to ^{210}Pb contained in some solder *inside* the copper cup of the detector: the rates are the same for all types of lead and for the blank, with some excess for the modern lead. The effect of ^{210}Pb in generating lead X-rays can be seen in Fig.2. Fig.3 shows that the peak at 803 keV can clearly be seen in the spectra with modern lead and with lead with certified content of less than 20 Bq kg^{-1} of ^{210}Pb, but is totally absent in the spectra for Roman lead .

The counting rates in all other peaks are similar , thus confirming the high purity of *all* types of lead. In fact there is a low excess of counts in the peaks corresponding to ^{60}Co *in the blank*. This effect is likely due to activation of copper by fast neutrons [2]. The constancy of counting rates in all peaks apart those due

to ^{210}Pb indicates that the background comes mainly from the detector itself. The limits of radioactive contamination from nuclei in secular equilibrium has been evaluated by means of a Monte Carlo method and are reported at 95% confidence level in Table 3. The counting rates in the peak at 803 keV allows a rough evaluation of the contamination by ^{210}Pb in the common modern lead and in the lead with a certified content of <20 Bq kg^{-1} . Values of 170±10 and 16±4 Bq kg^{-1}, are obtained, respectively.

4. BOLOMETRIC MEASUREMENT OF THE CONTAMINATION OF ^{210}Pb

The bolometric technique has been already used by us to determine the content of radioactive impurities in cryogenic thermal detectors. The internal contamination due to the uranium and thorium chains in TeO_2 crystals has been found to be definitely lower than a part per trillion from the absence of peaks corresponding to the total α decay transition energies of the nuclei in these chains [20]. We have also applied the same method in a preliminary measurement of ^{210}Pb contamination in lead using pieces of lead at the same time as sources and detectors of their own activity [13]. It is based on the counting rate at the peak at 5407 keV due to α decay of ^{210}Po.

One of the difficulties in the use of lead as a thermal detector is the expected large lattice contribution to the heat capacity, since the Debye temperature is low (about 80 K) . On the contrary the electron contribution is expected to be negligible in the superconducting state, at temperatures well below the transition temperature (about 7 K). Nevertheless a considerable part of the energy delivered by the interacting particle is spent in a superconductor in breaking Cooper pairs , which could not completely recombine in phonons within the time scale of the thermal pulse. It was however pointed out [21] and proved for lead in our preliminary experiment [13] that in materials with low Debye energy and high critical temperature the quasiparticle recombination rate should exceed the pair-breaking rate, leading to an acceptable thermalization. A series of measurements carried out by the Genoa group [22] shows that the thermalization efficiency is a universal function of the ratio between the operating and Debye temperatures of

the bolometer. This efficiency is about one when this ratio is higher than 3×10^{-3}, but decreases rapidly below this value.

A third difficulty comes from the fact that a single crystal is usually required as an absorber in a low temperature thermal detector. On the other way crystallization usually *cleans* a material from most of the contaminants including the radioactive ones. As a consequence the preparation of the crystal could cancel or decrease the primitive contamination in the original material to be studied. Lead however has a polycrystalline structure and even a piece of material simply cut from the sample to be investigated acts reasonably well as absorber in a thermal detector, as shown by our preliminary experiment [13].

In this new dedicated experiment of better sensitivity the ^{210}Pb contamination in the two types of Roman lead was measured and compared with the one of Johnson and Mathey commercial lead. This search was carried out in the underground Laboratori Nazionali del Gran Sasso. Four lead absorbers in form of cubes of 1 cm side were simply cut from samples from the Silvia bay (two), from Oristano and from Johnson and Mathey lead. On each of these absorbers a Neutron Transmutation Doped (NTD) thermistor provided us by E.Haller [23] was glued. An array made by these four detectors (Fig.4) was installed in a dilution refrigerator, specially made with previously tested low radioactivity materials. The refrigerator is shielded against external radioactivity by layers of OFHC copper and modern lead of 10 cm minimum thickness each. The entire set-up was operated inside a Faraday cage to reduce the effects of electromagnetic interference. The pulses from the four detectors were read independently with a read-out system similar to those employed in a previous series of experiments [20].

