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APPLICATIONS OF GERMANIUM GAMMA-RAY DETECTORS

Berkeley, California

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D. A. Shirley

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Just over a year ago Ewan and Tavendale at Chalk River Laboratories announced that a single crystal of lithium-drifted germanium would serve, when operated at low temperatures and under a modest bias voltage, as a high-resolution γ-ray detector.[1] The nuclear physics community responded rapidly to this revolutionary development. Within two weeks Fred S. Goulding and W. L. Hansen had built our first Ge(Li) spectrometer at Berkeley. Soon after, because of their far higher resolution, Ge(Li) detectors replaced NaI(Tl) scintillation crystals for almost every serious investigation in γ-ray spectroscopy. In this article the applicability of Ge(Li) detectors to the various types of experimental investigations in the field of γ-ray spectroscopy is briefly discussed and evaluated. Illustrative examples are given from work in progress in the author's laboratory.

A little historical perspective is helpful in understanding the impact of the new semiconductor counters. Gamma-ray spectroscopy has long been handicapped by the low resolving power of the available spectrometers. Until 1950 most γ-ray spectra were studied by absorption techniques, in which absorbers of varying thicknesses were placed between the source and an energy-insensitive counter. The manner in which the transmitted intensity decreased with increasing absorber thickness was related to the energies and abundances of various components of the γ-ray spectrum. The analysis was rather similar to the resolution of decay curves into components with different half-lives, and the results

were also quite similar. If the spectra were very simple and the energies of the few components were quite different the technique was applicable, but for complex spectra it was completely inadequate.

In 1947 DuMond announced the completion of a quartz bent-crystal spectrometer.[2] This instrument has very high resolution for low-energy γ-rays (better than Ge(Li) below about 300 keV), but its application to nuclear spectroscopy is severely limited by low efficiency and by the fact that it is a single-channel device, i.e., the γ-ray spectrum must be scanned, with only one small energy interval being studied at a time. In a multichannel device, by contrast, signals from γ rays of different energies are sorted electronically after detection, and all energies may be studied simultaneously. For many research problems there is no substitute for very high resolution at the lowest energies, and bent-crystal spectrometers can be expected to make important contributions in such applications.

The workhorse in γ-ray spectroscopy for over a decade has been the thallium-activated sodium iodide crystal, and it is with this device that the new germanium detectors must be compared. Because of its multichannel character and its high efficiency, the NaI(Tl) spectrometer has enjoyed universal adoption in spectroscopy laboratories. Its low resolution, however, has made it a poor cousin in this respect among spectrometers, and the rush by spectroscopists from NaI(Tl) to Ge(Li) underscores the long-held view that this low resolution is simply inadequate for many purposes.

Performance of Ge(Li) Detectors

Because we are interested here mainly in applications, only a minimum description of the detectors is presented below. Goulding and co-workers have given detailed discussions of the fabrication and operation of Ge(Li) counters,[3] and Eugene Miner has designed Ge(Li) cryostats for many applications.[4]

The detectors are as yet small, ranging up to 6 cm^2 in area and ~1 cm in depth. Because capacitance should be minimized for high-resolution performance the area must not be too large compared to the depth. Increasing the depth is at present a major technological problem. The detectors are operated at liquid nitrogen temperature (77° Kelvin) to minimize noise. They are housed in specially-designed dewar vessels made in such a way as to minimize stray capacitance, and are operated under a bias of several hundred volts.

A γ ray is detected when it deposits energy in the Ge crystal, creating many electron-hole pairs. These are swept out by the bias voltage, and a pulse is developed in a charge-sensitive preamplifier. The large number of electron-hole pairs allows only a small percentage statistical energy spread, and this is the essence of the high resolution available with Ge(Li). Preamplifier noise is minimized by using an EC 1000 tube as the first stage. Another major problem is to lessen this noise still further; it presently accounts for over half the peak width below about 500 keV.

The spectroscopist is usually interested in three important properties: resolution, efficiency, and speed. Resolutions of the three γ-ray spectrometers are compared in Fig. 1. Sodium iodide is out of the running over the entire energy range, although in some applications it may be preferable to Ge(Li) at the very lowest energies because of low-energy noise properties. Above 500 keV Ge(Li) is about a factor of 20 better than NaI(Tl), a very substantial improvement indeed. For many applications in nuclear spectroscopy this margin can mean the difference between success and failure. Above about 300 keV Ge(Li) gives better resolution than the bent crystal. This crossing point should soon drop in energy as preamplifier noise is reduced, but it theoretically cannot drop below about 200 keV. For high resolution, then, the bent crystal will continue to have a definite advantage at very low energies.

The efficiencies of the two multichannel spectrometers are shown plotted against energy in Fig. 2. Here the roles are reversed, with NaI(Tl) possessing a clear superiority to Ge(Li). In applications where efficiency is necessary (e.g., angular correlations) NaI(Tl) will continue to be used, especially for high-energy work. While Ge(Li) can never make up the deficit of two orders or magnitude in efficiency at 1 MeV, it should be noted that the comparison made here is hardly fair, because a 3-inch thick NaI(Tl) crystal is being compared with a Ge(Li) crystal of only a few mm thickness. As the experimental curves of Ewan and Tavendale show, the efficiency of Ge(Li) improves with thickness.[5] There are two reasons for this: the increase of thickness means that more Ge atoms are in the path of incoming γ-rays and the probability for photoelectric absorption of the full γ-ray energy is increased; in addition the probability for full-energy absorption by multiple events such as Compton scattering and subsequent photoelectric absorption of the scattered γ-ray is also enhanced. Further improvements can be expected for future thicker counters. Comparison of the two properties resolution and efficiency shows that while the efficiency for a full-energy peak may be two orders of magnitude smaller for Ge(Li) than for NaI(Tl) the peak may still be one-fifth as tall because the improvement of a factor of 20 in resolution concentrates the counts near the nominal γ-ray energy.

With a rise-time in the neighborhood of 30 nsec Ge(Li) detectors are about an order of magnitude faster than NaI(Tl). Thus they are applicable for fast counting and fast coincidence work. E. Matthias has already obtained fast-coincidence resolving times with Ge(Li) that are superior to the best obtainable with NaI(Tl).

Fig. 3 shows a typical Ge(Li) count of the 130-keV region of the γ-ray spectrum in the decay of Co^{57}. The two well-resolved peaks at 122.0 and 136.4

keV arise from transitions from the 136.4 keV state to the 14.4 keV state and the ground state of the famous Mössbauer nucleus Fe^{57}. With NaI(Tl) the two peaks are unresolved. The two γ-rays have very different angular distribution characteristics, and N. J. Stone has used Ge(Li) detectors to study them separately. Peak widths as small as 2.0 keV have been obtained on several counters for the 122.0 keV γ-ray; this seems to be the practical limit for the preamplifiers. The photoelectric effect is very important at these low energies, and full-energy peaks dominate the spectrum.

The influence of processes other than photoelectric absorption is illustrated in Fig. 4, which shows the "signature" of a 2754 keV γ ray in a Ge(Li) detector. A broad plateau appears at lower energies. This arises through Compton scattering and incomplete deposition of energy in the detector. It is little use in analysis. For such high-energy γ rays pair production is also important, and the positron usually annihilates with an electron in the Ge(Li) crystal, creating two 511-keV γ rays. If both leave the crystal undetected a "double escape" peak appears at 1022 keV below the original γ-ray energy, at 1732 keV in this case. Either 511-keV γ ray may escape singly, leaving enough energy for a single escape peak at 2243 keV. Ge(Li) shares with NaI(Tl) the adverse effect on the low-energy γ-ray spectrum of a background arising from Compton scattering of higher-energy γ rays. This effect is less severe in Ge(Li) because of the higher resolution, which improves the signal-to-noise ratio for the low-energy peaks.

Applications to Gamma-ray Spectroscopy

Applications of Ge(Li) detectors to seven different types of experiments are discussed separately below.

