BOLOMETRIC MEASUREMENTS OF BETA DECAY SPECTRA OF ^{187}Re WITH CRYSTALS OF SILVER PERRHENATE

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ABSTRACT

Crystals of silver perrhenate have been used for the first time in the bolometric measurement of the spectrum of ^{187}Re with the aim of determining the electron antineutrino mass. The temperature rise has been measured by means of Neutron Transmutation Doped germanium and of P implanted silicon thermistors. The energy scale has been calibrated using the K lines of Mn and the fluorescence L lines of Sn. Bolometers made from crystals of masses ranging from ~250 to ~ 500 μg have been run with FWHM resolutions on the Sn lines ranging from 25 to 60 eV. The end point of the spectrum has been located at 2460± 5_{stat}±10_{syst} eV. The measured half lifetime is 43± 4_{stat}±3_{syst} Gyr in good agreement with the values obtained with mass spectroscopy from the buildup of ^{187}Os in rhenium samples. The sensitivity of the measurement on a non-zero neutrino mass achievable with these bolometers is discussed.

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1. INTRODUCTION

All recent determinations of the electron neutrino (in fact antineutrino) mass are based on measurements of the energy spectrum of the electron emitted in beta decay of tritium. Unfortunately they yield negative values for the square of the neutrino mass or unexpected distortion in the spectra [1-3]. A fraction of the decays could in fact occur to excited states of the daughter molecule, and as a consequence part of the energy delivered in the decay could be lost. Other distortions of the spectrum could be caused by backscattering or by the finite thickness of the source. It seems therefore essential to complement the above-mentioned spectrometric experiments with *calorimetric* ones, where the entire delivered energy, except the antineutrino one, is recorded. Particularly apt to these aims appears the bolometric technique suggested in 1984 [4,5] and now widely applied [6-8]. It is based on the fact that at low temperatures the heat capacity of a diamagnetic and dielectric crystal or of a superconductor operated well below the transition temperature is proportional to the cube of the ratio between the operating and Debye temperatures. At low temperature it becomes so low that even the tiny energy delivered by a particle to the crystal can be revealed and measured from its temperature increase. A drawback of the calorimetric measurements of β-decays, worsened by the detector slowness, is that all the decays are collected, and not only the interesting fraction close to the end point. For a massless neutrino this fraction is inversely proportional to the cube of the transition energy, which should therefore be as low as possible.

The preliminary results reported here refer to the decay

$$^{187}\mathrm{Re} \longrightarrow {}^{187}\mathrm{Os} + \mathrm{e}^{-} + \bar{\nu}_{\mathrm{e}} \qquad (1)$$

with a transition energy of ~ 2.7 keV and a half lifetime of ~ 43 Gyr [9]. The large isotopic abundance of ^{187}Re (62.8%) allows the use of natural material. While this nucleus is very favorable for the low transition energy, the calculation of the effective spectrum for its first forbidden unique transition is less straightforward than for the allowed decay of tritium, also due to the electron screening. We note that the precise measurement of the lifetime of this decay is of considerable importance in geochronology

to determine the age of a geological sample from the relative Re-Os abundance [10-14]. Measurements of this ratio have been performed on meteorites in order to "date" the origin of the Solar System. Separation of the daughter ^{187}Os atoms from large sources of initially osmium-free rhenium allowed determining the lifetime of ^{187}Re . The most recent measurement yields a value of (42.3 ± 1.3) Gyr [15]. The first direct measurements of the decay [16-18] indicated a rate lower by about 50%. This was attributed [19,20] to effects of electron screening [21], which could lead to a *bound state decay.* It would consist mainly [20] of an indirect exchange process where an electron is created in a previously occupied bound state, with the simultaneous transfer of the bound state electron to a previously unoccupied bound state. In this case the electron would not be recorded in a conventional detector. Bound state decays were, however, found to be negligible in a later calculation [22]. A recent experiment carried out by K. Ashkorab et al [23] with a high-temperature quartz proportional counter indicates in fact a half-life in agreement with that determined by mass spectroscopy.