Measurements were carried out with operating temperatures of the four detectors ranging from 15 to 20 mK. One of the detectors with Silvia lead performed definitely worse than the other , due to microphonism. Only the latter was therefore used, together with the other two, for further analysis. All bolometers worked quite well even if their *apparent* heat capacity, naively deduced from the pulse amplitude, corresponded to values about one order of magnitude larger than the ones expected from the Debye law. A ^{60}Co calibration

spectrum (the first obtained with a lead detector) is shown in Fig. 5 . The resolution at these energies for the three detectors ranges from 35 to 48 keV.

The array has been run for about 120 hours of effective running time. The spectrum in the high energy region for the bolometer with the absorber of Johnson and Mathey lead shows a relevant peak around 5407 keV , corresponding to the transition energy for α decay of ^{210}Po (Fig.6). We have at present no final explanation for the asymmetry of the peak: it could be due to a position effect determined by the polycrystalline nature of the absorber. The counting rate in the peak yields a contamination of 260± 9 mBq kg^{-1} of ^{210}Po and consequently of ^{210}Pb . This value is in agreement with that found with a bremsstrahlung analysis for a similar lead used in the Heidelberg-Moscow experiment [12]. No peak appears in the corresponding spectra obtained with the bolometers with Oristano and Silvia lead. We can therefore set at 95% confidence level upper limits of 4 and 7 mBq kg^{-1} for the activity of these samples of Roman lead, respectively. These are the lowest contents of ^{210}Pb ever found in lead, measured with a sensitivity almost two orders of magnitude better than in any other experiment.

5. CONCLUSIONS

Our neutron activation analysis shows that both samples of Roman lead are almost 99% pure. The impurities are however very different in composition. Large amounts of antimony and silver are present in the ingot from the Silvia bay, while the main impurity in the lead from Oristano is copper. The low content of silver in the lead from Oristano indicates that this metal was desilvered.

The analysis by γ - spectroscopy shows no evidence for radioactive contamination in secular equilibrium, not only for the Roman lead, but also for three modern samples of this metal.

The bolometric analysis of the content of ^{210}Pb in Roman lead shows that its contribution to radioactivity is less than a few mBq , five orders of magnitude lower than in common modern lead , and also two orders of magnitude lower than for the best specially prepared commercial lead.

We are presently operating thermal detectors made with crystals of $PbWO_4$ *made with Roman lead* in view of a planned thermal experiment on direct interactions of WIMPS.

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Table 1: Impurity concentration in the four samples of Roman lead (ppm) Measured with the two procedures **a** and **b** reported in the text

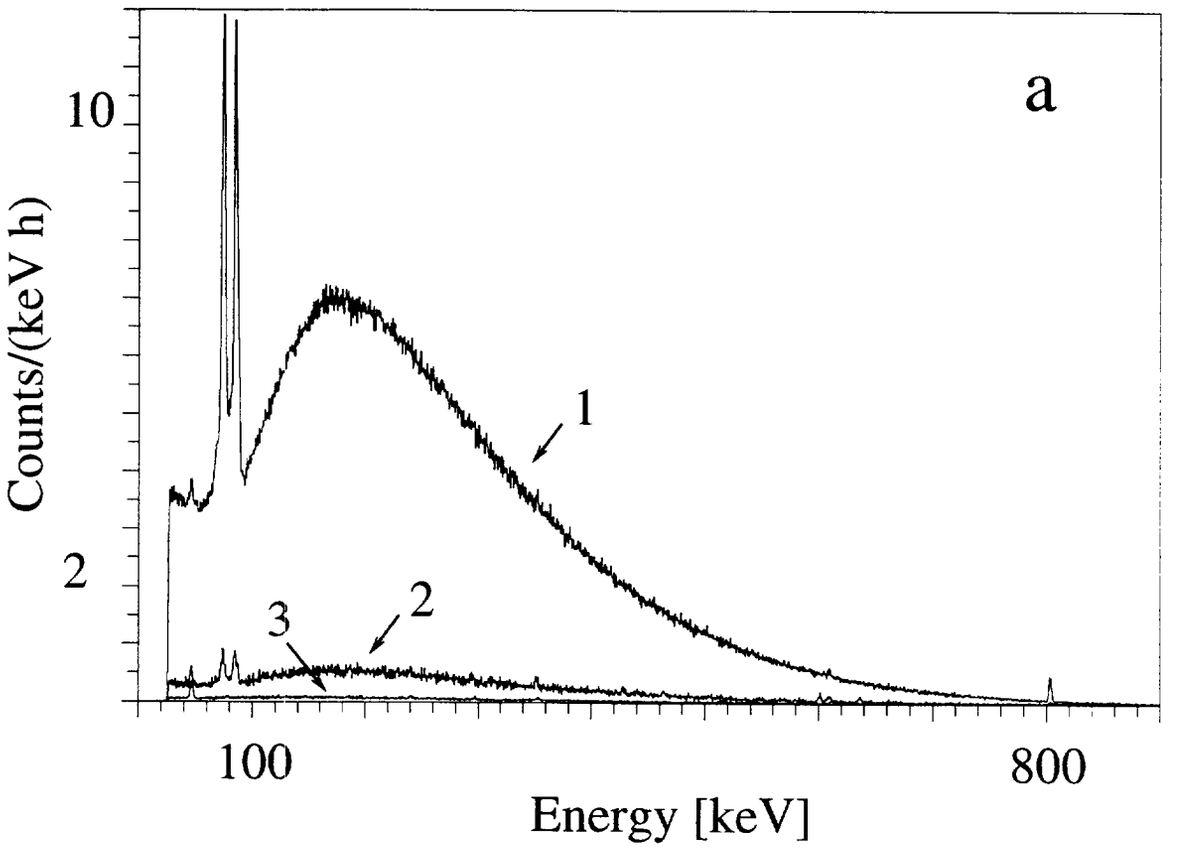
	Oristano a	Oristano b	Silvia a	Silvia b
Se	.98±.10	1.4±.2	2.0±.5	<1
Zn	.5±.1	<20	10-20	<20
As	.5±.2	.012±.004	15±5	40±5
Sb	.09±.01	.1±.01	900±30	960±40
Ag	45.0±2.5	58±5	140±30	240±40
Ni	4.5±.5	3. ±.2	30±10	18±6
Cu	-	1270±100	-	600±30
W	<.02	-	.3±.1	-

