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Ge(Li) DETECTORS FOR THE ENERGY RANGE 200-1700 keV

by

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A B S T R A C T

Gamma ray pulse height spectra for seven radioactive sources have been analyzed for the full energy peak areas. Using these areas in conjunction with the results of an extensive compilation of relative gamma ray intensities, a relatively simple efficiency function has been used to calibrate a coaxial Ge(Li) detector. The resulting relative intensities for each source have been used in investigating the consistency of the weighted averages of the compiled relative intensities. Possible systematic errors are discussed and recommendations are made with respect to the choice of gamma ray intensity standards.

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I - INTRODUCTION

Routine usage of cooled Ge(Li) detectors in gamma ray spectroscopy requires a measurement of the efficiency of the detector as a function of the gamma ray energy as well as the calibration of the pulse height in terms of energy. An efficiency measurement may be either absolute or relative depending on the necessities of the experiment. In principle, a relative efficiency measurement is simpler since it does not require the use of calibrated intensity sources. In the simplest case, one convenient gamma-ray source with conveniently spaced lines of known relative intensity would suffice. In most cases, several sources are used to measure the relative efficiency over the energy range of interest (which in our work is 200- 1700 keV). While this does add slightly to the complexity of fitting the efficiency function, the simplicity of data taking is greater, since source to source variations in count rate, geometry, etc. are minimized with a net advantage over absolute measurements.

The accuracy of the efficiency measurements will depend on the spectral analysis (the functional form of the energy response used to determine the areas and, of course, the counting statistics), the "reliability" of the efficiency function used to interpolate to energies between those actually measured, and the accuracy of the intensity values of the calibrating sources. A great deal of work has been carried out in spectral analysis¹⁾, development of suitable efficiency functions²⁾, and measurements of absolute and relative intensities for a large number of sources. Up to the present time several authors²⁻⁴⁾ have claimed accuracies of 1 or 2% for absolute efficiencies over the energy range \approx 150 to 1400 keV for several types of cooled Ge(Li) detectors. The main interest of this work is an investigation which could provide a consistent set of relative intensity values for a number of widely used gamma-ray sources.

II - SPECTRAL MEASUREMENTS AND ANALYSIS

A liquid nitrogen cooled true-coaxial Ge(Li) detector of 56 cm³ (resolution of 2.3 keV at 1.33 MeV) was used to measure the gamma-ray emission spectra of seven radioactive sources. The detector was dc-coupled to a Philips uncooled FET preamplifier followed by an Ortec 451 Spect.Amp. and a model 720 Northern Scientific 4096-channel pulse height analyzer (PHA). Because the efficiency of the detector was to be used in work with fixed source-detector distances, we tried to maintain this distance at 10 cm in the present set of measurements. Although relative measurements don't require dead-time analysis, other count rate effects may complicate the spectral analysis (e.g. pulse pile-up will broaden the peak shape and large count rate variations may be more than the electronic correcting circuits can handle). Thus we used sources which gave only minor variations in the PHA dead time rate. Since measurements of ²⁰⁷Bi and ¹³³Ba between 5 cm and 15 cm showed no observable change in relative intensities we decided to take the data for ^{110m}Ag at 75 cm to compensate for the relative weakness of the source.

A background spectrum of gamma-rays coming from the concrete walls (essentially ²²⁶Ra, ²³²Th and ⁴⁰K) was measured, but the corrections never exceeded 2%. Because of the sizes of our sources, the self-absorption in the worst case (120 keV in ¹⁸²Ta) required less than a 1% correction.