A. Spectrum Analysis

The most important single application is the analysis of γ-ray spectra from radioactive isotopes. These spectra typically consist of several

components, each corresponding to a transition between two energy levels in the nucleus. Accurate determinations of these energies allows one to piece together the nuclear level scheme. With NaI(Tl) spectra peak widths were typically about 60 keV, and energy determinations to 1/10 the line width gave an accuracy of about 6 keV. With Ge(Li) counters these figures become 3 keV and 0.3 keV. We are presently measuring energies to an accuracy of ± 0.3 keV, although we hope soon to be able to reduce this to ±0.1 keV, an improvement that should be possible because of the small variation of background with energy under the narrow Ge(Li) peaks. We are presently limited by two factors unknown in NaI(Tl) spectroscopy. First, there are very few γ rays with energies known to 0.1 keV that can serve as calibration standards. Secondly, the pulse-height analyzers in use, with only a few hundred channels, do not provide a sufficiently detailed representation of the very detailed Ge(Li) spectra. Thousands of channels will be required to do the job adequately, and it is already clear that computers are going to plan an increasingly important role in Ge(Li) spectroscopy, both as successors to pulse-height analyzers in storing data and for data reduction.

In the decay of Ho^{166m}, one of the first cases studied with Ge(Li) detectors in our laboratory, several transitions were immediately evident that had been entirely overlooked in careful experiments with NaI(Tl) counters. In Fig. 5 spectra taken with both types of spectrometer are shown, normalized to equal counting times and with the same solid angle subtended by the counters. The high-energy doublets were unexpected, and two partially-resolved peaks in the 700-800 keV region of the NaI(Tl) spectrum became eight well-resolved peaks. This portion of the spectrum is discussed further in Section C below.

The nucleus Lu^{177m} has attracted considerable attention lately because of the decay of its high-spin "three quasi-particle" state through two rotational

bands in Hf^{177} and one in Lu^{177}. Alexander, Boehm, and Kankeleit[6] have worked out its decay scheme in detail, using the Cal Tech bent-crystal spectrometer. The γ-ray spectrum and their decay scheme are shown in Fig. 6. Johan Blok in our laboratory has studied this decay in connection with hyperfine-structure work, and has been able incidentally to add some confirmatory information on the level scheme. Taking advantage of the high signal-to-noise ratio in the Ge(Li) detectors he has found the missing 426-keV transition, from the 19/2- state in Hf^{177}. By coincidence measurements, using the 426-keV γ-ray as a gate, he has placed this transition unambiguously. Further coincidence experiments confirmed many details of the other cascades.[7] With NaI(Tl) spectra this work would have been completely out of the question.

B. Coincidence and Angular Correlation Measurements

In coincidence experiments two γ rays are detected with two counters, and appropriate circuitry determines whether they are nearly coincident in time and thus probably emitted successively in the decay of the same nucleus. Because two detectors are involved, the efficiency for coincidences goes as the square of the singles efficiency. Thus for low-efficiency Ge(Li) counters coincidence counting is somewhat difficult, particularly at high energies. In Fig. 7 the full-energy singles and coincidence efficiencies for NaI(Tl) and Ge(Li) detectors are plotted against γ-ray energy. In constructing the figure we have omitted the contributions to counting efficiency of solid angle(s) subtended by the counter(s); our efficiency is defined as the percentage of those γ rays incident upon the counter that give rise to a full-energy pulse. The dashed singles curves (labeled "NaI" and "Ge") are simply squared to produce the solid coincidence curves ("NaI-NaI" and "Ge-Ge").

Several conclusions regarding the applicability of Ge(Li) detectors for this type of experiment can quickly be reached on perusal of Fig. 7. First

we note that coincidences between two γ-rays at 1 MeV are observed with about 4 orders of magnitude less efficiency using a Ge-Ge system than with NaI-NaI. For experiments such as angular correlations, in which the number of coincidences is a prime factor, it is unlikely that Ge(Li) will find much application at high energies. For ordinary analytical coincidence work, on the other hand, even this factor of 10^4 might be made up for by the fact that a Ge-Ge coincidence at 1 MeV embodies some (20 × 20 =) 400 times more energy information than does a NaI-NaI coincidence at the same energy, because of the higher resolution. It is easy to imagine situations in which even a few events with well-defined energies can be much more valuable than many events of uncertain energy. This was particularly true, for example, in Blok's work on Lu^{177m}.

Going lower in energy the situation becomes relatively more favorable for Ge(Li) detectors, and at sufficiently low energies even angular correlation experiments are feasible. An interesting practical compromise between resolution and efficiency that often works is the use of an unsymmetrical coincidence circuit, with NaI(Tl) on one side and Ge(Li) on the other. Efficiency curves for such circuits are easily constructed from the singles curves by sliding them up or down appropriately, noting that the efficiency is plotted on a log scale. This has been done in Fig. 7 for the case of a "gate" counter set at 200 keV and the energy for the other counter left variable. These curves are labelled "Ge_{200}-NaI" and "Ge-NaI_{200}" for Ge(Li) and NaI(Tl) gate counters, respectively. For the cases Ge_{200}-NaI_{500} and Ge_{300}-NaI_{200} (extending the above notation) the efficiencies are about the same as that of the well-studied angular correlation in Ni^{60}, which we would denote NaI_{1173}-NaI_{1333}.

Using this "unsymmetrical" arrangement, E. Matthias, N. J. Stone, and R. B. Frankel have studied angular correlations of the coincidences between 400 keV γ rays (observed with NaI(Tl)) and 35.5-keV γ rays in the decay of

fission-product Sb^{125}. A coincidence spectrum is shown in Fig. 8. As the angle between the two detectors is varied through 90 degrees the x-ray intensities remain constant but the 35.5-keV γ ray varies in intensity by 30%. They have also studied the rotation of this correlation pattern in a magnetic field, operating the Ge(Li) detector in a field of several thousand gauss.

C. Low Temperature Nuclear Orientation

A field that is greatly enhanced by the use of semiconductor detectors is nuclear orientation, in which the angular distributions of radiations from oriented nuclei yield more subtle information, such as spins and moments, about nuclear energy levels than is available from simple counting and coincidence experiments. In the past α and β particles and conversion electrons from oriented nuclei had been detected with germanium and silicon counters: the advent of Ge(Li) detectors now extends this application to γ rays. Intensities are measured as in angular correlation, but, unlike angular correlation, singles counting is done, and the efficiency is not prohibitively low. On the other hand Ge(Li) detectors resolve the γ ray peaks sufficiently well that definitive angular distribution measurements can be made on each peak.

E. Matthias and S. S. Rosenblum have studied angular distributions of γ rays following the decay of oriented Ho^{166m} nuclei. In Fig. 9 the 700-800 keV portion of the spectrum is shown, for angles of 0°, 45°, and 90° between the direction of orientation and the Ge(Li) detectors. The γ-ray intensities vary greatly with angle, and the angular dependence is different for each γ ray. In Fig. 10 are shown the complete angular distributions for three γ rays in Er^{166}, along with their positions in the decay scheme. The angular distributions, when quantitatively analyzed, yield the spins of the levels as shown. Nuclear orientation is a far more generally applicable method than is commonly realized, and its usefulness is greatly enhanced by the introduction of Ge(Li) detectors.

D. Mössbauer Spectroscopy

A technique in γ-ray spectroscopy that has enjoyed great popularity since its adoption over the past few years is the application of the Mössbauer Effect to problems in chemistry and solid-state physics. A major limitation of this technique has been a lack of suitable resonant γ-ray transitions. In many cases the transitions are there, but the γ-ray peaks are obscured by other (nonresonant) radiations. Ge(Li) detectors make most of these cases tractable simply by resolving the resonant γ rays and improving thereby the signal-to-noise ratio. In Ir^{191}, Mössbauer's original nucleus, we have such a case. The 83-keV γ ray gives both a narrower resonance and a larger effect than does the 129-keV γ ray that Mössbauer studied, but until now it has been obscured by x-rays accompanying the decay of Pt^{191}. Fig. 11 shows part of the γ-ray spectrum of this isotope, taken with a Ge(Li) counter. The 83-keV γ ray is clearly resolved from the x-rays, and J. Huntzicker and S. S. Rosenblum have observed the Mössbauer Effect with this γ ray.