The advantage of calorimetric experiments on β-decays stays in their property to measure *all* the energy released to the absorber and not the electron energy only. Even transitions to intermediate states would be in principle revealed if the further decay occurs within the time response of the detector. In the field of solid state physics and particularly of material science, considerable interest could arise from deformations of the spectrum as a consequence of atomic and environmental effects [24-26]. This beta environmental fine structure (BEFS) in the low energy β-decay spectrum of ^{187}Re has been recently detected in metallic rhenium in a pioneering experiment by the Genoa group [27].

2. EXPERIMENTAL DETAILS

Our microbolometers have been operated both with Neutron Transmutation Doped (NTD) germanium thermistors provided by the Berkeley group [28] and with phosphorus implanted silicon (Si:P) thermistors fabricated in collaboration with the IRST Institute in Trento [29] . The NTD Ge thermistors, of 300x100x20 μm^3 linear dimensions, have two boron ion implanted contacts covered by Au pads (100x50 pads μm^2) deposited on both ends of the same 300x100 μm^2 side of the thermistor. Two 17 μm diameter pure

aluminum wires are ultrasonically bonded to these pads. The Si:P chips are formed by cutting from the implanted wafer a silicon dice (about 700x600x100 μm^3) containing one single 300x300 μm^2 thermistor. Four 17 μm diameter aluminum wires are ultrasonically bonded to the (Al:Si) metallized contacts.

Microbolometers operated with Sn absorbers and NTD Ge thermistors yielded a FWHM resolution of ~5eV for the K lines of Mn [30], the best ever obtained in X-ray spectroscopy with an energy dispersive detector. A resolution of ~13 eV was obtained with a similar tin absorber and a not-yet-optimized Si:P thermistor. The Si:P chips are, however, easier to handle and still far from optimization. As a consequence the results reported here refer to measurements carried out both with NTD Ge and with Si:P thermistors.

The microbolometers were made by gluing the NTD Ge or Si:P thermistors to the absorbers and mounted in an high cooling power Oxford Instrument dilution refrigerator with an experimental space of about 4 dm^3, a base temperature of 6 mK and a cooling power of 200 μW at 100 mK.

Since both NTD Ge and Si:P temperature sensors are high impedance thermistors, the parasitic capacitance between the two thermistor terminals must be minimized in order to avoid excessive signal integration. The variation of this capacitance is furthermore responsible for microphonic noise generated by wire vibrations. The front-end electronics was therefore operated a few centimeters away from the detector at cryogenic temperatures. The cold amplifier consists of a common drain silicon JFET. Ten of these devices are mounted on an aluminum plate, suspended by means of low conductance, tensioned Kevlar fibers inside a copper box. This box is mechanically connected to the 50 mK plate of the Oxford Instruments dilution refrigerator and is placed below its coldest point (6 mK). A thermal connection is established by means of a copper rod between the JFET box and the 1.2 K stage of the refrigerator. During JFET operation the total power dissipated on the JFET plate is around 20 mW. All the thermal connections are calibrated in such a way that this power rises the plate temperature to about 120 K, the optimum JFET operating temperature. The copper JFET box temperature is around 2 K, and all the 20 mW power flows to the 1.2 K stage. The detector base temperature is near 20 mK under these conditions, well below the optimum operation temperature of the bolometer.

The detector holder, which can house 10 microbolometers, is placed between the JFET box and the coldest point of the refrigerator to which is thermally connected. The

signal wires which connect a detector terminal to the JFET gate (the other terminal is grounded) are tensioned by 33 μm diameter, ~5 cm long manganine wires, which provide electrical connection and negligible thermal conductance. Great care is taken to shield the black body radiation emitted by the 120 K plate, which could heat up the detectors.

The JFETs are Interfet devices selected in order to have a white noise of about 5 nV/√Hz. New devices with about 3 nV/√Hz white noise have been tested and will replace the present ones in the future.

Pulses are digitized and saved to disk for off-line analysis. The spectra are obtained by optimum filtering. An artificial neural network approach has been implemented for signal discrimination.