The full energy peak (FEP) areas were determined with a least squares fitting program. Initially the shape of the FEP was fit to a simple Gaussian with a quadratic background. Fits with this simple function were unsatisfactory for the more intense lines, and more complex forms⁵⁻⁶⁾ for the peak shape were tried. The best results, smallest $\chi^2_{d.f.}$ (see discussion eq. 3), were obtained with a Gaussian with matched exponential wings as suggested by Routti and Prussin⁶⁾. The parameters of

this form varied smoothly with energy, gave reasonable values of $\chi_{d.f.}^2$ for nearly all peaks, and the error estimates, from the combination in quadrature of the total number of counts and the standard deviation of the fitted background

$$\sqrt{\sigma_{\text{peak area}}^2} = \left(\sqrt{\sigma_{\text{total area}}^2 + \sigma_{\text{BKG}}^2} \right) f ; f = \begin{cases} 1; & \chi_{d.f.}^2 \leq 1 \\ \sqrt{\chi_{d.f.}^2}; & \chi_{d.f.}^2 > 1 \end{cases}$$

behaved reasonably. In order to avoid introducing possible systematic errors between peaks with large and with small areas and at the same time get the error on the small peaks to a reasonable level we determined the areas of the ^{182}Ta , ^{154}Eu , $^{110\text{m}}\text{Ag}$, and ^{125}Sb sources by fitting the spectra with the parameters of the matched wings fixed according to the gamma-ray energy. While this makes only a minor difference in the areas of the small peaks it makes a large difference in the error estimates. It happens that the shape parameters and the background interact in a correlated way such that the area remains nearly constant, but the errors become large if the background and shape parameters are all free during the fittings.

III - INTENSITY AVERAGES

The selection of appropriate radioactive sources will depend on the energy region of interest, availability of the sources, and the reliability of the relative intensity information for the sources. Based on these criteria the seven frequently used sources have been divided into two groups. The first group consists of ^{133}Ba , ^{94}Nb , and ^{207}Bi . This group of sources is characterized by relatively simple decay schemes with at most four strong lines and a high degree of internal consistency. (See Tables 1-3). These were

used in the initial efficiency calibration of the detector.

(See Section IV) . The second group (^{182}Ta , ^{154}Eu , $^{110\text{m}}\text{Ag}$, and ^{125}Sb) is characterized by rather complicated decay schemes, low internal consistency and complicated spectral shapes (e.g. the doublets in ^{125}Sb), but they have the compensating advantages of many strong lines emitted over a wide energy region. Thus, with a single source, the efficiency function could be accurately determined over a wide energy region.

The intensities of the lines in the first group have been averaged by weighting the values taken from the literature:

$$\bar{I}_w = \sum \frac{1}{\sigma_i^2} I_i / \sum \frac{1}{\sigma_i^2} \quad (1)$$

$$\frac{1}{\sigma_{I_w}^2} = \sum \frac{1}{\sigma_i^2} \quad (2)$$

The consistency of the data can be estimated from the χ^2 test simply by calculating

$$\chi^2 = \sum \frac{1}{\sigma_i^2} (I_i - \bar{I}_w)^2 \quad (3)$$

and using the tabulations of the χ^2 probability for $|N-1|$ degrees of freedom. A rough rule of thumb is that $\chi_{d.f.}^2 (= \chi^2 / |N-1|)$ should be between 0.75 and 1.5 around 2/3 of the time. This is one test for "good" data.

Also, if the data have about the same errors then

$$\bar{I}_s = \frac{1}{N} \sum I_i \quad (4)$$

$$\sigma_{I_s}^2 = \frac{1}{N(N-1)} \sum (I_i - \bar{I}_s)^2 \quad (5)$$

should give about the same values as eq.(1) and (2), respectively.

The results for the first group are given in Tables 1-2 with the individual values for the lines listed in the first columns, followed by I_s , I_w and $\chi_{d.f.}^2$ given in the last three columns. The errors of I_s and I_w are given by the corresponding equations (5) and (2). The values of $\chi_{d.f.}^2$ for each line and for each author are in the

appropriate row and column, respectively. Except for the weak lines of ^{133}Ba , which are included in the group 1 tables only for the sake of completeness, one sees that all of the standard deviations of the I are $\leq 1\%$ with values of $\chi^2_{\text{d.f.}}$ indicating a very high internal consistency. The percentage range of the intensity values for one of the strong lines varies up to 7% eq. in ^{207}Bi . The low values of $\chi^2_{\text{d.f.}}$ for the authors reporting on ^{207}Bi [$\chi^2_{\text{d.f.}} \leq 0.446$ only 20% of the time] are probably indicative of error estimates which are larger than one standard deviation. The relative intensities for ^{94}Nb (Table 3) come from existing decay scheme information.