E. Conversion Coefficients

A sensitive indicator of the multipole character of a γ ray transition is its conversion coefficient, which is the ratio of the intensity of electrons ejected from atomic shells in lieu of γ-ray emission to the γ-ray intensity itself. The comparison of these two intensities is greatly facilitated by semiconductor counters, using Si(Li) for the electrons and Ge(Li) for the γ rays. Dr. Jack M. Hollander has built a "conversion coefficient machine" at the Lawrence Radiation Laboratory in Berkeley.[8] To use it one simply places a source in a fixed position and counts, doing simultaneous multichannel analysis on both electrons and γ rays. Comparison of the relative intensities with a known calibration factor for the machine then given conversion coefficients directly. This is a welcome contrast to the laborious point-by-point scanning of an

electron spectrum with a magnetic spectrometer. Although a Si(Li) detector gives poorer resolution for electrons than do the best magnetic spectrometers, its efficiency is orders of magnitude higher, and for many decay scheme studies it is preferable.

F. Neutron-Capture Gamma Rays

When a nucleus captures a neutron the binding energy is given up by γ-ray emission. The transition energies and intensities convey information about the structure of the resultant nucleus. Application of Ge(Li) detectors will greatly enhance the feasibility of capture γ-ray studies by providing more definitive measurements of the number, energies, and intensities of γ rays emitted. O. A. Wasson, K. J. Wetzel, and C. K. Bockelman of Yale University have made a survey of the (n,γ) spectra for several elements, using Ge(Li) counters.[10] They have demonstrated the feasibility of this type of experiment and have already found many new transitions.

G. Mu-mesic X-rays

A very exciting application of Ge(Li) detectors is their use in μ-mesic x-ray studies. Mu mesons are very much more closely bound to nuclei than are electrons. In lead, for example, a mu-meson in the lowest stable orbit is actually inside the nucleus. For this reason mu mesons can serve as very sensitive probes of the charge distribution within the nucleus. Until now studies of the structure of mu-mesic atoms have been very severely handicapped by the poor quality of detectors used to observe the characteristic x-rays. Now Ge(Li) detectors are beginning to revolutionize this field. H. L. Anderson, C. K. Hargrove, E. P. Hincks, and A. J. Tavendale at the University of Chicago have already applied Ge(Li) detectors to μ-mesic x-ray studies[11] with considerable success, and the future holds great promise for this application.

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5. G. T. Ewan and A. J. Tavendale, Canadian Journal of Physics 42, 2286 (1964).
6. P. Alexander, F. Boehm, and E. Kankeleit, Phys. Rev. 133, B284 (1964).
7. J. Blok and D. A. Shirley, Phys. Letters 13, 232 (1964).
8. J. M. Hollander, Lawrence Radiation Laboratory, Berkeley, private communication.
9. C. K. Bockelman, Electron Accelerator, Laboratory, Yale University, private communication.
10. H. L. Anderson, University of Chicago, private communication.

FIGURE CAPTIONS

Fig. 1. Resolution vs. energy for three types of γ-ray counter.

Fig. 2. Relative peak efficiencies for Ge(Li) and NaI(Tl) detectors.

Fig. 3. Multichannel analysis of a Co^{57} spectrum taken with Ge(Li) detector.

Fig. 4. Energy response of Ge(Li) detector to a 2754-keV γ ray.

Fig. 5. Ho^{166m} spectrum with NaI(Tl) and Ge(Li) detectors, normalized to equal counting times and solid angles.

Fig. 6. Lu^{177m} γ-ray spectrum taken with Ge(Li) counter, and decay scheme of Alexander et al.

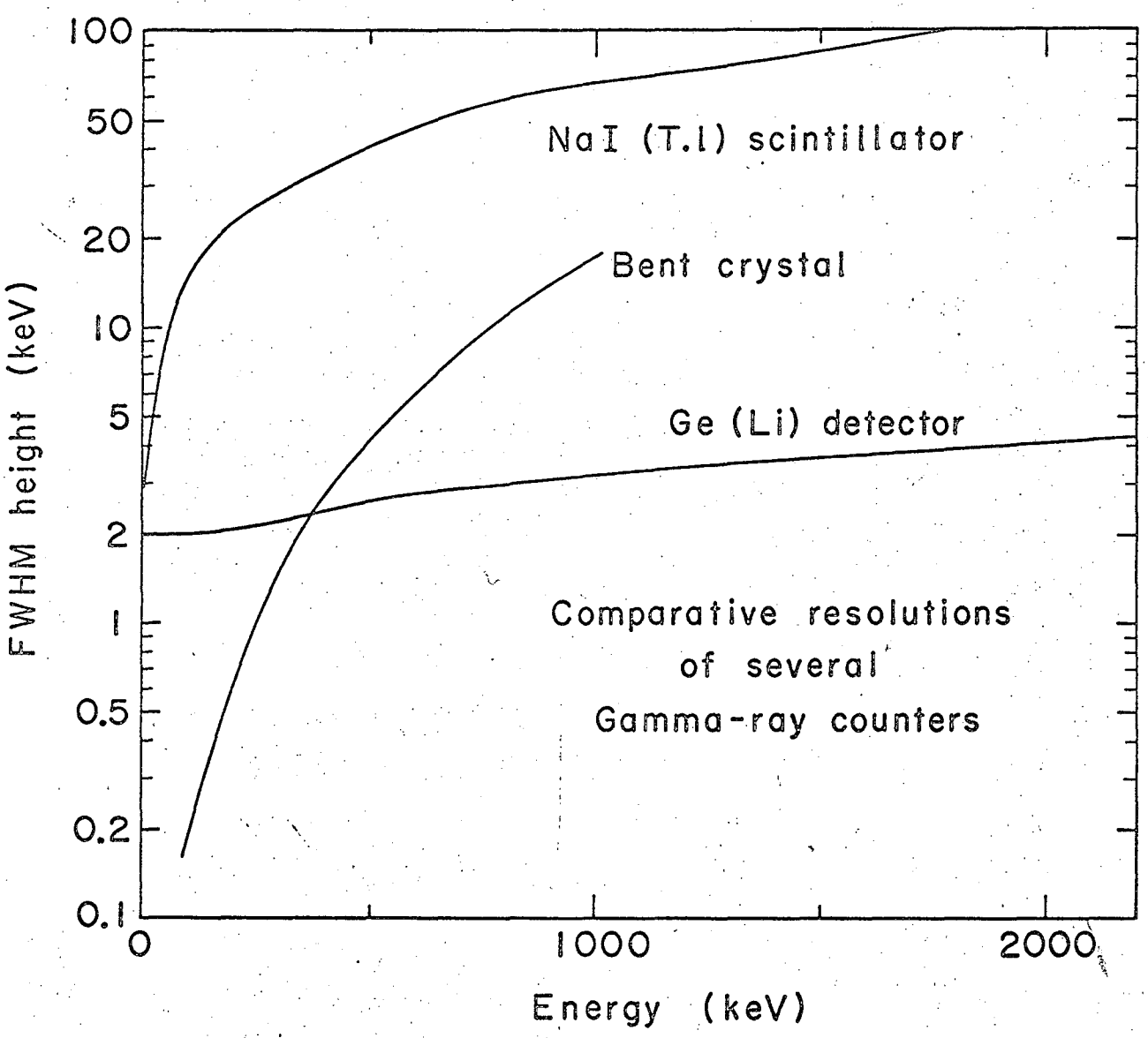
Fig. 7. Singles and coincidence efficiencies for Ge(Li) and NaI(Tl) detectors.

Fig. 8. Ge(Li) coincidence spectrum in the decay of Sb^{125} to Te^{125}, using a 400-keV gate.

Fig. 9. Partial γ-ray spectra from the decay of oriented Ho^{166m} nuclei, at various angles from the orientation axis.

Fig. 10. Angular distributions of three γ rays following the decay of oriented Ho^{166m} nuclei, with their positions in the level scheme.

Fig. 11. Partial γ-ray spectrum in Ir^{191}, following the decay of Pt^{191}. Ge(Li) detector makes 83-keV Mössbauer γ ray easily visible. The 129-keV γ-ray was Mössbauer's original transition.