3. RESULTS

Only the Genoa [18,31,32] and Milan [29,33,34] groups have reported so far calorimetric measurements on ^{187}Re decay. They have encountered considerable difficulties when using metallic rhenium since this superconductor has a low critical and a high Debye temperature [6-8]. In this type of material the energy released to the electron system by ionizing events can be trapped to quasiparticles for long times, and does not contribute to the phonon signal. In addition, rhenium nuclei have an electric quadrupole moment, which could be responsible for excess specific heat at low temperatures. This mechanism is particularly dangerous if, due to magnetic field trapping, some regions inside the rhenium samples remain in their normal state. In fact, the nuclear spin system couples to the phonon bath through normal electrons. Excess specific heat could be connected also to the presence of paramagnetic impurities. All these mechanisms tend to decrease the signal height with respect to the values predicted by the Debye law and to introduce long time constants in the pulse development.

Our group has carried out a systematic study with absorbers made from metallic rhenium samples from different suppliers. After purity tests performed using the RRR and the Inductively Coupled Mass Spectroscopy method, rhenium with high impurity levels was found to be inadequate as bolometric absorber because of a large excess specific heat. The effect of proper chemical etching to remove surface impurities and of different cutting techniques for the polycrystalline samples was also carefully

investigated. After the purity selection, samples of rhenium in polycrystalline and crystalline forms were studied, both with and without a cryoperm magnetic shield. The general result is that the pulse amplitudes are always smaller than expected from the naïve application of the Debye law. In addition, different behaviors have been observed at low (<100 mK) and high (100-250 mK) temperatures.

This was clearly shown by the results obtained with a crystal of rhenium of about 66 μg glued to a Si:P implanted thermistor. A spectrum obtained at ~ 55 mK showed an apparently good resolution of ~ 70 eV FWHM, but clear distortions of the Mn lines [34]. Pulses had a decay structure made by an initial fast component, probably of non-thermal origin, followed by a long tail. The amplitude ratio between the fast and slow components changed from pulse to pulse resulting in a deterioration of the resolution. At higher temperatures this effect generally diminishes and finally disappears, but at the expense of a larger heat capacity. No distortion was observed when operating this bolometer at ~250 mK, but the resolution became ~ 95 eV.

In order to overcome the thermal detection difficulties due to the superconductivity of metallic rhenium, we are pursuing an alternative approach: the use of absorbers made from dielectric compounds of this element. Crystals of NH_4ReO_4, K_2ReCl_6, $KReO_4$, $AgReO_4$ and $Re_2(CO)_{10}$ have been tested. The best results have been obtained so far with four crystals of silver perrhenate ($AgReO_4$) for which the spectral line widths do not seem to be substantially larger than expected from the signal to noise ratio. We have used both the NTD and the Si:P implanted chips described before.

Three detectors have been exposed independently to three weak fluorescence sources. The primary X-ray sources were obtained by drying on a mylar disk a 0.1 molar water solution of $N(OH)_3$ with $^{55}FeCl$ dissolved in it. An 8 μm thick tin sheet has been placed between each source and the corresponding detector to produce the fluorescence L lines of tin. A fourth detector was operated for comparison without this sheet. No special care was put in the selection of the materials for the preparation of the sources. As a consequence some fluorescence lines other than the manganese and tin ones were present in some spectra. A signal which could be attributed to the K line of calcium has been found in the spectrum of one of the first three detectors. The K titanium line was clearly present in the spectrum of the fourth and was in fact used for its calibration. We would

like to stress that these lines were not observed in test measurements carried out with the same detectors and without source.

All spectra have been corrected for temperature variations [35]. The use of the above mentioned fluorescence lines allowed to check "on line" the linearity in the energy spectrum as needed to obtain a correct Kurie plot. The properties and performance of the four detectors are reported in Table I. The difference in these performances is mainly due to the quality of the absorber. The mass of the absorber has been precisely measured in detector 2 for a determination of the ^{187}Re lifetime.