The intensity values for these sources from the present work were measured using the efficiency function determined from a weighted fit of all 81 lines in the seven sources as described in section IV (Separate fits of the group 1 and of the group 2 sources gave essentially the same results). The fits to the intensity values were done twice, with and without our results included in the averages and the resulting fits were essentially the same. This is what one would expect when the results are as consistent as the values in Tables 1-2 .

IV - RELATIVE EFFICIENCY FUNCTION

The choice of an efficiency function for interpolating the measurements over a large energy region is critical since good intensity information and careful spectral measurements may be seriously degraded by a poor choice. The disadvantage of using only the nine reliably known lines of the first group is the possibility of selecting and fitting an efficiency function which does not close to the "true" function in the regions where the data is missing or sparse (e.g. between the 1064 and 1770 keV lines). Although it should be possible to arrive at the efficiency function of a detector of known dimensions by a Monte Carlo procedure⁷⁻⁸⁾, given sufficient

computing time, many researchers^{3,9)} have turned to approximations of a semi-empirical nature in which the photoelectric absorption and Compton scattering events are explicitly considered in terms which depend on the cross sections for the events, i.e.

$$\epsilon(E_\gamma) = p_1 \left[1 - \exp(-p_2 \tau_{pe}) \right] + p_3 \tau_{ce} \exp(p_4 E_\gamma) \quad (6)$$

or of a purely empirical form²⁾ such as

$$\epsilon(E_\gamma) = p_1 \exp(-p_2 \ln E_\gamma) - p_3 \exp(-p_4 E_\gamma) + \dots \quad (7)$$

The present data has been fitted with functions essentially like eq.(6) with 5 parameters and eq.(7) with 4 parameters(see Fig.1). In order to decide which of these forms to use, fits were done employing only group 1 data and with group 1 and group 2 data. The 4 parameter fits gave $\chi^2_{d.f.}$ of 1.45 for the group 1 data and 1.26 for the combined group 1-group 2 data. Two versions of functions like eq.(6) were tried. Eq. (6) fits to group 1 data gave a consistently higher $\chi^2_{d.f.}$, e.g. 1.97 vs 1.45 for eq. (7). A second version of eq. (6) gave a large $\chi^2_{d.f.}$ (≈ 7) for all of the lines. It should be noted that even though eq.(6) gives a poorer fit to the data, one can not use a χ^2 test to prove that it is "wrong" for our detector. One can not be sure that the errors for the group 1 intensities are not slightly too small, or that one of the intensity values isn't slightly wrong with a biased effect on the fit to eq.(6). The consistency of the group 1 source intensities suggests that eq.(7) is better than the others which were tried. Another aspect of the fit to eq.(7) is the fact that 70% of the data deviate by less than 4% from the fitted curve while for eq.(6) the deviations reach 8%. Another test of the analysis was to do separate fits to data with poor statistics and data with good statistics using eq. (7) with essentially the same result for the efficiency function. This was done primarily to test the spectral analysis for bias

between the large and small peaks.

It must be noted that the two functions differ by as much as 3% in some regions (and diverge below 150 keV). This means that we must allow for the possibility of systematic errors. However, the fractional deviations between the present results and the fitted function (eq.7), shown in figure 2, show no obvious trends above 200 keV. [See section V.3] Figure 2a shows that the efficiency is not well known for the region $100 \text{ keV} < E < 200 \text{ keV}$ with ^{182}Ta and ^{133}Ba quite low with respect to ^{125}Sb and ^{154}Eu . While ^{182}Ta has been measured with a crystal spectrograph it has the problem of getting accurate intensity values jumping in energy from 264 keV to 928 keV. There was also a problem with the spectral analysis of the 123 keV line of ^{154}Eu which had consistently poor fits and areas larger than expected. The discrepancies in the region from 100 to 200 keV could be avoided by adopting a cutoff at 200 keV or by stating errors which "patch up" the problems. The former is convenient for us since the detector is to be used for higher energies; however, it seems to imply that nothing is known below 200 keV. While we must increase our errors to avoid biasing the averages (there is no way to resolve the problems without more measurements), we have no explanation for the small apparent source to source variations below 200 keV.