Table 2 : Counting rates for different types of lead (count h^{-1})

Energy (keV)	Nucleus	Modern	Mod. <20	Mod.<5	Oristano	Silvia	Blank
46.4	^{210}Pb	1.4±.1	.74±.08	.83±.10	.88±.05	.93±.10	.88±.06
74.8	X-rays	2.7±.3	.8±.1	.25±.06	<.08	<.1	<.08
77.1	"	35±1	1.4±.2	.40±.07	<.1	<.15	<.1
85.2	"	25.0±.5	1.4±.2	.44±.07	<.1	<.14	<.1
87	"	9.1±.8	.7±.2	.15±.06	<.1	<.13	<.1
238.6	^{212}Pb	<.5	.2±.1	.17±.05	.15±.05	.2±.1	<.2
295.2	^{214}Pb	<.4	.2±.1	.4±.2	.10±.03	<.3	.09±.03
352.0	^{214}Pb	<.7	.4±.1	.6±.2	.2±.1	.2±.1	.15±.05
583.1	^{208}Tl	<.2	.07±.03	.07±.03	<.1	<.08	.07±.02
609.4	^{214}Bi	.13±.05	.3±.1	.5±.15	.14±.03	.08±.04	.12±.03
661.6	^{137}Cs	<.1	.06±.02	.07±.02	.05±.01	<.07	.08±.04
803.1	^{210}Pb	1.27±.07	.12±.03	<.2	<.1	<.15	<.1
911.1	^{228}Ac	.05±.02	.05±.02	<.07	.018±.08	<.06	.03±.01
1120.4	^{214}Bi	.09±.02	.08±.02	.09±.03	.07±.02	.06±.02	.04±.01
1173.2	^{60}Co	.09±.02	.06±.02	<.1	.082±.005	.09±.03	.12±.02
1132.5	^{60}Co	.08±.02	.11±.02	<.1	.08±.0	.08±.02	.13±.02
1460.8	^{40}K	.20±.03	.30±.05	.14±.03	.19±.02	.17±.04	.26±.05
1764.7	^{214}Bi	.07±.01	06±.02	.10±.03	.04±.01	<.06	.04±.01
2614.6	^{208}Tl	.042±.006	.059+.011	.042±.010	.042±.006	.03±.01	.034±.005

Table 3 : Limits (95% c.l.) on contamination of different lead samples (mBq kg^{-2})