MUB-4857

Fig. 1

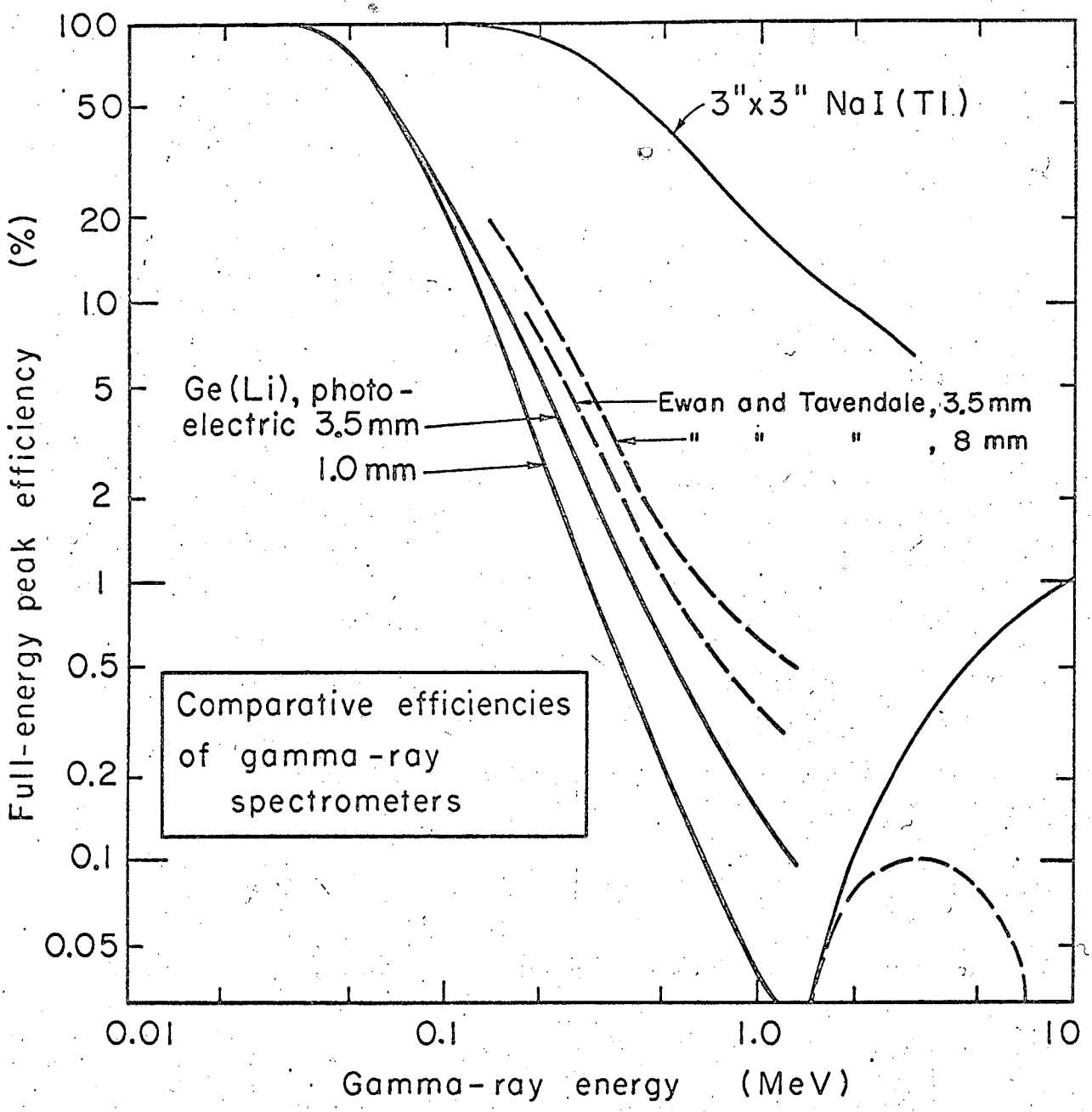

100
50
20
10
5
2
1
0.5
0.2
0.1
0.05
Full-energy peak efficiency (%)
3"x3" NaI(Tl)
Ge(Li), photo-electric 3.5mm
1.0mm
Ewan and Tavendale, 3.5mm
" " " , 8 mm
Comparative efficiencies of gamma-ray spectrometers
0.01
0.1
1.0
10
Gamma-ray energy (MeV)
MUB-4865

Fig. 2

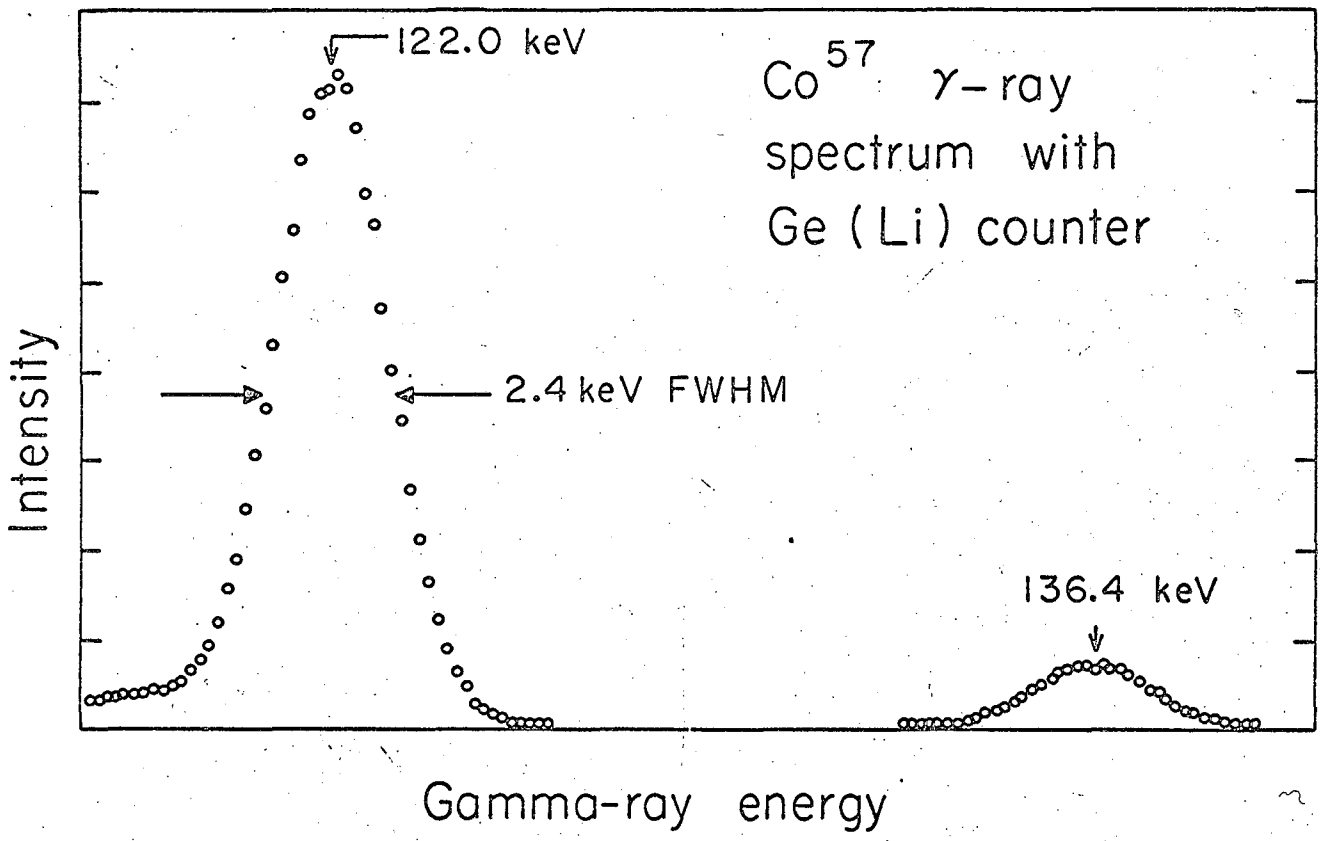

122.0 keV
Co 57 γ-ray
spectrum with
Ge (Li) counter
2.4 keV FWHM
136.4 keV
Intensity
Gamma-ray energy
MUB-4867