V - RESULTS AND DISCUSSION

V.1 - Group 2 Results and Reliability

The results of this work for the intensities of the group 2 lines have been incorporated in Tables 4-7 along with the values from the literature averages, etc. in the same format as the group 1 data. The intensity values from the literature have been "censored" in that work which appeared to be systematically different

(not wrong but different) has been omitted. While a simple human error could induce a systematic error (say an error in the spectral analysis of the 100 line) it is just as possible that the censored works have used better efficiency functions. They have been omitted simply because they are not consistent.

The spectral analyses which were performed on the present data were compared for consistency. While any comparison with the simple Gaussian fits may show some bias (since those fits had large $\chi^2_{d.f.}$ and may have had distorted background shapes), it is suggestive that the areas of the matched exponential wings analysis were consistently 1-2% larger than those from the Gaussian analysis when the errors were $\leq 1\%$. While this trend seemed to be observable up to 2% errors it was washed out by the errors above 2%. The cause of the differences in the areas was not apparent in the region of the background itself, but rather in the region under the peak where the backgrounds were observably different. While it is desirable to fit doublets with a common background, such an analysis may cause (hopefully small) bias in the areas unless the function used is very close to the true peak shape. Any unusual feature of the background data, e.g. a Compton edge, may cause fitting errors even for a single peak. Unless careful, elaborate, and time consuming peak stripping is used to do the spectral analysis, it would appear that some bias could be caused in almost any analysis.

A potentially more serious source of systematic errors arises from the choice of the efficiency function itself. Reflection will show that even if an estimate of the potential systematic error were available it would have to be specified as a function of energy to be useful. In the case of the area analyses, one can say that the "strong" lines (say $I=0.2$ or larger) should have very small systematic errors and weaker lines may have errors of 2%. But any systematic error in the efficiency function is energy dependent and, in the case of

relative intensity values, is a very complicated function of the two energies involved. In the present work eq.(6) and eq.(7) best fits had two regions of significant differences. If one assumes that the true efficiency function lies somewhere between eq.(6) and eq.(7), the relative deviation ranges from +3% to -2%. For an arbitrary pair of lines, the systematic error in the intensity ratio may be as much as $\pm 5\%$. In the absence of an "a priori" way of determining the efficiency function the problem of estimating the systematic errors has no solution, nor even an apparent way of approximating the effects in any single experiment. Fig.1 shows a plot of the efficiency function data and fits to both eq.(7) and eq.(6). One can argue that, since there are no apparent systematic differences above 200 keV (see Fig. 2b), it is unlikely that significant systematic errors exist in the choice of eq.(7) for the efficiency function.

V.2 - Quality of the sources for calibration

Of the group 2 sources, ^{110m}Ag and ^{182}Ta appear, on the basis of internal consistency, to offer at present the best prospects for a good efficiency determination. They have good consistency and the range and density of lines is good. There is a lack of low energy lines in ^{110m}Ag , while ^{182}Ta has a gap in lines between 264 keV and 928 keV. Except for the problem of "connecting" the ^{182}Ta lines they would be a good choice. The source ^{133}Ba with ^{110m}Ag would be a good choice to cover the energy range 200-1500 keV and ^{182}Ta could then be used to extend the energy range if needed.

^{125}Sb would be useful at the lower energy range except for the difficulties which arise in extracting the areas of the doublets. The errors in the intensities of the more intense lines are on the order of 1.5% ($\sqrt{\chi^2_{d.f.}}$ is greater than 1.) whereas the strong lines of ^{110m}Ag and ^{182}Ta have errors around 1%.

^{154}Eu has poor internal consistency, but it would be the best choice for a single source from the standpoint of the spacing (except at low energy) and of the range of the strong lines. While the present work is primarily concerned with the energy region above 200 keV, it was unpleasant to observe that the 123 keV line was very high with respect to the other sources' low energy lines. (See fig. 2a). However, it should be pointed out that even the $\chi^2_{\text{d.f.}}$ was still very large when it was fit with the matched wings function. It is also the case that the 248 keV line has a Compton edge under this peak. Its omission here is on the basis that it is not relevant to our work and for our experimental conditions it would require special analysis.