	^{232}Th	^{186}U	^{40}K	^{137}Cs	^{214}Bi	^{60}Co
Modern	**.0**93	1100	1.37	.23	.35	.062
Modern <20	.15	1000	2.11	.14	.41	.049
Modern<5	.10	340	1.32	.14	.68	.090
Oristano	.076	700	1.35	.14	.19	.038
Silvia	.23	710	4.16	.38	.50	.140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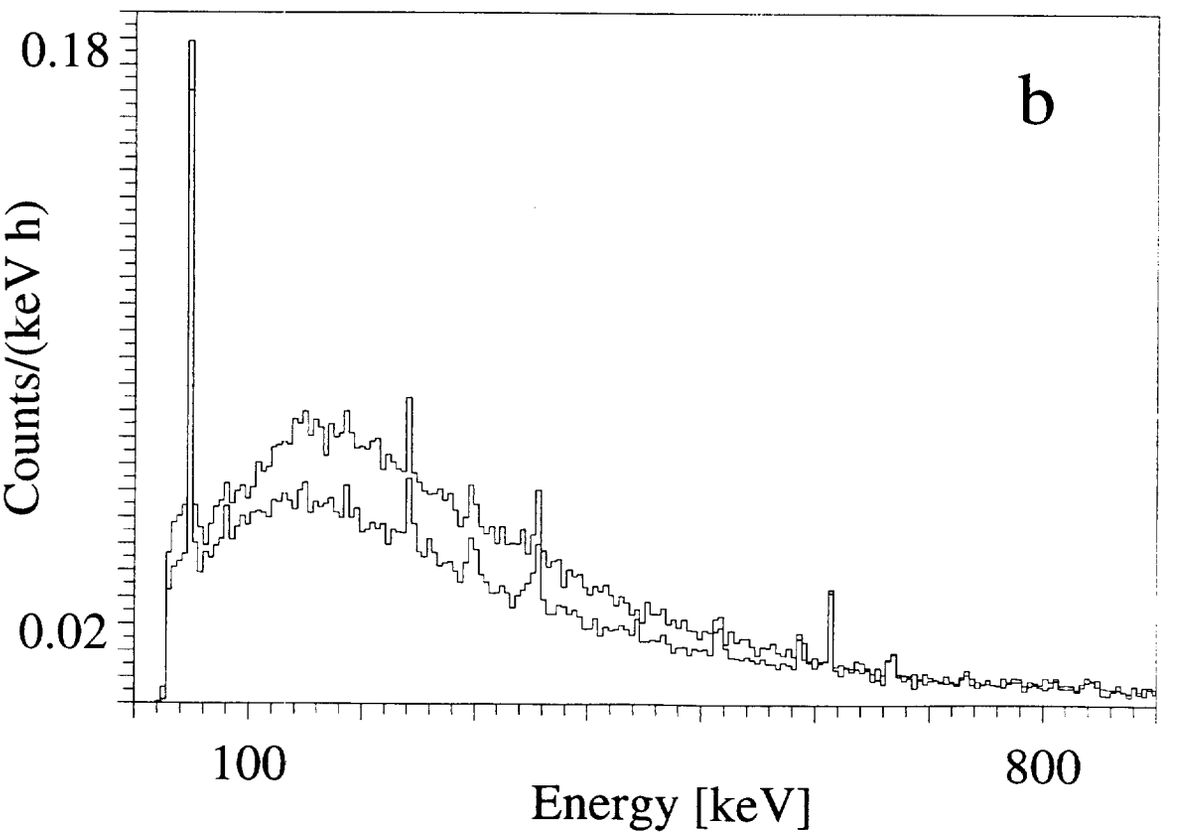
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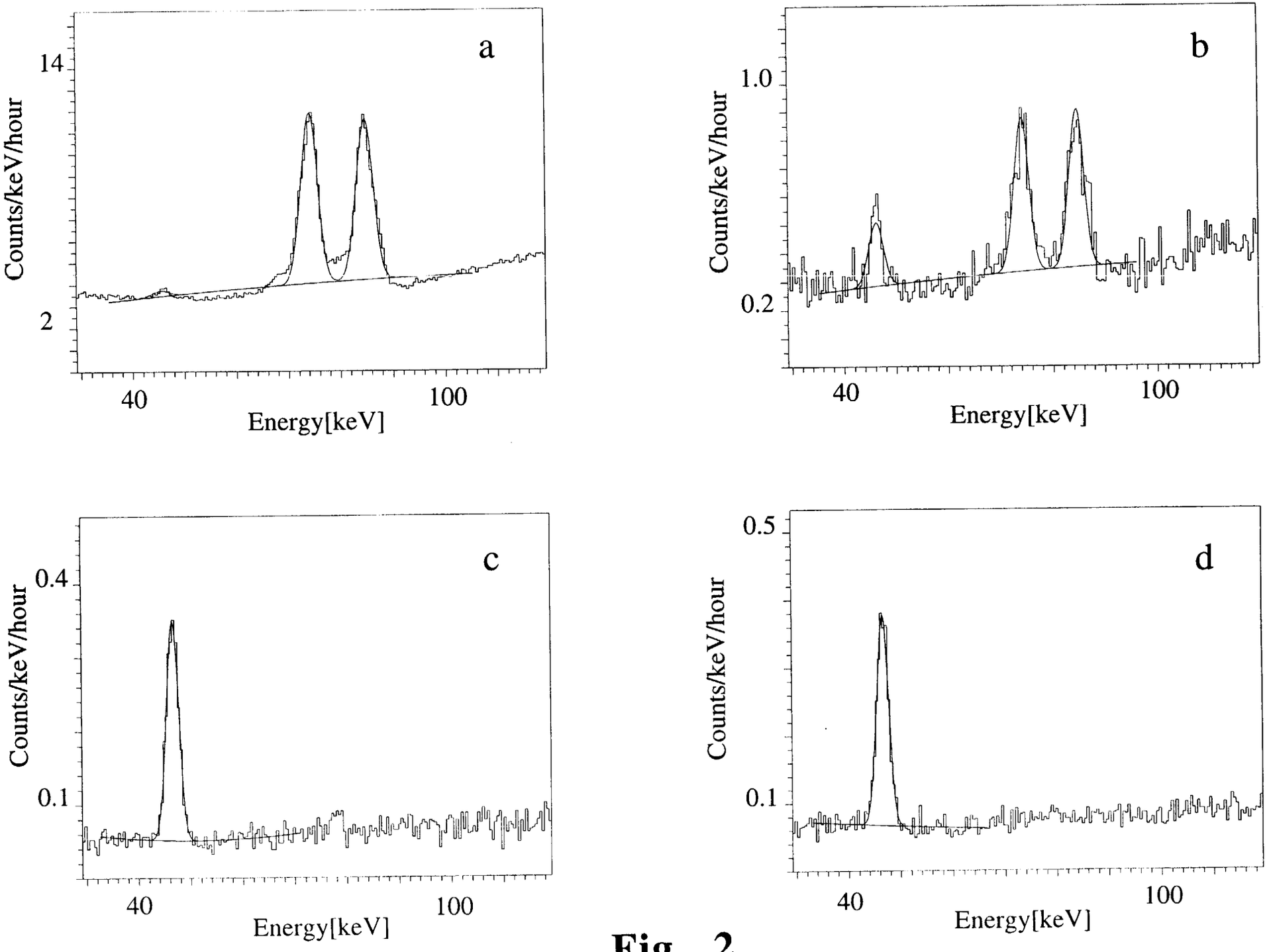
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c
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Fig. 2