Fig. 3

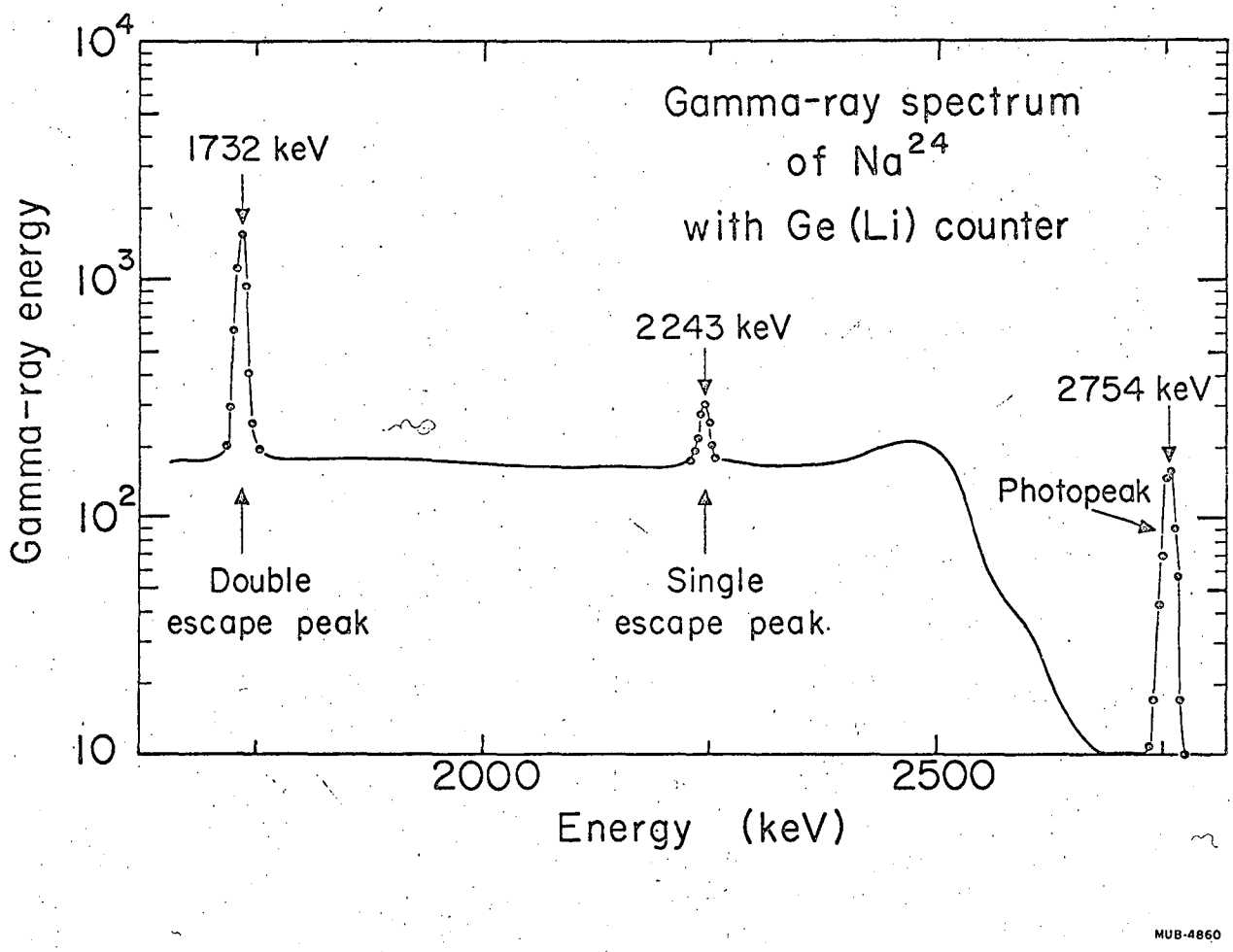

Gamma-ray spectrum
of Na^{24}
with Ge (Li) counter
1732 keV
2243 keV
2754 keV
Photopeak
Double
escape peak
Single
escape peak
Gamma-ray energy
10^4
10^3
10^2
10
2000
2500
Energy (keV)
MUB-4860

Fig. 4

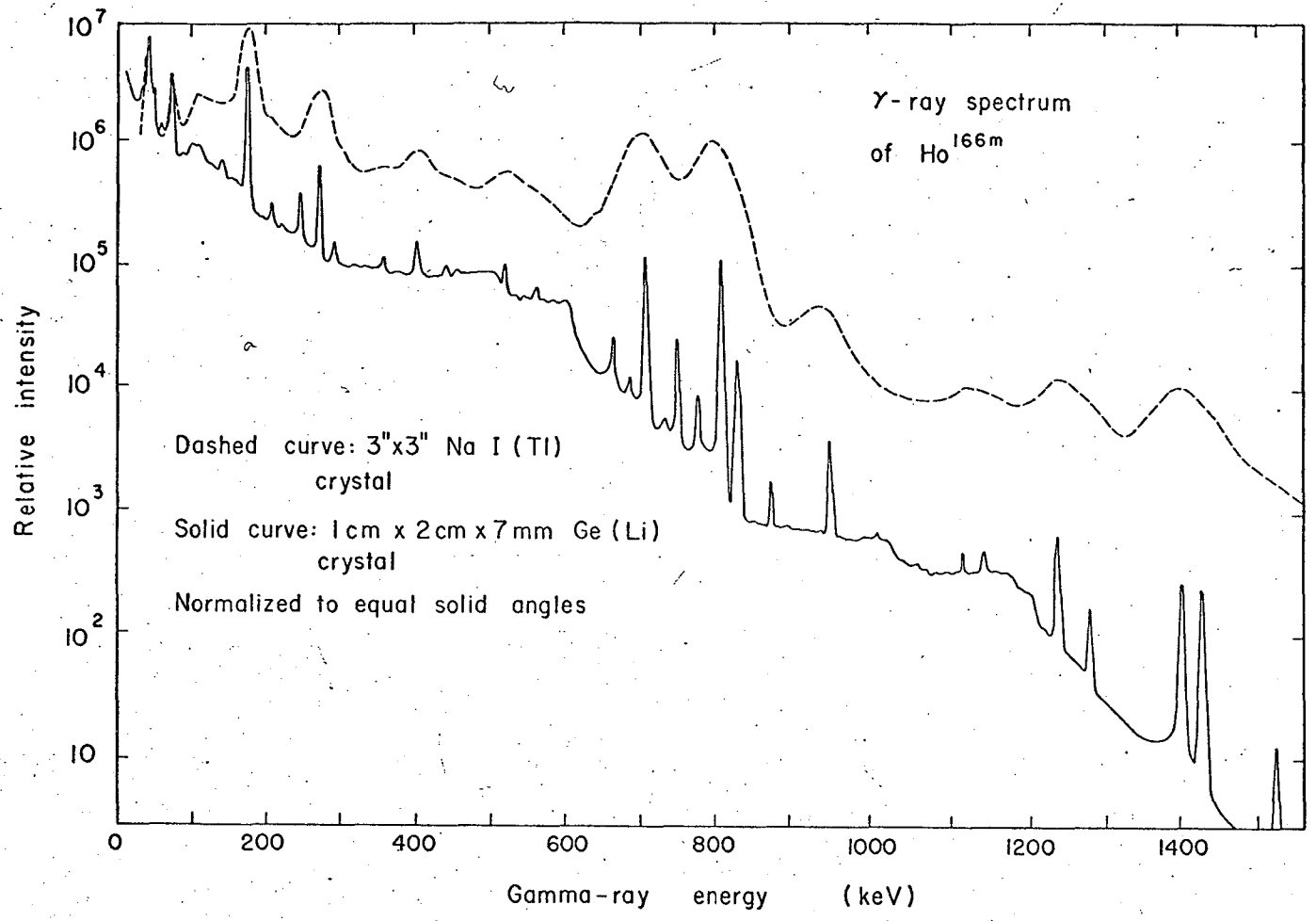

γ-ray spectrum
of Ho166m
Dashed curve: 3"x3" Na I (Tl)
crystal
Solid curve: 1cm x 2cm x 7mm Ge (Li)
crystal
Normalized to equal solid angles
Relative intensity
Gamma-ray energy (keV)
10^7
10^6
10^5
10^4
10^3
10^2
10
0
200
400
600
800
1000
1200
1400