V.3 - Recommendations for intensity values

Without both a "definitive" work on efficiency functions for Ge(Li) detectors and consistent values for the multiline sources, the best approach would seem to be to adopt a statistical approach similar to that presented here. One assumes, that in averaging over several sources and several different authors for each of the sources, that biases in the averages will tend to decrease since different spectral analyses and different efficiency functions have been used. Tables like those presented here also give information about consistency and quality of the data, presence of "safety" factors in errors reported, etc. Even for a single source it would be a prohibitive effort to evaluate the individual author's data assuming that one could get sufficient information to do so.

The danger in a statistical approach is illustrated in fig. 2a where the data at low energies is shown in comparison to the fit to eq.(7). On average the fit seems fine, until one notices that the deviations for the individual sources are correlated, i.e. some

of the sources are above the fit and others below it. While for the present study this isn't a vital point, it does show that even taking one source may cause a serious error. Of the sources studied here, only ^{182}Ta has a measurement by means other than solid state detectors with high resolution. One would hope that these low energy relative intensities would be somewhat more accurate.

It is also interesting to note that fits to the semi-empirical efficiency function eq.(6) resemble the Monte Carlo results of Aubin et al. This suggests that they do have a reasonable physical basis. The problem would seem to be to find a way to include geometrical effects for different types of detectors so that one doesn't have to perform a Monte Carlo simulation of each detector.

In summary, the quality of any individual report of intensity values is likely to be dominated by the systematic errors induced by the choice of efficiency function representation. These errors, which may be as much as 5 % compared with 1-2% for the intensity values, will dominate until a good "a priori" description of the efficiency function is found.

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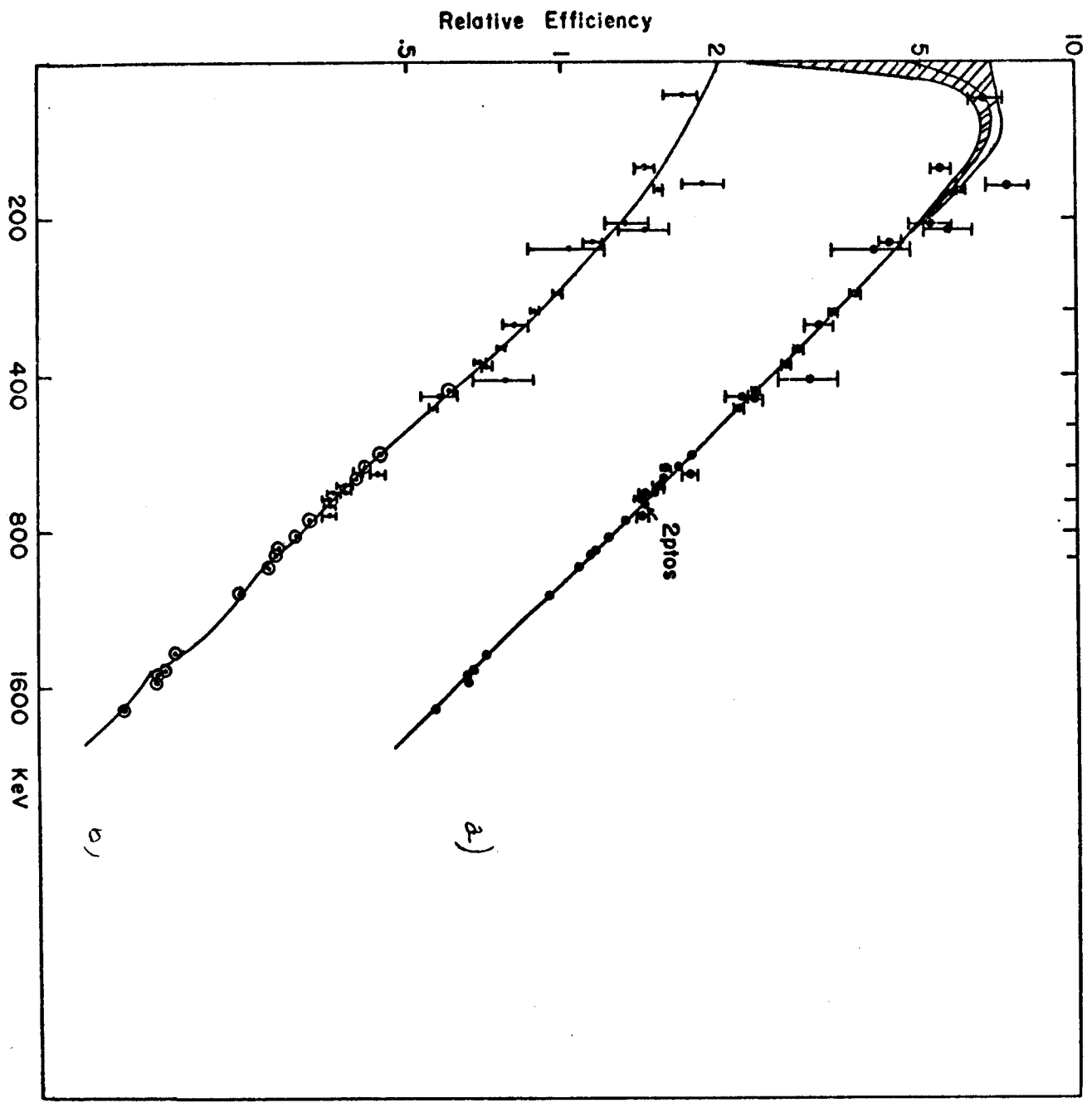