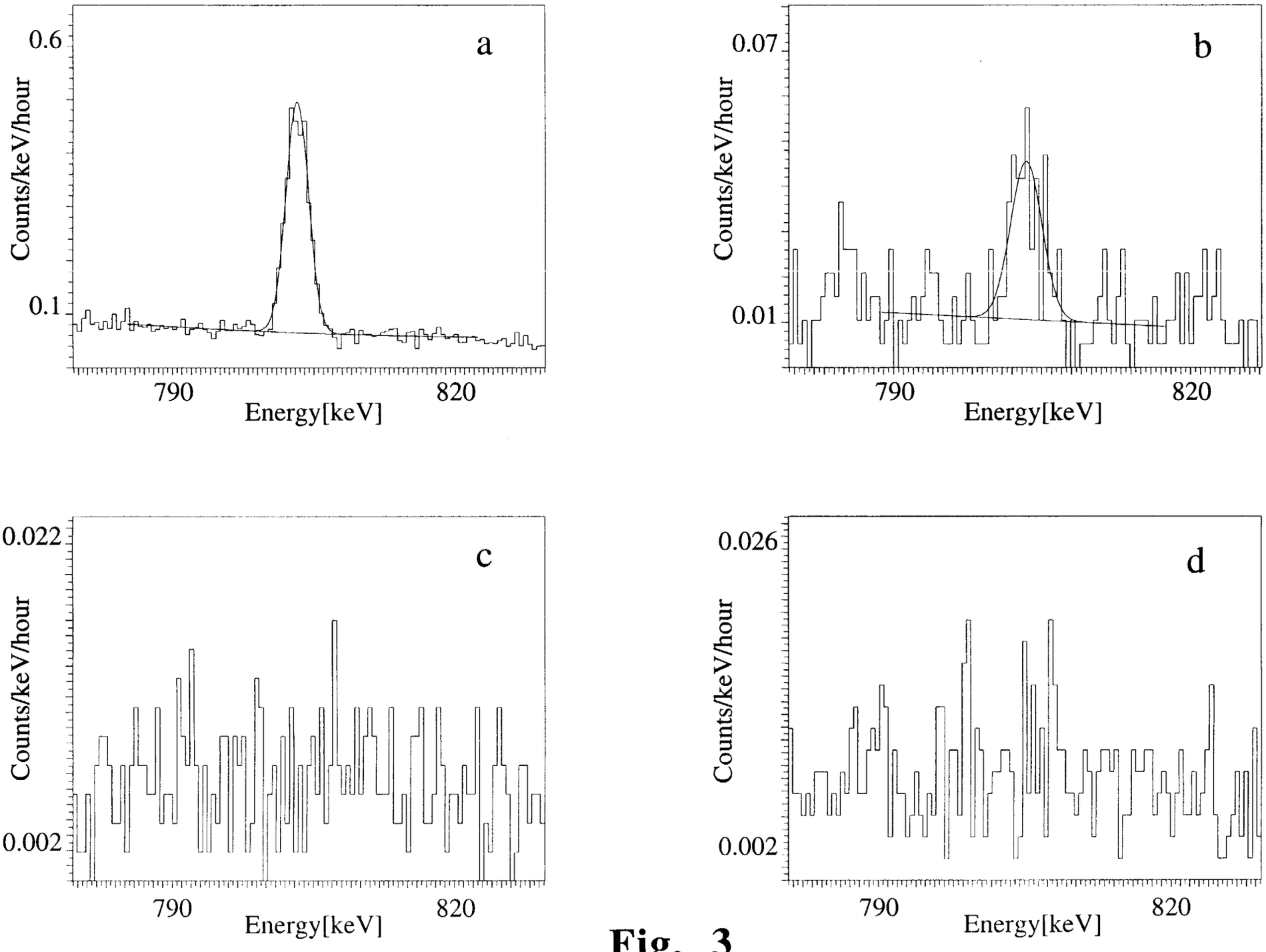
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Fig. 3