Fig. 5

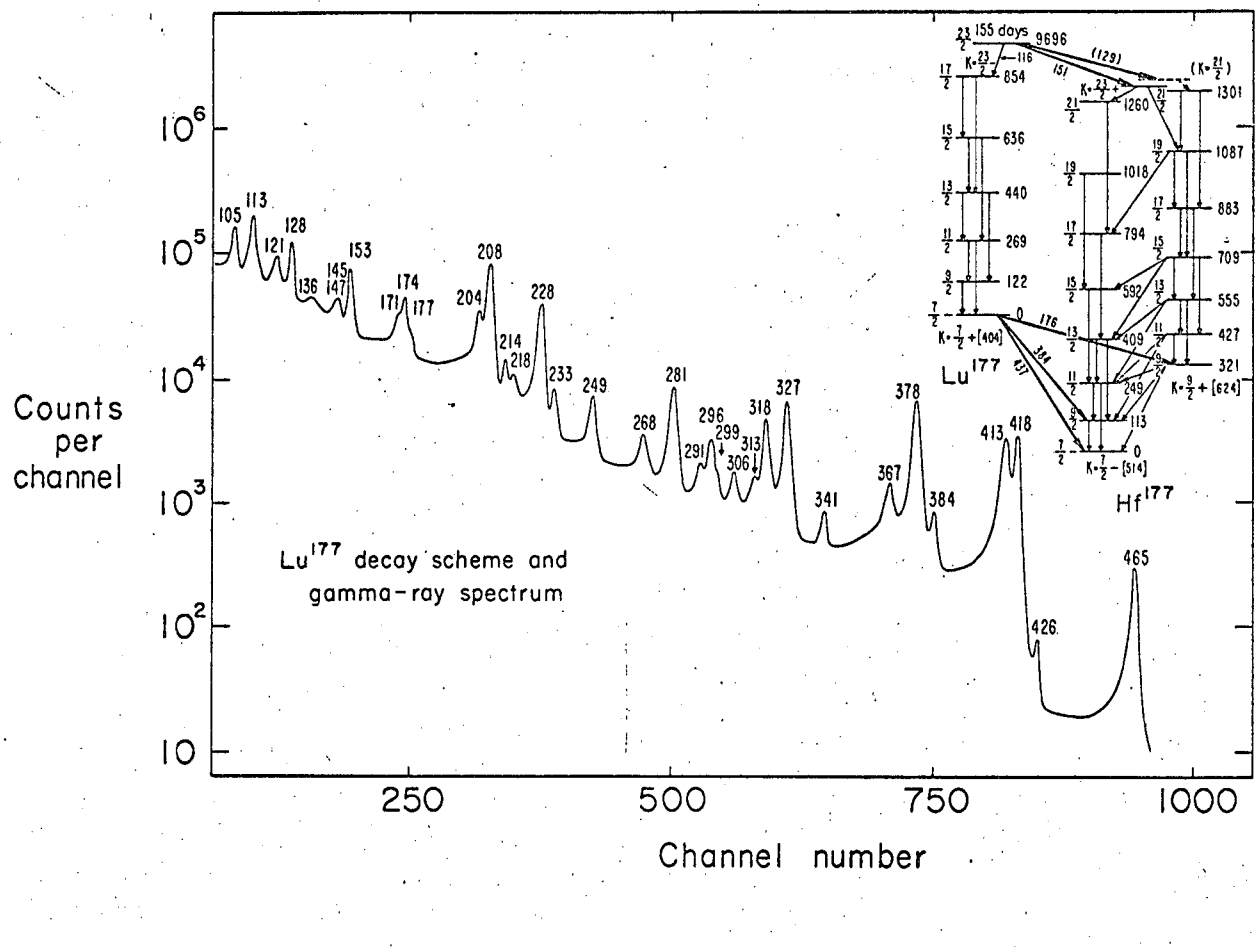
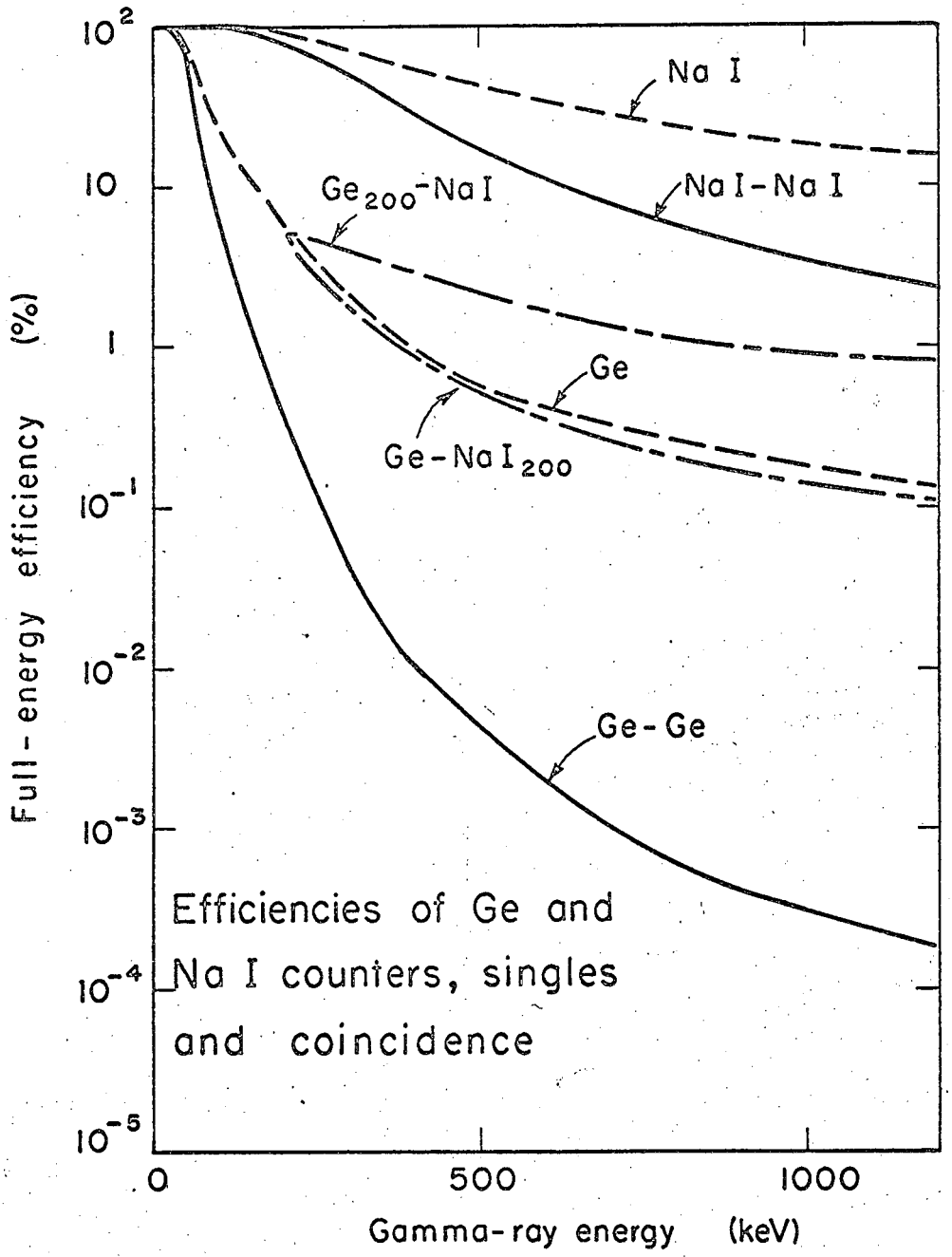
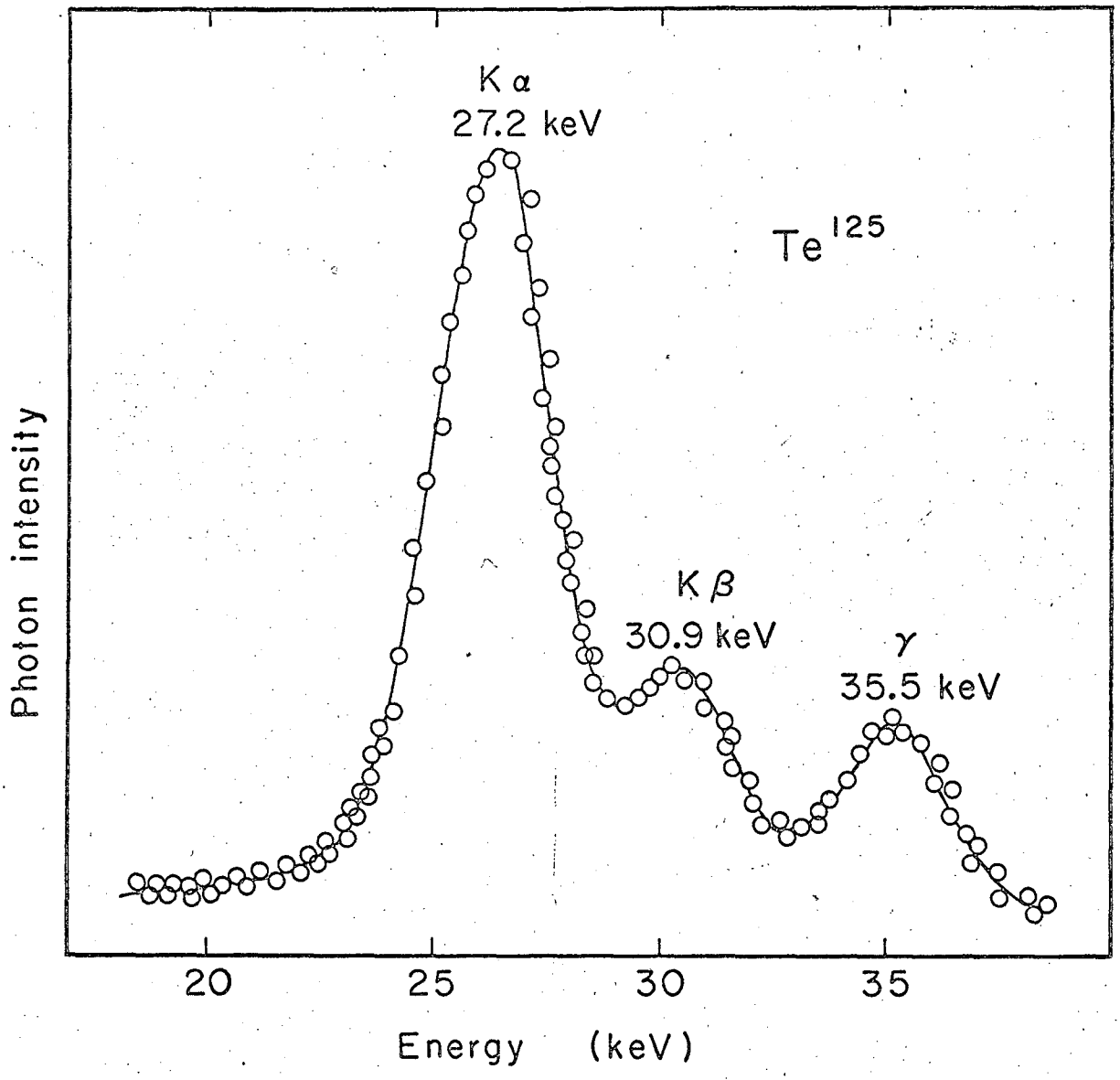