FIG. 1

Figure 1 ;

Relative gamma ray intensities as a function of energy:

a) eq.(7) b) eq.(6) For clarity

b) has been displaced by a factor of 0.4 .

Figure 2 ;

Gamma ray intensities (relative) in relation to least squares fit to eq.(7) E_{exp}/E (eq.7) for an energy range of :

a) 100 to 400 keV

b) 400 to 1800 keV

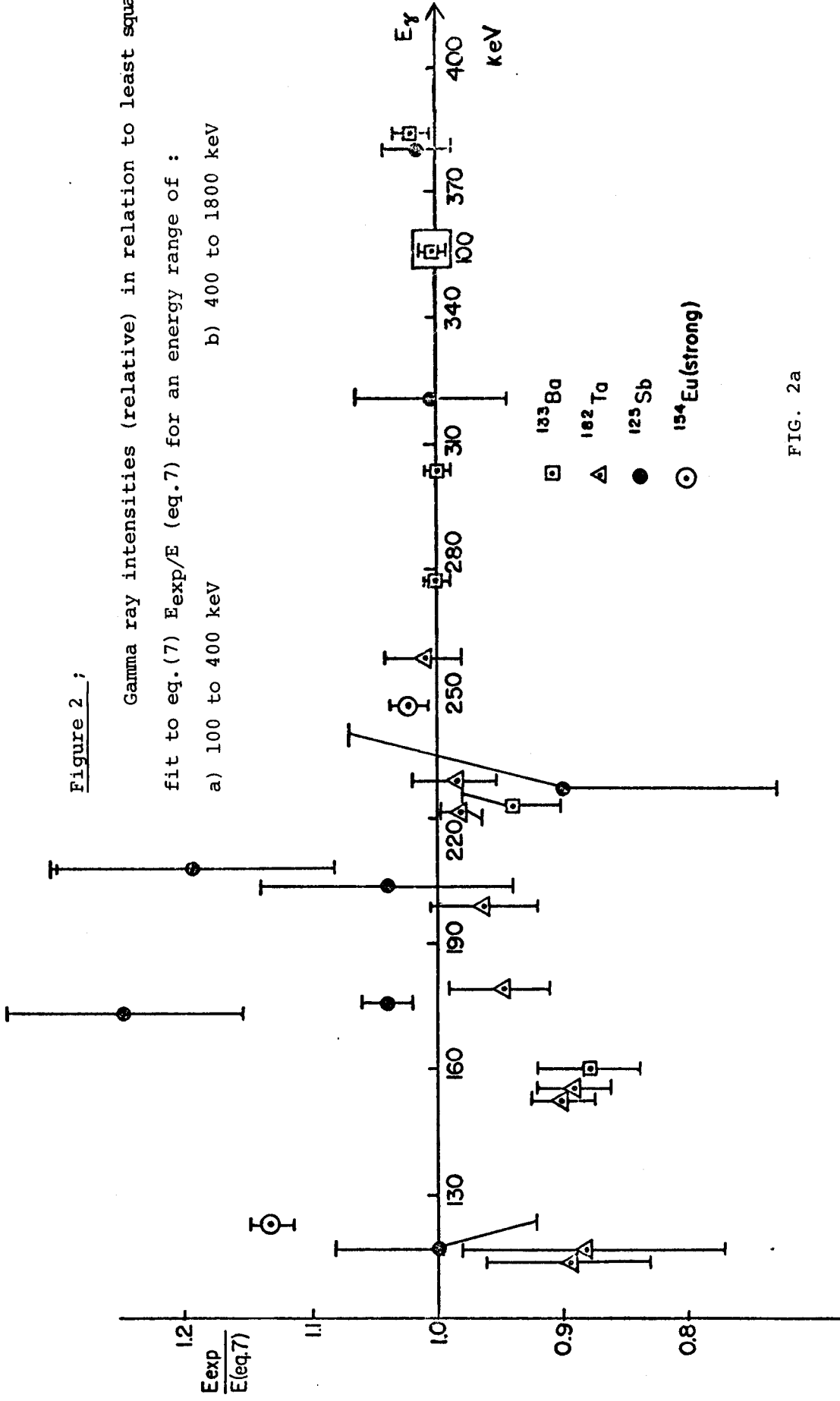


FIG. 2a

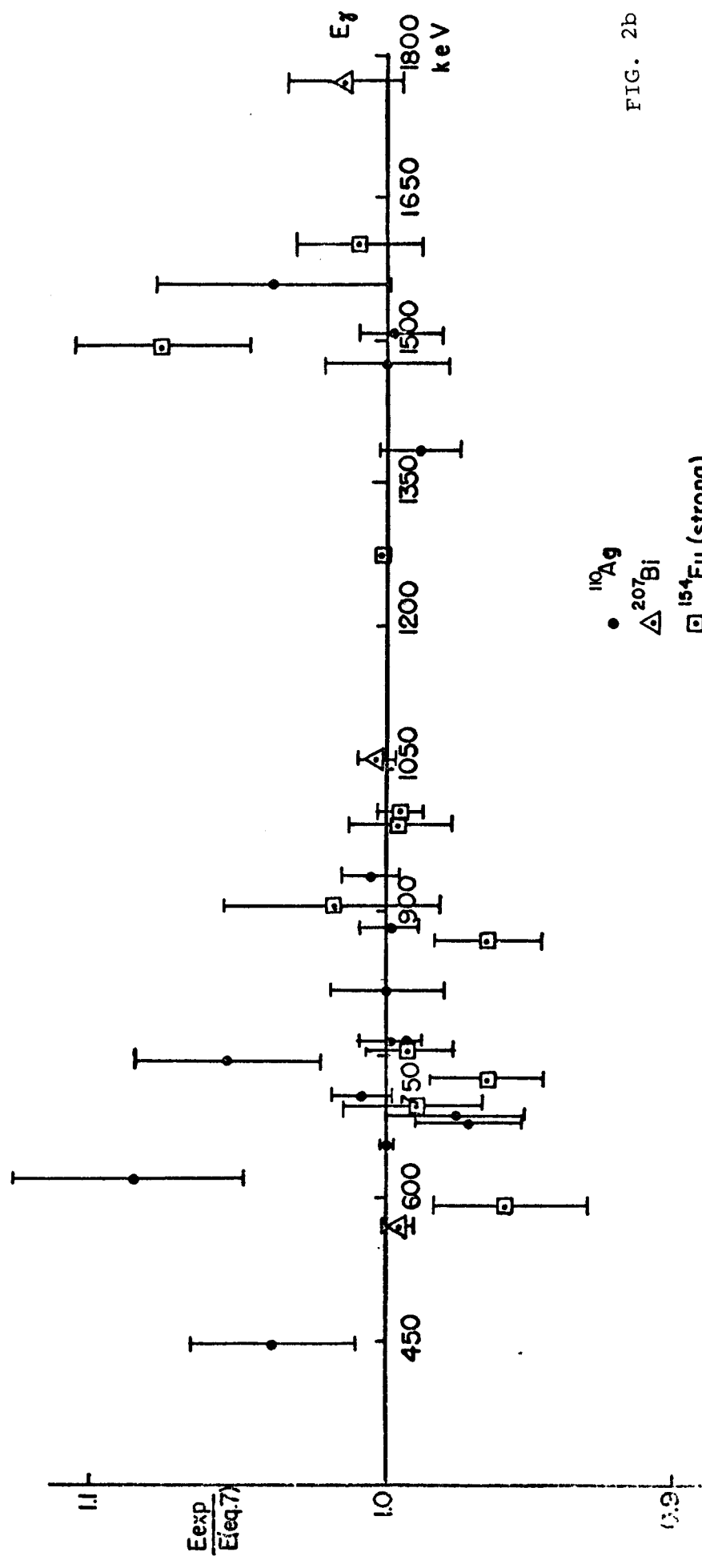


FIG. 2b

- ^{110}Ag
- △ ^{207}Bi
- ^{154}Eu (strong)

TABLE 3

Relative gamma ray intensities in the decay of ⁹⁴Nb

Energy relative intensity

(keV)

702.6 98±2
871.1 100

TABLE 4

Relative gamma ray intensities in the decay of ¹⁰²Ta