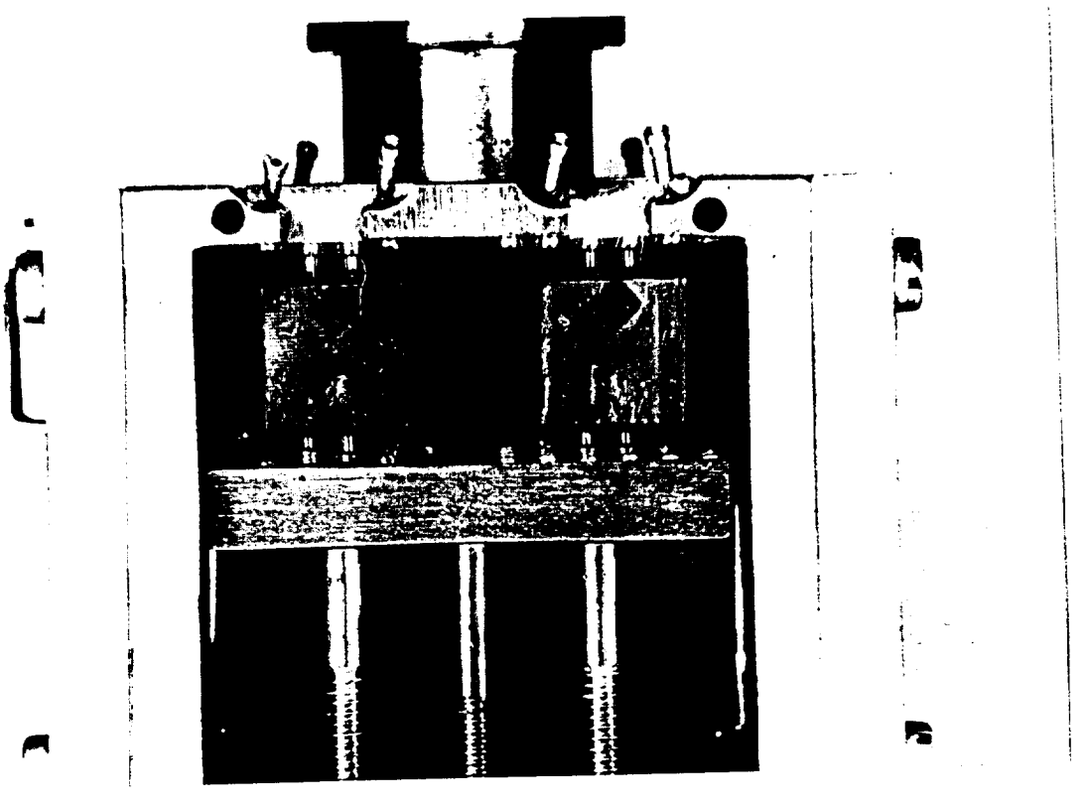
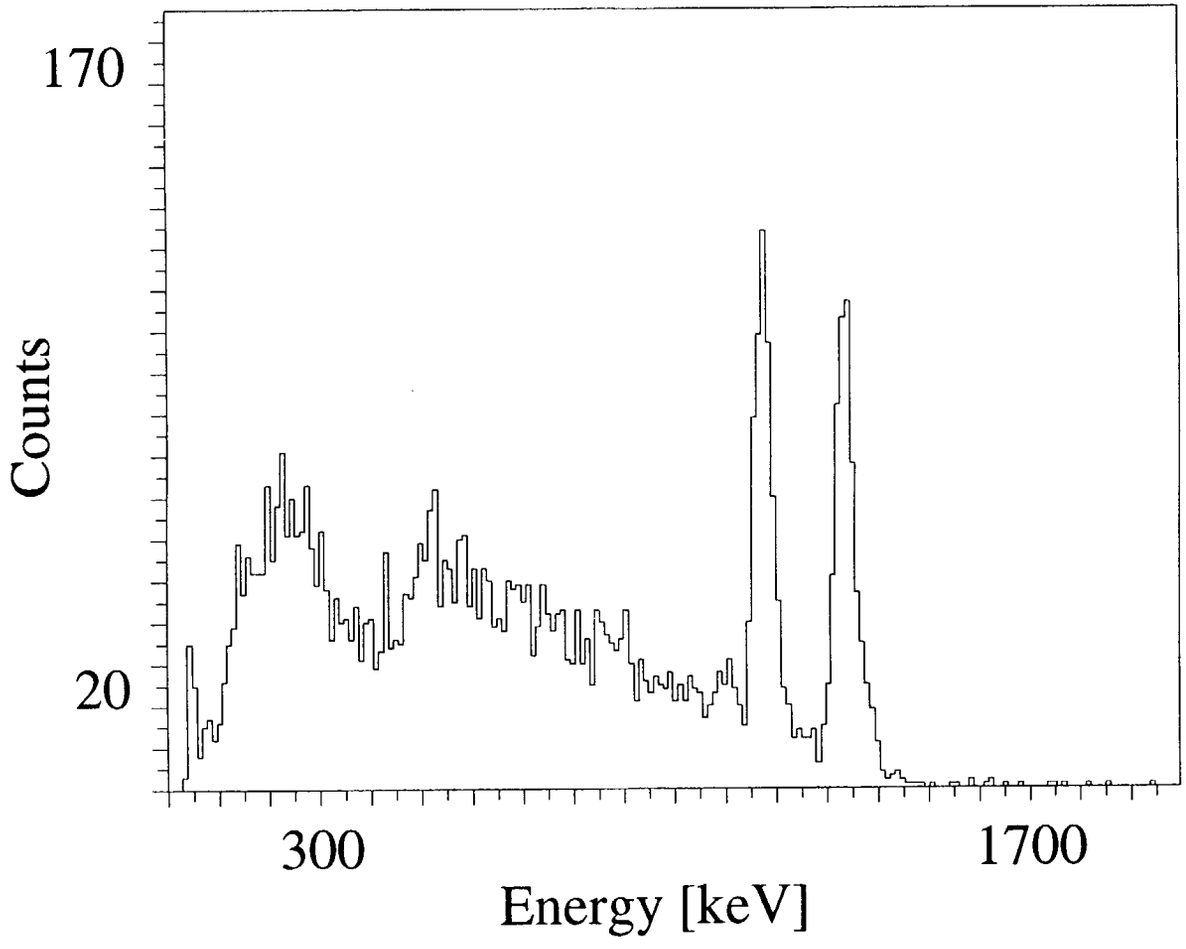