The spectra obtained with the three bolometers exposed to the Sn fluorescence lines are shown in Fig.1. The peaks at 3.44, 3.66, 3.71, 3.75 and 3.91 keV (Fig.2) are due to the Sn fluorescence lines $L_{\alpha_{1,2}}$, L_{β_1}, L_{β_4}, L_{β_3}, and L_{β_2}, respectively. The shoulder at 3.69 keV which appears in the spectrum of detector 3 (Fig.2c) is due to the possible K_α fluorescence line of calcium mentioned above. The conversion of the pulse height to energy has been obtained using a naïve preliminary model which accounts for the non-linear response of the detector. The deviation from the expected line position is shown in Fig.3, where the error bars include both the statistical and systematic contributions. The somewhat larger deviations of the Mn peaks is due to the inadequacy of the model at higher energies, which becomes less relevant at low energies. Forth order polynomials fit much better the data, but are less reliable below 3 keV , due to the lack of X-ray lines in this energy region. Even with our naive model, however, the deviations from the expected line positions are within few electron volts. Near the ^{187}Re β end point the calibration accuracy is fully adequate to the aims of the present paper. Future work include the realization of a source with fluorescence lines closer the end point and a better modelling of the calorimeter response.

The Kurie plots obtained with the four detectors presented in fig. 4 show a considerable similarity even if obtained with different bolometers and in particular with thermistors of different type. They are corrected for the screening effect according to the calculations of W. Buhring [21]. We would like to note that the mean free path in silver perrhenate for electrons with energies from 100 to 2600 eV should range from 4 to 18 monolayers, respectively [36]. The distortion of the spectra due to escaping electrons is therefore negligible.

The Kurie plot obtained with detector 1, which has by far the largest statistics, yields a transition energy of 2460 ± 5_{stat} ± 10_{syst} eV, where the systematic error is mainly due to the source induced background. This energy is definitely lower than the values [9] obtained in previous direct measurements [16-18]. It is on the contrary in reasonable agreement with the value of 2481 eV, reported recently in a cryogenic experiment with metallic rhenium by the Genova group [32].

The Kurie plot obtained with detector 2, where the mass of the absorber was carefully measured, yields a half lifetime of (43 ± 4_{stat} ± 3_{syst}) Gyr for the decay of ^{187}Re. This figure is in excellent agreement with the above-mentioned values determined with mass spectroscopy, and with the latest result obtained with a proportional counter [23]. Yu.A . Akulov and B.A. Mamyrin [37] have recently studied the possibility that the rate of beta decay depends on atomic or molecular structure for tritium decay. In ^{187}Re this effect should however be largely within our experimental errors.

Due to insufficient statistics and the role played by the background, we prefer not to give for the moment a value for the mass and for the square of the mass of the electron antineutrino.

4. CONCLUSIONS

Our measurements are still preliminary, but show the potentiality for an experiment on ^{187}Re β decay with dielectric crystals of rhenium to overcome the difficulties inherent to the superconductivity of this element.

We plan to develop our further activity in view of the measurement of the neutrino mass by:

a. optimization of the NTD , and improvement of the Si:P implanted thermistors
b. optimization of the quality of different crystals of perrhenate and other compounds of rhenium.
c. completion of the calibration system with fluorescence lines obtained with a common source with shutter
d. fabrication of large arrays of microcalorimeters with crystals of rhenium compounds. The electronics for the contemporary read-out of ten channels has been already implemented. We believe that this approach is essential to

overcome the main difficulty of a microcalorimeter: pile-up due to the relatively large rise time that prevents the use of a single massive crystal. A Montecarlo code has been implemented to simulate the statistical sensitivity of such arrays. Description and results of this code are outside the aim of the present paper. We anticipate that a sensitivity definitely better than ~ 10 eV on m_ν can be reached with an array of 10 bolometers similar to the current ones, operating for a year. An improvement of the energy resolution, of the risetime and on the number of detectors could considerably increase this sensitivity.

The study of the BEFS effect in our bolometers would be of great interest and complementary to the search on metallic rhenium by the Genoa group. Comparisons of our spectra with the more complex calculations for silver perrhenate are in progress. We would like to note, however, that the nearest atoms dominate the BEFS effect. In our case they are those of oxygen with a low cross section. As a consequence the possible effect of BEFS on the determination of the neutrino mass considered by W. Stoeffl [38] should be less pronounced.