Counts per channel
Channel number
Lu177 decay scheme and gamma-ray spectrum
Lu177
Hf177
155 days
9696
(1291)
151
116
854
636
440
269
122
0
176
384
457
1301
1260
1087
1018
883
794
709
592
555
427
409
321
249
113
105
113
121
128
136
145
147
153
171
174
177
204
208
214
218
228
233
249
268
281
291
296
299
306
313
318
327
341
367
378
384
413
418
426
465

Fig. 6



MUB-4864

Fig. 7



MUB-4834

Fig. 8

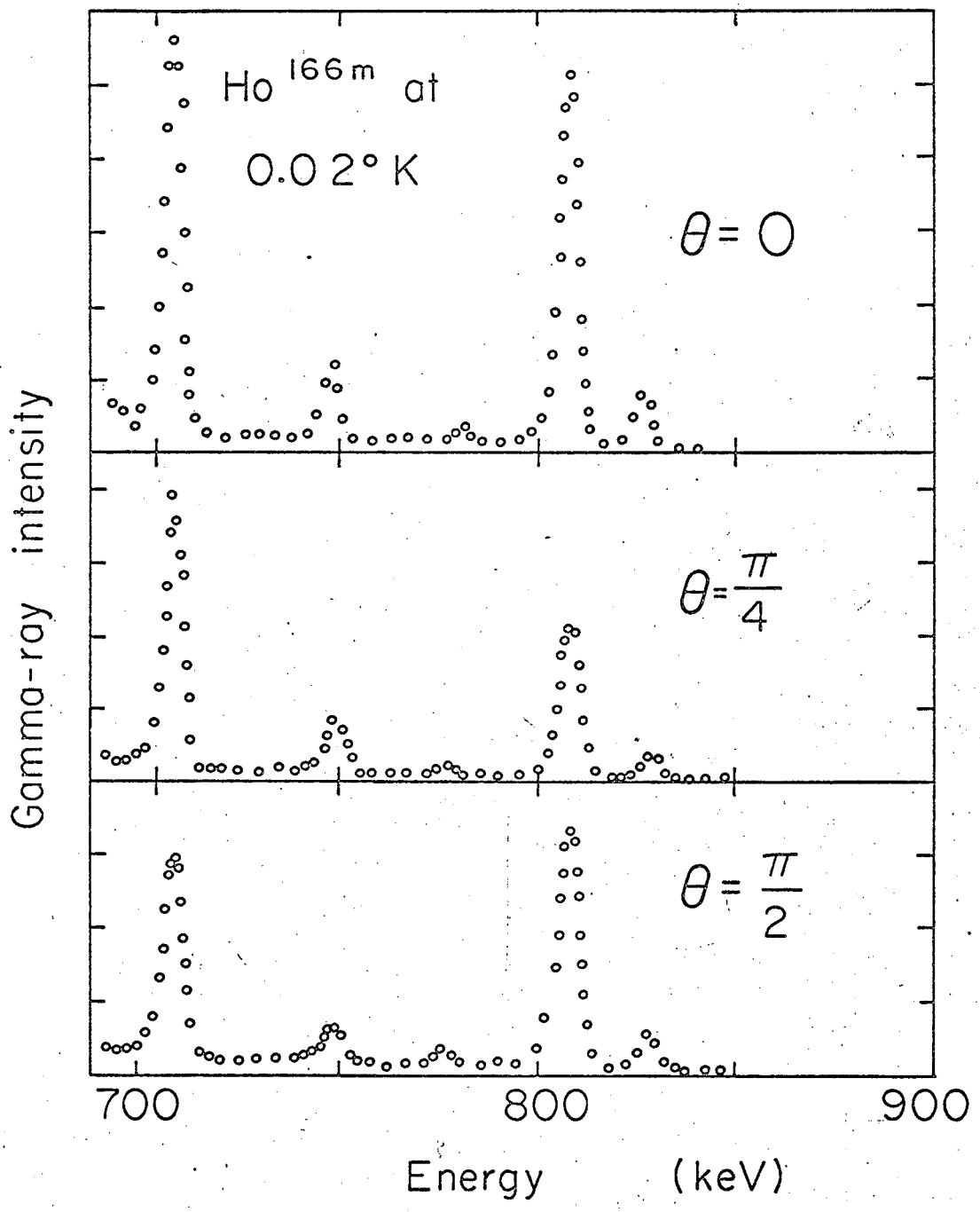

Ho 166m at
0.02° K
θ = 0
θ = π/4
θ = π/2
Gamma-ray intensity
700
800
900
Energy (keV)
MUB-4866