Fig. 4



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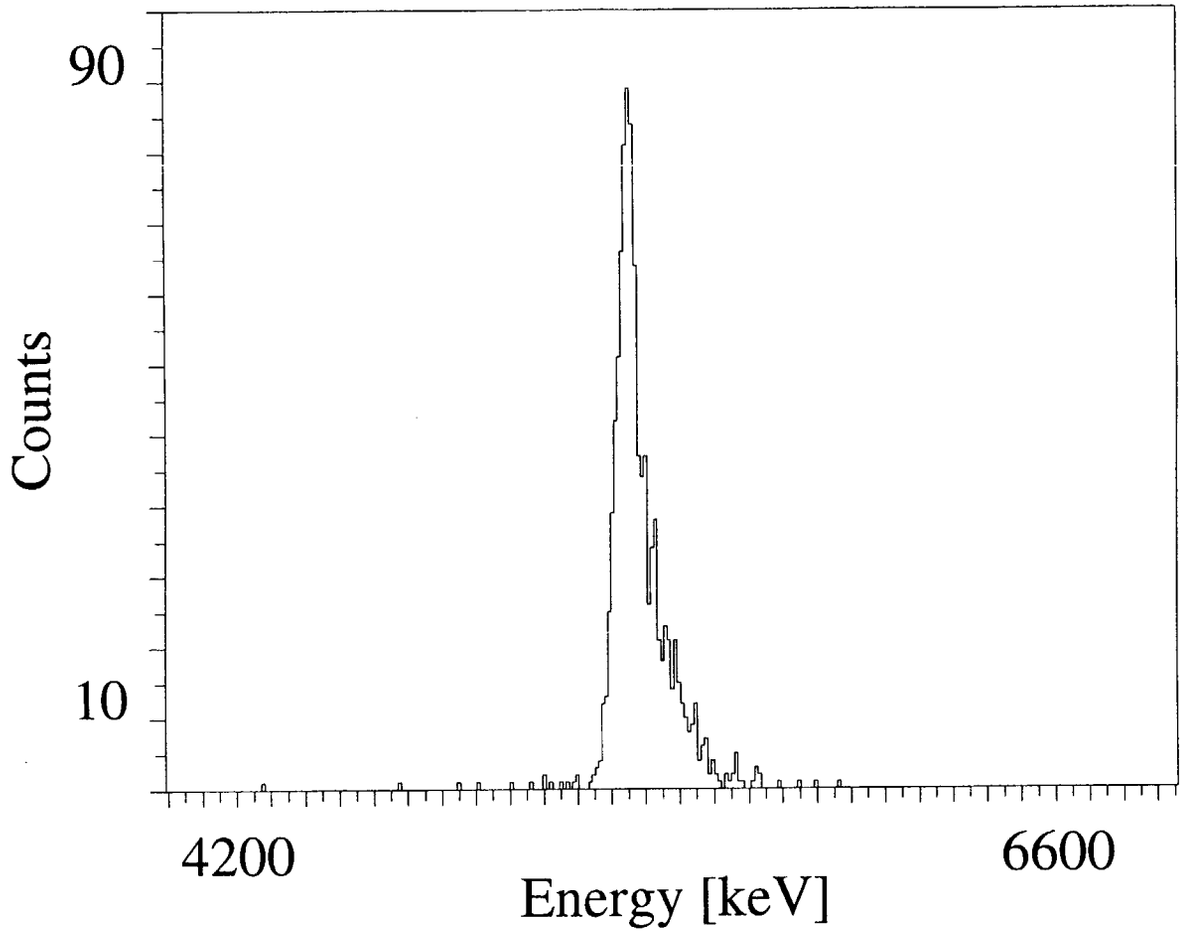

Fig. 6