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FIGURE CAPTIONS

Fig.1 Spectra obtained with the three detectors calibrated with the fluorescence L lines of Sn. The peaks due to the K lines of ^{55}Mn are reduced by a factor of ten for sake of clarity

Fig.2 Details of the spectra in the region of the Sn L fluorescence lines

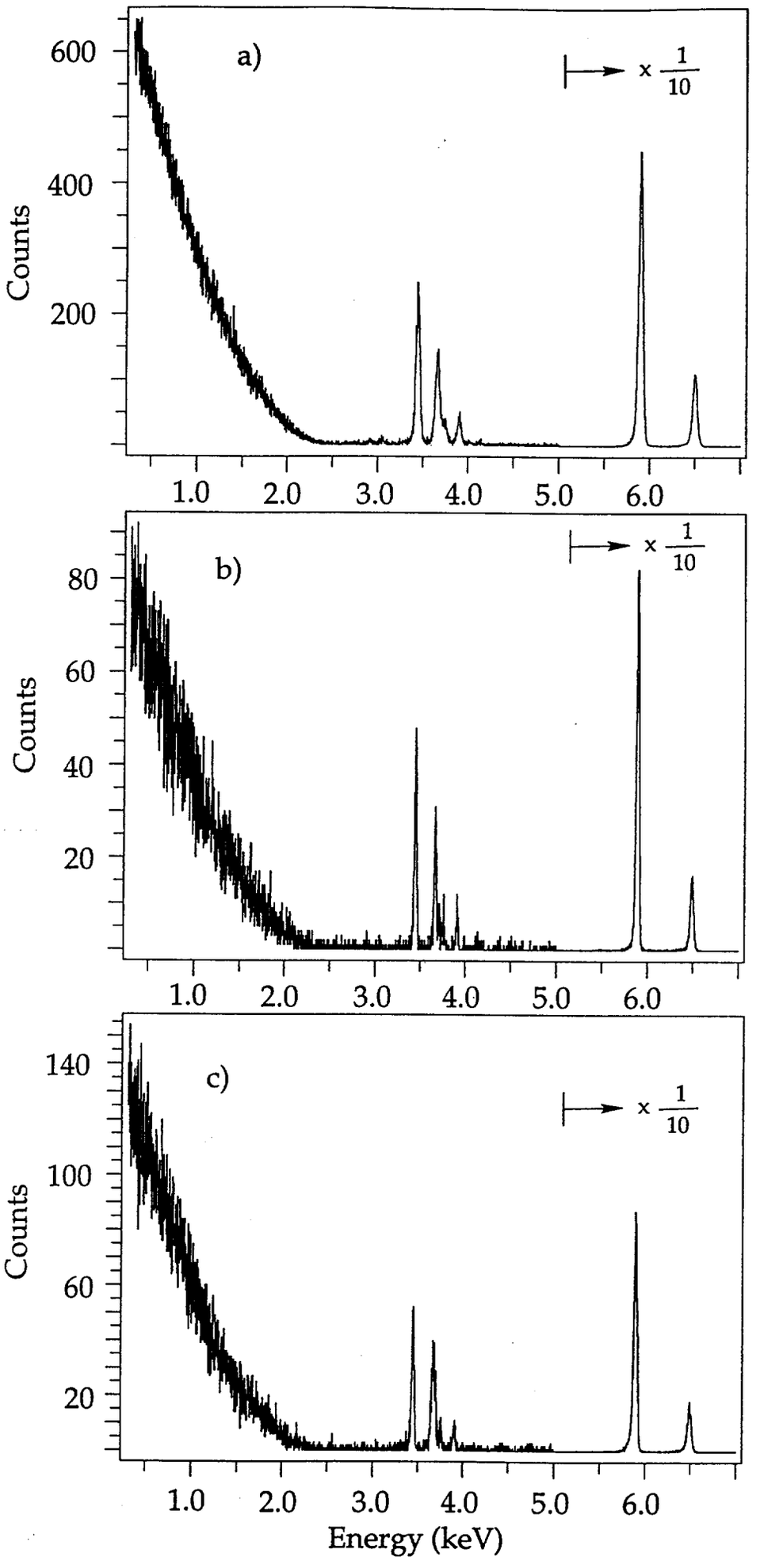
Fig.3 Deviations from the expected position of the Sn L and Mn K lines in the three detectors

Fig.4 Kurie plots with the four detectors. The plot of detector 4 is pratically coincident with that of detector 2. We have multiplied it by a factor of 1.5 for sake of clarity