Energy (keV)	REF. 23	REF. 24	REF. 25	REF. 26	REF. 4	Present work	$\bar{I}_{\gamma, \text{OTR}}$ exp. (1,2)	χ^2 d.f. (line)
100.1	40.65	1.79	0.0	0.0	40.80	40.60	40.59	0.67
113.7	5.53	0.30	0.0	0.0	5.43	4.95	5.28	0.04
116.4	1.28	0.08	0.0	0.0	1.26	1.18	1.23	0.54
152.4	20.70	0.80	21.30	0.0	20.50	19.59	20.35	0.12
156.4	8.13	0.40	8.10	0.20	7.77	7.43	7.73	0.44
175.4	9.31	0.41	9.60	0.30	9.10	8.88	9.06	0.59
198.4	4.35	0.25	4.40	0.0	4.31	4.13	4.30	0.51
222.1	22.80	0.90	22.60	0.0	21.90	21.75	21.92	0.14
229.3	11.30	0.50	10.90	0.60	10.60	10.39	10.66	0.49
246.1	10.90	0.50	10.60	0.30	10.50	10.36	10.46	0.74
928.0	0.0	0.0	1.79	0.09	0.0	1.53	1.64	0.47
959.7	0.0	0.0	1.02	0.06	0.0	0.91	1.08	0.50
1001.7	0.0	0.0	5.98	0.30	0.0	5.99	5.86	0.94
1113.4	0.0	0.0	1.13	0.10	0.0	1.18	1.17	0.18
1157.3	0.0	0.0	1.84	0.35	0.0	0.0	1.92	0.10
1158.1	0.0	0.0	0.99	0.28	0.0	0.0	1.93	0.12
1189.0	0.0	0.0	47.40	0.70	0.0	0.0	0.82	0.51
1221.4	0.0	0.0	79.30	1.20	46.50	47.61	46.86	0.80
1231.0	0.0	0.0	33.40	0.50	77.80	78.14	77.97	0.40
1257.4	0.0	0.0	4.33	0.70	32.96	32.32	32.84	0.54
1273.7	0.0	0.0	1.90	0.40	4.26	4.33	4.30	0.07
1289.1	0.0	0.0	4.04	0.70	1.86	1.66	1.80	0.48
χ^2 d.f. (author)	0.60	0.18	0.38	0.50	3.86	4.06	3.94	0.48
					0.09	0.44	0.06	0.30

1121.3 100.0