Fig. 9

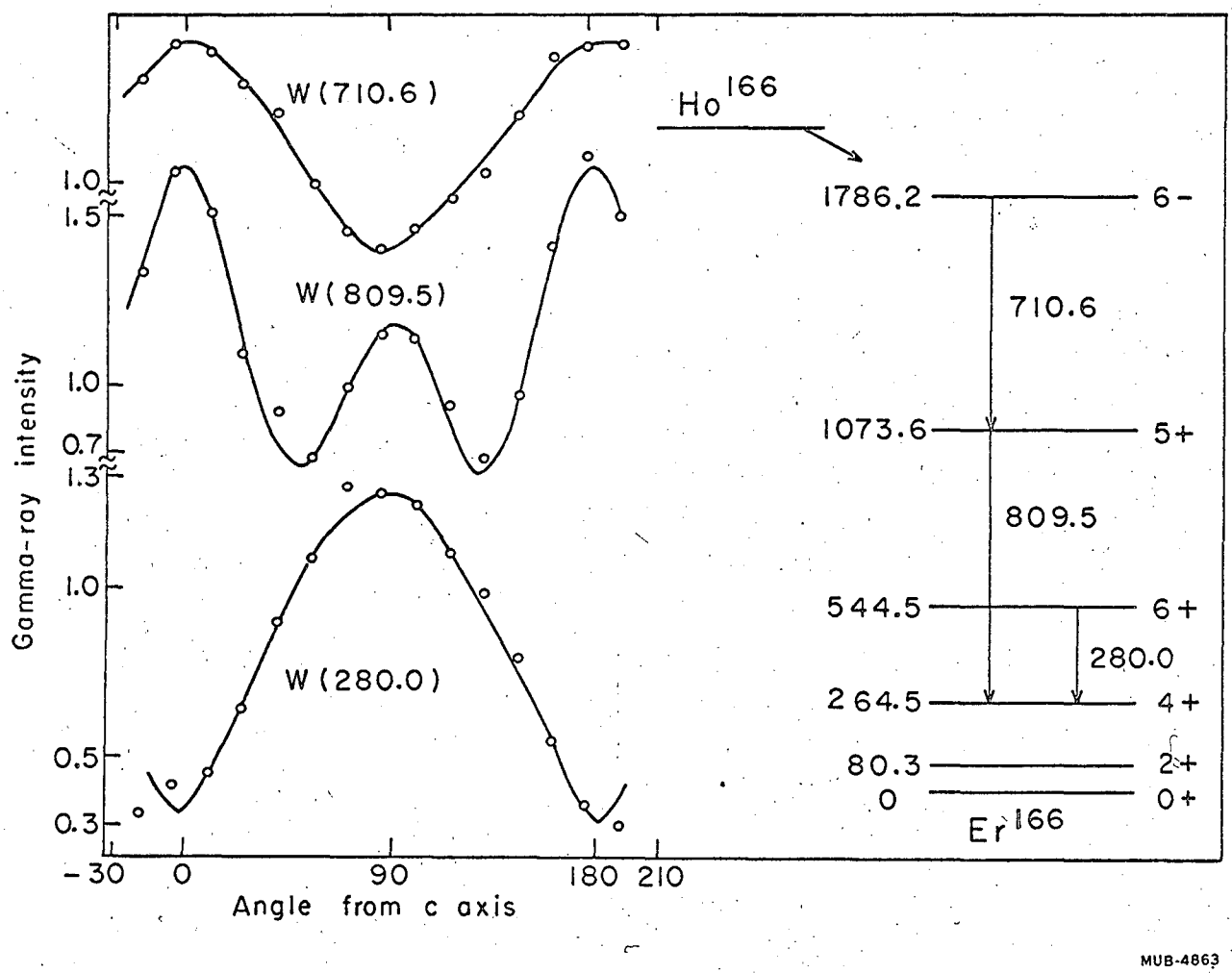

W(710.6)
W(809.5)
W(280.0)
Gamma-ray intensity
1.0
1.5
1.0
0.7
1.3
1.0
0.5
0.3
-30
0
90
180
210
Angle from c axis
Ho166
1786.2
6-
710.6
1073.6
5+
809.5
544.5
6+
280.0
264.5
4+
80.3
2+
0
0+
Er166
MUB-4863

Fig. 10

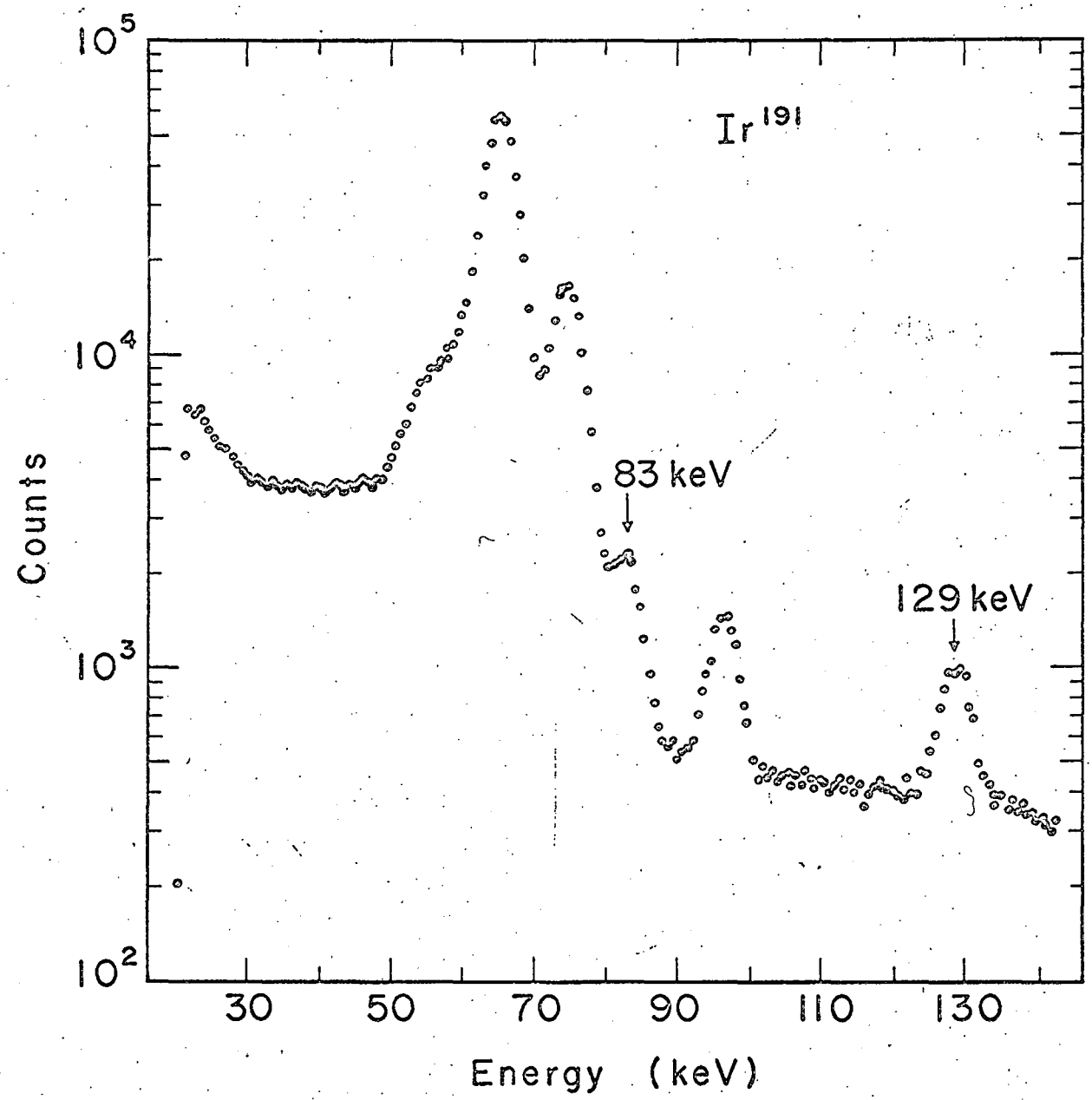

10^5
10^4
10^3
10^2
Counts
Ir^{191}
83 keV
129 keV
30
50
70
90
110
130
Energy (keV)
MUB-4839

Fig. 11