Table I : Properties and performance of the three detectors

	Detector 1	**Detector 2**	**Detector 3**	**Detector 4**
Thermistor	Si:P	NTD	Si:P	Si:P
Absorber mass (μg)	~250	434 ± 13	~250	~500
Running time (h)	720	66	135	144
Temperature (mK)	60	70	60	60
Risetime (ms)	1.5	0.5	1	4
FWHM baseline width (eV)	55	25	24	55
Δ_{FWHM} **for Sn L lines** (eV)	60	29	25	62*

*This detector has been roughly calibrated with the K_α Ti lines


a)
b)
c)
x 1/10
Counts
Energy (keV)
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400
200
80
60
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20
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100
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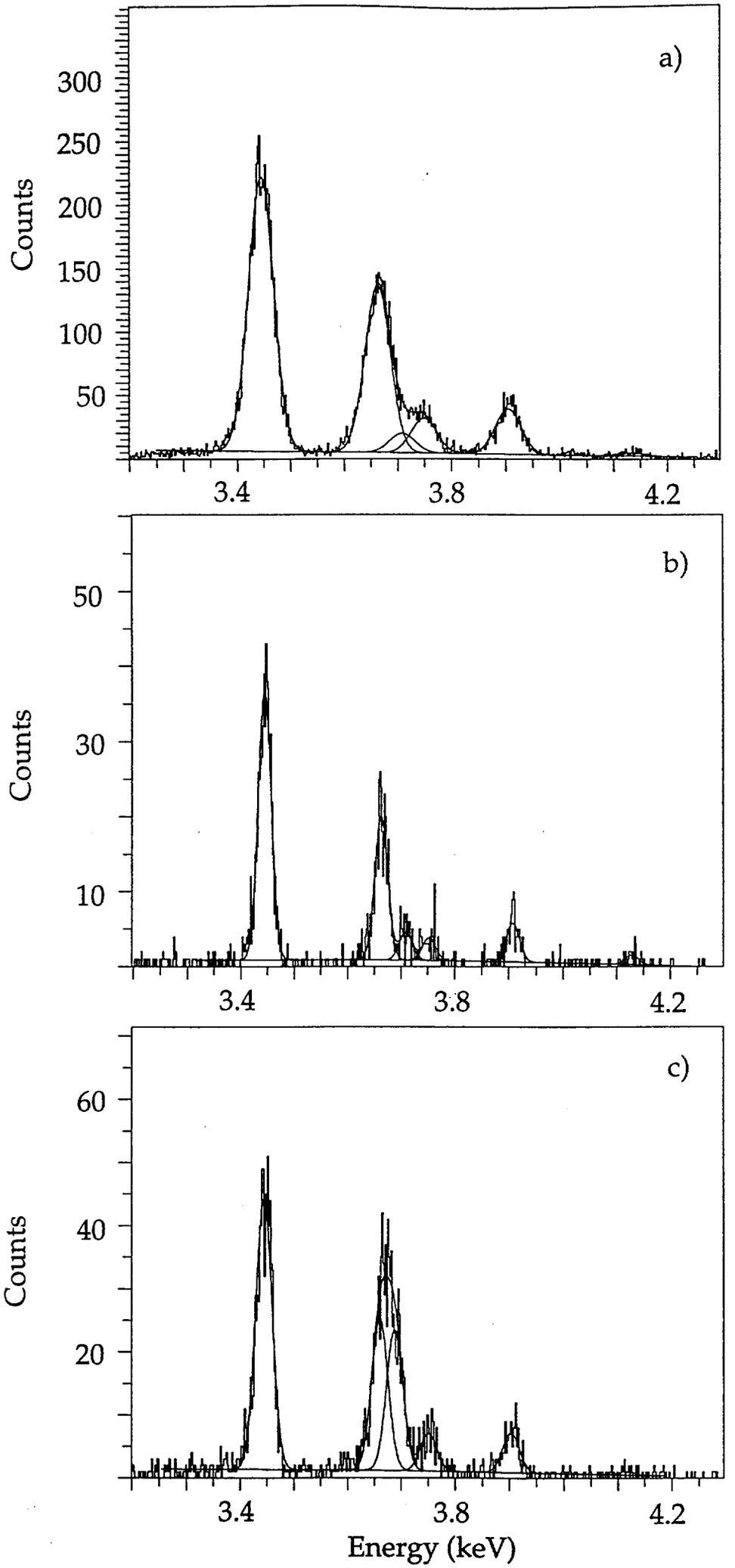
3.0
4.0
5.0
6.0

Fig. 1


a)
b)
c)
Counts
Counts
Counts
300
250
200
150
100
50
50
30
10
60
40
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4.2
Energy (keV)

Fig. 2

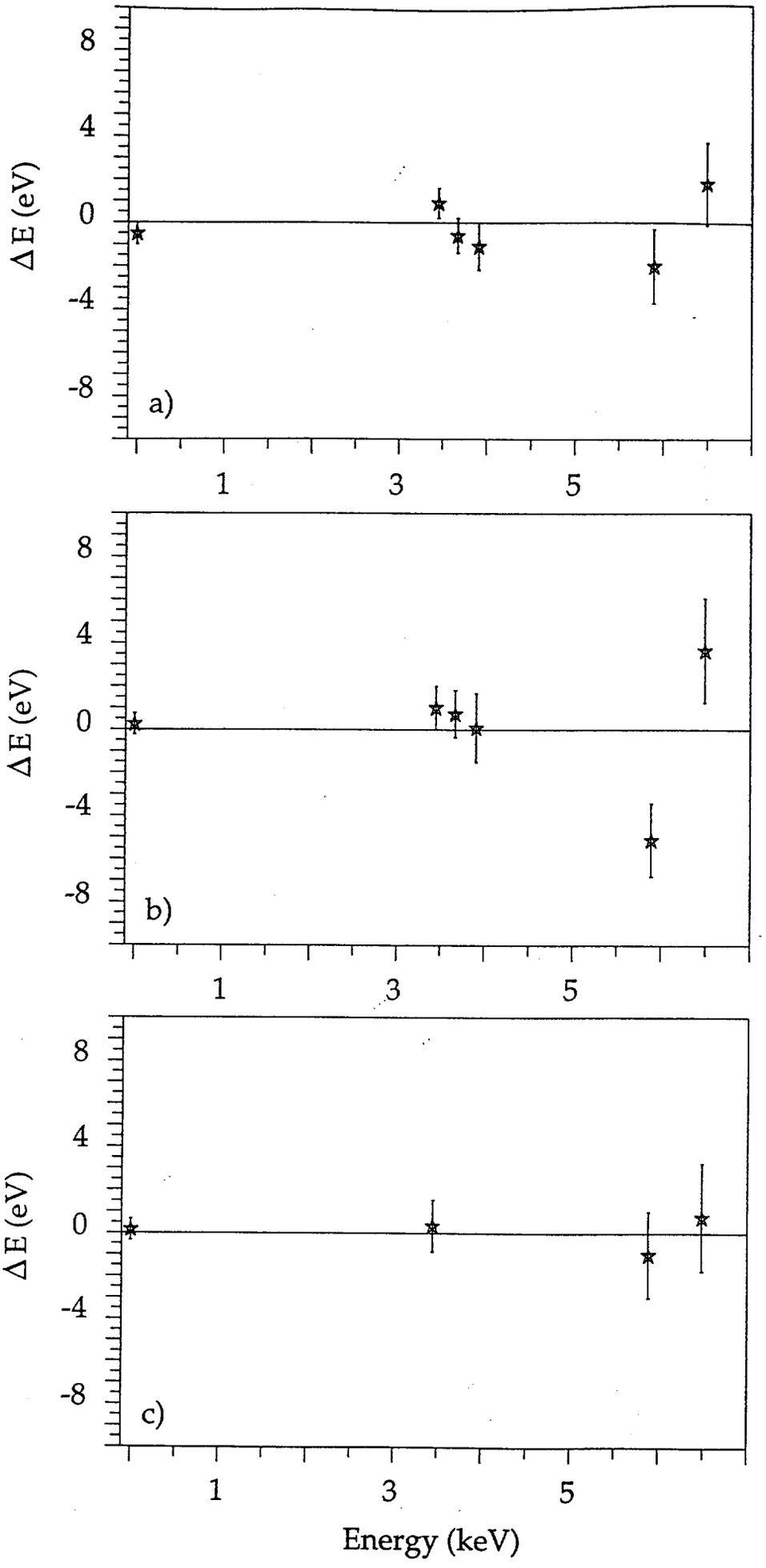

Fig. 3

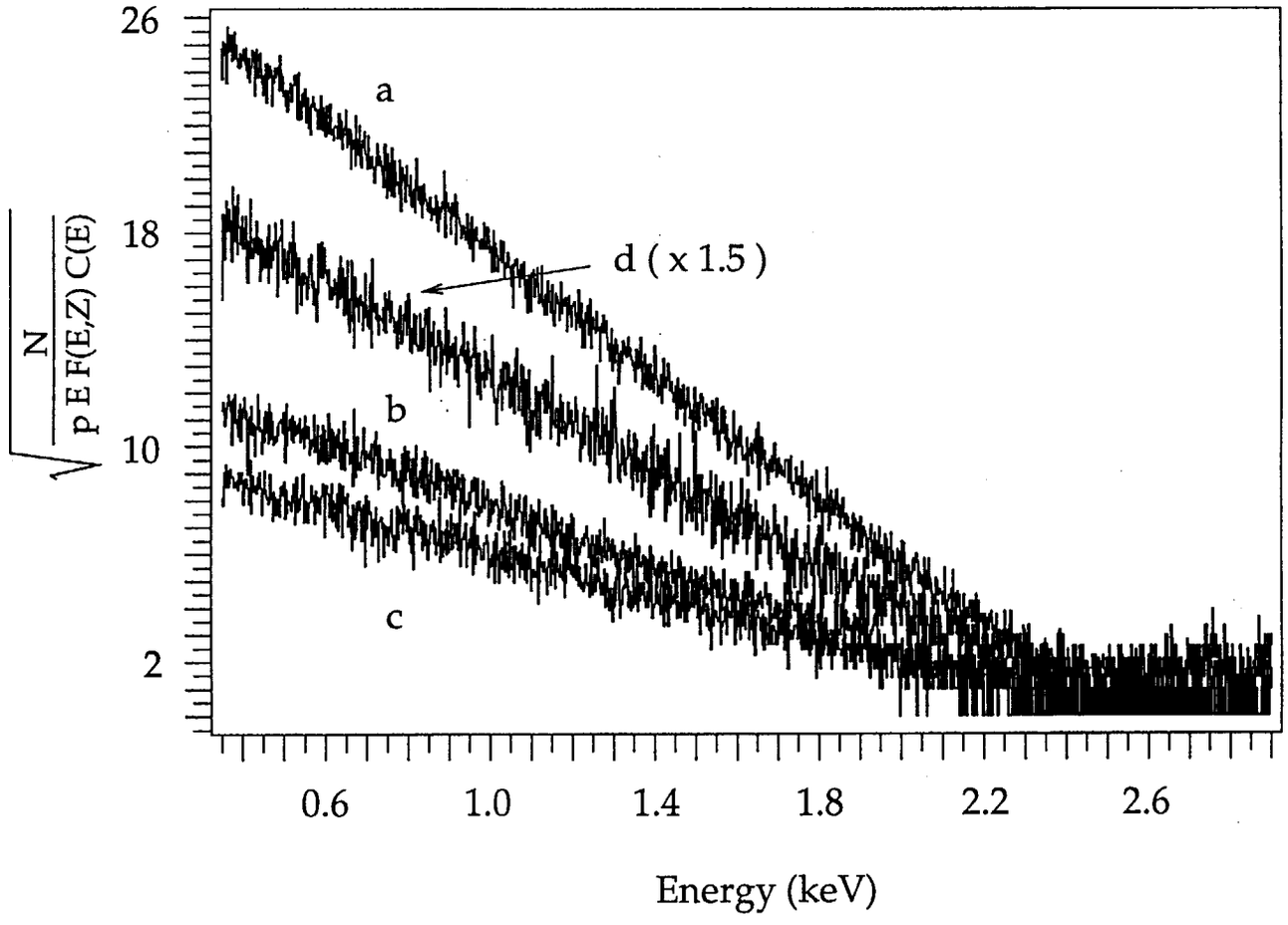

Fig. 4